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**Proceedings of the
THIRD INTERNATIONAL CONGRESS
of the
INTERNATIONAL RADIATION
PROTECTION ASSOCIATION**

**September 9-14, 1973
Washington, D.C.**



INTERNATIONAL RADIATION PROTECTION ASSOCIATION

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THIRD INTERNATIONAL CONGRESS OF THE
INTERNATIONAL RADIATION PROTECTION ASSOCIATION

Held at the Washington Hilton Hotel,
Washington, D.C., September 9-14, 1973

Editor

W. S. Snyder

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February 1974

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PREFACE

The Third International Congress of the International Radiation Protection Association (IRPA) was held in Washington, D.C., September 9-14, 1973, and was hosted by the Health Physics Society. Two previous Congresses were held in Rome, Italy, in September 1966 and Brighton, England, in May 1970.

The scientific program, consisting of 237 papers, was truly international in scope. Scientists and administrators of 33 countries from all corners of the world actively participated in the Congress. In addition to the scientific program, refresher courses were held as part of a continuing professional development program. Scientific tours, an exhibition of radiation protection equipment and services, and a lively social program completed the Congress activities. A modified rapporteur system of presentation was used, and a relatively few papers were given orally by individual authors. This system, not in great use in the United States, received some criticism. However, it does provide an efficient system of presenting in a limited time a large number of papers, with only the pertinent facts being offered by the rapporteur, thus saving the audience listening to details.

The Scientific Program Committee under chairmanship of Dr. Bo Lindell had a most difficult task in developing the technical program. Perhaps I am somewhat biased; however, I believe the committee did an outstanding job. At its first meeting in Budapest, Hungary, in May of 1972, the committee made the decision that all projection slides presented at the Congress would receive severe screening and inspection in order to prevent the presentation of poor slides and incomprehensible data. Some authors resented such an inspection procedure; however, I am certain that the audience appreciated the fact that the committee instituted this. Such a procedure should be an integral part of all scientific meetings.

The IRPA Executive Council agreed to allow publication of the Congress Proceedings after receiving a most welcome invitation from the U. S. Atomic Energy Commission to print the papers in two volumes at no cost to IRPA. The task of editing and working with the USAEC, of course, fell to the IRPA Publications Director, Dr. Walter S. Snyder, and his staff at the Oak Ridge National Laboratory. The publication of such an extensive body of scientific papers requires a tremendous amount of effort on the part of Dr. Snyder and his staff, and the IRPA Executive Council sincerely appreciates this effort. These Proceedings will provide a valuable record of the Congress and serve as a significant reference in the future.

The papers in these Proceedings are arranged by the sessions as specified by the Scientific Program Committee and as presented at the Congress. Unfortunately, it was impossible to include the rapporteur papers in this publication. Nevertheless, the Program Committee and the International Meeting Commission gratefully acknowledge the effort that went into the preparation and presentation of their reports. Only a few papers for various reasons could not be obtained for publication. I am sorry for those authors who will not have their papers published, after going to the effort of presenting their information at the Congress. The Editor and I both acknowledge with gratitude and express our thanks for the cooperation of the authors of the papers presented here.

At the IRPA General Assembly, we observed the passing of the Presidential Bell from Dr. W. G. Marley to the capable hands of Dr. Carlo Polvani. We all express our most grateful thanks to Gregg Marley for a job well done; to Carlo Polvani, we wish great success in leading IRPA these next three years. It was indeed gratifying to welcome to the fold of IRPA our most recent affiliated society--the Radiation Hygiene Section of the Scientific Society of Hygienists of the USSR. We also observed that IRPA now has a reasonable bank account and will wisely use its funds to support international and regional meetings and appropriate international organizations concerned with radiation protection. The Assembly also accepted the invitation of the Czechoslovak Society of Nuclear Medicine and Radiation Hygiene to hold the Fourth International Congress in 1976. We will be pleased to assist Dr. Vladislav Klener, the new Vice President for Congress Affairs, in preparing for that Congress.

Another outstanding event of the Congress was the first presentation of the Sievert Award to Dr. Bo Lindell of Sweden. Rolf M. Sievert was a giant in the field of radiation physics and protection and a gentleman. Bo Lindell is of the same mold and a most worthy recipient of the award. His lecture on the assessment of radiation risk was an outstanding contribution to the Congress.

The Congress would not have been a success without the financial contributions and assistance of many national and international groups and agencies. These are acknowledged on page of this volume. Also our deep appreciation is given to the scientific and technical exhibitors who gave us an opportunity to observe their equipment and learn of their services to our fields of endeavor. Due thanks is also given to the exhibitors for contributing to the financial success of our Congress.

It is an impossible task to thank personally each individual who worked hard and long, behind the scenes and at the forefront, to make the Congress the success that it was. To each of you, it was a pleasure for me to share in the effort and to work with so many fine people from so many lands.

Claire C. Palmiter
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FOREWORD BY THE EDITOR

The Editor hopes these Proceedings will be a useful summary of the Third International Congress. Every effort was made to hold authors to their allotted number of pages, and the compromises finally reached were related to the total number of pages for each category of papers. In a few instances, authors did arrange for use of supplementary pages originally assigned to another author from the same institution. No retyping has been done to achieve a uniform style, and thus the presentation of the material is due entirely to the authors. Some papers were withdrawn prior to the Congress, and some authors were unable or unwilling to deliver papers in time for inclusion in the Proceedings. The Table of Contents consists of the complete program as presented for the Congress. The reader may wish to contact individual authors for missing papers or for further details on the work presented here. The Editor wishes to acknowledge the work of James Hickey and Sally Stockstill who helped in the editorial work for these Proceedings.

Walter S. Snyder
Publications Director
IRPA

Oak Ridge, Tennessee
February 1974

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OPERATIONAL HEALTH PHYSICS - 1

OPERATIONAL HEALTH PHYSICS DURING DISMANTLING OF THE ELK RIVER REACTOR

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Abstract

The Elk River Reactor (ERR), a 58 MW (th) boiling water reactor, was operated for four years and shut down in 1968 for economic reasons. The facility is now being dismantled such that the reactor site will be returned to approximately the condition that existed prior to installation of the reactor with all vestiges of the reactor plant (except for subgrade foundations) having been removed and disposed of. This is the first time that decommissioning of a power reactor has involved complete removal and disposal of large radioactive structures, rather than in-place entombment.

Removal operations and radiological conditions during dismantling of the highly radioactive portions of the reactor are described. The removal sequence of these structures started with the reactor internals and inner thermal shield, progressed through the pressure vessel and outer thermal shield and ended with the inner sections of the biological shield. Segmentation of the structures, either by mechanical or hot-cutting techniques, and packaging for transportation were done remotely to control personnel exposure with the addition, at times, of temporary shielding. To prevent the spread of contamination to work areas, operations were carried out underwater, or where this was not possible, in a specially designed containment structure around the work area.

Introduction

The Elk River Reactor (Figure 1), an indirect-cycle, natural circulation, boiling water reactor, was constructed by Allis Chalmers under a United States Atomic Energy Commission contract as part of the Commission's second round power reactor demonstration program and was operated by the United Power Association of Elk River, Minnesota under contract to the Commission. The total thermal capacity of the plant was 58.2 megawatts with a rated net electrical output of 22.5 megawatts. The reactor plant was constructed adjacent to an existing conventional electrical steam generating plant on land owned by the United Power Association and leased to the Atomic Energy Commission.

Initial reactor criticality was achieved in November, 1962, with power operation commencing in July, 1964. The plant was operated by the United Power Association from June, 1965 until final shutdown in January, 1968. Since that time, the Elk River Reactor has been maintained in a shutdown status with a limited number of systems being operated. All fuel was removed from the reactor and shipped from the site in 1968 and 1969.

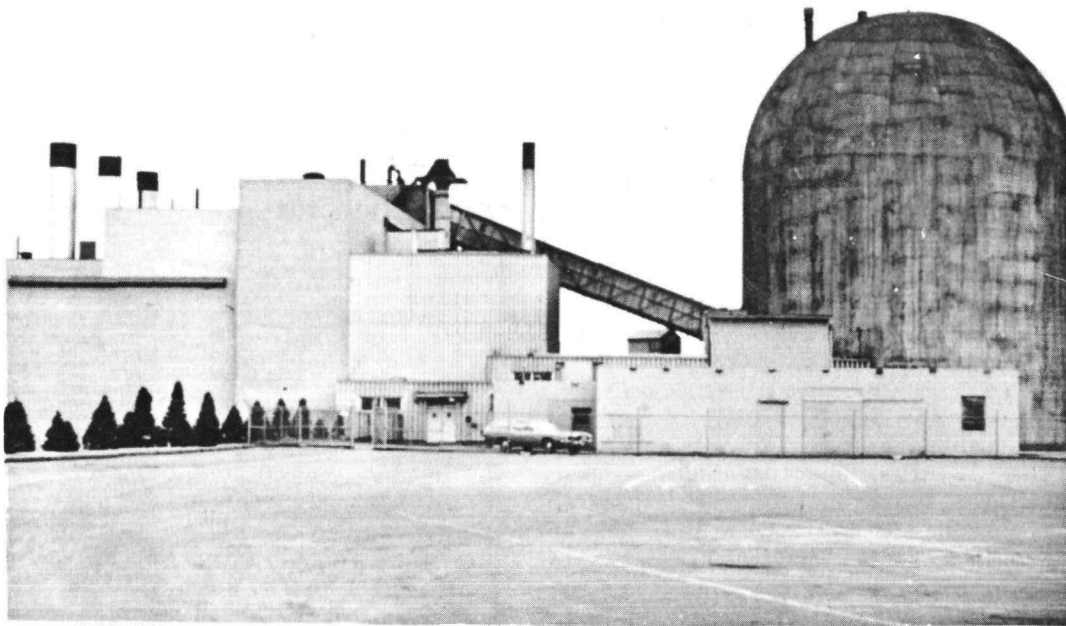


Figure 1: General View of the Elk River Reactor Site and Electrical Steam Generating Plant

Upon expiration of the contract with the Atomic Energy Commission for operation of the reactor, the United Power Association, for economic reasons, waived its option to purchase the plant. Pursuant to the terms of a modification to the operating contract, the Elk River Reactor Plant is being dismantled with all structures being removed approximately to grade level. Upon completion of the dismantling operation which is estimated to require approximately 2-1/2 years, the site will be available on an unrestricted basis to the United Power Association for installation of facilities associated with its conventional steam-electrical generation plant.

Planning of dismantling operations began in March, 1971 and a document summarizing planned operations⁽¹⁾ was submitted for approval to the Atomic Energy Commission in August, 1971. An Environmental Impact Statement on the project⁽²⁾ was issued by the Commission in May, 1972. An order to proceed with dismantling of the facility was received from the Commission in June, 1972 when dismantling operations began. To date, all highly radioactive structures including the reactor internals, pressure vessel and outer thermal shield have been removed and disposed of. The current phase of dismantling involves removal and disposal of the concrete biological shield.

Removal Operations

The general arrangement of reactor internal structures is shown in Figure 2. The removal sequence of highly radioactive structures proceeded from the inside out. That is, the first structures removed were the components contained within the reactor pressure vessel, then the pressure vessel itself, and finally the outer thermal shield which was a cylindrical lead and steel sandwich providing a radiation barrier and a thermal barrier between the pressure vessel and the biological shield concrete. Estimates of the total inventory of long-lived radioactivity in each of the structures and measurements of the resultant maximum contact radiation levels are shown in Table 1. The total inventory of radioactive material in the facility at the start of dismantling was estimated to be 10,000 curies of which greater than 99% was contained in the structures listed in Table 1. These structures are those which have been removed and disposed of thus far in the dismantling program. Contact radiation levels as measured underwater varied from 1 to 8000 R/hr.

All internal structures were attached each to the other either mechanically by means of machine screws or bolts or they were welded in place. Specially designed remote tools were used to separate those components which were mechanically united while a remotely-operated plasma arc cutting torch was used to separate components welded together. Separation of all components was done underwater. With the exception of the core and shroud support plate, components were transferred in-air (Figure 3) from the reactor cavity to a fuel element storage well located near the reactor cavity where preparation for shipment was completed. Because of high radiation levels, the core and shroud support plate was transferred underwater through the fuel

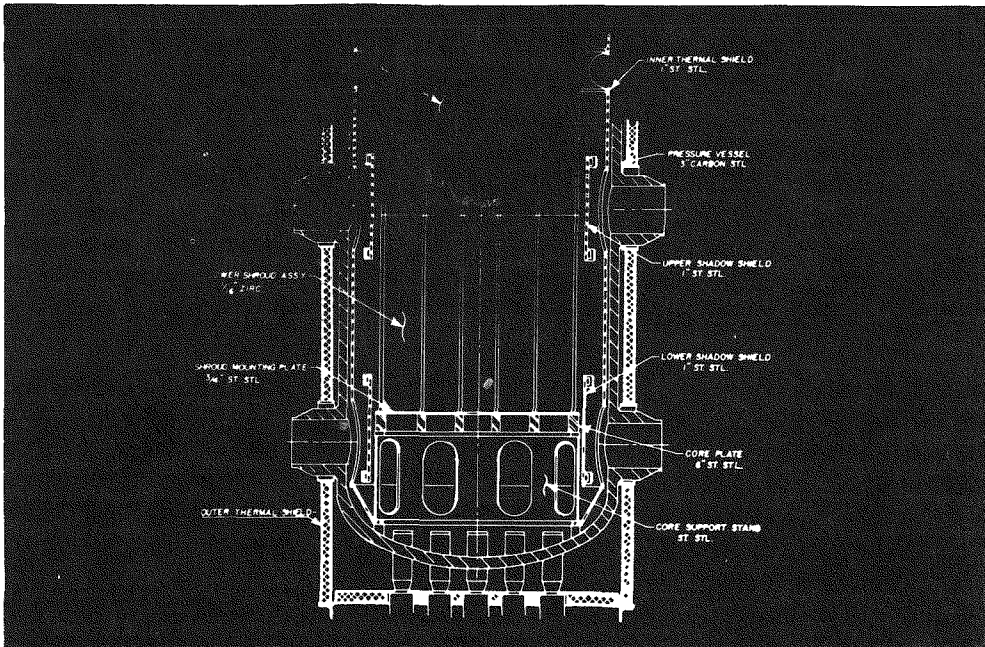


Figure 2: Elk River Reactor Pressure Vessel Internals and Outer Thermal Shield

transfer canal to the fuel element storage well. The packaging and disposal of the reactor internals depended upon the size of the component and its external radiation level. Components were segmented, if necessary, and loaded underwater (Figure 4) into various types of steel or lead and steel shielded cask liners (Figure 5) which in turn were then transferred in-air (Figure 6) to one of two shipping containers for transportation to the burial facility. The LL-50-100 container (Figure 7) weighs 50,000 pounds and has an internal volume of 100 cubic feet. Shielding is provided by 3-1/2 inches of lead and 2-1/4 inches of steel. The BC-48-220 container (Figure 8) weighs 48,000 pounds, has an internal volume of 220 cubic feet and has 7 inches of concrete and 2-1/2 inches of steel for shielding.

Table 1

Radioactive Inventory and Maximum Contact
Radiation Levels by Reactor Component

<u>Component</u>	<u>Inventory (Curies)</u>	<u>Maximum Contact Radiation Level - R/hr</u>
Internals		
Upper Shroud	770	2800
Lower Shroud	35	175
Core & Shroud Plate	2370	8000
Core Support Stand	100	150
Inner Thermal Shield	3090	1000
Shadow Shields	2330	3000
Feedwater Distribution Ring	75	60
Pressure Vessel	1110	115
Outer Thermal Shield	75	1
	<u>9955*</u>	
Total		

*As of April 30, 1971

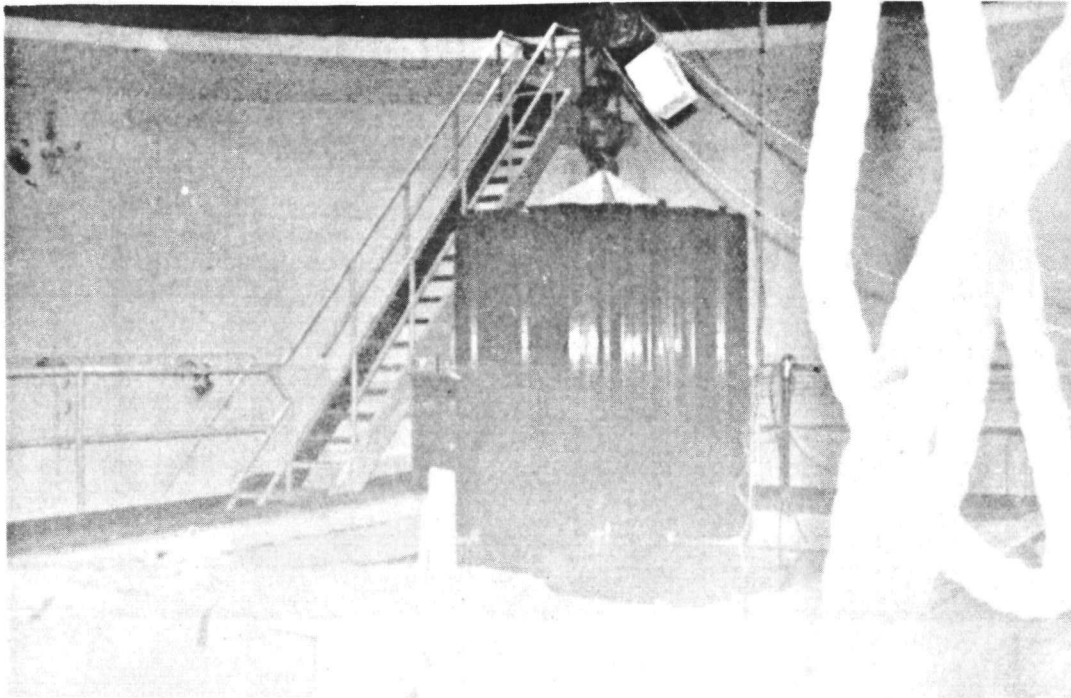


Figure 3: In-Air Transfer of the Reactor Upper Shroud Assembly

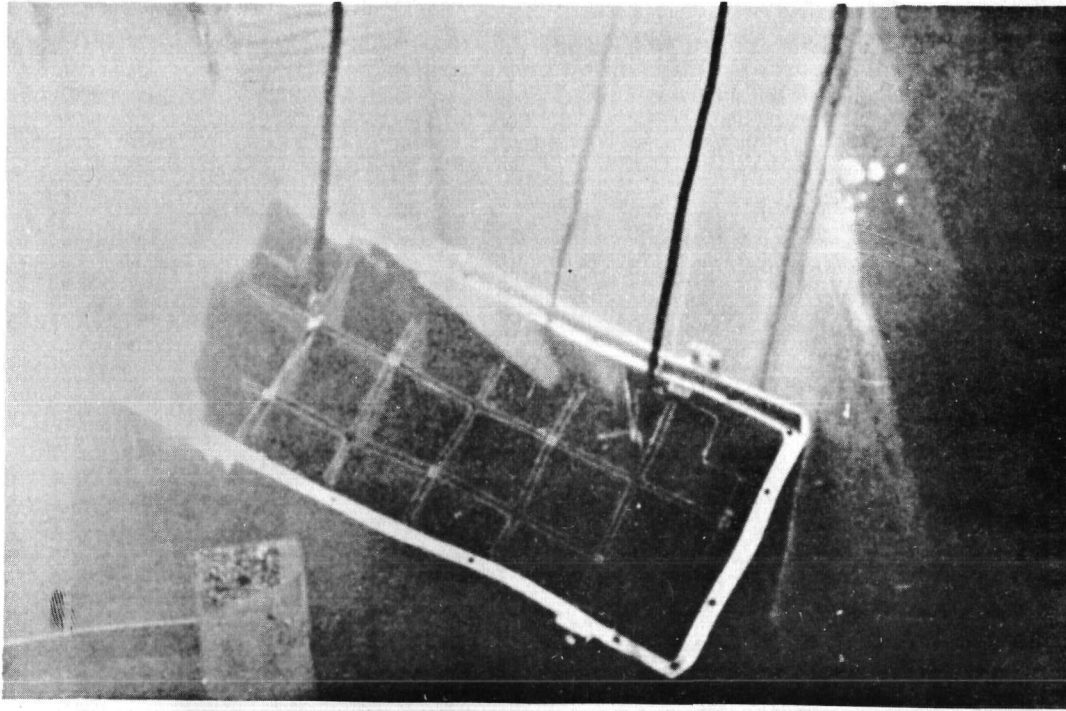


Figure 4: Segmented Upper Shroud Assembly in Shielded Cask Liner Underwater

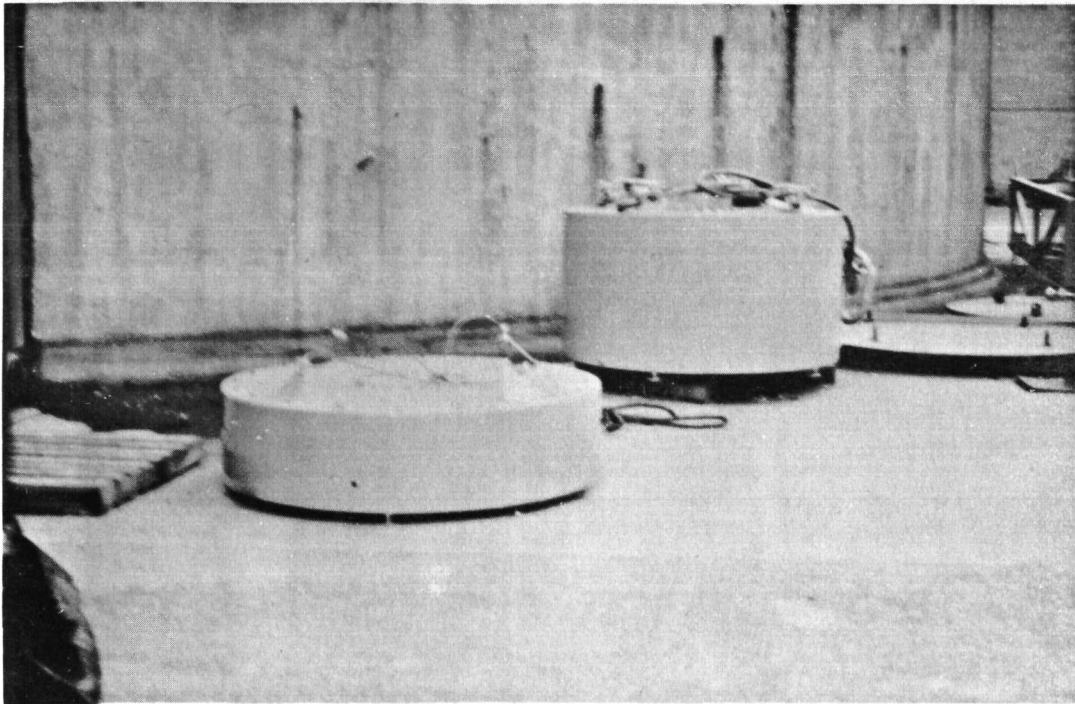


Figure 5: Typical Shielded Cask Liners Used for Packaging of Reactor Internal Components

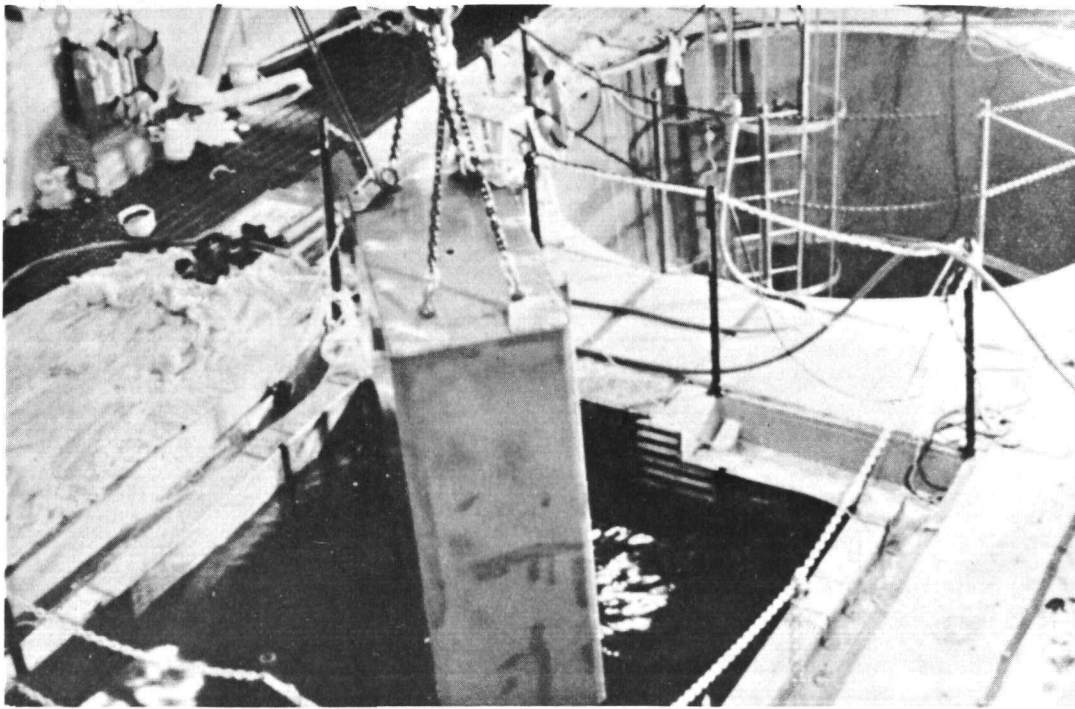


Figure 6: In-Air Transfer of a Shielded Cask Liner Containing a Segment of the Upper Shroud Assembly

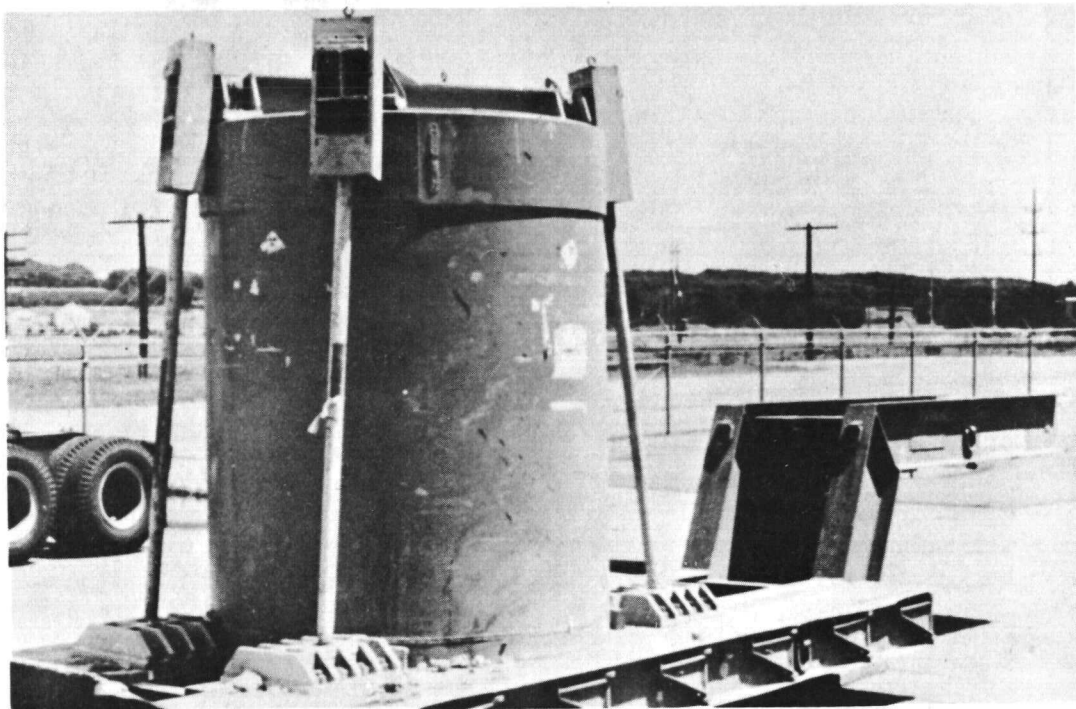


Figure 7: The LL-50-100 Shipping Container

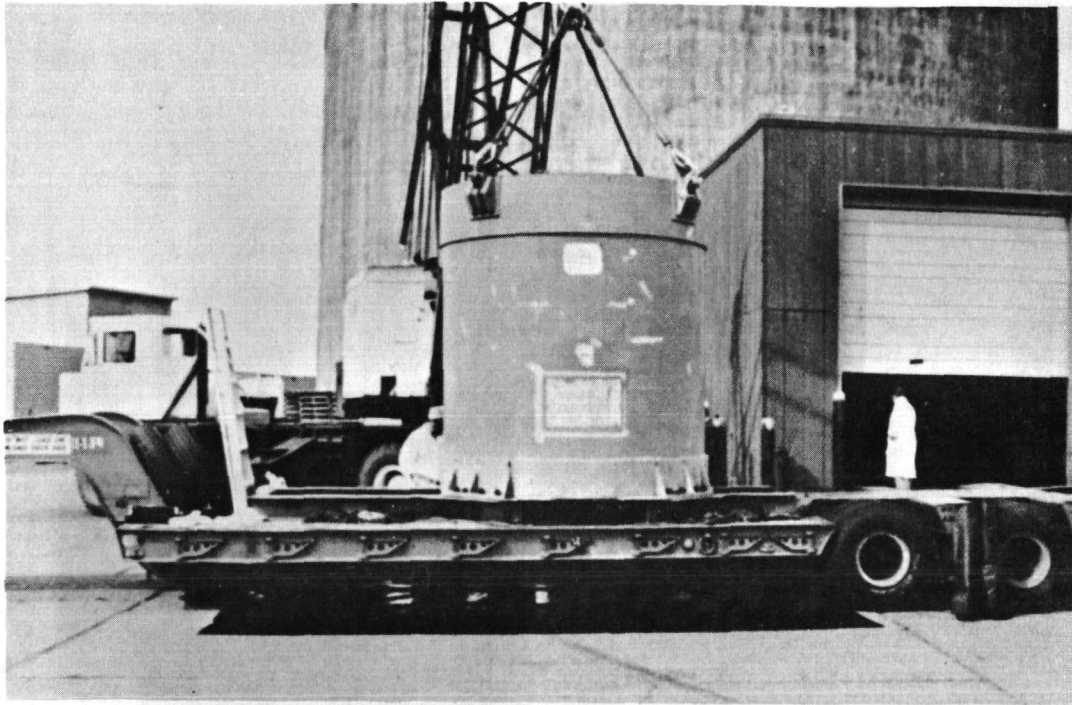


Figure 8: The BC-48-220 Shipping Container

During in-air movements of radioactive components or loaded liners, all unnecessary personnel were cleared from the reactor building and those engaged in the transfer were located in a shielded area for the control of radiation exposure. During underwater operations, personnel exposure rates usually varied between 5 and 15 mR/hr while in-air movements caused exposure rates of from 100-200 mR/hr to personnel in the shielded area. During loading and preparation of the shipping container, personnel exposure rates as high as 2 R/hr were experienced for short periods of time.

Segmentation of the pressure vessel and inner and outer thermal shields was done in place. The inner thermal shield was segmented with a remotely-operated plasma arc cutting torch designed for underwater operation. The pressure vessel and the steel portions of the outer thermal shield sandwich were segmented remotely in-air, again with a plasma arc torch. Lead was melted away from the outer thermal shield with an acetylene torch. All of these operations were done within a containment enclosure formed by the side walls of the reactor cavity. All penetrations into the cavity were sealed and a metal cover containing a filtered air inlet was placed over the cavity (Figure 9). The containment enclosure had a separate exhaust fan and filtering system (Figure 10) containing a prefilter and a high efficiency particulate filter capable of removing most of the airborne contamination generated during cutting operations. Samples of airborne contamination from within the containment enclosure taken during cutting operations indicated concentrations on the order of 10^{-7} $\mu\text{Ci/cc}$ of gross beta activity. On the other hand, samples of reactor building air outside the containment structure rarely exceeded 10^{-11} $\mu\text{Ci/cc}$ indicating the effectiveness of the exhaust filtering system. Following segmentation, the pressure vessel and outer thermal shield segments were packaged for shipping as outlined previously (Figure 11 and 12).

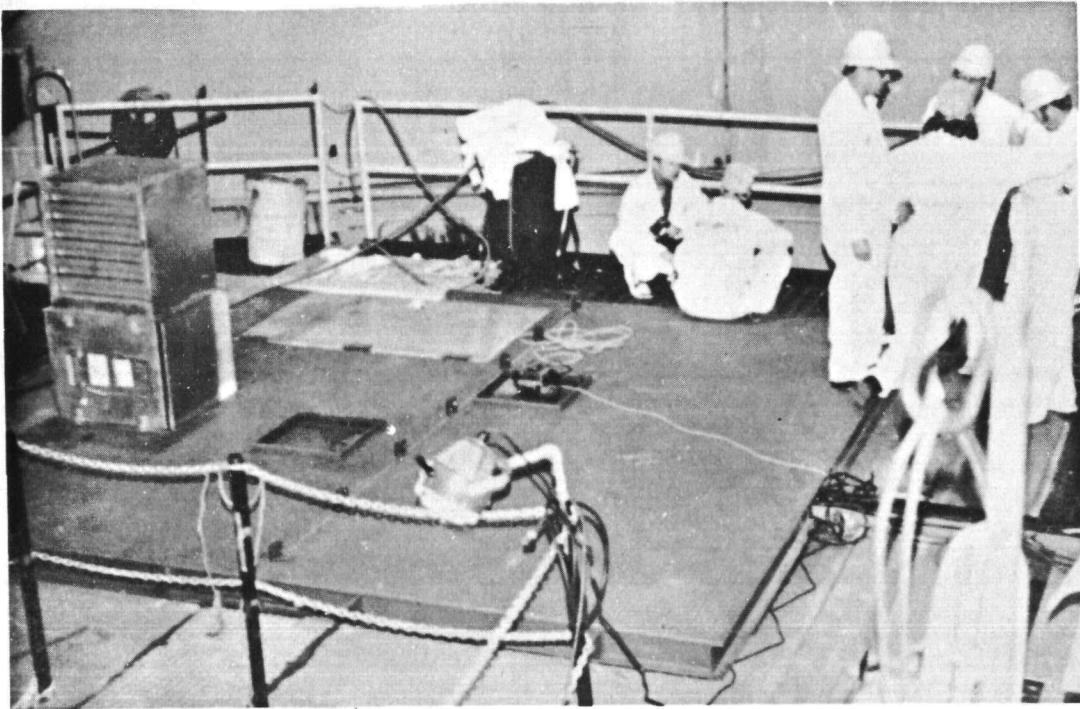


Figure 9: Metal Cover over the Containment Enclosure Showing Filtered Air Inlet, Viewing Windows and Access Plate

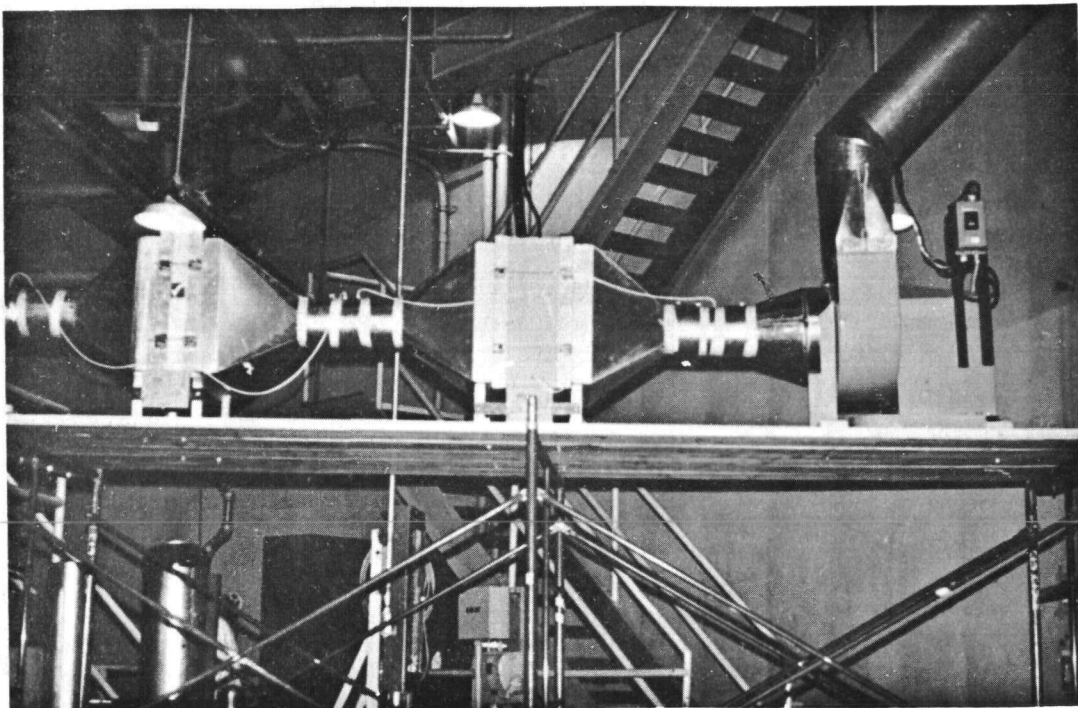


Figure 10: Containment Enclosure Exhaust System Containing Pre-Filter, Absolute Filter and Blower

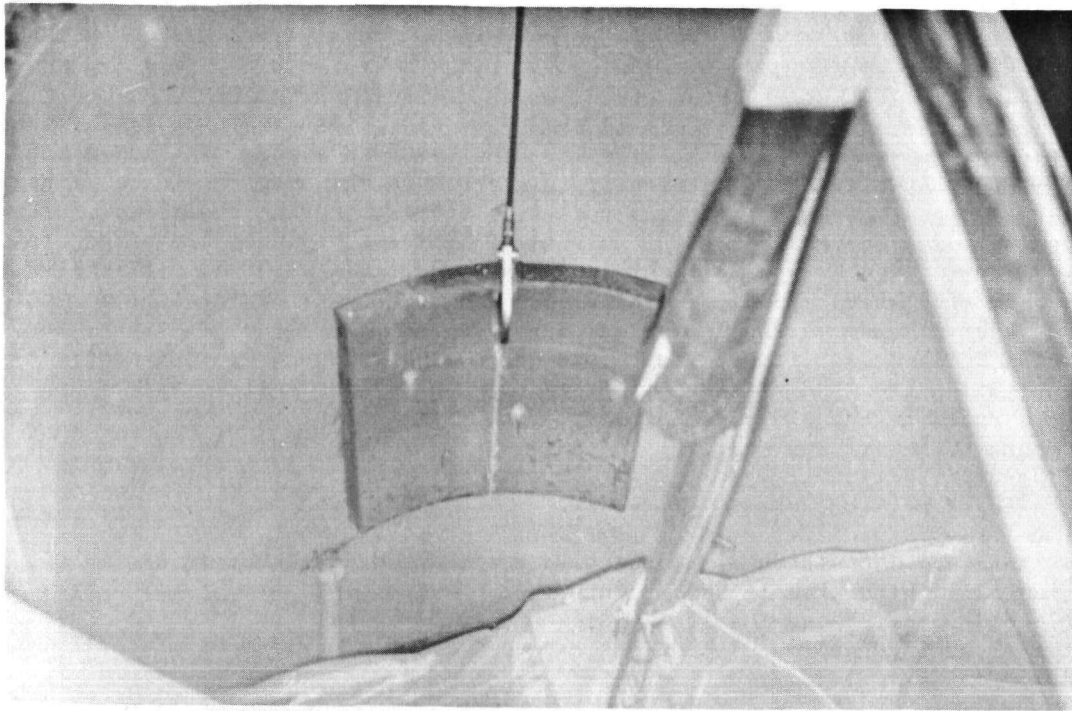


Figure 11: In-Air Transfer of a Pressure Vessel Segment

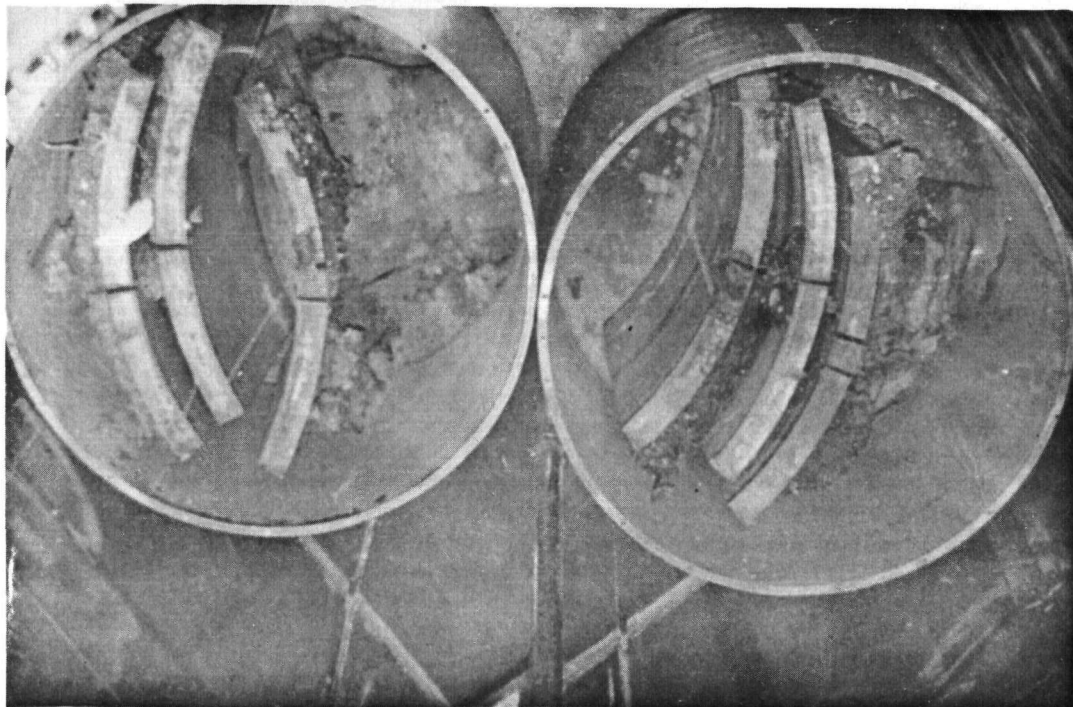


Figure 12: Pressure Vessel Segments in Cask Liners during Preparation for Shipment and Disposal

Personnel Exposure

In spite of the attention given during dismantling to remote or underwater operations or the use of temporary radiation shielding, significant radiation exposure has been received to date by those engaged in dismantling activities. The greatest portion of this exposure has been received during almost constant work in low level radiation fields. During the first fourteen months of dismantling, a total of 62 rem of whole body penetrating exposure has been received by approximately 80 people connected with the dismantling project. The average whole body exposure since the start of dismantling has been about 0.8 rem while the maximum total exposure received to date by any workman has been 4.8 rem. Even though 99% of the radioactive inventory has been removed from the site and disposed of, that which remains, primarily concentrated in the biological shield, will require additional chronic exposure to low radiation levels. It is estimated that perhaps another 20-30 rem of personnel exposure may be necessary to complete dismantling of radioactive structures.

Internal deposition of radioactive material in personnel is estimated periodically with a whole body counter. The radionuclides identified by whole body counter examination have been almost exclusively ^{137}Cs and ^{60}Co . To date, no internal deposition has exceeded 1% of a maximum permissible body burden.

Liquid and Airborne Waste Management

The restraints placed upon the dismantling project with respect to the release of liquid and airborne wastes to the environment were outlined in the project's Environmental Impact Statement. A commitment was made to control waste releases below the numerical guidelines proposed in Appendix I of Title 10, Part 50 of the Code of Federal Regulations as shown in Table 2. The annual average concentration of liquid effluents at the site boundary is not to exceed 20 pCi/l of gross beta activity and 5000 pCi/l of tritium. The annual average airborne emissions are not to exceed the numerical values contained in Appendix B, Table II, Column I of 10 CFR 20 divided by a factor of 100,000. For the radionuclides estimated to be present in Elk River Reactor structures and given the meteorological conditions of the area and site layout, the annual average concentration at the worst location of the site boundary is not to exceed 7.5×10^{-3} pCi/m³. The intent of these numerical guidelines is to limit public exposure in the vicinity of the project to less than 5 mrem per year. Since June, 1972, the average concentration at the site boundary of liquid effluents has been 5.5 pCi/l of gross beta activity and 350 pCi/l of tritium while airborne waste concentrations has averaged 4% of the applicable limit. Using very pessimistic assumptions, it is estimated that the maximum dose received by an individual in the vicinity of the site from effluents generated during dismantling operations has been 0.4 mrem.

Table 2

Proposed Numerical Guidelines of
Appendix I, 10 CFR 50 for Release
of Radioactive Waste Products to the Environment

<u>Type of Waste</u>	<u>Annual Average Concentration</u>
Liquid	20 pCi/l 5000 pCi/l for Tritium
Airborne	<u>Appendix B, Table II, 10 CFR 20</u> 100,000
(For Elk River Dismantling)	0.0075 pCi/m ³

Operations to be Completed

Since the United Power Association's contract with the Atomic Energy Commission calls for complete removal and disposal of the reactor building including all structures contained therein, there is a considerable amount of dismantling yet to be completed. However, all highly radioactive structures have been removed from the site. From a radiological standpoint, the only structures yet to be removed which contain significant quantities of radioactive material are the concrete biological shield and certain reactor systems such as the liquid waste disposal system. It is estimated that removal of these structures will take approximately six months following which demolition of remaining structures by conventional methods will begin.

References:

- (1) AEC Elk River Reactor Dismantling Plan (Rural Cooperative Power Association (UPA) and Gulf United Nuclear Fuels Corporation), SS-836, August 27, 1971.
- (2) Environmental Statement, Elk River Reactor Dismantling (U. S. Atomic Energy Commission), WASH-1516, May, 1972.

СПЕЦИФИЧЕСКИЕ ВОПРОСЫ РАДИАЦИОННОЙ БЕЗОПАСНОСТИ И
САНИТАРНО-ДОЗИМЕТРИЧЕСКОГО КОНТРОЛЯ
ПРИ РАБОТАХ С ТРАНСПЛУТОНИЕВЫМИ ЭЛЕМЕНТАМИ

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Abstract

The report shows that the factors of radiation exposure at operations with transplutonium elements are in quantitative dependence on the physico-chemical properties, the time of target irradiation in the reactors and the degree of their purification from the fragment fission products. The doses of external and internal irradiation have been estimated.

В процессе радиохимической переработки облученных мишеней при получении тяжелых изотопов трансплутониевых элементов (ТПЭ) радиационная обстановка характеризуется как внешними потоками ионизирующих излучений, так и газоаerosольным загрязнением воздушной среды радиотоксичными веществами.

Особенность работ с ТПЭ состоит в том, что приходится сталкиваться с одновременным действием практически всех типов ионизирующих излучений. Вследствие этого требуется дифференцированный подход к комплектации защитного оборудования в зависимости от рода проводимых работ, изотопного состава тяжелых элементов, их весовых количеств и степени очистки от продуктов деления. Данные положения могут быть иллюстрированы материалом, представленным в табл. I и 2.

Из табл. I видно, что при работах с малыми количествами ТПЭ, когда используются защитные экраны малой толщины, важное значение приобретает степень очистки ТПЭ от изотопов осколочного происхождения.

Таблица I

Относительный вклад в суммарную мощность дозы
различных типов ионизирующих излучений
смеси оксидов америция с кюрием

Тип излучений	Относительный вклад в %	
	Без защиты	Защита из чугуна 15 см
Нейтроны спонтанного деления	5,2	77,0
Нейтроны (α , n)-реакции	0,2	3,0
Собственное гамма-излучение ТПЭ	13,3	-
Собственное гамма-излучение продуктов деления	21,7	19,5
Тормозное излучение	59,6	0,5
ВСЕГО	100,0	100,0

Таблица 2

Относительный вклад в суммарную мощность дозы
различных типов ионизирующих излучений
оксида калифорния

Тип излучений	Относительный вклад в %	
	Без защиты	Защита из чугуна 15 см
Нейтроны спонтанного деления	95,5	99,9
Собственное гамма-излучение калифорния	3,5	0,1
Собственное гамма-излучение продуктов деления	0,2	-
Тормозное излучение	0,8	-
ВСЕГО	100,0	100,0

При работах с калифорнием практически вся радиация формируется нейтронами спонтанного деления.

Одновременно с изучением уровней радиации за защитой была проведена оценка индивидуальных квартальных доз от нейтронного и гамма-излучений при работах с изотопами америция и кюрия. Для операторов величины доз внешнего гамма-облучения составляли $0,05 \pm 0,15$ рентген, а дозы от нейтронов - до $0,05$ бэр. Это указывает на то, что работы проводились, главным образом, с небольшими количествами ТПЭ в боксах с легкой защитой.

Введение в работу изотопов калифорния повышает дозы нейтронного облучения до $0,36$ бэр за квартал. При этом доза гамма-облучения остается без изменений.

Представляло интерес оценить также для операторов горячей лаборатории примерную величину инкорпорированной активности, накопленную за время работы с ТПЭ. Характеристика воздушной среды рабочих помещений определялась ниже следующими данными.

Распределение среднемесячных концентраций радиоактивных аэрозолей ТПЭ при выполнении различных операций соответствовало логарифмически нормальному закону с параметрами: медиана - $1 \cdot 10^{-15}$ кюри/литр, стандартное геометрическое отклонение - $2,4$. С вероятностью 95% все значения концентраций находились в интервале $1 \cdot 10^{-16} \pm 1,4 \cdot 10^{-14}$ кюри/литр.

Характерный изотопный состав аэрозолей: америций-241 - 10% , америций-243 - 5% , кюрий-244 - 75% , калифорний-252 - 10% . Как правило, получаемые соли были окислами ТПЭ и относились к классу нерастворимых соединений.

Дисперсность аэрозолей, определяемая радиографическим методом с использованием жидких ядерных эмульсий, характеризовалась параметрами: $СМАД = 3 \pm 5$ мкм, $\sigma_g = 1,5 \pm 2$, $АМАД = 13 \pm 20$ мкм.

Величины накопленной активности и доз внутреннего облучения различных областей дыхательного тракта (носоглоточной, трахеобронхиальной, легочной и лимфоузлов) при непрерывном поступлении аэрозолей ТПЭ в течение 5 лет работы приведены в табл. 3.

Таблица 3

Нагрузки и дозы
для различных областей дыхательного тракта

Изотоп	Накопленная активность в пикокюри		Дозы облучения в бэрах			
	P	L	N - P	T - B	P	L
Америций-241	4	3	$2,4 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,04	0,2
Америций-243	2	1,5	$1,2 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,02	0,1
Кюрий-244	27	23	$19 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,3	1,7
Калифорний-252	3	3	$2,5 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,04	0,2
ВСЕГО	36	30,5	0,025	$1 \cdot 10^{-3}$	0,4	2,2

Оценка проводилась согласно динамической модели легких¹ с использованием расчетных формул и номограмм из работы². Коэффициент использования средств индивидуальной защиты был принят равным 90% . Рассчитанные дозы внутреннего облучения легких за

5 лет соизмеримы с дозами, полученными за счет внешнего облучения в течение этого же времени.

При переработке реакторных мишеней, когда необходимо выделить ценные короткоживущие изотопы типа кюрия-242, берклия-249, ейнштейния-253, облученный материал поступает с малым временем выдержки. Вследствие этого значительную роль начинают играть изотопы осколочного происхождения. Следует учитывать возможный выдох в воздух радиоактивных газов и аэрозолей продуктов деления и, в первую очередь, йода-131. Проведена оценка вкладов газовой и аэрозольной фаз йода-131 при некоторых способах переработки мишеней (табл. 4).

Таблица 4

Относительный вклад аэрозольной и газовой фаз
в суммарную активность йода-131 в воздухе горячей камеры

Вид переработки мишени	Относительный вклад, %	
	Аэрозольная фаза	Газовая фаза
Механическая резка	99,8	0,2
Щелочное растворение	81,0	19,0
Кислотное растворение	48,0	52,0

Из табл. 4 видно, что при кислотном растворении существенно возрастает процент газовой фазы, которая играет ведущую роль в загрязнении воздушной среды радиоактивным йодом-131 и увеличивает вероятность проникновения его в зону пребывания персонала.

Выявляемая специфичность радиационных условий при работах с ТПЭ требует дифференцированного подхода к решению задач санитарно-дозиметрического и медицинского контроля персонала.

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ENSURING RADIATION SAFETY WHEN USING LARGE ISOTOPIIC
SOURCES OF IONIZING RADIATIONS FOR BRINGING ABOUT
RADIATION PROCESSES.

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ABSTRACT.

Results of an investigations series for ensuring personnel radiation safety when bringing about various radiation processes are presented. The investigations include: studying radiation penetration regularities in large isotopic plants shielding structures such as labyrinths, technological channels etc. with design formulae; studying environment contamination mechanisms formation using the design formulae which allows to prognosticate the forseen wasres amount. Analysis of the accidents occurred during large isotopic plants operation and proper procedures developed to prevent their occurrence are given. The All-Union standards and methodical rules regulating radiation safety conditions in design, construction and operation of large radiation apparatus on the basis of the authors investigations are considered. The many-years experience showed that the observance of the standards developed by the autors reduces and in a number of cases completely excludes the possibility of the personnel over-exposure, the environment and irradiated products contamination.

ENSURING RADIATION SAFETY WHEN USING LARGE ISOTOPIC
SOURCES OF IONIZING RADIATIONS FOR BRINGING ABOUT
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Ensuring radiation safety when using large isotopic sources is a necessary condition for bringing about any radiochemical and other investigations and processes (1). The urgency of studying such matters is caused by the necessity of solving engineer and technological problems in developing radiation plants and processes excluding any possibility of undersirable consequences of the over-exposure of the plants personnel, the neighbouring area population and irradiated products consumers (2,3). The development of a series of such plants which are in full accord with the present day radiation protection demands became possible due to fundamental and experimental investigations including (4):

- protection from external radiation - studying radiation penetration regularities in large isotopic plants shielding structures and developing design methods;
- protection from internal radiation - studying the environment contamination mechanisms' formation;
- radiation processes safety - studying the conditions of accident-free radiation processes;
- formulation of standards and methodical rules - development of sanitary rules for radiation plants design, construction and operation.

The following hazard factors arising from the use of large ionization sources in radiation technology were considered:

- a) radiation hazards defined by the physical nature, activity and type of radioactive substances and the character of their technological use; quantitatively these hazard factors are defined by the doses of the personnel external exposure and internal exposure stipulated by the radioactive substances entering the organism of workers;
- b) non-radiation hazards characterized by the quantity and type

of toxic substances intaken from irradiated objects or formed by irradiation processes, or as a result of a fire or explosion or by aggressive media formation. Sudden or gradual destruction of hermetic shells in the sources, shieldings and apparatus may be observed afterwards (5).

Transportable and movable units may have biological shield in the form of a cylinder or a sphere. The information about the larger efficiency of such type shielding in comparison with flat shielding having the same thickness could be found only in single publications (6-9). However the physical nature of this phenomenon was not analyzed. The authors have shown that for a point on the shielding surface the share of the scattered radiation from the flat-type shielding is larger than from the spherical shielding of the same thickness. Energy distribution studies of scattered gamma-radiation intensity showed the maximum the energy of which was approximately equal to the energy of gamma-quanta single scattered at an angle θ . Calculation and experimental results allowed to determine that the dependence of the angle distribution of the intensity (I_θ) of gamma-quanta scattered in a spherical or flat shielding from the angle θ is of exponential character and can be given in the considered angles range (with an error $\pm 10\%$) as follows:

$$I_\theta = A_{fl}(\cos\theta) \cdot \exp\left(-\frac{\theta}{\theta_{fl/sph}}\right) \quad (1)$$

where: A_{fl} ; A_{sph} - are the proportionality coefficients for flat and spherical shieldings respectively;

$\theta_{fl/sph}$ - the constants of the angle distribution of the intensity of gamma-quanta scattered in flat and spherical shieldings respectively.

Moreover the decrease of the shielding material atomic number and gamma-quanta energy leads to the increase of the difference $\delta^{(s)}(A)$ in the energy B_E and dose B_D build-up factors:

$$\delta^{(s)}(A) = \frac{B_E^{fl}(A) - B_E^{sph}(A)}{B_E^{sph}(A)} \cdot 100\% \quad (2)$$

Shielding geometry effect becomes more essential for gamma-quanta energy less than 1 Mev and the shielding made from the low atomic number materials (10,11). The physical characteristics studies of multiply scattered gamma-quanta in the lead and steel shielding in the standard many-elbowed channel of the serial agricultural units showed that the curve from of gamma-ra-

diation intensity reduction (I) along the axis of the channel is slightly dependent on the initial energy of radiation and can be given as the following dependence:

$$I(l) = I(l_0) \cdot C \cdot l^{-K} \quad (3)$$

where: C and K are the empirical coefficients equal to 58,0 and 2,6; 22,0 and 3,0 for steel and lead respectively. It is necessary to note that K is slightly dependent on the shielding material; l - is the geometrical parameter of the many-elbowed channel.

As the investigation results of spectral composition of ^{60}Co and ^{137}Cs gamma-radiation passed through the many-elbowed channel show there is the maximum in the energy range of 150 keV. When the channel is filled by water-equivalent material the spectra maximum is changed to the energy range of 100-120 keV, whereas the portion of multiply scattered gamma-quanta increases, radiation intensity for ^{60}Co and ^{137}Cs decreases on 20-30% accordingly. The obtained data on scattered gamma-radiation spectral composition allowed to recommend technological channels schemes and choose detectors with corresponding characteristics for biological dosimetry.

The studies of the field of the exposure dose rates in a labyrinth and its spectral distribution gave a formula (4) that is of great importance not only for constructing labyrinths but also for the evaluation of exposure doses during the anticipated accidents (12,13):

$$P_{\text{зад}} = 8,4 \cdot A \left[\frac{e^{-\mu x} \cdot B(h^i, \mu x, z)}{R^2} + \frac{2}{(2R)^n R_i^2} \prod_{i=1}^n \frac{S_i \cos \theta_i \cdot \alpha_i(\theta, E)}{R_{i+1}^2} \right] \quad (4)$$

where: A - the irradiator gamma-equivalent, mg-equiv. Ra;

θ - the normal/ R_i angle;

R_i - the distance (cm) from the centre of the irradiator to the centre of the area S_i (cm^2);

R_{i+1} - the distance from the area S_i where i - reflection occurs to the corresponding calculated point (or to the next area S_{i+1});

μ - the linear coefficient of gamma radiation decrease, cm^{-1} ;

x - the concrete shielding thickness, cm;

R - the irradiator - to calculated point distance, cm;
 $\omega_i(\theta, E)$ - the albedo of the dose from concrete (from
i-area);

$B(\omega, \mu, z)$ - the dose build-up factor.

The formula (4) is just under the following assumptions; the irradiator is of point type, linear dimensions of the areas are comparatively small to the distance R_i .

Realizing the future prospects of using beta-radiation sources in the large radiation plants construction the authors have experimentally studied beta- and gamma-radiation penetration through the shielding non-homogeneities peculiar to any plant. As a result they got empirical dependence which allowed to evaluate mixed radiation dose from technological slits and openings. The dependence received takes into account the slit depth (t), its height (δ), and the outer edge of the slit-to-detecting point distance (x). The formula connecting the dose rate ($P_{\beta+\gamma}$, $\text{mrad}\cdot\text{sec}^{-1}$) of beta-gamma-radiation at the outlet of the technological slit with its geometry, $^{90}\text{Sr} + ^{90}\text{Y}$ irradiator activity (Q , curie) and the effective atomic number (z) of the material from which the inner lining of the irradiation chamber is made, is the following:

$$P_{\beta+\gamma} = \frac{K \cdot \delta \cdot (\bar{z})^{0,27} \cdot Q}{t^2} (4e^{-0,6x} + e^{-0,25x}) \quad (5)$$

It can be assumed with sufficient accuracy for practical aims that $K (\bar{z})^{0,27} = 3,4 \cdot 10^2$ (k - the proportionality coefficient) under the following conditions: the irradiator is flat and parallel to the technological slit; the irradiator and the shielding are made of aluminium, stainless steel, lead. In cases corresponding to the given assumptions made on the basis of analyzing the technological schemes of beta-irradiators use, the calculated by the formula (5) radiation dose rate exceeds the measured one on 15 + 20% (14).

Studies of the ways of radioactive contaminants formation during the operation of the so-called "closed" radiation sources were made. It was caused by the facts of the unsealment of the radiation sources used in large gamma-installations that was confirmed by a number of other papers.

According to the technological scheme of reciprocal movements of an irradiator and irradiated object large isotopic radiation

installations may have fixed and mobile irradiators. By the type of shielding we can classify them as the installations with dry, liquid (water) and mixed shielding, that allowed detailing the mechanisms of radioactive contaminants formation during the operation of different installations.

The radioactive contaminants at the gamma-installations with dry shielding can appear as a result of the source unsealment (plants with the fixed irradiator) or residual radioactive contaminant dispersion from the sources shell (plants with mobile and immobile irradiator). In this case the dispersion coefficient of surface contamination (\mathcal{E} , m^{-1}) can be used as a qualitative characteristic of aerosol formation intensity. As a result of model and production research we have got an expression for defining total activity of wastes Q (curie) formed at the installations with dry shielding for the time t (15,16):

$$Q = \mathcal{E} \cdot t \cdot V \left\{ (n - n^1) \bar{C} + \left[\sum_i C_i S_i + a_0 e^{-\frac{0,693 \cdot t}{T_{1/2}}} \cdot \frac{S}{S_0} \right] n^1 S_0^{-1} \right\} \quad (6)$$

where: \bar{C} - the average surface contamination of one sealed source, curie/ m^2 ;

n - the total number of sources in the installation, pieces;

n^1 - the number of unsealed sources, pieces;

V - the volumetrical rate of the cooling air, $m^3 \cdot h^{-1}$;

S_0 - the surface area of the source, m^2 ;

C_i - the radioactive contamination of separate areas of the upper and lower surface parts ($i=1,2$) and side surface of the sources ($i=3$), curie. m^{-2} ;

S_i - the area of the upper and lower parts ($i=1,2$) and side surface of the sources ($i=3$), m^2 ;

a_0 - the initial activity of one source, curie;

$T_{1/2}$ - the half-life period of an isotope, h^{-1} ;

t - the time, which has passed from the moment of the source initial activity definition, h ;

S - the defect area in the source shell, m^2 .

By means of experiments the dispersion coefficient $\mathcal{E} = C_0 / C$ was found, where C_0 - the quantity of the radioactive material, blown from the source, curie. m^{-3} ; C - the radioactive contamination of the radiation source surface shell, curie. m^{-2} . The authors have shown that this coefficient value is equal to $1,5 \pm 0,9 \cdot 10^{-6}$, m^{-1} . Moreover it was found that practically this coefficient depends neither on the surface contamination of the sources nor on

the air flux speed above the surface.

For the gamma-installations with water shielding the transferred to water activity value Q (curie), can be defined from the relation $Q=b.K.s.t$, where b - the specific activity of the radioactive material, curie/g; K - the coefficient characterizing the solution of radioactive contaminants in the liquid from the surface unit for the time unit, $g/cm^2.day$ (paper review shows that for large gamma-installations this coefficient is accepted to be $5 \cdot 10^{-6} g/cm^2.day$); s - the defect value in the source shell, m^2 ; t - the time of the source contact with the water medium, day.

While prognosticating the quantity of the radioactive material released into the environment it is possible to define beforehand the value of the defects. For preliminary calculations the value of a defect can be accepted in the range of $0,5 + 1,0 mm^2$, that approximately corresponds to the pin-hole area of the sources under testing and is similar to real conditions connected with the moment of detecting the considerable leakage of radioactive material into environment caused by the local corrosion of the source shell.

Our investigation results caused stopping the serial production of ^{60}Co , ^{137}Cs radioactive isotopes and beta-sources in aluminium shells and initiated their production in stainless steel. The use of highly active sources in hermetic stainless steel shells excluded environmental radioactive contamination.

As many-years experience of using large radioisotopic installations showed our recommendations concerning organizational-technical procedures and shielding constructed in accordance with our calculations provide bringing down irradiation levels to the safe values under the normal mode of radiation plants operation. However, the accidents caused by various reasons can occur during the plants operation. By the analysis it was stated that accident reasons can be the following: the radiation sources unsealment, the shut down of the devices for the sources movement, design and construction drawbacks of the shielding and braking the safe operating procedures. Very often combination of the two or more abovementioned reasons can be the cause of an accident (17).

Taking into account that the shut down of the blocking and signalization systems would cause radiation accidents with serious consequences, the system of blocking the entrance door "according

to the dose" was developed. This system uses automatic commutation of the dosimetry instruments sensors (18) and is designed for large isotopic installations. The proposed blocking system excludes the possibility of the personnel entering the irradiation chamber when the sources are under working conditions.

But still the existence of the smallest probability of the blocking and signalization system shut down made us to develop the dose evaluation express-method for the case of an accident exposure of the large radioisotopic installations personnel that is necessary for the timely and proper medical protection actions (19). Probability evaluation of the average and limit accident occurrence during the operation of one or a group of large radiation installations having the same constructional peculiarities showed that the most reliable are the installations with dry shielding and fixed irradiator and the installations with mechanical devices for irradiator movements; whereas the less reliable are the installations with pneumatic devices for the sources movements.

One of the possible causes of non-radiation accidents at the large isotopic installations is the personnel poisoning by the radiolysis products (ozone and nitrogen oxides) or by toxic substances either from irradiated objects or formed as a result of irradiation process. Production and model research conducted allowed to give recommendations concerning the arrangement, size and construction of inflow and outflow vent holes, and rational ventilation modes of operation in the working area of large stationary gamma-installations, that exclude the possibility of such type accidents occurrence (20).

Our investigations results for ensuring radiation safety when using large radiation sources were used by the USSR Ministry of Health which exercises state sanitary monitoring in our country.

The sanitary monitoring system adopted in the USSR supposes the development of the all-Union normative documents regulating the radiation safety conditions when carrying out design, construction and operation of radiation-hazardous objects including large radiation installations. Such regulatory documents developed for the first time in the world practice are: "Sanitary rules of the construction and operation of large isotopic gamma-installations", "Sanitary rules of the construction and operation of the reactor primary circuits", "Sanitary rules of the construction

and operation of large isotopic gamma-installations with fixed irradiator", "Sanitary rules of the construction and operation of large beta-installations", "Methodical instructions on carrying out radiation monitoring when loading, adding to load and replacing the ionizing sources of large isotopic installations". All these documents developed with our help are approved by the USSR Ministry of Health and thus became a sanitary law for all the institutions and enterprises connected with the development and operation of large radiation installations.

Such unification of demands to the large radiation installations allows not only to use optimal solutions received by research and development but to provide the united system of sanitary radiation monitoring of radiation workers, employed at these installations. The collected dosimetry data including those received by the use of the various methods of personal monitoring and counters for determining whole-body exposure allow to evaluate annual exposure doses to be in the range up to 0,5 rem/year which envisages the high reliability of the developed radiation protection system and shielding of the large isotopic installations.

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К ОЦЕНКЕ ВЛИЯНИЯ АДСОРБИРОВАННОЙ ФАЗЫ ЗАГРЯЗНЕНИЯ
ЗАЩИТНЫХ ПОКРЫТИЙ И СРЕДСТВ ИНДИВИДУАЛЬНОЙ ЗАЩИТЫ
НА РАДИАЦИОННУЮ ОБСТАНОВКУ В ПОМЕЩЕНИЯХ

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ABSTRACT

This report gives a method for accounting dependence of radioactive contamination hazard on the function type of its distribution in the depth of material. The method is based on describing time dependence of distribution functions by Green's operators. The proposed procedure is simple enough and can be used by the personnel of dosimetric service which has not special mathematical training.

В ряде случаев основным фактором, определяющим опасность, которую представляют поверхности защитных покрытий и средств индивидуальной защиты, загрязненные радиоактивными веществами, является поступление этих веществ в воздух помещения. С этой точки зрения загрязнение, проинфильтровавшее в глубь материалов, может считаться фиксированным. Но течением времени, выделяясь на поверхность оно создает опасность рецидива загрязнения. Вероятность перехода адсорбированного вещества в адсорбированное состояние существенно зависит от того, как это вещество распределено в объеме покрытия. В данной работе предлагается способ количественного определения зависимости степени опасности адсорбированного загрязнения от вида функции его распределения по глубине материала.

Эта задача представляет особый интерес при работе с некоторыми альфа-излучателями. С аналогичными вопросами приходится встречаться, когда имеют дело с нерадиоактивными токсичными веществами, об-

ладающими высовой проницаемостью. Следует отметить, что функция распределения альфа-активных излучателей по глубине загрязненного объекта может быть измерена по спектру альфа-излучения у поверхности, причем исследуемая загрязненная поверхность не подвергается какой-либо специальной обработке и в процессе измерения не разрушается^{1;2}.

В данной работе использованы следующие определения. Под поверхностью понимается часть материала, доступная непосредственному воздействию моющих средств или контакту с воздухом помещения.

Считается, что токсичное вещество, сосредоточенное на поверхности или перешедшее с нее в воздух помещения, находится в опасном состоянии или опасной фазе. Токсичное вещество, недоступное непосредственному контакту с внешней средой, мы будем называть квазификсированным загрязнением.

Для оценки степени опасности требуется по заданному распределению квазификсированного вещества определить вероятность рецидива в течение времени t . В связи с прикладным характером задачи в данной работе использован приближенный полуэмпирический способ описания процесса, позволяющий на конечном этапе избежать громоздких вычислений.

Обозначим распределение квазификсированного загрязнения в начальный момент времени через $\rho_0(x)$, где x — пространственная координата, направленная вглубь загрязненного объекта. Пусть процессы массопереноса и обмена веществом между опасным и квазификсированным состоянием являются линейными. В этом случае зависимость функции распределения абсорбированного вещества от времени может быть представлена как результат действия оператора Грина на первоначальную функцию распределения.

Практически в результате измерения функция $\rho_0(x)$ получается в форме набора дискретных чисел, определяющих количество активного

активного вещества, сосредоточенное в различных слоях материала. Поэтому операторы записываются в матричной форме.

$$[\rho(t)] = [G] * [\rho_0] \quad (1)$$

Необходимо подчеркнуть, что матрица $[G]$ может быть получена непосредственным измерением или полуэмпирическим путем. В реальных условиях невозможно учесть все факторы, влияющие на перераспределение вещества в объеме материала и выделение его во внешнюю среду. Поэтому для оценки степени опасности используется оператор $[G]$, который описывает не реальное развитие абсорбированной фазы, а некоторый фиктивный критический процесс, а именно, при его построении полагают, что все неопределенные составляющие процесса протекают таким образом, что они максимально способствуют выделению вещества в опасную фазу. Аналогичным приемом можно учесть также и нелинейные процессы.

Количество токсичного вещества, перешедшего в опасное состояние, определяется разностью поверхностных плотностей квазификсированного загрязнения в начальный и заданный моменты времени. Оно находится суммированием всех компонент вектора $[\rho]$ (см. выражение (1)). Изменив порядок суммирования, результат можно представить в виде произведения двух векторов.

Окончательно: Если в начальный момент времени $t = 0$, в материале имеется квазификсированное вещество, распределенное по закону $[\rho_0]$, к моменту t в опасное состояние перейдет не более чем

$$Q = [R_{ec}(t)] * [\rho_0] \quad (2)$$

$$[R_{ec}(t)] = [1] - [N(t)],$$

где:

$$N_k(t) = \sum_i G_{ik}$$

$[1]$ – вектор, все компоненты которого равны 1.

Следует подчеркнуть, что при умножении вектора начального распределения на вектор $[R_{ec}(t)]$ как правило требуется выполнить 5÷6 (не более 10) операций арифметического умножения с последующим

сложением результатов.

Необходимые значения $[R_{ec}(t)]$ получают заранее при изучении материала в лаборатории.

В качестве примера рассмотрим зависимость степени опасности, которую представляет токсичное загрязнение, от вида функции его распределения по глубине материала при условии, что процесс массопереноса в объеме материала подчиняется закону Фика, причем поток на поверхности стремится к бесконечности.

На рис.1 показаны три графика функций распределения одного и того же количества токсичного вещества по глубине материала. Там же приведены величины, пропорциональные максимально возможной скорости перехода вещества в опасную фазу при соответствующем начальном распределении. Хорошо видно, что степень опасности, которую представляет токсичное вещество, существенно зависит от его распределения по глубине материала. В этой связи можно несколько уточнить формулировку задачи дезактивации по отношению к квази-фиксированному загрязнению. А именно, вместо удаления токсичного вещества, что как правило практически невозможно, достаточно изменить функцию распределения так, чтобы уменьшить вероятность перехода загрязнения в опасную фазу.

В качестве примера рассмотрим дезактивацию загрязненного материала, процесс переноса в котором подчиняется закону Фика. На рис. 2 показаны графики функций распределения токсичного вещества по глубине материала, получающихся при непрерывной длительной обработке сильным моющим средством. Зависимости получены расчетным путем. На том же рисунке дана таблица, в которой приведены полное количество загрязняющего вещества (столбец Q), оставшееся в материале к моменту времени t , величина, пропорциональная максимальной скорости перехода вещества в опасную фазу (столбец $\pi \frac{Q'}{Q_0}$). Видно, что в период времени от $t = 0,01$ до $t = 0,4$ степень опасно-

сти убывает в 9,1 раза, и при этом удаляется только 30% загрязнения.

Применение формализма операторов Грина часто облегчает вычисление приближенных матриц $[G]$, а также упрощает синтез данных, если известны отдельные составляющие процесса. Например, при изучении эрозии материала в результате его истирания трудно провести эксперимент, учитывающий одновременно диффузию в объеме материала и разрушение его поверхности. Поэтому соответствующие данные удобнее получить в независимых опытах. Результирующая матрица Грина может быть найдена методом теории возмущения как оператор Грина для уравнения

$$\rho_t = H[\rho] - v \frac{\partial \rho}{\partial x},$$

где: H — оператор, учитывающий процесс массопереноса внутри материала. Как указывалось, его функция Грина предполагается известной;

v — скорость разрушения поверхности.

Получение конкретных алгебраических выражений выходит за рамки данного доклада.

В результате анализа при помощи таблиц $[Rec(t)]$ можно количественно определить степень фиксации радиоактивного загрязнения поверхности, т.е. измерить, какая часть токсичного вещества представляет реальную опасность.

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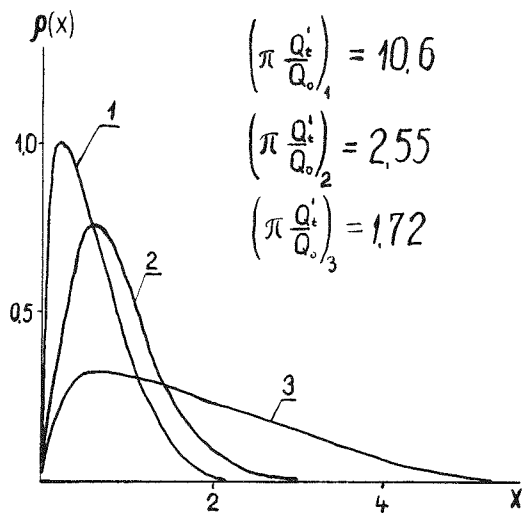


Рис. 1. Зависимость степени опасности загрязнения от функции распределения вещества по глубине материала. По оси абсцисс - глубина материала; по оси ординат - плотность радиоактивного вещества. Величина $(\pi \frac{Q_t^i}{Q_o})$ - степень опасности для распределений вида 1, 2 и 3.

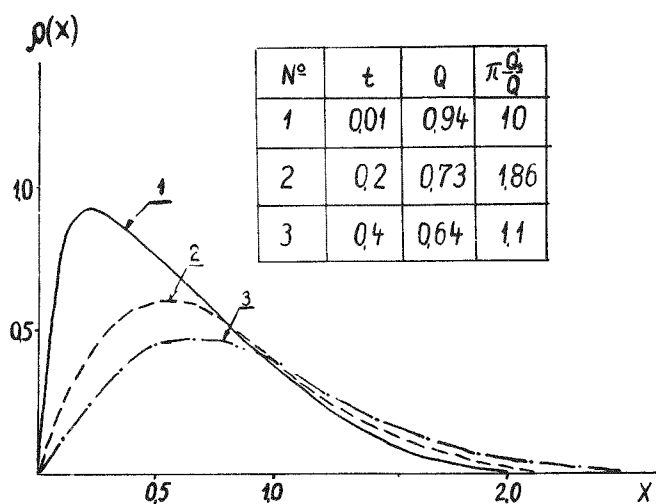


Рис.2. Изменение степени опасности загрязнения при дезактивации. По оси абсцисс - глубина материала. По оси ординат - плотность радиоактивного вещества. Пояснения даны в тексте.

ОБЕСПЕЧЕНИЕ РАДИАЦИОННОЙ БЕЗОПАСНОСТИ ПРИ ИСПОЛЬЗОВАНИИ
ПРИБОРОВ С РАДИОИЗОТОПНЫМИ ИСТОЧНИКАМИ ИЗЛУЧЕНИЙ.

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Summary.

The description of measures is given as far as prevention and current sanitary control is concerned providing radiation security using radioisotopic instruments. The demands are indicated to the sources of radiation, from instrument and their comparison with the doses of radiation.

Приборы с радиоизотопными источниками ионизирующих излучений используются во все расширяющихся масштабах в целях контроля, автоматизации и интенсификации технологических процессов, элиминации электростатических зарядов, предупреждения несчастных случаев, пожарной безопасности и т.д. Большой контингент лиц, контактирующих с приборами, большое разнообразие и рассредоточенность приборов по объектам, широкий диапазон условий их эксплуатации возлагают повышенную ответственность на профилактический контроль качества приборов и источников, их соответствия современным требованиям радиационной безопасности.

Установившаяся в СССР система профилактического санитарного контроля новых типов радиоизотопных приборов и других изделий радиационной техники включает следующие этапы: лабораторные испытания опытных образцов, выдачу разрешений на производственные испытания опытных образцов, выдачу разрешений на изготовление и эксплуатацию установочной партии, контроль источников и приборов установочной партии на соответствие их опытным образцам, экспертизу технической документации, выдачу разрешений на серийное изготовление.

При лабораторных испытаниях опытных образцов источников производится измерение или контроль параметров, указанных в таблице 1.

Наиболее полной программе испытаний подвергаются источники альфа- и мягкого бета (β_M , $E \leq 0,25$ мэв)-излучений. Механическая, термическая и коррозионная устойчивость источников гамма, нейтронного, рентгеновского и жесткого бета (β_H , $E \geq 0,25$ мэв)-излуче-

ния, имеющих одинарные или двойные ампулы из нержавеющей стали или сплавов алюминия, определяются свойствами этих материалов, которые достаточно хорошо известны. Однако, если при эксплуатации источники могут подвергаться воздействию коррозионных сред или таким механическим нагрузкам, которые способны вызвать устойчивые явления, испытания производятся по полной программе.

Таблица 1.

Параметры, контролируемые при испытаниях источников.

№№	Контролируемый параметр	Источники излучения:					
		γ	n	x	$\beta_{ж}$	$\beta_{м}$	α
1.	Выход рабочего излучения	+	+	+	+	+	+
2.	Выход сопутствующих излучений	-	-	+	+	+	+
3.	Герметичность	+	+	+	+	-	-
4.	Загрязненность поверхности	+	+	+	+	+	+
5.	Скорость выделения радиоактивного изотопа	-	-	-	-	+	+
6.	Устойчивость к ударам и вибрации	+	+	+	+	+	+
7.	Устойчивость к истиранию	-	-	-	-	+	+
8.	Термоустойчивость	-	-	-	-	+	+
9.	Коррозионная устойчивость	-	-	-	-	+	+

Что касается испытаний на устойчивость к ударам, вибрации и температуре, то принимаемые при этом испытательные нормы практически не отличаются от испытательных норм, предлагаемых в проекте 1. Испытания на устойчивость к проколам не производятся. За критерий устойчивости источников проникающих излучений принимается сохранение герметичности ампул.

У источников альфа- и мягкого бета-излучения механическая, термическая и коррозионная устойчивость обеспечивается в основном свойствами материала (керамика, эмаль, металл-матрица, окислы металлов, слоенки и т.д.), в котором заключен радиоактивный изотоп. Герметизирующие покрытия имеют вспомогательное значение, т. е. их толщина не превышает нескольких мкг/см². Благодаря радиационно-физическим и радиационно-химическим процессам происходит ослабление связи поверхностного слоя препарата с ниже-лежащими слоями и появление на поверхности источника слабофиксированной загрязненности. К таким источникам неприменимы методы контроля герметичности, предлагаемые в проекте¹, так же как и непривлечено определение закрытого источника, принятое в рекомендации ISO².

В настоящее время в СССР принято следующее определение закрытого источника³. "Закрытый радиоизотопный источник излучения... источник, в котором радиоактивный материал заключен в оболочку (ампулу или защитное покрытие), предотвращающую контакт персонала с радиоактивным материалом и его рассеяние свыше допустимых уровней в условиях, предусмотренных для использования источника".

Это определение позволяет количественно оценивать источники на соответствие их требованиям радиационной безопасности по ско-

рости выделения радиоактивного изотопа при его эксплуатации в допустимых условиях. Важной характеристикой таких источников является также количество радиоактивного изотопа, которое может выделяться из источника при аварийных нагрузках.

Исследования радиационной обстановки на производствах, использующих приборы с источниками альфа- и мягкого бета-излучения и лабораторные исследования процессов дефиксации изотопов из этих источников позволили установить предельно-допустимые скорости дефиксации (таблица 2).

Таблица 2.
Предельно-допустимые скорости дефиксации изотопов.

№№	Тип источника	Предельно-допустимая скорость дефиксации мкюри/см ² месяц
1.	Из ²³⁹ Pu на основе эмали для нейтрализаторов электростатических зарядов.	$1 \cdot 10^{-4}$
2.	Из ²³⁸ Pu на основе эмали для альфа-активационных анализаторов.	$3 \cdot 10^{-3}$
3.	Из ²³⁹ Pu на основе глазури для дымоизвещателей.	$5 \cdot 10^{-4}$
4.	Из трития на титане для нейтрализаторов электростатических зарядов.	1

При таких скоростях дефиксации изотопов из источников уровни загрязнения производственной среды остаются в пределах допустимых для неконтролируемых зон.

Источники альфа- и мягкого бета-излучения в процессе эксплуатации требуют периодической очистки от пыли и загрязнений. В связи с этим они подвергаются испытаниям на устойчивость к истиранию сухой фильтровальной бумагой под давлением до 0,5 кг/см² поступательным или вращательным движением в количестве до 1500 циклов.

Коррозионные испытания таких источников проводятся в основном для выяснения их устойчивости во влажной атмосфере при повышенных температурах (98% относительная влажность при 40°C).

При лабораторных и производственных испытаниях опытных образцов приборов производится определение уровней излучения от прибора и при необходимости - измерение доз облучения персонала, занятого эксплуатацией оборудования с установленными приборами.

По максимально допустимым уровням излучения приборы разделены на 2 группы: к 1-ой группе относятся приборы, нормальная работа с которыми может привести к облучению в дозах, превышающих предел дозы для категории "отдельные лица из населения" (гамма-дефектоскопы, гамма терапевтические аппараты, приборы для исследования буровых скважин).

Во второй группе - приборы, при работе с которыми доза облучения не выходит за предел дозы для указанной категории. В эту группу входит большинство приборов технологического контроля, нейтрализаторы электростатических зарядов с источниками из плуто-

ния-239 и др. Целесообразно также выделить третью группу - такие приборы (дымоизвещатели и др.), которые в допустимых условиях эксплуатации не могут дать дозу облучения свыше генетически значимой дозы для населения (таблица 3).

Таблица 3.
Максимально-допустимые уровни излучений
от приборов

Группа №	Мощность дозы, мбэр/час.	
	на поверхности	на расстоянии 1 м
1	-	3
2	10	0,3
3	0,3	0,01

Фактические дозы облучения персонала, занятого эксплуатацией приборов 1 и 2 группы (таблица 4) значительно ниже соответствующих нормативов.

Таблица 4.

Дозы облучения персонала при работе с
радиоизотопными приборами.

№№	Приборы (аппараты)	Группа	Доза облучения, бэр/год
1.	Нейтрализаторы электростатических зарядов с источниками из плутония-239	2	0,1 - 0,2
2.	Приборы технологического контроля с источниками бета-излучения.	2	≤ 0,1
3.	Приборы технологического контроля с источниками гамма-излучения	2	0,1 - 0,3
4.	Гамма дефектоскопы стационарные.	1	0,5 - 2
5.	Гамма дефектоскопы переносные	1	2 - 3
6.	Приборы для гамма и нейтронного каротажа скважин	1	1 - 3

Помимо профилактического контроля, органы санитарной службы СССР осуществляют текущий (периодический) надзор за радиоизотопными приборами: за соответствием условий их эксплуатации допустимым условиям, за правильностью учета и проведением дозиметрического контроля (для приборов 1 группы), а также проводит расследование каждой аварийной ситуации, т.е. любого отклонения от нормального процесса эксплуатации прибора, которое может создать повышенную радиационную опасность для работающих или населения.

Анализ аварийных ситуаций с приборами показывает, что основными причинами их являются отклонения фактических условий эксплуатации от допустимых условий для данного типа источников и приборов (главным образом по механическим и коррозионным воздействиям).

По мере накопления опыта в эксплуатации приборов данного типа происходит снижение числа аварийных ситуаций. Так при десятикратном увеличении ежегодных поставок приборов абсолютное число аварийных ситуаций практически не увеличилось.

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HEALTH PHYSICS AND OPERATIONAL EXPERIENCES IN A TREATMENT AND PACKAGING
FACILITY FOR SOLID AND SEMI-SOLID WASTES AT E.I.R.

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1. Summary and conclusions

Solid or semi-solid wastes have either to be stored for long periods or to be disposed by dumping into the ocean or enclosure into suitable geological formations. Each case requires an appropriate treatment and packaging of the wastes. Despite the relatively low specific activities there exist considerable radiation hazards for the operating personnel due to incorporation risks. At EIR Würenlingen a waste treatment facility was built that reduces such hazards to acceptable levels. The main installation is a large combination of steel boxes, well ventilated and held at reduced pressure. Operations that can be carried out include sorting of wastes, compression of loose wastes or drums, dismantling or reduction to small pieces, solidification of sludges with cement, casting of concrete mantles and covers etc. All direct contacts between personnel and radioactive material are avoided. Either the men work from outside the box with gloves or remotely operated hydraulic tools or machines, or they work inside the box with respiratory protection or in ventilated plastic suits. An incinerator plant under construction will bring further volume reduction of burnable waste, even for α -activities. Our experiences are reported and show that radioactive wastes of low or medium specific activity can be treated and packed according to any requirements without hazards for the operating personnel, the surrounding population or the environments of the treatment facility or the storage sites.

2. Purposes of the laboratory and choice of the concept

Most wastes are produced and collected in forms that do not allow storage for more than a few years. For storage over extended periods of years or final disposal a treatment of the wastes is indispensable. Years of improvised waste handling experience and an extensive literature search resulted in specifications and an operations plan for each treatment method considered, from which the necessary working conditions and installations could be derived. Thus we developed a concept for an installation that should best suit our needs. The laboratory came into operation in 1970 and can be used for the following methods :

- sorting of mixed wastes, mainly into four groups: combustible, non-combustible, compressible, non-compressible;
- baling of wastes by pressing for volume reduction;
- solidification of liquid or semi-liquid wastes (sludges) with cement;
- dismantling and cutting of large waste items (filters, components, plastic sheets) into small pieces that fit into 100 or 200 liter drums;
- enclosure of wastes in cement or concrete.

The wastes must be packaged into steel drums or prefabricated concrete containers according to international transport regulations and requirements for fi-

nal disposal such as dumping into the deep ocean. Most of the wastes were low- and medium-level wastes from EIR operations (average input $100 \text{ m}^3/\text{yr}$), but also wastes from other sources (some $40 \text{ m}^3/\text{yr}$) such as industry, research institutes, hospitals and in smaller amounts from nuclear power stations were treated. About 10 percent were medium-level wastes. The laboratory is only in operation during the regular working hours. The crew consists of a supervisor, a health physics technician and four to five operators. In 1971/72 wastes were treated during an effective average of 30 weeks per year, the rest of the time was used for revisions, decontaminations, vacations etc. With an average yearly production of 650 drums (200 liter) of conditioned low-level waste and 80 concrete containers of medium-level wastes the capacity of the laboratory is not fully utilized. By increasing the number of operators and working during two shifts a day an input of $400 - 500 \text{ m}^3/\text{yr}$ of unconditioned wastes could be handled.

3. Description of the laboratory

The laboratory was installed in an already existing massive one-story reinforced concrete building, $22 \times 17 \times 4.5$ meters. An annex contains the installations for cement mixing and the feed pump. The building (plan, Fig.1) is divided into a large storage, loading and operating hall, entrance and exit lock areas, active and inactive change rooms and office, and a personnel access and control lock. The ventilation system is housed in two rooms on top of the change rooms, together with the breathing air supply and the continuous exit air monitor.

About half of the operating hall serves as a temporary storage and transfer area for incoming wastes. The other half is the main working area with a large ($8 \times 6 \times 3.3$ meters) combination of massive airtight steel boxes, consisting of a transfer box for loading wastes either into the sorting or the operations box, a sealed operations box, a sorting box, a baling press box and a cement box with an exit lock. The boxes are connected by hydraulic sash doors with electrical interlocks between the inner and outer doors in order to avoid operating errors and spreading of contaminations. The sorting box (Fig.2) contains a foot-pedal-controlled conveyor belt with three external working places and sealed connections for drums that can be removed into the operations box. The conveyor discharges into the press box. The sorting box is protected by two independent fire extinguishing systems with CO_2 and water. The baling press box (Fig. 3) accommodates a 100 ton press where loose waste or filled 100 liter drums can be compressed to 1/5 or less of the original volumes. A lucite hood is lowered on top of the baling container during pressing and the air expelled from the compressed waste is directly drawn into the exit air system. In a feed loop prefabricated cement is circulated by a pump between the cement preparation annex and the cement box where it can be filled by simple and safe means into the steel drums containing the conditioned waste and compacted by a vibrator. Sorting, press and cement box operations are done from outside from the operating hall by means of glove openings or hydraulic controls (Fig.2). All other operations such as dismantling, cutting, filling of compressed drums into larger drums are carried out in the operations box (Fig. 3). Access for working inside the sealed operations box is only possible through the personnel access lock (Fig.4) and the operators are dressed in proper protective clothing. The breathing air equipment can supply up to five operators in fully ventilated suits. In case of a power failure two reserve tanks with compressed air allow an evacuation of all five workers without any hurry or hazard by the usual procedures through the access lock. The latter serves as the control room for the supervision of operations inside the operations box with all the necessary equipment for voice-controlled intercommunication with and between individual or all operators. The intercom cables are located in the air hoses of the ventilated suits. Special quick-connect contamination-proof couplings and parallel air and intercom connections in the control lock and the box permit individual adjustment of intercom and air supplies before the operators enter the box.

All parts of the building are also connected by a loudspeaker intercom system.

The whole laboratory is very well ventilated with about 20 air changes/hr. Filtrated and if needed warm air is fed into all rooms and is then sucked through three sets of absolute filters into the operations box and from there into the other boxes. The exit air ventilators draw the air from the sorting, press and cement boxes through two filter stages with glass fiber absolute filters, of which one filter unit is located directly after each box while the second stage unit is in the exit air duct before the ventilators. Ventilation control keeps the boxes at reduced pressures of 10 - 30 mm water gauge pressure difference.

4. Equipment for radiation protection

Incoming "hot" waste drums and components can be temporarily stored in the operating hall behind a 60 cm thick concrete wall. Shielded transport containers are available for transfers of medium-level wastes. The boxes are made of 5 mm thick steel sheets without additional shielding, but for the treatment of medium-level waste mobile lead shields of 5 cm thickness can be installed at the respective working places. The baling press is connected to the cement box by a rail track with a hydraulically operated trolley (Fig.3) carrying a container with a cylindrical 6 cm thick lead shield which takes up the drums for pressing, concreting and unloading through the exit lock of the box.

At the three most exposed locations outside the boxes GM counter γ -dose rate monitors are mounted. In the storage/loading area and the exit area of the operating hall and in the control lock sets of portable instruments for β/γ dose rate and β/γ and α surface contamination monitoring are available together with shielded detectors for wipe tests and air samples. Exit contamination control of the personnel is done by background-compensated β/γ hand/foot monitors in the active change room and the operating hall, a scintillator α hand monitor in the active change room and by very sensitive β/γ and α hand/foot monitors at the exit of the inactive change room. The air in the operating hall is continuously sampled and the filters are periodically checked for α and β/γ . The exit air activity from the boxes is continuously monitored and registered for α and β/γ activities. The exit of the operations box and the active change room are equipped with showers. A complete and sufficient stock of radiation protection materials is maintained.

5. Operational and health physics techniques

The wastes are delivered into the operating hall in closed steel drums, in some special cases in sealed plastic bags. All treatment operations are executed inside the boxes. The interlocked sash doors permit safe transfers without leakage of activities. If the large door of the operations box has to be used, a plastic tent is connected as a temporary lock for the transfer of large equipment or waste items. All rooms are regularly controlled by wipe tests. Special care is given to the transfer of filled containers from the box to the exit lock area. Drums are washed with water in the box and transferred wet. When leaving the exit lock of the cement box they are immediately checked for contamination at the entire surface. This is done in the operating hall where final decontamination can be done if necessary, before a crane transports the containers to the exit lock area.

Working in the operating hall and at the sorting, press and cement boxes is done in ordinary coveralls without additional protective garments. For work inside the operations box the operators change completely into special underwear, coveralls, rubber boots, hoods and gloves, for wet work also disposable plastic suits. The normal respiratory protection is the army gas mask, fully ventilated protective suits of EIR designs are only worn for Tritium, Carbon-14 or Radium wastes. All protective clothes offer relatively comfortable working conditions for periods of 2 1/2 to 3 hours between half-hour or longer breaks. When leaving the operations box the operators take off clothes and respiratory protection in the access lock and undergo there a coarse contamination control. After a shower

in the active change room they check again for contamination and leave for the inactive change room where they put on their own underwear and ordinary working or street clothes. A final contamination check follows at the exit of the inactive change room. When using ventilated suits the men take a shower with the suits on at the exit of the operations box (Fig.4) before entering the access lock. There the suit is taken off with the help of another man wearing mask, hood and gloves, who makes the first contamination check. All complicated or hazardous operations are supervised and monitored by the crew's health physics technician. But each crew member completed a 4 weeks' radiation protection course at EIR and has to take care of his own radiation protection and monitoring. The supervisor of the laboratory is also a fully trained health physics technician. Independent controls of working environment and methods are performed by the working place survey group of the Health Physics Division.

For personnel monitoring direct reading pocket chambers (200 mR) and film badges with quarterly evaluation are used, supplemented by TLD chips in finger-rings which we designed to stand heavy mechanical work without damage to chips or gloves. Incorporation monitoring is done by periodical urine analyses, after risky operations or incidents by special investigations and whole body counting.

6. Operational experiences

Radionuclide composition, physical and chemical forms of the wastes vary a great deal and require very different treatment and packaging methods which, except the pressing, must be manually executed. A rationalized conditioning by special remotely or automatically operated equipment is out of reach for an installation of this size for space and cost reasons. In 1971 and 72 wastes with the following main activities were treated :

α-emitters (mostly Pu)	ca. 10 Ci	⋮	Tritium	ca. 260 Ci
Radium	ca. 1 Ci	⋮	Carbon-14	ca. 3 Ci
mixed fission products + ⁶⁰ Co	ca.660 Ci	⋮		

The average whole body doses of the personnel due to external exposure were about 1.5 rem/yr and were similar for the entire crew. The incorporation monitoring showed no values above the investigation levels, most of them could be interpreted as representing less than 1 percent of the MPBBs. These minor incorporations resulted from Iodine-131 or Tritium work in the operations box.

No air contaminations outside the boxes in the operating hall have been found, and surface contaminations outside the boxes are extremely rare events at harmless levels less than five times the operational guides for uncontrolled zones. This proves that practically no contaminations are spread from the interior of the boxes to the outside. The reasons for this are: an excellent ventilation and relatively large negative pressure differences in the airtight boxes, rigorous and disciplined controls of personnel and material at the exits of the boxes, and frequent coarse decontamination of the boxes before the contamination levels become too high. A certain hazard exists for the hands of the operators due to relatively frequent damaging of gloves. If no wounds are inflicted, washing is in most cases sufficient for decontamination. Only in three cases the hands had to be decontaminated by a specialist from our first aid and personnel decontamination team. One operator received a cut into the hand from an Iodine-131 contaminated item. The wound was washed with saline solution and surgically cleaned by a physician. This was the only, minor incident and had no lasting consequences.

Our experiences have been excellent, the concept chosen is well suited to our needs. Summing up : good protection of the personnel depends on a reasonable combination of installations that are appropriate to the tasks and a good and disciplined working technique with reliable radiation protection and monitoring.

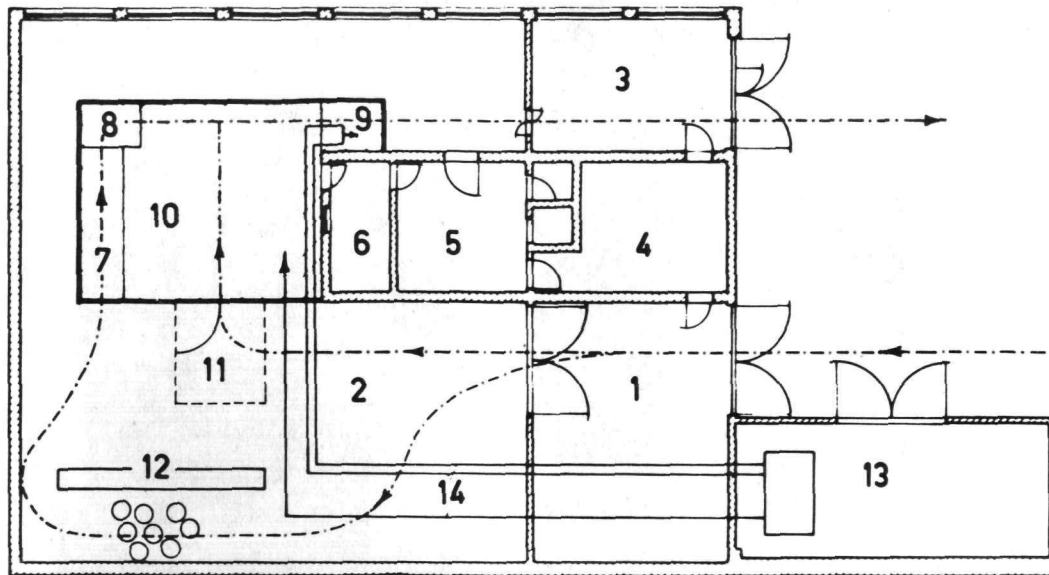


Fig. 1 : EIR waste treatment laboratory, floor plan

- | | |
|--------------------------------------|--------------------------------------|
| 1 vehicle + waste entrance lock area | 8 baling press box |
| 2 storage, loading + operating hall | 9 cement box |
| 3 waste exit lock area | 10 sealed operations box |
| 4 inactive change room | 11 protective tent at large box door |
| 5 active change room, shower | 12 shielding of incoming waste store |
| 6 personnel access + control lock | 13 cement preparation annex |
| 7 sorting box | 14 cement feeding tubes |



Fig. 2 : Exterior of the sorting box and the transfer box. An operator is loading a plastic bag into the sorting box, where another operator is sorting the waste from the conveyor belt into several drums or the press box (left background). The lower, closed sash door is used for the transfer of drums into the operations box, visible through the window at the right. γ -dose rate monitors are mounted at the working face of the boxes.

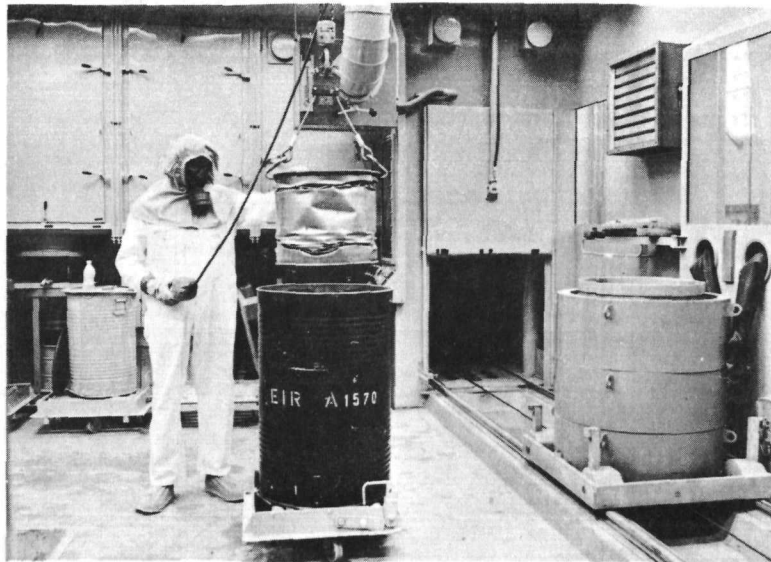


Fig. 3 : Interior of the operations box. A 100 liter drum (only partially compressed for the sake of clarity) has been removed from the baling and transfer trolley with its shielded container (at the right on the track). The sash door of the baling press box is kept open, normally it would be closed during that phase (right background). The right hand wall has a series of windows and glove ports for the external operator of the press and the transfer trolley. An entrance air filter is also visible. In the background at left the wall of the sorting box is seen with an exit air duct and the attachments and trolleys for the sorting drums below. The operator wears the usual dress for work inside the box.

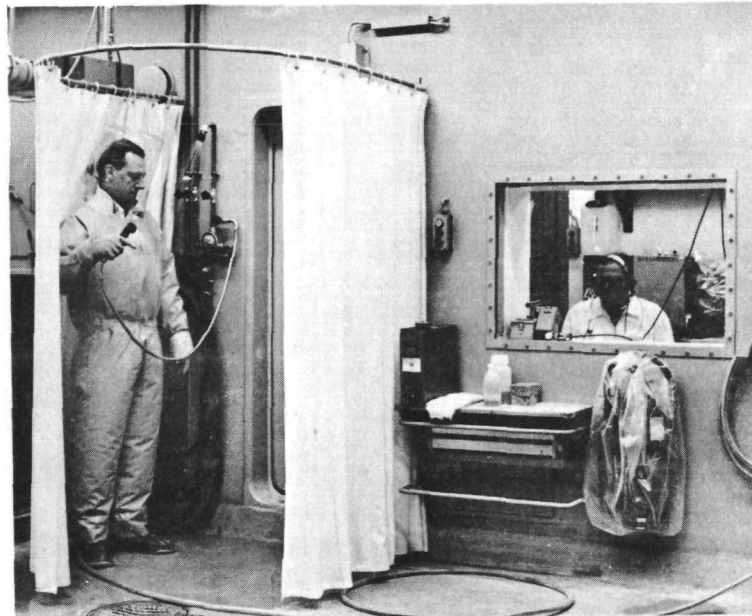


Fig. 4 : Exit of the operations box to the personnel access and control lock. An operator in a fully ventilated PVC-suit with clear hood and intercom takes a shower before leaving the operations box through the door into the access lock. Behind the window the operator in the control lock has intercom contact with the operators in the box and regulates the air supplies. Five air and intercom hoses are connected at the right of the window. The hoses are disconnected from the suits before leaving the box. Decontaminating material and a contamination-protected fire extinguisher are below the window.

SOME PRELIMINARY INVESTIGATIONS ON THE CONTRIBUTION
OF MUONS TO THE STRAY RADIATION LEVEL AROUND THE
CERN 28 GeV PROTON SYNCHROTRON

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Abstract

Below a few GeV the predominant radiation in the environment of accelerator installations is stray neutrons. Muons may however become dominant if pions and kaons produced by the interacting primary beam are allowed to decay in the course of free flight and the resulting muons in forward direction are not sufficiently shielded.

Measurements with a counter telescope allowing for the determination of their angular distribution behind the shield around the beam direction are reported. The attenuation length for muon spectra from the decay of pions of a few GeV in several materials was determined and is compared with theoretical values. The measurements show in addition the contribution to stray radiation levels by other components penetrating the main shield of the accelerator. Their relative importance at different distances and their environmental impact are discussed.

Introduction

Radiation protection measurements around multi-GeV proton accelerators are difficult due to the largely unknown mixture of stray radiation outside the shielding. This is caused by the interactions of primary protons with target and shielding materials giving rise to a variety of secondary radiations covering a wide energy range. The predominant penetrating component of the dose measured is generally neutrons.

The contributions of muons, however, may become important if pions and kaons produced in the interaction of the primary beam are allowed to decay in the course of free flight into mu-mesons. A certain attenuation of the latter component projected in the forward direction requires more shielding than is needed for the parent pions, since the muons -- up to the energy range which is important for our problem -- only interact electromagnetically whereas the hadrons in addition undergo nuclear interactions.

The attenuation of hadrons is predominantly exponential in the shielding and may be described by a single parameter, i.e. the attenuation length λ which is about 130 g cm^{-2} in iron shielding for proton beams of 10-300 GeV.

Although the attenuation curves for muons (calculated and measured) turn out not to be simply exponential, they still may be characterized

over certain depths by one single parameter. Apparent attenuation lengths reported¹⁻³ for muons from primary proton energies of 25 GeV range between 430 and 900 g cm⁻². These values are influenced by the choice of the production formulae of pions, their decay path (length and angle) and the resulting muon spectrum but seem less influenced by the shielding materials used in the calculations.

Experimental Equipment

A counter telescope was constructed to detect muons resulting from the decay of pions and kaons in the forward direction downstream of targets and after penetrating the shielding wall of the accelerator enclosure.

Three scintillation counters in triple coincidence of 5 cm width, 20 cm height and 2 cm thickness are mounted on a supporting bar at a distance of 75 cm from each other. The supporting bar pivots around a central axis in the horizontal plane and can be moved in the vertical plane as well. The pulses observed from the telescope are counted and are stored according to their height in a 256 multi-channel analyzer with a 100 MHz ADC. All measurements are made with reference to a two-fold coincidence monitor counter left in a fixed position in the radiation field during the measurements. This is done in order to take into account intensity variations of the accelerator.

Results

The equipment was tested first in a series of measurements performed at an angle of 6° with respect to an internal target at a distance of 45 m. The total shielding thickness between target and detector amounted to only 3000 g cm⁻². A typical recorded spectrum is shown in Fig. 1. This consists of a peak at low channel numbers corresponding to a small stopping power or minimum ionizing particles and a long tail with higher stopping power.

When additional shielding material (lead, iron) is placed in front of the first counter, a second peak at higher channel numbers appears and the first peak decreases (Fig. 2). The conclusion of this effect is as follows: the radiation penetrating the shielding wall is muons "contaminated" with hadrons. Different kinds of particles in the GeV range however have roughly equal stopping power, about 2.2 MeV g⁻¹ cm⁻², in a scintillator and will thus be found grouped around the same channel numbers. By introducing shielding the hadrons produce forward-peaked secondaries of lower energies that build up the second peak as these cause a higher energy loss in the detector.

By increasing the shielding thickness this second peak is found to be attenuated with a λ corresponding to hadrons, whereas the first peak decreases with an attenuation length which approaches a value expected for muons.

The apparent attenuation lengths for muons determined from this experiment were 379 g cm⁻² for lead and 457 g cm⁻² for iron.

A second series of experiments was performed using a test beam tuned to 19 GeV/c pions. The same build-up phenomenon was observed. Attenuation lengths for muons gave in this case values of 417 g cm⁻² for lead and 491 g cm⁻² for iron, in other words slightly higher than in the preceding experiment. From these measurements a superiority of lead of

about 20% compared to iron was found for the attenuation of muons, whereas the error in the determination of the parameter λ is of the order of 10%.

In a third series of measurements an attempt was made to detect muons behind a beam stop in an experimental hall. An extracted beam from the PS interacted with a target located 23 m upstream of the shielding (Fig. 3). Spectra were recorded in seven positions and all of these showed a typical Landau distribution with no "tail" towards higher channel numbers. Figure 4 is shown as an example.

When the counter telescope was turned in the horizontal plane around its vertical axis, the maximum count rate was observed when the instrument pointed in the direction of the target. The angular dependence of the muon intensity, for example for position 6 (Fig. 3), is shown in Fig. 5. The angular response of the telescope for monodirectional radiation has the shape of a triangle and is also given in the figure. It was shown by calculations that the measured angular distribution is not distorted by this response function and thus corresponds to the actual one.

A more complicated intensity distribution of the muons, which could be decomposed into three peaks, is observed at for example position 2 (Fig. 6).

Figure 3 indicates the positions in which observations with the telescope were carried out. The arrows point in the direction of measured maximum muon intensities; their lengths have been drawn proportional to the height of the measured peaks. The extensions of these arrows pass through the target -- the main source of pions and subsequently muons -- or point to some weakness in the shielding, for example in the direction of reduced thickness of iron.

Using the CERN radiation survey method⁴ a dose rate of 1.36 mrem/h was measured in position 1. According to the results 42% of the dose rate is due to fast neutron and high energy particles, the rest is made up of charged particles and γ rays. The integration of the angular distribution for muons amounts to 0.33 mrem/h with the usual conversion of 10 muons $\text{cm}^{-2} \text{s}^{-1}$ corresponding to 1 mrem h^{-1} . Their share of the total dose rate passes from one fourth to one half at greater distances, i.e. outside the experimental hall along the axis of propagation for the muons, as the fast neutron and high energy particle component falls off rather rapidly. Muons, although rather localized, will become the dominant component of stray radiation at even greater distances.

Acknowledgements

The authors would like to thank Mr. L. Andersson for his work concerning the electronics of the equipment, Dr. Z Khawza and Mr. M. Nielsen for their participation in the measurements and evaluation of the results, and Dr. B. Schorr for his mathematical help in the unfolding of the angular distribution.

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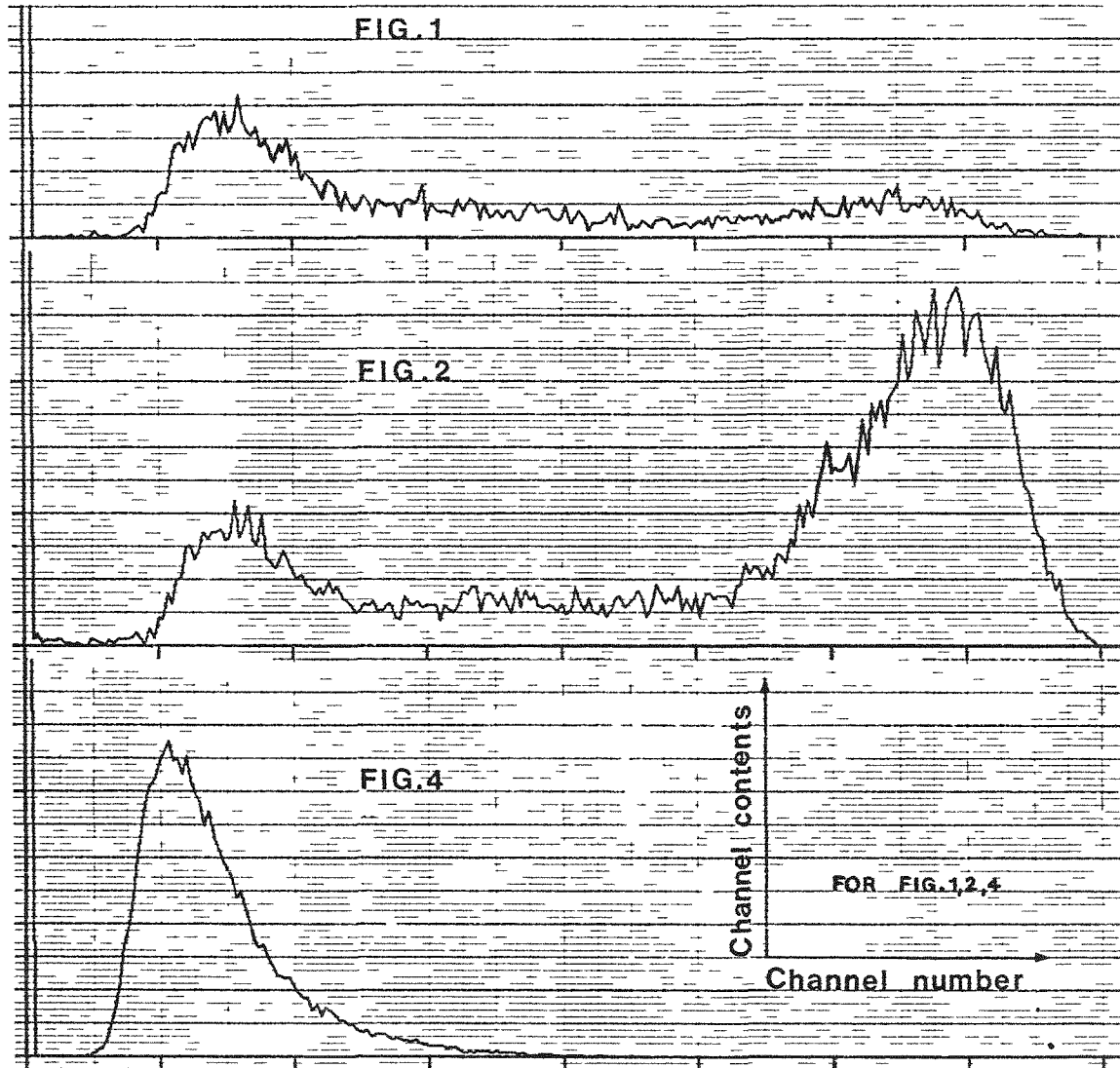


Fig. 1. Pulse height spectrum of events recorded with counter telescope behind 3000 g cm^{-2} shielding material at 60° from an internal target.

Fig. 2. Same as Fig. 1, but with 5 cm of lead in front of the first scintillator.

Fig. 4. Pulse height spectrum of events from mu-mesons as measured in position 5 shown on Fig. 3.

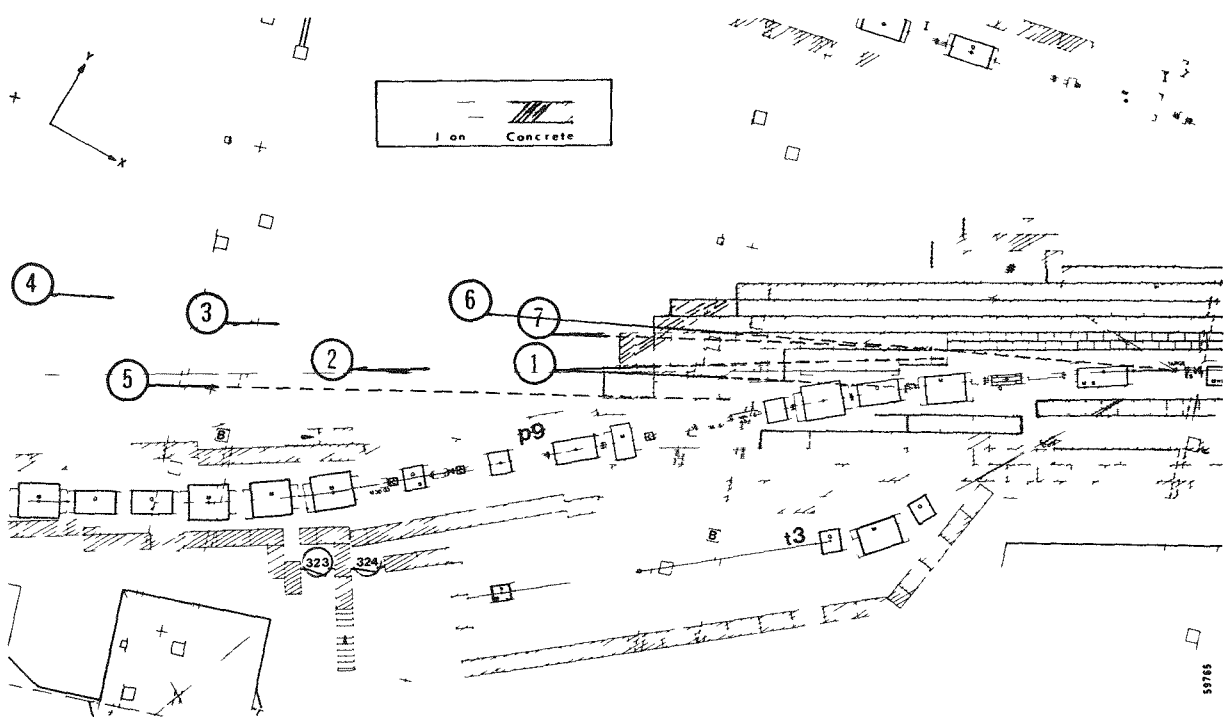


Fig. 3. Mu-meson measurements in the west experimental area downstream of an external target behind an end-stop made out of iron and concrete shielding material. The length of the arrows corresponds to the muon intensity; they are pointing in the direction of the maximum.

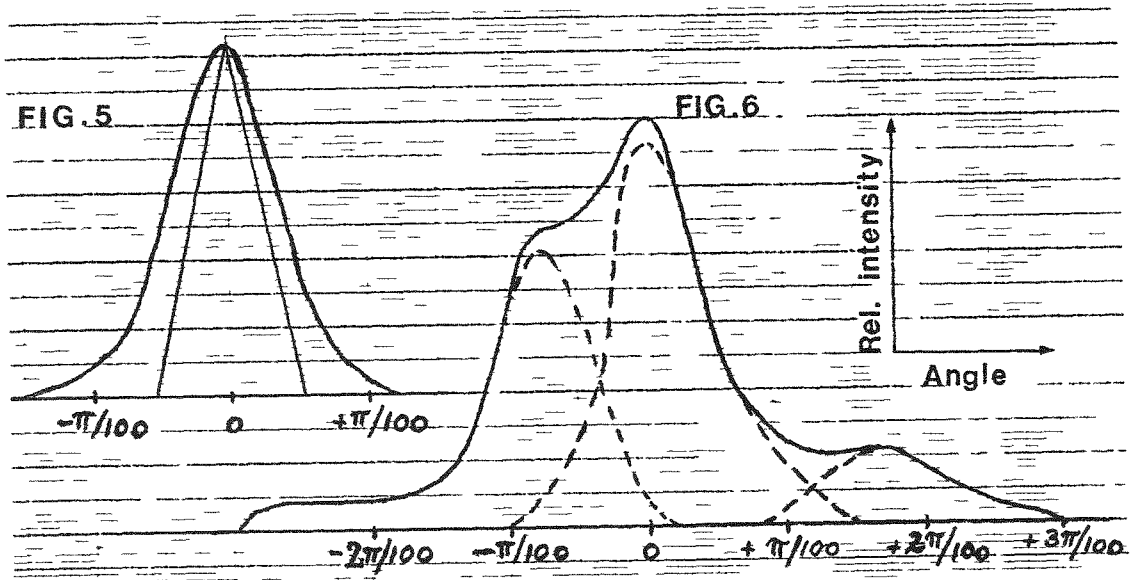


Fig. 5. Angular distribution of mu-meson intensity in position 5 of Fig. 3. The angular response function of the counter telescope having the shape of a triangle is also shown in this figure.

Fig. 6. Angular distribution of mu-meson intensity as measured in position 2 of Fig. 3.

RADIOLOGICAL SAFETY EXPERIENCE IN HANDLING AND FABRICATION OF PLUTONIUM FUEL ELEMENTS

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Abstract

The problems and experience gained in radiological safety during PuO_2 fuel fabrication for 'PURNIMA' reactor are discussed in this paper. Safety aspects in design, construction and commissioning of metallurgical facilities and glove boxes, measures for contamination control, air, area and criticality monitoring instrumentation and their calibration are dealt with. Radiological health data are summarised to reflect the adequacy of precautionary provisions followed in fuel fabrication work. Evolution of the methods for related safety problems like assessing neutron emission from PuO_2 fuel pins and Pu in solid wastes are outlined.

Introduction

Radiometallurgy Laboratory at the Bhabha Atomic Research Centre has been operating a plutonium fuel fabrication facility during the last two years involving handling and storage of kilogram amounts of plutonium. The facility has fabricated the complete core charge of PuO_2 pins for the zero power fast reactor 'PURNIMA' at Trombay.

The fuel pin of 'PURNIMA' has stainless steel cladding and consists of a central PuO_2 core of 180 mm length followed at either end, by a molybdenum plug of 80 mm length and stainless steel end plug to serve as axial reflectors in the reactor. The pin is of 11 mm diameter and of 495 mm overall length and has a plutonium loading of 123.5 g. The complete core loading called for production of 178 full fuel pins, 4 half and 4 quarter fuel pins. In addition, seven Pu-Be start-up source pins, identical in shape to the PuO_2 fuel pins, were also fabricated. This paper deals with the safety aspects in the design of the handling facilities and the experience gained in the radiological safety during the fabrication campaign.

Fuel Pin Fabrication

The feed powder PuO_2 , after milling, granulation, etc. is loaded in a suitable die and pressed with a 10 ton hydraulic press, located in a glove box. The green pellets, thus obtained, were sintered in an argon and 8% hydrogen atmosphere in a molybdenum furnace inside a glove box. After sintering the length and the weights of the pellets were checked and the geometrical density compared with that obtained by an immersion method using dibromo ethane. Acceptable pellets were then inserted in the stainless steel clad tube, whose top end plug was already welded and radiographed. The tube was inserted into the glove box; a holding spring was pushed in, followed by a molybdenum plug, PuO_2 pellets and another molybdenum reflector. The loaded clad tube was held in a special welding chamber in a glove box and the chamber was evacuated and filled with helium. After inserting the lower end plug, welding was carried

out remotely by argon arc welding. The fuel pins were then decontaminated, subjected to radiography and helium leak test. Approved fuel pins were loaded in birdcages and transferred to the Plutonium Store or reactor site.

Design Safety Features

The long biological half-life and the high energy of the emitted alpha particles together with selective localisation in bone or lung makes plutonium one of the most toxic materials when deposited inside the body. With the maximum permissible lung burden for insoluble plutonium (e.g. PuO_2) being as low as $0.016 \mu\text{Ci}$ ($\approx 0.26 \mu\text{g}$) the glove box design called for stringent built-in safety features with regard to containment capability. Since a mass as low as 500 g of Pu could lead to a nuclear excursion under unfavourable conditions, criticality safety was to be considered through mass control, safe separation of Pu units and safe design of birdcages for storage and transport, with administrative control at each stage of handling.

Safety Features in Laboratory Design

The fuel fabrication facility is housed mainly in two high active area halls with entry from an active corridor. The corridor has a decontamination room at one end, and the other side, opens out to a personnel corridor, leading to a change room. A plutonium store room is located in the personnel corridor. Equipments for the various metallurgical operations are housed in glove boxes, located in the high active halls. Services of a high order of integrity, required for a class A laboratory have been provided. Laboratory area and glove boxes are provided with separate air supply and exhaust system, the equipments of which, located in a filter house, discharge the effluents through a 76 m stack, after filtration through high efficiency particulate filters. Ventilation for the active halls and corridors have been designed to give respectively 10 and 7 air changes per hour and pressure differentials have been maintained between the areas to enable air-flow from low active areas to high active ones.

Glove Boxes and their Safety Features

Many design safety features have been incorporated in the fabrication of glove boxes from the point of view of containment. Glove box frame and floor are made of s.s 304 for ease of decontamination. Filter boxes of aluminium are conveniently located so as to enable replacement of the inlet and outlet filters by a single hand operation through the upper port. Absolute filters of MSA Honey-comb type with an efficiency of 99.9% for $0.3 \mu\text{m}$ particles are used. Transfer and posting-in operations are carried out through air locks with double doors and bagging-in ports. Normal atmosphere was found adequate for PuO_2 fuel work; however, for Pu metal handling, the boxes could be turned to an argon system, provided with a purifier and a recirculation unit. Operations were carried out in glove boxes under -1 in.WG.

Air enters through an isolation valve, a ball valve, rotameter, a regulator and an inlet filter; and vents via an exit filter, a ball valve and a three-way solenoid valve. A mechanical pressure controller and a bellow adjust minor pressure variations but in case of accidental overpressurisation or any rupture of glove or failure of recirculation system, the three-way solenoid valve initiates emergency control by opening the box directly to the -10 in. WG. main glove-box exhaust line. The inlet regulator closes and a pressure differential switch flashes an alarm in the form of a red light on glove-box board, warning the operating staff of an emergency situation. Neoprene gloves, 0.8 mm thick, were considered adequate against the soft radiation emitted by plutonium. Apart from installed CO_2 extinguishers in the halls, eutectic salt mixture in sealed PVC bag was kept handy in glove-boxes to smother any fire. The furnace coolant water is normally on main water supply. An emergency water tank has been provided to take care of failure of main water supply or loss of pressure. Filtered water from a pool was also connected to the line as an alternative for sustained supply.

Pre-commissioning Tests

The primary responsibility of the health physics staff at the time of commissioning of the facility was to check the adequacy of the protective features and assess operational safety.

Glove Box Containment Evaluation. The gloves and glove boxes were checked for leakage before commissioning for Pu handling. The box leakage rates were found to be less than 0.05% box volume/hour, as is prescribed for inert atmosphere boxes.

Glove Box Filter Efficiency Tests. Filter efficiency checks were carried out with uranine aerosols. Filters were approved for use only when they conformed to 99.9% efficiency for 0.3 μ m particles.

Effluent Drains Checks. High and low active drains were checked with inactive cold runs using rhodamin dye to ensure proper pump connections and valve operations.

Breathing Air Line Checks. Compressor air was checked for presence of oil mist, moisture and CO to ensure that their levels were below the tolerance limits. The minimum requirement of 3 cft/m at the breathing points was checked.

Operational Safety and Hazards Control

Mass of Pu, handled was initially limited to 75 g per batch to acquire experience and later, the batch size was progressively increased to 500 g Pu, after reviewing the safety aspects. In all about 85 sintering runs and about 200 in-box welding operations, covering fuel pins and start-up source pins were carried out.

Constant health physics surveillance was provided for the operations. Access control to the fuel laboratory was enforced through change rooms. In potentially active areas like filter room, decontamination room, entry was effected under health physics supervision or after obtaining special work permits. Use of protective clothing consisting of overalls, overshoes, head caps and surgical gloves for handling pellets and pins was recommended. Further, TLD's on forehead and chest, normal beta-gamma and fast neutron film badges, criticality badge and pocket dosimeters were worn while at work with PuO₂. Air line respirators connected to 15 lbf/in² airline via quick connection couplings were kept readily available for emergency use.

Equipments for sintering and weighing operations were located in a train of interconnected glove boxes to preclude the necessity of intermittent bagging out operations and consequent external exposure. Different phases of work were segregated to avoid contamination spread. The welding operations, metallography work and source pin fabrication were grouped separately in another train of glove boxes to facilitate flexibility and control of radiation exposure and contamination.

As the quantity of PuO₂ handled was progressively increased, extensive radiation survey was conducted to control personnel exposure using conventional radiation monitoring instruments. The area and air monitors were strategically located in the laboratory. Provision was also made to monitor the effluent streams. The monitors along with a remote read-out on a Central Health Console give alarm at pre-set limits for initiating corrective action.

External Hazards and Control

PuO₂ powder was obtained from reprocessing nat.U fuel from Cirus reactor.

Radiation survey data of the first seven sintering runs, with 75 g Pu per batch, indicated high beta-gamma dose rates from pellets; the pellets showed a gamma dose rate of 300-450 mR/h and the beta dose rate was 1-2 R/h. The glove box panels registered a gamma dose rate of 50-75 mR/h. This also

indicated a ratio of about 6 between contact and chest level dose rates. Analysis of an aliquot sample of PuO₂ by health physics staff indicated mainly ⁹⁵Zr-⁹⁵Nb activity and a total activity of about 5 µCi/g of PuO₂. Subsequently PuO₂ was therefore obtained from spent fuel rods with lesser fission product content. As a result, the gamma dose rates on the sintering glove box panel came down to 12-30 mR/h even with increased quantities of 200-550 g PuO₂.

The fabrication of fuel pins as well as start-up source pins did not call for special shielding to boxes. As a measure of radiation safety, fuel pin welding and Pu-Be source pin fabrication jobs were carried out by rotation of staff. This was necessary as the beta-gamma dose rate from a full fuel pin at 1 cm was nearly 40-50 mR/h while the neutron dose rate was 130-140 mrem/h. Fuel pins were checked individually for loose contamination by an alpha probe inside the box and also with swipe counting. After radiography and helium leak test, the weld-zones were checked for fixed contamination. Before machining of the welds for most pins, the counts varied in the range of 100-1000 dpm/cm², maximum being 64000 dpm/cm² while after machining and polishing, the levels for most pins came down to 200-400 dpm/cm².

For start-up source pins of 0.9 Ci strength, the method of fabrication was to mix nearly 16 g of Pu as PuO₂ with nearly equal quantity of Be followed by pressing and sintering in a high vacuum induction furnace. The neutron exposures incurred during fabrication of 2 source pins were of the order of 50 mrem on chest and 400 mrem on wrist per man. The contact gamma dose rate of the source pin was nearly 150 mR/h while the dose rates at 30 cm from the pin were 16 mR/h due to gammas and 25 mrem/h due to neutrons.

Cumulative dose(beta-gamma-neutron) received by a few members of the staff, directly involved in the fabrication work during the campaign period 18.6.70 to 21.3.72 are indicated below:

Persons	Exposure(mrem)	Person	Exposure(mrem)	Person	Exposure(mrem)
A	177	F	658	L	763
B	482	G	867	M	820
C	199	H	541	N	379
D	827	I	1069	O	473
E	166	J	1540	P	569

From the estimated ratio of contact to chest level dose rates, maximum extremity exposure could be of the order of 9 R.

Air Contamination Control

Each glove box premise has a suction port with an air sampling head connected to a central air sampling pump. Filter paper samples obtained with this system as well as with annular impactors when analysed for long lived activity, did not show any air contamination in the laboratory. In addition a Pu-in-air monitor located in the laboratory detects air borne Pu by alpha spectrometry. The detector is of a silicon surface barrier type. The unit is pre-set to sound an alarm at 8 MPC hours in presence of natural radioactivity whose spill-over in 4.1 - 5.1 MeV plutonium channels is estimated to be less than 10% of the total.

During glove changing operations, respirator area was maintained. Maintenance work was carried out once on an induction furnace for which frog suit and air line respirator were prescribed. Only one instance of air contamination due to a small tear on glove arose. Due to immediate corrective action, no personnel exposure occurred.

Bioassay and whole body counting of the operating staff showed that there was no internal exposure. About 34 members of staff were monitored, after completion of the programme for Pu deposition in lung with a thin NaI(Tl) crystal with a Be window. The count rates obtained were of background levels after repeat monitoring.

Environmental Contamination Control

Air-borne effluents were discharged after monitoring downstream through a stack. The glove box and laboratory exhaust had negligibly small long lived activity. The liquid wastes, both high active ($>10^{-4}$ $\mu\text{Ci}/\text{cm}^3$) and low active ($<10^{-4}$ $\mu\text{Ci}/\text{cm}^3$) ones were collected in separate tanks and sent for disposal. Maximum levels of alpha and beta-gamma activity of the liquid effluents discharged from the fuel fabrication facility for processing were nearly 1.2×10^{-6} $\mu\text{Ci}/\text{cm}^3$ and 5.6×10^{-6} $\mu\text{Ci}/\text{cm}^3$ respectively; the net activity figures over a year for alpha and beta-gamma were nearly 2 mCi and 10.5 mCi respectively. Solid wastes, suspected to contain Pu were segregated in standard containers marked 'active' while non suspect wastes were handed over to the waste management facility. Low active solid wastes generated were to the extent of 15 - 20 packets, each of 2 c.ft. volume and the packets had a maximum surface dose rate of 1 mR/h and these were also sent for disposal.

Criticality Safety

Preliminary clearance was limited to 250 g Pu in the sintering furnace glove box, taking into account the possibility of the coolant line rupture. Presence of two batches, each of 250 g Pu, was permitted in either of the high active halls at any time. Later, on the basis of operating experience, the quantity of Pu for sintering was progressively increased to 500 g Pu and the same handling limit, was enforced for the welding box too. Administrative control ensured that water or other hydrogenous materials were not brought inside the box; however small quantities, required for specific operations, were permitted after special clearance.

Birdcages have been fabricated to store and transport the fuel pins. The birdcage consists of a mild steel slotted angle frame work with an aluminium container, rigidly fixed at its centre. Inside the Al container is a square cluster of nine aluminium tubes welded together at the top and bottom to form a bundle. Each of the tubes accommodates one fuel pin in a PVC bag; thus nine pins, amounting to 1.26 kg PuO_2 can be stored in the birdcage. The central Al container is provided with a tight fitting cap with a neoprene gasket to render it leak tight. A prototype birdcage was subjected to drop and water leakage tests and was approved for use.

The birdcages carrying the complete core charge of 'PURNIMA' reactor (≈ 22 kg Pu) were stored in the Pu store room in a plane array. The birdcages of size $60 \times 40 \times 40$ cm^3 have been designed to maintain between the central Al containers a minimum surface-to-surface separation of 30 cm, to isolate the containers in the event of flooding. Effective neutron multiplication factor of a birdcage with 9 fuel pins, under flooding conditions (including internal flooding) has been estimated as about 0.67. Thus the nuclear safety of the individual birdcage as well as the array was ensured in the event of flooding.

Criticality Monitor

Criticality monitors are located in the two high active halls and Pu store room. The sensing device consists of an ion chamber, connected to a period amplifier. The amplifier gives an indication of the rate of rise of the gamma field during an excursion. The criteria for alarm setting of the system were fixed as follows:

- (i) the system shall sound a positive alarm if a criticality burst of 10^{15} fissions occurs at a distance of 30 ft. from the detector and delivers prompt gamma dose in 100 milli-seconds,
- (ii) the system shall not give an alarm as a result of handling 10 Ci ^{60}Co source at a distance of about 10 ft from the chamber.

The above criteria will be satisfied if a change in radiation level by 6 decades (i.e. 10 mR/h background to 10^4 R/h) triggers the alarm.

In order to study the response of the monitor, a criticality event was simulated by shooting a ^{60}Co capsule of 1 Ci strength past the ion chamber. The source was ejected with compressed air over a distance of $8\frac{1}{2}$ ft in 0.25 sec, giving a change of field from 10 mR/h to 10^4 R/h. The alarm limit was set at 50% of the maximum deflection obtained during calibration.

Estimation of Pu in Waste and Fuel Pins/Pellets

As a measure of inventory control, instruments were developed to estimate Pu content in solid wastes, PuO_2 pellets and finished fuel pins.

Assessment of Pu in solid waste was carried out by counting low energy X-rays from Pu in the channels, corresponding to 11.0 to 21.5 KeV, with a 1 mm thick and 25 mm diam. NaI(Tl) crystal, having a Be window. Measurements with 1 μCi Pu source, in a 125 mm diam and 175 mm high standard waste container, gave twelve times the background counts in four minutes and this indicated a feasibility of estimating μg levels of Pu in solid wastes.

A BF_3 filled annular counter was developed to measure the neutron emission from PuO_2 pellets and fuel pins. Since Pu has been obtained from reprocessing of low burn-up fuel, it was possible to estimate ^{239}Pu and ^{240}Pu content in the pellets and pins knowing the neutron yields from spontaneous fission and (alpha, neutron) reactions in PuO_2 . An approximate assessment of the neutron dose rate could also be made from the measurements.

Conclusion

Safe operation of the facility has been amply demonstrated by low personnel exposure and absence of unsafe incidents and this has given an incentive to fast reactor fuel development programme.

Acknowledgement

Thanks are due to Dr. A.K. Ganguly, Head, Health Physics Division for his directions on safety aspects during the various stages of the fuel fabrication programme.

CELLULOSE NITRATE PLASTIC FILM - ITS PREPARATION
AND APPLICATIONS IN HEALTH PHYSICS

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Abstract

The paper deals with the standardisation of Cellulose nitrate(C.N.) films (thickness ≈ 3.0 mg/cm²) for the detection of alpha particles from a ²³⁹Pu source and some of its applications. The etchant used was 6 N NaOH. The optimum etching time for the development of tracks was found to be 4.5 hours at room temperature (24-25°C). The track formation efficiency under these conditions works out to 17.9%.

Results of some of our studies are included here. It is now possible to detect low alpha activities of the order of 10⁻³ dpm, thereby making many types of extremely low level alpha measurements possible.

Introduction

With the advent of solid state track detectors in 1958¹ it has been possible to detect heavy charged particulate radiation such as fission fragments, alpha particles, deuterons or protons in the presence of other radiation such as beta-gamma, neutrons etc.

Of the various plastic films used so far in such measurements, C.N. plastic film has been reported to be the most sensitive. This paper describes the preparation and standardisation of the C.N. plastic films in our laboratory and some of its applications in health physics work through the registration of alpha particle tracks.

Preparation of the C.N. plastic film

Chemicals used

- a) Cellulose nitrate (commercial grade) (N₂=12.1 wt%)-17.0 g
- b) ethyl acetate (A.R.) - 61.9 g
- c) Isopyropyl alcohol (A.R.) - 5.1 g
- d) Butyl alcohol (A.R.) - 4.0 g
- e) Cellosolve acetate (commercial grade) - 8.0 g
- f) Dioctyl phthalate (DOP) (commercial grade) - 4.0 g

Using these chemicals in the proportions shown above the plastic films were prepared in the following manner by Benton's technique²:

Cellulose nitrate was first dissolved in ethyl acetate to which the other three solvents were added in the order shown above. To the solution, 4.0 g of DOP was added as a plasticiser. The mixture was then permitted to age for four days to allow it to attain chemical equilibrium before films could be prepared.

For preparation of the films, the above solution was used as such; and for obtaining thinner films - after dilution with different volumes of ethyl acetate. These solutions were poured on glass plates of size 10" x 4" which were kept in a slanting position with a slope of about 1 in 7. The glass plates were left undisturbed overnight.

The next day, the films were gradually peeled off from the glass plates by inserting water drops between the films and the glass plate. This was followed by annealing of the films at a temperature of 100°C for about 16 hours after which they were ready for experimental use.

The above films were in thickness range of 2 to 6 mg/cm². The chemical composition of the films in wt. % was found to be: C = 37.8, H=4.8, N=9.45 and O = 47.95 (by difference).

Track formation by chemical etching

The final shape of the tracks formed by chemical etching is influenced by numerous factors such as the direction of entrance of the particle into the film, the physical structure of the substance, choice of the etchant and its concentration, temperature of etching, etching time etc.^{3,4}. Under ideal conditions, i.e. when the track recording material is homogeneous and the diffusion effects are absent, the etched tracks are conical in shape with sharp terminal ends².

The alpha particle tracks registered in our C.N. films have been found to be of two types - sharp conical and pit type (Fig.1). The sharp conical tracks indicate that ideal conditions of film preparation are not beyond reach. The etchant in our case was 6 N NaOH solution at room temperature.

Standardisation of our C.N.film

²³⁹Pu is one of the hazardous isotopes which is being handled in BARC laboratories. It was therefore decided to carry out the standardisation of these films with respect to this isotope. The films were exposed to alpha particles from ²³⁹Pu sources on stainless steel planchets and the following parameters were studied:-

Etching temperature

It was found that the best track shapes could be seen when the etching was carried out at room temperature (24-25°C). At higher temperatures the track shapes tended to be pit type.

Useful thickness of the film

This was found to be about 3 mg/cm². At smaller thickness the film becomes difficult to handle after etching and at larger thickness the etching times are very long (8 hours or more) at room temperature.

Optimum etching time

This parameter was determined for the films of thickness ≈ 3 mg/cm² (exact thickness of the film = 2.9 mg/cm²) and was found to be 4.5 hours at room temperature. Beyond this etching time the number of tracks was found to diminish.

Track formation efficiency

Under the optimum conditions as given above it was found that the films (≈ 3 mg/cm²) record 17.9 tracks per 100 disintegrations in the sample, in close contact with the source, the etching time being 4.5 hours at room temperature.

Detection of low levels of alpha contamination

Radiometallurgy section, handling Kg amounts of PuO_2 , does not show any detectable floor contamination by normal monitoring techniques. To check the presence of extremely low levels of contamination, if any, five spots were chosen on the floor for the exposure of the C.N.film. These spots had not shown any loose or fixed contamination by scintillation probe monitoring. They were then covered with C.N.film^s followed by PVC covering and left undisturbed for about 11 days.

After exposure the films were washed with soap and water to remove the attached loose dust, if any. The films were then etched in 6 N NaOH solution for 4.5 hours at room temperature. After etching, all the films were washed, dried and examined under the microscope. A control film was also processed in the same fashion.

The experimental films showed the presence of many groups of tracks- each group representing one active speck. The minimum and maximum number of tracks in any group was found to be 6 and 238 respectively.

Discussion

If we assume that the track formation efficiency for alpha particles here is the same as that calculated in the standardisation experiments we can calculate the activity of each speck responsible for a group of tracks. It is seen that activity of any individual speck encountered in these experiments lies in the range from 2.14×10^{-3} dpm to 8.5×10^{-2} dpm. The table below shows the levels of contamination as detected by each film.

Place of exposure for the film	Area of the film	Total No. of tracks on the film	Corresponding activity in		
			dpm	dpm/cm ²	*M.P.L. of surface contamination
Below the bagging port of welding glove box	7.28	637	0.23	0.03	1.3×10^{-2}
Below the transfer port of balance glove box	5.28	330	0.12	0.02	8.1×10^{-3}
Below the bagging port of induction furnace glove box	6.44	1312	0.47	0.07	2.9×10^{-2}
In front of decontamination fumehood	6.34	210	0.03	0.01	4.7×10^{-3}
in front of decontamination fumehood	9.28	209	0.07	0.01	3.2×10^{-3}

* 1 M.P.L. of alpha surface contamination at BARC = 2.5 dpm/cm^2

This study shows that the new technique goes a long way in lowering the detection limit for alpha contaminations. It can be adopted for routine use also as the technique is simple.

Detection of low levels of alpha activity in bioassay samples

Encouraged by the success of C.N.films in the detection of low levels of alpha contamination, an attempt was made to study their utility for bioassay samples as well. The results of our studies with plancheted plutonium alpha activity of less than 1 dpm, from urine samples, are described below.

Two such stainless steel planchets (diameter = 2.0") of activity 0.16 and 0.593 dpm were kept in contact with C.N.films for about 11 and 7 days, thus exposing the films to 2520 and 3920 disintegrations respectively as per the counting data. Exposures were carried out in a vacuum dessicator to eliminate the interference due to natural radioactivity in air.

After exposure, the films were washed with soap and water to remove the attached loose precipitate and etched as described earlier. The number of alpha particle tracks recorded on each film were counted during scanning and found to be 3841 and 5667 respectively.

Discussion

The number of tracks recorded on the C.N.films are higher by factors of 1.52 and 1.44 than the total number of disintegrations in the samples as calculated from their respective dpm values. The discrepancy in the two results can partly be assigned to following reasons:

- i) High statistical errors in counting of the bioassay samples which were of the order of $\pm 39.6\%$ and $\pm 20\%$ in our two samples respectively.
- ii) The ZnS scintillation counter is being standardised by sources having a diameter of about 20 mm or less while the area covered by the precipitate on the bioassay planchet samples may be anywhere from 40 to 50 mm in diameter. Our preliminary investigations have shown that correction for this area would enhance the dpm value of the sample by about 25%.
- iii) The precipitate on the planchets leads to degradation of energy of alpha particles. The exact degree of degradation cannot be ascertained for such low activity samples. But it has been observed that the track registration efficiency increases as the energy of the alpha particles decreases. And for ZnS scintillation counter the efficiency of counting falls off as the energy of alpha particles decreases. These factors will therefore further help to bridge the gap.
- iv) The amount of precipitate was found to vary on the two planchets under investigation.

All these factors will help in partially explaining the discrepancy. But a detailed investigation is separately necessary to explain it completely.

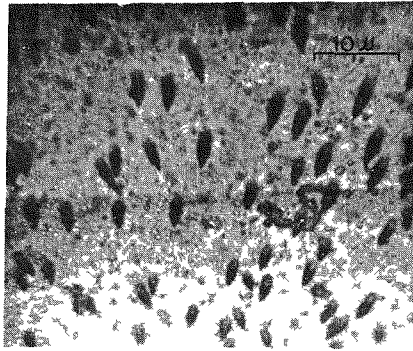
Other applications

The C.N.films prepared in our laboratory have also been used in the following studies:

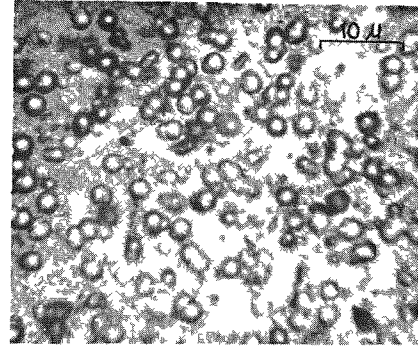
- 1) Autoradiography of electrodeposited planchet type sources. Of the few sources studied, some have revealed the presence of many clusters suggesting extreme nonuniformity of deposition (Fig.2). A detailed investigation into the causes leading to such non-uniform deposition is being carried out.
- 2) Autoradiography of single drop planchet sources. It has been observed that the activity along the edges (width of the edge $\approx 500 \mu\text{m}$) is about 60% of the total for source diameters of 6 to 7 mm.
- 3) Autoradiography of UO_2 - PuO_2 pellets (weight proportion 97.4:2.6) to study the uniformity of distribution of plutonium in Uranium (Fig.3). The information obtained should be of great help to metallurgists in evaluating
 - i) degree of non-uniformity in mixing
 - ii) sizes of individual particles of plutonium
 - iii) formation of aggregates of plutonium during mixing, if any.
- 4) Autoradiography of Uraninite mineral to locate the active grains of Uranium. This film makes the location of active grains very easy when compared with the photographic film because it is not sensitive to beta-gamma radiations

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(a)



(b)

Fig. 1 Alpha particle tracks (a) conical, (b) pit type.

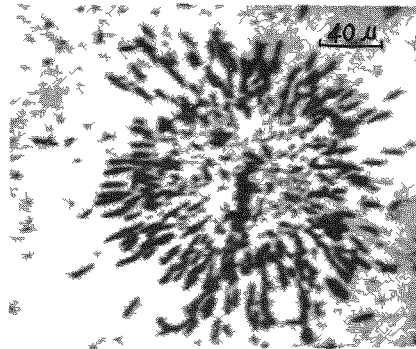
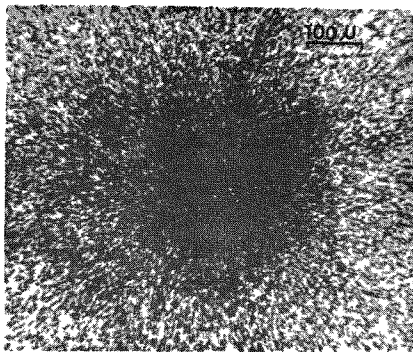
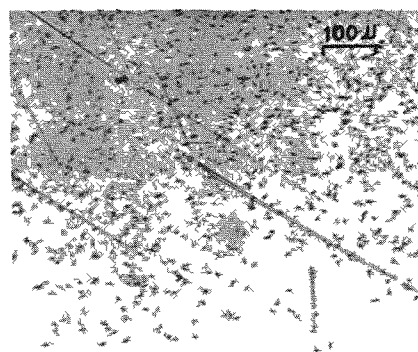


Fig 2 A cluster from electrodeposited ^{239}Pu source



(a)



(b)

Fig 3 Distribution of PuO_2 in UO_2 (a) cluster showing non-uniformity, (b) an area showing uniformity

RETENTION OF IODINE ON DUCT SURFACES

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The behaviour of fission-product iodine in a reactor exhaust duct and filtering system is of continuing interest. At CRNL parts of the research reactors are cooled with air which is released from a stack located 770 m beyond the absolute filters. Gases are transferred to the stack through a large duct. Occasionally, small amounts of iodine are released to the system from an experimental reactor loop. These nuclides have been used in the present study to measure the hold-up of iodine in the system.

Introduction

The behaviour of airborne radioactivity is of interest in evaluating the hazard from continuous or accidental discharge of fission products from a nuclear reactor. One of the factors affecting the amount released to the environment is the fractional deposition on containment surfaces. This report deals with the mechanism of deposition on specific surfaces and with re-emission from them.

The research reactors at CRNL are operated as test facilities in which the fuel material, cladding and the cooling conditions can be altered. A test loop, inserted in a single NRX fuel channel, is isolated from the reactor cooling system. In some cases the loop circuit may be vented to the main air duct; rare gases and halogens from a failed fuel element can then be discharged from the stack. The remaining fuel rods in NRX are water cooled and do not vent to the atmosphere.

Iodine-131 is particularly important in assessing the hazard from nuclear reactors. It has a fairly long half-life and escapes readily from ruptured fuel and the element is essential to man. The remaining fission product iodines have shorter half-lives and contribute proportionally less to the integrated dose, but they are useful for investigating the behaviour of the element in a dynamic system.

Various chemical forms of gaseous ^{131}I have been recognized in addition to the elemental form^{1, 2}. Perhaps the most troublesome of these is methyl iodide^{3, 4}, but hypiodous acid, HOI, is released under moist conditions at high pH^{5, 6}. Both of these are difficult to remove from the gas phase.

Elemental iodine should deposit on duct surfaces but the more penetrating forms, which generally comprise about half the total, are unreactive and are not retained on surfaces to an appreciable extent. Assuming that the retention of I_2 is reversible and that the mean life on the surface is the order of days, there should be an observable difference between the 8.065 d ^{131}I and the other isotopes which are retained on the surface. At the stack one should see the sum of the following components: a) All isotopes present in the

penetrating form; b) ^{131}I which has been deposited on the surface and subsequently released; c) ^{133}I and possibly ^{135}I in the elemental form which has undergone partial or substantial decay before escaping from the surface. Iodine-134 (52.3 m) should only be transported to the stack in the penetrating, i. e. non-elemental form.

Methods

A flow diagram of the ventilation system is shown in Figure 1. The absolute filters are located at a short distance from the NRX reactor. The duct is constructed of mild steel 1.2 m diameter by 770 m long and is painted on the inside with one coat of Zincilate 410. This provides a sacrificial coating of zinc which protects the underlying metal.

Samples were collected by drawing a measured amount (0.1 to 2 m³) of air through a canister. Collection times varied from five minutes to two hours depending on the nature of the experiment. In experiments designed to measure the chemical form of iodine the sample was collected with a freshly prepared May pack². In all other cases a polyethylene capsule, 12 mm x 50 mm, filled with activated charcoal, was used to collect the sample. The order and efficiency of components in the May pack is given in Table 1. Uptake of hypoiodous acid by copper screens and charcoal was determined in this study. McCormack⁷ reports that more than 70% of the HOI is retained by the carbon paper. In the present case less than 11% is found on the other components of the May pack, so more than 89% must be retained by the paper.

Table 1

		I ₂	Particulate	HOI	CH ₃ I	Ref.
4 Copper screens	100 mesh	97%	nil	<3%	nil	8, 9
1 Glass fibre filter	Type GFA	3%	99%	<2%	nil	9
1 Gelman carbon paper	Type ACG/B	nil	nil	>70%	1%	7
Barnebey-Cheney #513	60g	nil	nil	<6%	99%	7
Activated charcoal						

Air samples were taken at the four locations shown in Figure 1. In order to minimize loss of iodine prior to counting, the individual components were sealed in polyethylene envelopes. Sample 1 was collected through a metal tube inserted six inches into the duct so is probably not representative of the mean concentration at that point.

All the nuclides were determined simultaneously by gamma spectrometry. A 125 cc Ge(Li) detector with 2.3 keV resolution was used for most of the experiments. This was coupled to a 4096 channel analyser. Details of the method of computation were given in an earlier report¹⁰. With this system the isotopes of iodine, bromine, xenon, rubidium and cesium could be resolved easily in a single count. Repeated measurements were made to verify half-life and to check for interfering lines such as the 529.5 keV gamma transition of 2.4 h ^{83}Br . In this case the maximum error in the determination of the 20.8 h ^{133}I was 3% in the first hour. The energies of the lines which were selected for quantitative determinations are given in Table 2 together with the preferred gamma branching ratios.

Results

Chemical Composition of Iodine

A typical result from the analyses of the May packs is given in Table 3.

TABLE 2
Selected Gamma Ray Energies

Mass	Half - Life	Preferred γ Line	γ per Disintegration
131	8.065 d	364.49 keV	0.790
132	2.284 h	667.8	0.999
133	20.8 h	529.9	1.00
134	52.3 m	847.04	1.00
135	6.68 h	1260.45	1.00
82	35.34 h	554.23	0.73
		776.45	0.83

These samples were collected at location 2, immediately after the absolute filters. Corrections have been applied for incomplete uptake of I_2 on the copper screens (97%) and for retention by the particulate filter (see Table 1). The species held by the charcoal paper is largely hypoiodous acid⁷, though high molecular weight alkyl iodides may be collected with this fraction.

The five iodine isotopes show a remarkably similar distribution on the May pack. This is in contrast to Keller et al¹¹ who recently observed large isotopic differences under conditions of high humidity. However, in both instances the fraction found in the elemental form was lowest for ^{131}I .

TABLE 3
Chemical Composition of Iodines and Bromine

Mass No.	131	132	133	134	135	82
Elemental	26.1%	29.7%	31.1%	31.2%	29.7%	17.7%
Particulate	1.8	2.4	2.1	2.3	2.3	~2.0
HOI/HOBr	64.6	60.3	63.1	61.0	63.4	60.1
CH ₃ I/CH ₃ Br*	7.5	7.6	3.8	5.4	4.2	21.2

*This represents an upper limit; species which are not completely held by earlier components in the pack will be collected and counted as methyl iodide or bromide. Relative humidity = 35%.

Retention of Iodine-131

The concentration of ^{131}I at various points in the duct was measured in three separate experiments. The purpose of this series was to establish the extent to which this nuclide was retained on surfaces under conditions of near constant release. The first three runs in Table 4 followed long periods with the reactor operating at constant power.

Iodine-131, corrected for air flow at the sampling point, shows little variation along the length of the duct with the reactor operating (samples 2, 3 and 4); it follows that there is negligible removal of ^{131}I in the duct system. The results also suggest that ^{131}I is not removed by the absolute filters. The apparent increase from the first to second sampling point is caused by poor mixing and non-representative sampling at the first location.

There tends to be a release of extra fission products at shutdown which is still evident 15 hours later (Table 4). Some of the excess ^{131}I is retained on the duct surface, thus at $t_0 + 15$ hours the concentration is lower at the stack (location 4) than at the filters (location 2).

TABLE 4

Iodine-131 Concentrations at Various Points in the Duct

Experiment	Sampling Point				Time from Shutdown
	1	2	3	4	
1	4.35	6.85	6.64	6.59	nCi/m ³ *
2	3.88	6.48	6.15	6.31	*
3	3.78	5.39	-	5.46	*
3a	5.54	8.61	-	7.28	15 h
3b	3.19	5.04	-	5.18	20 h
3c	2.03	2.45	-	2.85	40 h

*Reactor Operating, Flow = 52,670 m³/h.

TABLE 5

Iodine-133/Iodine-131 Activity Ratios

Experiment	Sampling Point				Time from Shutdown
	1	2	3	4	
1	5.2	5.0	4.1	4.2	*
2	5.4	5.2	4.6	4.5	*
3	10.2	9.3	-	5.0	*
3a	1.8	1.1	-	1.7	15 h
3b	1.3	1.6	-	2.0	20 h
3c	1.0	1.7	-	2.3	40 h
3d	0.8	2.6	-	2.4	60 h

*Reactor Operating. Ratios are corrected for radioactive decay from time of shutdown as applicable.

Other Isotopes of Iodine

Table 5 shows the retention of ¹³³I, one of the four isotopes of iodine with half-lives shorter than eight days. Ratios to ¹³¹I permit use of results from the first sampling point. In Table 6 the data for ¹³³I and other nuclides are normalized to facilitate intercomparison.

Under steady state conditions at constant reactor power there is moderate retention of ¹³³I on the duct. The results in Table 6 show little dependence on half-life though ¹³³I is slightly higher than ¹³⁴I and ¹³⁵I at the stack end of the duct. Tellurium-132 (78 h) is found beyond the absolute filters and this causes ¹³²I to be high and variable.

Release Following Reactor Shutdown

The foregoing were steady state experiments; the reactor loop had operated at constant flux for periods of up to three weeks. Following shutdown, it was observed that ¹³¹I levels remained relatively constant (see Table 4), but the 6.68 h ¹³⁵I and 20.8 h ¹³³I dropped by an order of magnitude (Table 5). This rapid change provided an opportunity for observing desorption from the duct surface. Results are given in Table 7.

Bromine-82

Bromine-82 is a shielded nuclide which has a very low fission yield so would not normally be observable with the other halogens. In NRX it is probably produced by an n, γ or n, p reaction on bromine or krypton.

TABLE 6

Concentrations of Iodine and Bromine Isotopes Relative to ^{131}I (Normalized)

Isotope	Half-Life	Exp. 1 Sampling Point				Exp. 3 Sampling Point			
		1	2	3	4	1	2	3	4
133	20.8 h	1	0.95	0.78	0.80	1	0.91	-	0.58
135	6.68 h	1	0.99	0.75	0.74	1	0.92	-	0.52
132	2.28 h	1	1.1	1.0	1.0	1	1.6	-	2.0
134	0.87 h	1	0.97	0.74	0.75	1	0.93	-	0.50
82	35.34 h	1	0.67	0.69	0.68	1	0.53	-	0.51

TABLE 7

Duct Concentrations After Shutdown Relative to ^{131}I (Normalized)

Isotope	Experiment	Sampling Point			*Reactor Operating Time from Shutdown
		1	2	4	
133	3	1	0.91	0.58	*
	3a	1	0.62	0.90	15 h
	3b	1	1.2	1.4	20 h
	3c	1	1.7	2.3	40 h
	3d	1	2.6	2.4	60 h

Discussion

Morris and Nicholls^{1,2} measured the deposition velocity on copper and galvanized steel and found that there was rapid uptake with a much slower rate of removal. In the present case the metal surface of the duct has a coating of granular zinc onto which the iodine deposits. This layer of ^{131}I can be displaced with inactive iodine⁹, showing that the exchange is reversible.

The data indicate that only the elemental form is exchanging. If R is the fraction retained on the duct then T, the fraction which is not retained is given by $T = 1 - R$. Values of T can be obtained from ^{134}I and ^{135}I concentrations at location 4 (Table 6). The observed retention, R, is 0.26 for the first experiment and 0.49 for the third. May pack results show that 30% and 50% of the iodine present is in the form of I_2 at these times.

Most of the molecules on the surface of the duct at a given time will be ^{131}I because this isotope has the longest half-life. At an observed activity ratio of nine to one the numbers of atoms of mass 131 and 133 are equal in the gas phase. Normally the 52 min ^{134}I is a factor of ten less abundant.

The mean life of a molecule on the surface before escape by exchange can be estimated from the data in Table 6. Most, though not all, of the 20.8 h ^{133}I decays while held on the surface. Let us assume that the fraction exchanging is the same for all isotopes and that in the case of 52 min ^{134}I this fraction decays completely on the surface. Now if R_0 is the fraction retained on the duct which undergoes complete decay and R_t is the fraction undergoing partial decay before escape, the fraction of ^{133}I which remains is

$$\frac{N_t \lambda}{N_0 \lambda} = \frac{R_0 - R_t}{R_0} \quad \text{where } \lambda = 0.693/20.8 \text{ h}^{-1}.$$

$$\text{but } N_t \lambda = N_0 \lambda e^{-\lambda t}$$

N_0 = number of atoms at zero time.

N_t = number of atoms at t.

Solving for t gives values of 46 and 53 hours for the two experiments in Table 6. Thus the mean life of iodine on the surface is 2.1 days.

Iodine-135 has a half-life of 6.68 h and the amount remaining after 2.1 days decay is 0.6%. As practically all of the ^{135}I on the surface decays in situ, one would expect the same retention as in the case of ^{134}I . The data in Table 6 confirm this result.

It is evident from Table 7 that the ^{133}I concentration is increasing along the duct following shutdown. The relative level at sampling point 4 reaches a value $2\frac{1}{2}$ times the inlet concentration at $t_0 + 60$ hours. Hence this nuclide must be coming from the metal and filter surfaces. The release probably occurs by an exchange reaction involving ^{131}I . At $t_0 + 20$ h the abundance of this isotope in the gas phase is over 95%, while the abundance of ^{133}I is higher on the surface than in the gas phase. Exchange at this time will enhance the concentration of the isotope in the gas phase.

The outlet concentration of ^{133}I is again the sum of two components: a penetrating fraction reduced in concentration by a factor of 10 following shutdown, and an exchanging fraction which escapes from the surface after partial decay. Note that the latter is deposited before shutdown at the higher concentration. Then the increase at the stack is:

$$Q = \frac{(1-R) F + R e^{-\lambda t}}{F} \quad \text{where } Q = \frac{\text{[outlet]}}{\text{[inlet]}}$$

$$= \frac{(0.48 \times 0.1) + 0.52 \times 0.21}{0.1} \quad F = \text{decrease in } ^{133}\text{I} \text{ after shutdown}$$

$$= 1.6 \quad \lambda = 0.693/20.8 \text{ h}^{-1}$$

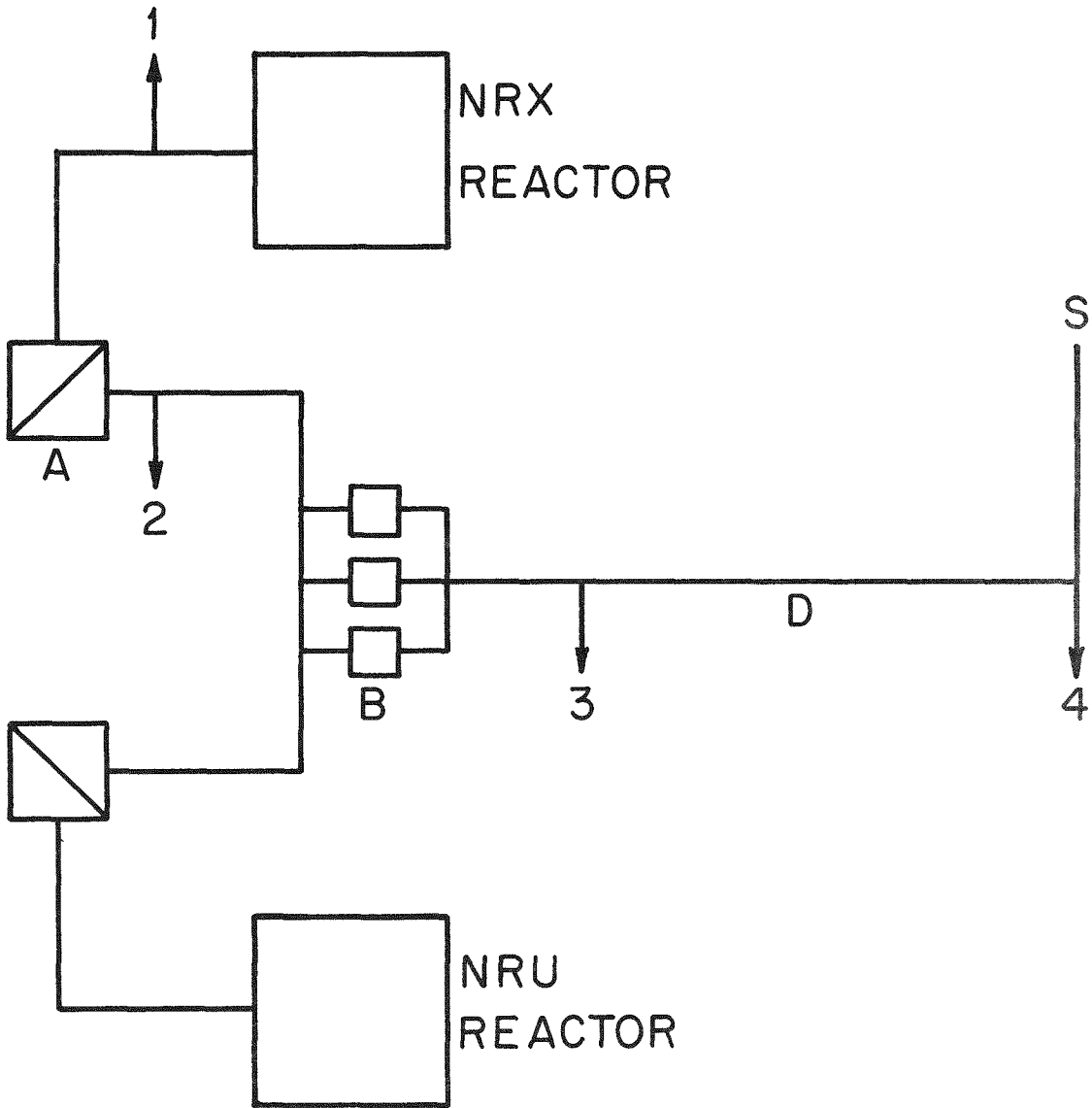
$$t = 2.1 \text{ d}$$

This value can be compared with an observed average of 1.7 from measured concentrations. The agreement supports the calculated mean residence time of 2.1 days.

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A - ABSOLUTE FILTERS
 B - MAIN FANS
 D - DUCT
 S - STACK
 1, 2, 3, 4 SAMPLE POINTS

Figure 1. Schematic diagram of the reactor ventilation system

EXEMPLE D'ORGANISATION DE LA SECURITE NUCLEAIRE
DANS UN ETABLISSEMENT UNIVERSITAIRE

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Résumé

Le Commissariat à l'Energie Atomique a été appelé à fournir à l'Université une assistance technique en matière de Sécurité Nucléaire.

Les auteurs font la synthèse des nouveaux problèmes qui se sont posés à eux en vue de mettre en place une organisation tenant compte des moyens dont ils disposaient.

Ils précisent ensuite les modalités d'action qu'ils ont élaborées et expérimentées dans un grand établissement universitaire.

Introduction

Parmi tous les établissements dans lesquels sont utilisées des sources de rayonnements ionisants, les établissements d'enseignement et de recherches sont ceux où l'organisation de la sécurité nucléaire est certainement la plus délicate à réaliser.

Après avoir mis en relief certains des problèmes qui se posent en milieu universitaire nous définirons le cadre dans lequel un organisme tel que le Commissariat à l'Energie Atomique (C.E.A.) peut apporter son concours en vue de mettre en place une telle organisation.

Ce cadre étant dressé, nous examinerons ensuite les modalités d'action telles qu'elles sont actuellement en cours d'expérimentation au sein d'un établissement universitaire.

I. Problèmes particuliers de sécurité nucléaire en milieu universitaire

Si les problèmes de sécurité liés au domaine de la recherche ne sont pas l'apanage exclusif de l'Université, il n'en reste pas moins que les problèmes de sécurité nucléaire liés au domaine de l'Enseignement ne se posent guère que dans ses établissements.

I.1. Problèmes de sécurité nucléaire liés au domaine de l'Enseignement

La mission de l'Enseignant étant d'éduquer ses élèves, ceux-ci se doivent d'être tenus informés des dangers attachés aux matériaux ou aux appareils qu'ils manipulent. De ce fait l'enseignant est généralement mieux attaché aux problèmes de sécurité que son collègue chercheur et, si l'individu est à la fois l'un et l'autre, nous pouvons être assurés qu'il prendra plus de précautions avec ses élèves qu'il n'en prendrait pour lui-même.

Il n'en reste pas moins que la mise en oeuvre de sources de rayonnements ionisants dans une salle de travaux pratiques astreint l'enseignant à prendre des dispositions particulières entraînant l'emploi de matériels ou d'équipements dont la maintenance et le contrôle périodique nécessitent le plus souvent la mise en oeuvre d'autres matériels tout aussi complexes et onéreux. En ce cas le soutien d'un technicien sera précieux à l'enseignant, surtout si ce technicien a une compétence particulière pour répondre à la saine curiosité des élèves et si de plus il prend le soin de les rassurer ou d'éveiller leurs craintes lorsque celles-ci ont un fondement réel.

I.2. Problèmes de sécurité nucléaire liés au domaine de la recherche

En ce domaine les problèmes se posent différemment du fait d'une complexité plus grande des moyens utilisés.

Toute Unité de Recherche dépendant d'un organisme, fut-il public ou privé, est une source pratiquement intarissable de problèmes de sécurité. Si de plus les travaux effectués entraînent la mise en oeuvre de radioéléments ou de générateurs électriques de rayonnements, ces problèmes peuvent s'amplifier très rapidement et nécessitent, dès leur apparition, la compétence d'un spécialiste en radioprotection.

La Recherche, surtout lorsqu'elle est fondamentale, ce qui est le cas le plus fréquent dans l'Université, n'a pas une voie tracée de façon intangible et ceux qui mènent de tels travaux le savent.

La multiplicité des techniques et des moyens dont la Science dispose actuellement, ajoutée ou plus exactement multipliée, par celle des radioéléments artificiels et de leurs nombreux composés disponibles, fait que le niveau du danger doit, ou devrait être, pratiquement estimé en permanence.

Pour que cette appréciation continue des risques puisse exister, il est non seulement indispensable que le chercheur possède une réelle compétence en matière de sécurité, mais également qu'il s'astreigne à mener ses travaux avec un souci constant des dangers qu'ils peuvent présenter.

Comme une telle association n'est ni forcément courante ni forcément bénéfique aux travaux du chercheur, la solution la plus sûre, tout en étant la solution de facilité apparente, est de faire seconder le chercheur par un cadre compétent : si la jonction est bonne entre les deux parties et si les échanges de vues sont suffisamment ouverts et fréquents, les résultats doivent être satisfaisants.

I.3. Problèmes liés à l'organisation générale de la sécurité nucléaire dans un établissement universitaire

Il paraît indéniable, au moins en France, que la réglementation actuellement en vigueur relative à la protection des travailleurs contre les rayonnements ionisants a surtout été élaborée en vue de son application dans des établissements à caractère industriel. De ce fait une adaptation devient indispensable lorsqu'il s'agit de l'appliquer en milieu universitaire.

A l'énoncé des critères pouvant être retenus en vue de définir les bases d'une organisation de la sécurité dans un établissement nous ne pouvons que mettre en évidence la nécessité de cette adaptation :

a/ responsabilité de l'employeur : si les notions de responsabilités civile et pénale restent très voisines dans les secteurs industriels et universitaires, il n'en est pas moins vrai qu'un étudiant ne peut être considéré comme un employé;

b/ structures hiérarchiques : si dans l'industrie les responsables sont désignés, dans l'université ils sont le plus souvent élus;

c/ diversité des sources de rayonnements, de leurs approvisionnements et de leurs emplois : il n'y a pas de commune mesure même entre un Centre industriel de recherches et une université, notamment en ce qui concerne la centralisation des approvisionnements;

d/ harmonisation des modalités de surveillance et de contrôle : dans l'industrie ces modalités sont imposées; dans l'université elles doivent être proposées et acceptées;

e/ choix et mise en oeuvre des moyens de prévention et d'intervention : l'industriel fait généralement le nécessaire car il est soumis à certains contrôles réglementaires et doit pouvoir offrir des garanties valables de sécurité à son personnel et à son assureur; l'universitaire n'a pratiquement

que sa seule conscience professionnelle pour prendre ou faire prendre les mesures nécessaires;

f/ discipline et diversité des personnels et visiteurs : s'il n'est nul besoin de s'étendre pour expliquer la différence de discipline dans l'un et l'autre secteur, il peut être bon de rappeler que dans l'université la diversité des personnels est très grande.

g/ information des personnels et réalisation d'exercices de sécurité : si la quasi totalité des personnels employés dans l'industrie nucléaire a suivi des conférences d'information ou des cours de formation en matière de radioprotection, seuls certains universitaires appelés à travailler sur des sources de rayonnements ont effectué un stage de formation à l'Institut des Sciences et Techniques Nucléaires; ce stage de formation sera complété d'une manière indispensable par des exercices de sécurité nucléaire dont seuls les établissements du secteur industriel ont bénéficié jusqu'alors à notre connaissance;

h/ concertation du plan général d'intervention avec les secours publics : si tout établissement détenant des substances dangereuses est classé et répertorié comme tel et si les Secours publics sont en possession d'un plan leur permettant d'intervenir dans ses installations, il est fort vraisemblable à l'heure actuelle que seuls les établissements du secteur industriel procèdent à une mise à jour rigoureuse de ces plans.

i/ sources de financement et mode de gestion des unités : si l'industrie représente un capital qui est, normalement, source de revenus, et si la gestion de ses différentes unités est coordonnée à un niveau élevé, l'unité d'enseignement et de recherches universitaires est pratiquement autonome, son financement étant presque exclusivement assuré par l'allocation de fonds publics transmis par l'intermédiaire de l'Administration : les fonds alloués étant forcément limités, il convient de reconnaître que l'affectation de crédits à l'achat de matériels ou d'équipements de sécurité ne peut être le premier investissement envisagé.

2. Cadre des activités C.E.A. - A.T.S.N. auprès de l'université

Les modalités d'action que nous avons élaborées et mises en place dans une grande université française l'ont été dans le cadre de l'ASSISTANCE TECHNIQUE EN SECURITE NUCLEAIRE (A.T.S.N.) apportée par le COMMISSARIAT A L'ENERGIE ATOMIQUE aux organismes publics ou privés qui lui en font la demande.

2.I. Assistance en matière de prévention

2.I.I. Aspect technique

Des conseillers techniques sont mis à la disposition de l'Université pour étudier en collaboration avec les responsables des différentes unités :

- a/ la conception particulière des installations,
- b/ l'aménagement et l'équipement des lieux de travail,
- c/ la délimitation des zones réglementées,
- d/ la rédaction de consignes générales de sécurité nucléaire,
- e/ l'organisation de la surveillance radiologique en milieu de travail,
- f/ le problème des déchets et effluents contaminés,
- g/ l'organisation de l'intervention,

h/ les problèmes de décontamination,

2.1.2. Aspects médicaux

Une assistance est également prévue mettant à la disposition du service médical universitaire des médecins-conseils du C.E.A. en vue d'organiser la surveillance médicale des différentes catégories de personnels exposés au danger des rayonnements ionisants :

a/ mise en oeuvre des examens médicaux,

b/ conduite à tenir en cas de contamination ou d'irradiation accidentelle,

2.1.3. Aspects réglementaires

La détention et l'utilisation de sources radioactives (scellées ou non scellées) étant soumises à une réglementation, il importe que les responsables des différentes unités d'enseignement et de recherche soient parfaitement informés des formalités à accomplir et des engagements qu'ils sont conduits à prendre et à tenir vis-à-vis des autorités compétentes. Cette information est faite également par les conseillers techniques du C.E.A.

2.2. Assistance en matière d'équipement

2.2.1. Aspects techniques

Le C.E.A. est amené, dans le cadre de son assistance en matière de prévention, à proposer un choix de matériels ou d'équipements individuels ou collectifs de sécurité.

Par ailleurs, il lui est souvent possible de fournir, sous forme de location, ces matériels ou équipements de sécurité.

2.2.2. Aspects économiques

Un avantage indéniable du système locatif est que l'Unité désireuse d'utiliser temporairement un matériel spécifique peut non seulement échapper aux délais courants d'approvisionnement, mais encore éviter un investissement souvent important qu'un emploi momentané du matériel ne justifierait pas.

2.3. Assistance en matière d'intervention

L'organisation du C.E.A. est telle que les moyens d'intervention nucléaire dont il dispose pour ses propres besoins peuvent très rapidement être mis en oeuvre. De ce fait, en cas d'accident grave à caractère radioactif ou de sinistre impliquant ou menaçant une quantité notable de substances radioactives, l'Université peut demander l'intervention du C.E.A. en alertant celui-ci suivant une procédure d'alerte parfaitement définie.

Dès son arrivée dans l'établissement accidenté l'équipe d'intervention nucléaire du C.E.A. se place sous l'autorité du représentant des autorités publiques ou, en son absence, se met à la disposition du responsable de l'établissement pour l'assister de ses conseils et de ses moyens.

De plus le chef de cette équipe C.E.A. a pour mission de proposer toute assistance complémentaire qu'il juge nécessaire et possible et de provoquer la mise en oeuvre de celle-ci lorsqu'elle est demandée soit par le responsable de l'établissement, soit par le représentant des autorités publiques.

Par la suite le C.E.A. rend compte au Ministère de la Santé Publique (Service Central de Protection contre les Rayonnements Ionisants) des dispositions qu'il a été amené à prendre au cours de son intervention.

3. Modalités d'action du C.E.A. au sein d'un établissement universitaire

3.1. Modalités d'action sur le plan technique

Il nous est avant tout apparu nécessaire de constituer un dossier suffisamment précis pour que chacun des responsables concernés de l'Université tant à l'échelon central qu'à l'échelon des unités soit systématiquement tenu informé des problèmes de sécurité nucléaire se posant dans leurs installations.

Le premier travail des conseillers techniques du C.E.A. étant de faire un bilan des dangers radioactifs et d'en déduire soit la conception particulière d'installations à l'état de projet, soit les aménagements à conseiller si ces installations sont déjà réalisées, nous avons mis au point deux documents techniques originaux :

- une fiche de sécurité nucléaire collective dite "fiche de zone"
- une fiche de sécurité nucléaire individuelle dite "fiche individuelle".

3.1.1. Fiche de zone (cf. annexe I)

Cette première fiche de sécurité nucléaire permet de rassembler sur un seul document tous les renseignements utiles relatifs à une zone de travail déterminée.

Une fois établie cette fiche permet non seulement de conseiller sur le choix ou la validité des équipements et aménagements de la zone considérée, mais également de déterminer le classement de celle-ci conformément à la législation en vigueur en vue de son balisage.

Le dossier constitué par l'ensemble de ces "fiches de zone" permet de faire le recensement à un instant donné de toutes les sources de rayonnements présentes dans un établissement; donc d'ouvrir un état qu'il suffira de tenir à jour pour évaluer ultérieurement le potentiel des risques radiologiques dans les installations.

Ce dossier permet également de préparer, sur des bases valables, le plan général d'intervention, notamment en ce qui concerne l'intervention nucléaire.

3.1.2. Fiche individuelle (cf. annexe II)

Cette seconde fiche de sécurité nucléaire vise à définir les risques auxquels est exposé un individu : elle est donc nominative. Ce document peut d'ailleurs être utilisé soit dans un cadre préventif, ce qui est évidemment conseillé lorsque l'on peut prévoir à l'avance l'affectation ou les affectations successives d'un individu, soit sous forme de récapitulation des travaux effectués par celui-ci durant une période antérieure déterminée.

Etablie à titre préventif, cette fiche permet au conseiller technique C.E.A. :

- de classer l'individu en regard de la législation sur la protection des travailleurs contre les rayonnements
- de fixer le choix des dosimètres individuels, de la tenue de travail et des divers équipements individuels
- de faire le point quant à la nature des risques d'exposition auxquels sera soumis l'individu en vue de sa surveillance systématique éventuelle et notamment de sa surveillance médicale.

3.2. Modalités d'action sur le plan médical

L'assistance du C.E.A. en ce domaine se traduit essentiellement par un rôle de conseil.

A cet effet des médecins-conseils C.E.A. se tiennent à la disposition de leurs collègues du service médical universitaire.

Afin d'établir un lien entre les conseillers techniques et le corps médical, nous avons mis au point, en collaboration avec les médecins-conseils du C.E.A., un troisième document (cf. annexe III) intitulé "FICHE DE CONSEILS MEDICAUX" qui est une fiche de sécurité nucléaire individuelle faisant suite à la "FICHE INDIVIDUELLE". Sur la base des renseignements portés par le technicien-conseil C.E.A. sur la fiche individuelle, le médecin de l'établissement universitaire ou, sur la demande de ce dernier, le médecin conseil du C.E.A., est en mesure de définir quelles doivent être la nature des examens que doit subir l'intéressé et la périodicité de ces examens.

Comme on peut le constater ce troisième document établi et exploité par des médecins reste entièrement confidentiel et peut être versé au dossier médical de l'individu avec, en pièce jointe, l'exemplaire de la fiche individuelle adressé au corps médical.

3.3. Modalités d'action sur les plans administratif et financier

3.3.1. Modalités administratives

Sur le plan contractuel nous avons mis au point, en collaboration avec les services administratifs centraux du C.E.A., le texte d'une "CONVENTION D'ASSISTANCE TECHNIQUE EN SECURITE NUCLEAIRE" qui rassemble tous les éléments indispensables à un tel contrat. Ce modèle de convention peut d'ailleurs être utilisé aussi bien avec un établissement industriel ou universitaire qu'avec un établissement relevant du milieu médical mais, dans ce dernier cas, un accord préalable du Ministère de la Santé Publique (S.C.P.R.I.) est nécessaire.

3.3.2. Modalités financières

Depuis 1972 nos prestations sont facturées et leur règlement est effectué par un Service Central de l'administration universitaire qui prend donc à sa charge la sécurité de toutes les unités placées sous sa tutelle.

Cette centralisation confère une grande souplesse à notre action auprès des différentes unités, celles ayant le plus de besoins n'étant pas forcément les mieux financièrement dotées.

Conclusions

Le début de notre action est encore trop récent pour que nous puissions considérer que les modalités proposées seront définitivement retenues.

Compte tenu de notre première expérience en milieu universitaire, nous espérons avoir jeté les bases d'une organisation qui permettra d'attendre avec plus de quiétude d'autres expériences de même nature.

Restant soucieux d'améliorer sans cesse tant la qualité de nos rapports avec l'université que celle de nos services, nous souhaiterions pouvoir profiter de l'expérience de ceux qui se sont, ailleurs dans le monde, penchés sur ce même problème.

EXPOSURES FROM RADIATION SOURCES
OF NATURAL ORIGIN

ИССЛЕДОВАНИЕ И ИСПЫТАНИЕ
РАДИОАКТИВНОСТИ СТРОИТЕЛЬНЫХ МАТЕРИАЛОВ

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Abstract.

Ionizing radiation in dwellings is one of the main sources of radiation effect on the population. Limitation or decrease of this factor may be done by standardization of radioisotope concentration in building materials. Gamma-spectrometric analyses for Ra-226, Th-232 and K-40 in 300 samples of building materials from various regions of the USSR have been performed and predicted values of gamma radiation doses in buildings made of these materials have been calculated. To evaluate the parameters determining air concentration of Rn in dwellings Rn accumulation and mechanism of its release have been studied. For limitation of external gamma radiation levels in dwellings the following values of specific radioisotope permissible concentration have been suggested: 10, 7, 125 pCi/g for Ra-226, Th-232 and K-40 respectively. For limitation of Rn concentration in dwellings permissible level of effective Ra-226 concentration (the product of Ra-226 concentration by emanation factor) has been recommended 0.6 pCi/g.

Резюме

Ионизирующее излучение в жилых помещениях является одним из основных источников радиационного воздействия на население. Ограничение или уменьшение этого фактора возможно путем нормирования содержания радионуклидов в строительных материалах.

Проведены гамма-спектрометрические исследования содержания радия-226, тория-232 и калия-40 в 300 образцах строительных материалов из различных районов СССР и рассчитаны ожидаемые дозы гамма-излучения в помещениях, построенных из этих материалов. Для выявления параметров, определяющих содержание радона в воздухе помещений изучен механизм накопления радона и механизм радоновыделения.

Для ограничения внешнего гамма-излучения в помещениях предложены следующие значения допустимого содержания отдельных радионуклидов: Ra-226 - 10; Th-232 - 7; K-40 - 126 пКи/г. Для ограничения концентрации радона в жилых помещениях рекомендовано допустимое значение эффективной концентрации радия /произведение концентрации радия на коэффициент emanation/, равное 0,6 пКи/г.

Введение

В настоящее время наибольший вклад в облучение населения в целом дают источники ионизирующей радиации естественного происхождения. Величина дозы облучения населения за счет большинства естественных источников, таких как космическое излучение, излучение земных пород, а также калия и некоторых других радиоактивных изотопов, содержащихся в организме человека, практически не может быть изменена. Особое положение в этом отношении занимает излучение строительных материалов, используемых для сооружения жилых, общественных, промышленных зданий и дорожных покрытий. Величина дозы облучения за счет излучения строительных материалов варьирует в довольно широких пределах в зависимости от содержания в них естественных радиоактивных изотопов.

Резко выраженная урбанизация населения, наблюдающаяся во всех странах, приводит к расширению масштабов жилищного строительства. При этом происходит массовое переселение людей из деревянных в кирпичные или бетонные дома, уровни излучения в которых, как правило, выше, чем в деревянных. Кроме того, развитие массового жилищного строительства приводит к необходимости изыскания новых, более дешевых материалов. В качестве таковых все чаще используются отходы горнорудной, металлургической и химической промышленности. Эти отходы могут содержать радиоактивные вещества в больших количествах, чем традиционно используемые строительные материалы². В связи с этим возникает необходимость в ограничении использования таких материалов из-за возможности существенного повышения уровня облучения населения.

Ограничение или уменьшение уровня облучения населения за счет излучения строительных материалов возможно путем нормирования их радиоактивности, то есть путем отказа от использования материалов с высоким содержанием радиоактивных веществ. Для обоснования таких нормативов необходимо достаточно подробно изучить существующие уровни радиационного воздействия на население излучения строительных материалов, их зависимость от вида используемых материалов и т.д.

Гамма-излучение в жилых помещениях

В различных странах /Швеции, Англии, СССР, США, Австрии, Японии, Болгарии, ФРГ, ГДР/ проведены многочисленные измерения дозы гамма-излучения в помещениях. Значения гамма-дозы в помещениях из различных строительных материалов и на открытой местности согласно этим измерениям представлены в таблице I^{1, 3-6}.

Таблица I

Дозы гамма-излучения внутри и вне помещений
за вычетом космического фона /мрад/год/

Тип строительного материала	Внутри помещения			Вне помещения			Разность средних доз
	мин.	макс.	сред.	мин.	макс.	сред.	
Дерево	29	100	56	22	119	56	0
Кирпич	26	346	87	21	308	78	9
Бетон	48	202	115	22	120	70	45
Гранит	75	300	125	18	118	74	51

Из таблицы видно, что доза гамма-излучения в помещениях выше дозы на открытой местности /за исключением деревянных домов/.

Стены и перекрытия помещений являются источниками излучения и экранами от космического излучения и излучения земных пород. Экранирование космических лучей перекрытиями зданий невелико. После прохождения I, 2 и 3 перекрытия космическое излучение ослабляется на 13, 18 и 21 процент соответственно. Степень экранирования излучения земных пород зависит от толщины стен помещения. Для излучения естественных радиоактивных изотопов толщина слоя насыщения составляет 100 г/см², при этом слой 50 г/см² обеспечивает 80% насыщения. При толщинах стен и перекрытий, близких к слою насыщения и характерных для современного жилищного строительства, излучение земных пород полностью экранируется. В этом случае мощность дозы в помещении определяется концентрацией радиоактивных веществ в используемых строительных материалах. Для определения коэффициентов пропорциональности между этими величинами можно воспользоваться значениями, рассчитанными для полости в бесконечном пространстве. Эти значения вычислялись рядом авторов. И хотя ими применялись различные методы расчета, полученные коэффициенты оказались весьма близкими. Используя коэффициенты, приведенные в работе для максимально возможной мощности дозы в помещении, построено целиком из данного материала /за вычетом космического фона/ получаем:

$$D_{\text{мкр/час}} = 4,7 C_{Ra} + 6,7 C_{Th} + 0,37 C_K \quad /I/$$

где C_{Ra} , C_{Th} и C_K — удельные активности радия-226, тория-232 и калия-40 в строительном материале в пКи/г при наличии радиоактивного равновесия в рядах урана и тория.

В реальных помещениях мощность дозы может оказаться несколько меньше значения, полученного по формуле /I/ как за счет наличия окон и дверей, так и в ряде случаев недостаточной толщины стен и перекрытий. Кроме того, для сооружения стен и перекрытий могут использоваться разные строительные материалы. В этих случаях мощность дозы в помещении будет иметь промежуточные значения между результатами расчета для отчетных материалов.

Для определения диапазона изменений концентрации радиоактивных веществ в строительных материалах нами, с помощью низкоэнергетического гамма-спектрометра, было исследовано около 300 образцов различных строительных материалов из разных районов СССР. Результаты измерений представлены в таблице II. В ней также приведены величины мощности дозы в помещении, рассчитанные по формуле /I/. Величина годовой дозы, обусловленная излучением строительных материалов в предположении 18-часового пребывания людей в помещении и с учетом коэффициента экранирования гонад, равного 0,63, может быть вычислена по формуле:

$$D_{\text{/град/год/}} = 18,5 C_{Ra} + 26,7 C_{Th} + 1,47 C_K \quad /2/$$

При обосновании нормативов радиоактивности строительных материалов мы исходили из того, что за счет проживания в домах, построенных из материалов с повышенным содержанием радиоактивных изотопов допустимо дополнительное облучение гонад, равное 100 мрад/год, по сравнению с облучением в домах со средним содержанием радиоактивных веществ. 100 мрад/год это средняя доза облучения гонад за счет излучения естественных источников. Поэтому такое нормирование ограничивает годовую дозу людей, проживающих в домах, построенных из строительных материалов с повышенным содержанием радиоактивных веществ, величиной утроенного естественного фона. Годовая доза в домах из обычных материалов может быть подсчитана по формуле /2/ для концентрации радиоактивных изотопов, равной кларковой $C_{Ra} = 0,9$; $C_{Th} = 1,41$; $C_K = 21,5$ пКи/г. Она со-

Таблица II

Содержание естественных радионуклидов в строительных материалах
/пКи/г/ и мощность дозы в помещении /мкр/час/

Тип материала	Число проб	Торий-232			Радий-226			Калий-40			Мощность дозы в помещении			Суммарное содержание изотопов /в долях ЦДК/		
		мин.	мак.	ср.	мин.	мак.	ср.	мин.	мак.	ср.	мин.	мак.	ср.	мин.	мак.	ср.
Красный кирпич	55	0,66	1,5	1,0	1,1	1,6	1,5	11	25	20	13	25	20	0,27	0,55	0,42
Бетон тяжелый	87	0,4	3,7	0,8	0,6	2,9	0,9	7	24	15	8	45	14	0,16	0,95	0,31
Бетон легкий	16	0,6	2,1	0,9	1,4	3,3	2,0	5	26	14	12	37	19	0,26	0,8	0,41
Гранит	2	-	-	4,5	-	-	3	-	-	40	-	-	56	-	-	1,20
Т у ф	13	1,1	2,9	2,0	2,1	2,6	2,6	26	34	18	24	41	30	0,52	0,88	0,69
Песок природный	18	0,4	0,8	0,5	0,4	1,0	0,5	2	17	8	4	15	8	0,09	0,33	0,17
Песок хвостовой	14	-	-	0,5	-	-	0,8	-	-	6	-	-	8	-	-	0,19
Цемент	7	0,4	0,5	0,4	0,4	3,2	1,2	3	11	6	5	22	10	0,12	0,46	0,22
Шлаки доменные	29	0,3	1,2	0,6	1,0	3,4	1,8	0,9	13	13	7	28	16	0,14	0,59	0,34
Шлаки фосфорные	15	-	-	0,6	-	-	0,6	-	-	4	-	-	33	-	-	0,72
Щебень	8	0,4	5,0	2,1	0,4	0,4	0,4	6	8	7	6	38	18	0,14	0,81	0,38
Облицовочные материалы	35	0,4	4,7	2,3	0,4	3,4	1,9	22	47	39	11	61	36	0,24	1,3	0,76

стальмет 33 мрад/год. Увеличение годовой дозы за счет промывания в домах, построенных из материалов с повышенным содержанием радиоактивных веществ, определяется по формуле:

$$D \text{ /мрад/год/} = 18,5 C_{Ra} + 26,7 C_{Th} + 1,47 C_{K} - 86 \text{ /3/}$$

 ПДК радиоактивных изотопов в строительных материалах могут быть рассчитаны по формуле /3/ при условии, что доза дополнительного облучения не должна превышать 100 мрад/год. Они составляют:
 $C_{Ra} = 10$; $C_{Th} = 7,0$; $C_{K} = 126$ пКи/г. При наличии в строительном материале этих изотопов должно выполняться условие:

$$\frac{C_{Ra}}{10} + \frac{C_{Th}}{7,0} + \frac{C_{K}}{126} \leq 1 \quad /4/$$

В таблице II приведены суммарные концентрации радиоактивных веществ в исследованных материалах /в долях ПДК/. Из таблицы видно, что превышение допустимой концентрации обнаружено только у гранита и некоторых отделочных материалов.

Таким образом, предлагаемые величины допустимого содержания радиоактивных веществ не приведут к ограничению применения подавляющего большинства традиционных строительных материалов. Ограничения распространяются на относительно небольшое число материалов с повышенным содержанием радиоактивных веществ. Такие материалы могут применяться при их разбавлении слабо радиоактивными компонентами, а также для сооружения дорог, плотин, времяпрепровождение людей вблизи которых ограничено.

Радон в воздухе жилых помещений

Наличие радиоактивных веществ в строительных материалах, помимо дополнительного внешнего облучения приводит к повышению радиоактивности воздуха в помещении по сравнению с атмосферным. Параметрами, определяющими степень радиационного воздействия радиоактивности воздуха, являются концентрации радона, торона и их продуктов распада. В литературе неоднократно отмечались случаи довольно высоких концентраций радона в помещениях, особенно в домах из материалов, изготовленных из отходов горно-рудной промышленности /вплоть до 15 пКи/л/ ^{11, 12}. Наличие таких случаев свидетельствует о необходимости контроля за радиоактивностью воздуха в помещениях. Однако концентрация эманаций и их короткоживущих продуктов распада неудобны в качестве параметров для контроля, поскольку они связаны со степенью воздухообмена, которая варьирует в довольно широких пределах в зависимости от метеосудовий, длительности проветривания помещений и т.д.

Для выявления более удобного для контроля параметра достаточно точно характеризующего радиоактивность воздуха в помещении, нами был рассмотрен механизм накопления радона в помещении. Решение соответствующего дифференциального уравнения дает выражение для изменения во времени концентрации радона в воздухе помещения:

$$C = \frac{QS + V k C_{атм}}{\lambda + k} [1 - e^{-(\lambda + k)t}] + C_0 e^{-(\lambda + k)t} \quad /5/$$

где: C_0 — концентрация радона в момент $t = 0$ /Ки/м³/
 Q — удельное радоновыделение ограждений /Ки/м²сек/
 S — площадь ограждений помещения /м²/
 V — объем помещения /м³/
 λ — постоянная распада радона /2,1 · 10⁻⁶ сек⁻¹/
 k — кратность воздухообмена в помещении /сек⁻¹/

* х/ предельно допустимые концентрации

$C_{атм}$ — концентрации радона в атмосферном воздухе /Кл/м³/

Из формулы /5/ следует, что установившиеся равновесные концентрации радона в помещении отрезается величиной $\lambda + K$. При кратности воздухообмена, равной одному объему в час $K = 2,8 \cdot 10^{-4}$, равновесная концентрация достигается уже через несколько часов. Выражение для равновесной концентрации радона с учетом того, что $\lambda \ll K$, записывается в виде:

$$C_{равн.} = \frac{Q}{K} \frac{S}{V} + C_{атм} \quad /6/$$

Из формулы видно, что концентрация радона в помещении всегда выше концентрации в наружном воздухе. Разность этих концентраций пропорциональна удельному радоновыделению огражденной помещения, отношению S/V и обратно пропорциональна кратности воздухообмена. Отношение S/V для обычных помещений не сильно варьирует. Его значение для типичных жилищ дано в таблице III, из которой следует, что среднее значение этого отношения составляет величину 1,5.

Таблица III

Отношение площади ограждений к объему помещения / S/V /

высота M \ S /M ² /	15 = 4 x 3,75	20 = 5 x 4	30 = 5x6	40=6x6,7
2,5	1,82	1,70	1,53	1,43
3,0	1,70	1,57	1,40	1,30
3,5	1,60	1,47	1,30	1,20

Формула /6/ позволяет оценить поступление радона из почвы под зданием. В работе [13] приводятся величины удельного радоновыделения почв: среднее значение $4,5 \cdot 10^{-13}$, диапазон изменений от $2 \cdot 10^{-15}$ до $1,7 \cdot 10^{-12}$ Кл/сек.м². Расчетное значение концентрации радона в помещении размером 5 x 4 и высотой 3 м, в предположении, что почвенный радон полностью поступает в помещение, а радоновыделение ограждений отсутствует и концентрация эманации в атмосферном воздухе равна 0,1 пКи/л, приведено в таблице IV.

Таблица IV

Расчетные значения концентрации радона в помещении / пКи/л / при поступлении радона из почвы

Q /Кл/м ² сек/ \ K /час ⁻¹ /	$1,7 \cdot 10^{-12}$	$4,5 \cdot 10^{-13}$	$2 \cdot 10^{-15}$
0,1	20,5	5,5	0,12
1	2,1	0,64	0,10
10	0,2	0,15	0,10

Из таблицы видно, что в помещениях с плохой проветриваемостью поступления радона из почвы может играть существенную роль. В современном многоэтажном строительстве используются бетонные перекрытия, которые практически полностью предотвращают поступление в жилища помещений почвенного радона. В таких домах основным источ-

ником радона является радоноразделение стен и перекрытий. Удельное радоноразделение ограничено является более удобным параметром, поскольку его величина, в отличие от концентрации радона, практически не зависит от воздухообмена в помещении. Это было установлено в результате рассмотрения механизма радоноразделения. Процесс радоноразделения можно разделить на два этапа: эмансирование радона во внутренние поры материала /доля атомов радона, выходящих во внутренние поры, называется коэффициентом эмансирования/ и диффузию радона по этим порам с выходом из материала. Такое разделение оправдано тем, что внутри зерен минерала диффузия протекает крайне медленно /коэффициент диффузии имеет порядок 10^{-22} см²/сек [7]. Поэтому распространяются по материалу только те атомы радона, которые вышли за счет отдачи при альфа-распаде радия во внутренние поры материала. Процесс распространения радона по порам материала описывается уравнением диффузии. При рассмотрении радоноразделения стен можно считать, что диффузионный перенос радона осуществляется в направлении, перпендикулярном к поверхности стен /по координате x /. Поток радона, параллельные поверхности стены, взаимно уравновешивают друг друга, так как высота и ширина стены значительно больше ее толщины. В этом случае уравнение диффузии может быть записано в виде:

$$\frac{\partial C}{\partial t} = \lambda C_0 - \lambda C + \frac{b}{\delta} \frac{\partial^2 C}{\partial x^2} \quad /7/$$

где

$$C_0 = \frac{C_{\text{ра}} \rho b}{\delta} \quad /8/$$

- C_0 - максимально возможная концентрация радона /Ки/см³/,
- $C_{\text{ра}}$ - концентрация радия в материале /Ки/г/,
- ρ - плотность материала /г/см³/,
- b - коэффициент эмансирования /отн.ед./,
- δ - пористость материала /отн.ед./,
- δ - коэффициент диффузии /см²/сек/

За начало координат принята половина глубины стены. В силу симметрии задачи поток радона при $x=0$ равен 0. Отсюда следует первое граничное условие

$$b \frac{\partial C}{\partial x} \Big|_{x=0} = 0 \quad /9/$$

Второе граничное условие можно сформулировать, исходя из баланса активности вне стены. Будем считать, что диффузия радона из стены толщиной $2d$ происходит в ограниченный внешний объем глубиной l . Для простоты можно принять, что на выходе из стены происходит мгновенное выравнивание концентрации радона. При этом второе граничное условие может быть записано в виде:

$$-b \frac{\partial C}{\partial x} \Big|_{x=d} - l \lambda C \Big|_{x=d} = l \frac{\partial C}{\partial t} \Big|_{x=d} /10/$$

Решение уравнения /7/ с граничными условиями /9/ и /10/ для стационарного случая / $\partial C / \partial t = 0$ / имеет вид:

$$C_x = C_0 \left[1 - \frac{\text{ch} \left(\frac{x}{l_0} \right)}{\text{ch} \beta + \frac{l}{2\delta} \text{sh} \beta} \right] \quad /11/$$

где

- $l_0 = \sqrt{\frac{b}{\lambda \delta}}$ - длина диффузии,
- $L = l / d \delta$ - отношение внешнего к внутреннему объему воздуха
- $\beta = d / l_0$ - отношение половины толщины стены к длине диффузии.

Концентрация радона во внешнем воздухе $C \Big|_{x=d}$ выражается

$$C|x=d = C_0 \left[\frac{1 + \frac{th\beta}{\beta}}{1 + \frac{1}{\alpha} \frac{th\beta}{\beta}} \right] \quad /12/$$

Удельное радоновыделение равняется:

$$Q = \beta \frac{\partial C}{\partial x} \Big|_{x=d} = Q_0 \frac{th\beta}{\beta} \left[\frac{1}{1 + \frac{1}{\alpha} \frac{th\beta}{\beta}} \right] \quad /13/$$

где Q_0 - максимально возможное радоновыделение, то есть радоновыделение в случае, когда весь радон, попавший во внутренние поры, выходит наружу.

Оно равно:

$$Q_0 = C_{\text{ра}} \beta \lambda d \rho \quad /14/$$

Для жилых помещений были рассмотрены два случая:

1. Отношение объема к объему внутристенного воздуха $1/\alpha$ велико. В этом случае

$$\frac{C(d)}{C_0} \Big|_{d \rightarrow \infty} \rightarrow 0; \quad Q \Big|_{d \rightarrow \infty} \rightarrow Q_0 \frac{th\beta}{\beta} \quad /15/$$

Это приближение справедливо для случая узкой щели радона в сильно вентилируемом помещении, поскольку при этом эффективный внешний объем значительно превосходит объем помещения.

2. Для невентилируемого помещения внешний объем равен объему помещения и величина $1/\alpha$ в этом случае равна:

При $S/V = 1,5 \text{ м}^{-1}$, $d = 0,25 \text{ м}$, $\delta = 0,4$; величина $\alpha = 6,7 \frac{th\beta}{\beta}$ всегда меньше 1. Принимая $th\beta/\beta = 1$, получим: $Q = 0,37 Q_0$, $C|x=d = 0,13 C_0$. Если $th\beta/\beta < 1$, то Q будет еще меньше отличаться от Q_0 .

Таким образом, для помещений обычных размеров величина удельного радоновыделения практически не зависит от кратности воздухообмена, то есть этот параметр является удобным для контроля. Однако измерение радоновыделения возможно проводить либо в уже построенных помещениях, либо на макетах ограждений. Это создает определенные трудности при радиационно-гигиенической оценке новых строительных материалов. В последнем случае желательно уметь определять его величину путем исследования небольших образцов материала.

Из выражений /14/ и /15/ следует, что удельное радоновыделение пропорционально произведению концентрации радия в строительном материале на коэффициент эманирования, которое назовем эффективной концентрацией радия:

$$C_{\text{ра эфф}} = C_{\text{ра}} \beta \quad /17/$$

Кроме того величина радоновыделения зависит от β - отношения половины толщины стены к длине щели (узкой функции $th\beta/\beta$ при $\beta \leq 1$ не сильно отличается от единицы, а при $\beta \geq 2$ имеет вид $\sim 1/\beta$). Таким образом, если длина щели (узкой функции радона в материале ограждения больше половины толщины ограждения, то радоновыделение можно оценивать по его максимальной величине Q_0 . Следует отметить, что мы рассматривали чисто диффузное приближение. Наличие же подпора ветра, перепад температур между комнатным и наружным воздухом, могут привести к появлению конвекционных потоков, т.е. к уменьшению эффективного значения β . При этом радоновыделение будет возрастать, но в любом случае оно не может превысить величину Q_0 . Следовательно, в реальных условиях возможны измене-

ния удельного радиовыделения в пределах от $Q_0 \text{ th } \beta/\beta_0$ до Q_0 . Если β невелико, то эти изменения будут незначительны. Результаты экспериментального определения длины диффузии радона в строительных материалах и характерные для этих материалов величины β приведены в таблице У.

Таблица У

Длина диффузии радона в строительных материалах

Вид материала	Длина диффузии l_0 /см/	Характерная толщина ограждения $2d$ /см/	$\beta = \frac{d}{l_0}$
Бетон тяжелый	15; 10; 13;	10	0,4
Бетон легкий	29; 28; 22	12 ÷ 35	0,2 - 0,7
Красный кирпич	15	50	1,7

Принимая во внимание данные таблицы У можно заключить, что для большинства ограждений радиовыделение будет определяться максимально возможной величиной, которая является функцией только эффективной концентрации радия в строительном материале. Эффективная концентрация радия является параметром, удобным для контроля за радиоактивностью воздуха в помещении, поскольку ее можно определить путем исследования небольших образцов материала.

Эманационным методом /при измерении до нескольких мм/ нами измерена эффективная концентрация радия в нескольких десятках образцов различных строительных материалов и рассчитаны равновесные концентрации радона в помещениях из данных материалов. Расчет концентрации радона проводился для постоянного воздухообмена, равного 1/час, $S/V = 1,5$. Толщина ограждения и плотность для тяжелого и легкого бетонов принимались соответственно равными $2d = 10$ и 20 см; $\rho = 2,1$ и $1,6$ г/см³. Для всех остальных материалов принималось $2d = 50$ см; $\rho = 1,5$ г/см³. Радиовыделение считалось максимально возможным, концентрация радона в атмосферном воздухе не учитывалась. В таблице VI представлены также значения коэффициентов эманации, рассчитанные с привлечением результатов гамма-спектрометрического определения концентрации радия в данных образцах.

Таблица VI

Эффективная концентрация радия в строительных материалах и расчетное значение концентрации радона в помещениях

Тип материала	Эффект. концентрация радия $C_{Ra \text{эфф}} \times 10^{-2}$ /пКи/г/			Коэффициент эманации ϵ %/			Концентрация радона в помещении C_{Rn} /пКи/г/		
	мин.	макс.	ср.	мин.	макс.	ср.	мин.	макс.	ср.
Бетон тяжелый	0,8	12	4,1	0,4	10,0	3,3	0,02	0,29	0,10
Бетон легкий	2,0	13,2	5,4	0,4	10,0	2,1	0,7	0,4	0,2
Красный кирпич	0,8	5,1	2,1	0,4	5,7	1,7	0,03	0,220	0,09
Кирпич силикатный	2,4	6,2	3,8	4,5	8,0	6,7	0,10	0,27	0,16
Туф	3,5	10,2	6,5	1,8	6,0	3,6	0,15	0,43	0,28
Кирпич пумици-товый	-	-	6,1	-	-	5,8	-	-	0,26
Пумицит	-	-	28,0	-	-	9,0	-	-	1,2

Допустимую величину эффективной концентрации радия можно установить, исходя из условия, чтобы при постоянном воздухообмене, равном 1 час⁻¹, не было превышения СДК^X короткоживущих продуктов распада радона /С_T/ для населения /1.10⁻⁵ Ки/м³ /5/. Принимая, что радоновыделение ограждений определяется максимально возможной величиной /время пребывания в помещении 18 часов в сутки/ и пренебрегая величиной С_{атм}, выражение для С_{раэфф} можно записать в виде:

$$C_{Raэфф} /Ки/г/ = 10^{-6} \frac{C_{Ra} \cdot K}{0.75 \alpha \rho \lambda \cdot S/V} \quad /18/$$

При воздухообмене 1 час⁻¹ концентрация короткоживущих продуктов распада в помещении составляет 0,53 С_{РН}. С учетом этого:

$$C_{Raэфф} /Ки/г/ = 10^{-6} \frac{C_1 \cdot K}{0.4 d \rho \lambda \cdot S/V} \quad /19/$$

Значение допустимой величины С_{раэфф} можно получить из формулы /19/ принимая α = 0,25 м; ρ = 1,5 т/м³; S/V = 1,5 м⁻¹

$$C_{Raэфф} = 0.6/n Ки/г/ \quad /20/$$

Как видно из таблицы У1 среди исследованных материалов не оказалось ни одного, имеющего эффективную концентрацию радия, близкую к допустимой величине. Сопоставление предлагаемых нормативов концентрации радия, определяющей внешнее облучение людей, и эффективной концентрации радия, определяющей радиоактивность воздуха в помещении, показывает, что первый норматив является, как правило, более жестким. Нормирование по эффективной концентрации радия может быть лимитирующим фактором только для материалов, радиоактивность которого обусловлена, в основном, радием и обладающих повышенным эманированием /ε > 6%/. Наблюдающиеся рядом авторов концентрации радона в помещении, превышающие 1 пКи/л, во многих случаях могут быть связаны с тем, что измерения производились при пониженном воздухообмене. Кратность воздухообмена, как правило, при этом не измерялась. Величина же допустимой эффективной концентрации радия рекомендована, исходя из среднегодовой величины кратности воздухообмена, и по этой причине ограничивает только среднегодовую концентрацию радона в помещении.

Исходя из меньшей значимости радиоактивности воздуха по сравнению с внешним облучением, контроль за эффективной концентрацией радия целесообразно проводить только для материалов с повышенным содержанием радия /более 5 пКи/г/. Следует иметь в виду, что материалы с эффективной концентрацией радия, превышающей допустимую, можно применять при условии использования противорадоновых покрытий. Наши исследования показали, что двухкратное покрытие стен масляной краской снижает радоновыделение на порядок. Кроме того концентрацию радона в помещении можно уменьшить путем увеличения воздухообмена.

С целью проверки результатов теоретического рассмотрения механизмов накопления в помещении и радоновыделения, точности методов измерения параметров, характеризующих эти процессы, а также соотношения между дозой внешнего гамма-излучения в помещении и концентрацией радиоизотопов в строительных материалах нами проведены исследования в модельном помещении. Помещение было построено из материалов, содержание радиоактивных веществ в которых, коэффициенты эманирования и другие параметры были предварительно исследованы. В помещении

X/ средняя допустимая концентрация

был обеспечен регулируемый воздухообмен. Рассчитанные и измеренные значения удельного радоновыделения различных ограждений представлены в таблице УП.

Таблица УП

Радоновыделение ограждений экспериментального помещения

Тип ограждения	Удельное радоновыделение /Ки/м ² сек/10 ⁻¹⁴ .	
	Расчетные значения	Измеренное значение
Внешняя стена	1,8	2,0 ± 0,1
Внутренняя стена	1,1	1,14 ± 0,06
Пол	0,7	1,20 ± 0,07
Потолок	0,9	0,80 ± 0,08

Из таблицы видно, что наблюдается хорошее согласие расчетных и экспериментальных значений удельного радоновыделения для всех ограждений, кроме пола, для которого измеренное значение в 1,5 раза выше расчетного. Это расхождение обусловлено частичным проникновением радона из подвального помещения.

Расчетные и измеренные значения концентрации короткоживущих продуктов распада радона при различных кратностях воздухообмена в экспериментальном помещении даны в таблице УШ, из которой видно хорошее согласие между этими величинами.

Таблица УШ

Концентрация продуктов распада радона в модельном помещении /пКи/л/

Воздухообмен К /час ⁻¹ /	К о н ц е н т р а ц и я	
	Расчетная	Измеренная
1	0,08	0,10
1,8	0,021	0,017

Расчет максимально возможной мощности дозы в модельном помещении дал величину 19,8 мкр/час. Для сопоставления этого значения с измеренной дозой необходимо внести 2 поправки: на наличие окон и дверей и на отсутствие слоя насыщения по гамма-излучению /толщина ограждений составляла 20 г/см²/. Величины поправок составляют 0,87 и 0,65, соответственно. С учетом их расчетная мощность дозы гамма-излучения в помещении составила 11 мкр/час. Измеренная величина равна 10 мкр/час. Таким образом, наблюдается вполне удовлетворительное согласие.

Заключение

Нормативы радиоактивности строительных материалов, разработанные на основе проведенных исследований, предназначены для ограничения радиационного воздействия на население за счет этого фактора. Предложенные нормативы не ограничивают использование в жилищном строительстве основной массы традиционных материалов. Строительные материалы, содержание радиоактивных изотопов в которых превышает нормативные величины для жилищного строительства, могут использо-

ваться для других целей. Способ ограничения радиационного воздействия на население при таком использовании указанных материалов требуют дальнейших исследований.

Широкое внедрение нормативов радиоактивности строительных материалов требует разработки оперативных и экспертных методов контроля. В качестве оперативного метода контроля целесообразно использовать измерение мощности дозы в массивах строительных материалов /карьеры, склады, отвалы и пр./ Из сопоставления формул /2/ и /4/ следует, что предложенные нормативы соответствуют мощности дозы в 4-й геометрии 47 мкр/час. Для экспертного контроля перспективно использование высокочувствительных гамма-спектрометров, а при обнаружении содержания радия-226 более 5 пКи/т — измерение эмаширования образцов

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EYE EXPOSURE FROM THORIATED OPTICAL GLASS

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Abstract

During a routine radiation survey of equipment with some optical components the Health Physicist was surprised to find a large reading on an ionization survey instrument. Significant radiation was also observed on a beta-gamma instrument. The source of the radiation was identified as thorium in high quality optical glass. Similar equipment was examined, but not all items produce readings on the survey instruments.

Thorium is added to the glass, in amounts up to 30% by weight, to provide improved optical properties. Similar results may be obtained by using other heavy elements. Thorium is carried as an impurity with some of these elements and the thorium concentration may be greater than 0.05% by weight.

Since the glass is used in an eyepiece, the amount of exposure to the eye should be investigated. The decay chain of thorium contains alpha, beta, and gamma components. Both the alpha and beta components are largely attenuated by the glass. The alpha particles that reach the eye will be absorbed in a thin surface layer, less than 100 microns, the beta components extend over a larger distance, and the gammas will produce almost constant exposure over the entire eye.

The beta-gamma exposure rate was determined by thermoluminescent dosimetry (TLD). The measured exposure rate was 1 mRem/hr averaged for 123 hours exposure at the surface of a lens which contained 18% thorium by weight.

A computer model for the emission of alphas from the glass, the absorption in air and the final absorption in surface layers of the eye provides a technique for examining the amount of exposure at different depths below the surface of the eye. The results of the model will be compared with experimental results from alpha spectroscopy.

Although the alpha particles come from only a small surface layer of the glass, the absorbed dose rate at the surface of the eye may be 50 to 1000 times greater from alpha than from beta-gamma radiation. The size of the lens, and the eye to lens distance determines the number and energy distribution of the alpha particles reaching the eye.

To reduce exposure to the eye, thin non-thoriated glass shields were inserted between the lens and the eye. The results of TLD and alpha counting with and without the shielding showed a complete removal of the alphas.

Introduction

Our introduction to thoriated glass came as a request to survey some electro-optical equipment with high internal voltages. There was no anticipation of external radiation, but the survey was conducted and 4-5mR of radiation was detected on an ionization type of survey instrument. The eyepiece of the equipment was removed to eliminate some shielding and to bring the survey instrument closer to the expected source of the radiation. However, the radiation level decreased. This process of elimination led to the optical glass as the source of the radiation. Significant radiation was also observed with beta-gamma instrumentation. Identification of the thorium as the source was accomplished by gamma ray spectroscopy.

After the thoriated glass was identified, similar electro-optical equipment was surveyed. Some of the equipment gave positive readings and others were within the natural background. The specifications of the optics were checked, but there were no requirements for thorium.

Thorium is added to glass, up to 30% by weight, to provide improved optical properties. Specifically, glasses with an index of refraction greater than 1.65 and with the product of Abbe constant and index of refraction greater than 70 are often made with thorium. Other heavy elements may be used to obtain similar results. When some of the lanthanide compounds are used, thorium is often contained as an impurity and the thorium concentration may exceed 0.05% by weight.

The use of thorium in optical glass raises few problems unless the glass is an eyepiece. When thorium is in the eyepiece, the eye is exposed to all of the radiation generated by the thorium decay chain - Alpha, betas and gammas - and the associated bremsstrahlung radiation. Initially we considered only the beta-gamma component.

The alpha exposure presented a special problem. The high concentration of thorium gave a high flux of alpha particles. Harvey¹ raised questions on the external radiation hazards of alpha particles. He evaluated plane sources of alpha activity on the skin surface. Witten and Sulzberger² investigated the mode of action of thorium on human skin. They found that the thorium was carried into the epidermis. In contrast to the broad surface contamination and the penetration effects, Dean and Langham³

concentrated on the exposure of the skin to particles of high specific activities. None of these studies provided direct data for evaluating eye exposure from externally originating alpha particles.

In the present study we investigate the exposure of the eye's surface from alpha particles emitted by small quantities of thorium uniformly distributed in glass. A model of the emission and absorption processes of alpha radiation will be developed and the exposure at various layers of the eye's surface will be calculated. Comparison of the theoretical values and some experimental data will be included.

Thorium Concentrations

Glass manufacturers publish catalogs of glasses. Some of the concentrations approach 30% by weight. The performance characteristics of the optical elements that were in use did not require the thoriated glass. However, it was recognized that some thorium may be included in trace quantities. The uncertainties associated with thorium in glass have led us to set a preliminary level of 0.05% thorium by weight. A survey meter identifies glass with large thorium concentrations, but is ineffective in identification of trace quantities. Gamma ray analysis was not effective on highly thoriated glass to yield the correct percent of thorium. This reflects the lack of secular equilibrium. For trace concentrations, the counting for gamma analysis becomes prohibitive. X-ray fluorescence provides a technique for obtaining the thorium concentration. To be accurate, thorium standards in glass matrices similar to the unknown are required.

Beta-Gamma Components

It was recognized very early that the radiation levels observed with the ionization chamber contained a contribution from the alpha particles. This was most easily observed by placing a thin sheet of paper between the glass and the gauge and noting the reduced instrument readings. In an attempt to obtain a better measure of the beta-gamma exposure rates, thermoluminescent dosimeters (TLD) were placed on two lenses. Total exposure time was 24 hours. These initial measurements included some alpha excitation of the phosphors. A second set of measurements was made with a borosilicate flat glass 2.8mm thick. The exposure time was 123 hours. The results of these measurements along with the alpha count rates are shown in Table 1. The radiation that can be assigned to beta-gamma is about 1mR/hr.

Table 1. Glass shielding effects
Lens area 6.77cm²

Lens	Th Concentration	TLD (mR/hr)		Alpha (counts/min)	
		No Shield	Shield	No Shield	Shield
82291	18.1	5.71	0.98	1700	0
86200	18.4	6.45	1.39	2100	0

These lenses have a mass of 27.2 grams. The gamma radiation will be proportional to the mass and thorium concentration. The beta component will be absorbed by the glass. Even the most energetic beta will not penetrate more than 2.5mm of the glass.

Alpha Radiation

The major portion of the survey instrument reading may be explained by alpha radiation. However, the relationship to absorbed dose does not follow from the instrumental reading. In this section, we will derive the number of alpha particles leaving the glass surface, identify the number reaching various depths within the eye, and the associated absorbed dose rates at these levels. Lens to eye distances, and thorium concentration strongly influence these results.

Range of Alpha.

The range-energy relationships for glass and tissue were calculated for specific energies by a computer program using the procedures outlined by Neufeld, et al.⁴ The program was checked by comparison with the tables for proton ranges in soft tissue given in the reference. The alpha range in soft tissue was compared with Walsh's⁵ results and demonstrates excellent agreement.

The range of alphas in glass is dependent on the glass composition. The chemical composition of a lanthanum glass containing a trace quantity of thorium is shown in Table 2. Small changes in the thorium concentration will have negligible effects on the range of the alpha radiation in the glass.

Table 2. Glass Chemical Composition

Density = 3.64 g/cm²

Element	Percent	Atoms/cm ²	Atomic No	Atomic Wt
La	35	5.51x10 ²¹	57	138.9
CA	35.7	1.95x10 ²²	20	40.08
O	23.2	3.17x10 ²²	8	16
As	3.8	1.11x10 ²¹	33	74.9
Zr	2.2	5.29x10 ²⁰	40	91.2
Th	0.1	9.45x10 ¹⁸	90	232.0

Even large interchanges between the lanthanum and thorium in percent by weight will have only small changes in the range provided the density remains constant. When the density changes, the range may be calculated as follows:

$$R_N = R \left(\frac{3.64}{P_N} \right)$$

where P_N is the new density and R_N is the new range.

The values for the range-energy for glass, air and tissue are given in Table 3. These values will be used in the subsequent calculations.

Table 3. Alpha - Range-Energy

Energy (MEV)	Glass (μm)	Range Air (cm)	Tissue (μm)
1	.25	.5	4.2
2	1.75	1.0	9.8
3	5.57	1.625	16.4
4	10.25	2.42	25.1
5	15.25	3.5	35.5
6	20.75	4.64	47.2
7	27.25	5.95	61.1
8	34.25	7.34	75.5
9	41.75	8.04	91.75

Alphas emitted from glass.

For the calculations that follow, the lens is considered as a flat circular glass disc with trace quantities of thorium uniformly distributed throughout. Thorium - 232 is taken in secular equilibrium with all of its daughter products. Although gamma analysis raised doubts that equilibrium exists, the use of this assumption will produce a maximum alpha emission.

Although the total number of alpha particles that leave the surface of the glass is important, we will calculate only those that are directed to an element of the eye's surface. Three distances then become important: X_a , the shortest distance between the eye and the lens; X_g , the distance the alpha travels in the glass; and the X_t , the distance in tissue below the surface of the eye.

Let $N(E)$ be the number of alpha particles emitted per cm^3 with an energy E . The number, dN_c , from an element of volume and directed toward the selected surface element of the eye, dA , is given by:

$$dN_c = \frac{N(E)dE(2\pi(X_t+X_a)\sin\theta/\cos\theta)((X_t+X_a)/\cos\theta)d\theta)\cos\theta dA\cos\theta dX_g}{4\pi((X_t+X_a)/\cos\theta)^2}$$

$$= (N(E)/2)(\sin\theta\cos^2\theta d\theta)dAdX_gdE$$

where θ is the angle between the eye to glass normal and the eye to volume element. This equation may be integrated over the volume of the lens and over the energies to give the total number emitted directed toward the surface element of the eye. All of the alphas emitted will not reach the eye. Absorption of energy begins in the glass, with no alphas originating at depths greater than $40\mu\text{m}$ every reaching the surface. Additional absorption

occurs in the air and finally in the surface layers of the eye. The energy distribution of the initial alphas is known, but as absorption occurs, the energy spectrum changes.

A computer program was written to perform the integrations and calculate the rate and energies of alphas reaching various layers of the eye from different sizes of lenses. This program also calculates the absorbed dose rates at each of the layers.

Results

Table 4 summarizes the output for the model of a lens 3cm in diameter containing 0.005% thorium.

Table 4. Eye Exposures from 3.0cm
Diameter Lens with 0.005% Thorium

Eye Penetration μm	0.1cm Eye to Lens		3.0cm Eye to Lens	
	Alpha Count $\text{cm}^{-2}\text{hr}^{-1}$	Absorbed dose rate $\mu\text{ rad hr}^{-1}$	Alpha Count $\text{cm}^{-2}\text{hr}^{-1}$	Absorbed dose rate $\mu\text{ rad hr}^{-1}$
0.	5.52	155.62	.612	16.220
5.	5.03	150.50	.443	11.632
10.	4.14	124.97	.320	8.480
15.	3.25	97.44	.223	5.888
20.	2.48	74.34	.158	4.173
25.	1.84	55.95	.114	3.006
30.	1.32	39.97	.089	2.577
35.	.91	27.30	.069	2.144
40.	.62	18.43	.043	1.473
45.	.41	12.29	.011	.376
50.	.29	8.46	0.0	0.0
55.	.20	6.05	0.0	0.0
60.	.14	4.42		

The 3.0cm eye to lens distance is typical of the operation of some of the systems we investigated. The 0.1cm data are included to obtain some indication of radiation levels at the surface of the glass.

The energy spectrum at the surface of the eye changes with the eye to lens distance. The peak of the energy occurs around 4 Mev and 0.5 Mev for the 0.5cm and 3.0cm distances respectively.

Discussion

The 1700 alpha counts/min for the lens in Table 1 may be compared with surface count rate in Table 4 by proportions of percentages and correction for area. For the first lens the calculation is as follows:

$$\frac{(1700 \text{ counts/min}) (.005\%) (60 \text{ min/hr})}{(18.1\%) (6.77 \text{ cm}^2)} = 4.76 \text{ counts hr}^{-1} \text{ cm}^{-2}$$

In a similar way, the second lens yields 5.06 counts hr⁻¹cm⁻². Both of these numbers are lower than that given in Table 4. The alpha survey instrument used to measure the count rate has a 1.5mg/cm² window. This window will be effective in shielding low energy alphas from the detector. The agreement between the measured and computed values is excellent considering the window thickness and the differences in concentration.

About half of all incoming alpha particles are stopped and two-thirds of the energy is deposited in the first 15μm of the surface. This includes the tear layer (7μm) and part of the epithelium. The first mitotic layers occur about 45μm below the surface. Even with the lens at the surface of the eye, only one particle will reach this depth every two hours for each cm² of surface.

All of the data was reported for 0.005% thorium. The data may be multiplied by 10 to obtain the results for 0.05% or other appropriate factors to obtain values for other thorium concentrations. Smaller diameter lenses will reduce the alpha flux, but not in a simple relation to area.

In comparison with the alpha absorbed dose, using the data from Table 1, the beta-gamma component is 0.3μ rad when corrected to .005% thorium. At the surface, this represents a factor of 100-1000 less. At the first mitotic cells the two dose rates are about the same.

Summary

Over 90% of the alpha radiation is absorbed before it reaches the first mitotic layer of the eye. From the data presented, the absorbed dose in the mitotic layer may be calculated and integrated over exposure times. No attempt has been made to relate these values to exposure criteria.

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DOSIMETRIE DES RAYONNEMENTS COSMIQUES A BORD DU TRANSPORT SUPERSONIQUE
CONCORDE

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Abstracts

Dosimetry of radiation on-board the French prototype supersonic transport Concorde 001 was performed during the aircraft's test flights from its Toulouse base.

The supersonic transport medical subcommittee of the General Secretariat for Civil Aviation directed these radiation measurement activities. Actual dosimetric operations were conducted by different specialized laboratories employing the following systems and techniques :

I - "Passive" integrator dosimetric systems

- a/ Nuclear emulsions in which each event is individually analyzed.
- b/ Stacks of nuclear emulsions of varying sensitivity which permit obtaining ionizing-particle distribution as a function of their TEL.
- c/ Radiothermoluminescent dosimeters.
- d/ Fast-neutron dosimeters.

2 - Dose-rate recording systems.

The authors analyze, compare, and comment upon results obtained. In addition, the dosimetric problems posed by possible solar flares and their effects upon flight plans, are discussed.

Conclusions are drawn relative to the possible dosimetry techniques to be employed in identifying radiation protection problems on-board the SST.

Introduction

Les avions qui évoluent à des altitudes supérieures à 15 000 mètres sont soumis à une irradiation naturelle différente de celle habituellement observée dans les couches basses de l'atmosphère.

La très prochaine mise en service des avions de transports supersoniques commerciaux (T.S.S.) accroîtra le nombre des personnes exposées car l'efficacité de l'écran que forme l'atmosphère terrestre aux particules du rayonnement cosmique est très diminuée à l'altitude de croisière de ces appareils.

Ce problème radiobiologique se présente sous deux aspects : d'une part il faut connaître le rayonnement cosmique permanent auquel l'avion est soumis dans les circonstances normales, d'autre part, il faut pouvoir détecter sans délai les irradiations anormales consécutives à certaines éruptions solaires. Ces dernières indications doivent pouvoir être exploitées très rapidement par les équipages.

Nous résumerons dans la première partie de cet exposé les connaissances acquises sur le rayonnement cosmique et nous exposerons ensuite quelles sont les

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mesures dosimétriques que nous avons entreprises et réalisées à bord du prototype CONCORDE 001.

I - Composition physique du rayonnement cosmique à l'altitude de croisière de Concorde

Le rayonnement cosmique qui atteint l'atmosphère terrestre peut avoir deux origines : l'une galactique, l'autre solaire.

A/ Rayonnement cosmique galactique

Ce rayonnement est constitué de particules de grande énergie :

- 80 à 85 % de protons

- 15 à 19 % d'hélium

et des particules lourdes Fe, Mg, C, O. .

Lorsqu'ils pénètrent dans l'atmosphère les noyaux sont désintégrés au cours de réactions nucléaires avec l'oxygène et l'azote de l'air et il apparaît des particules secondaires : neutrons, mesons, hyperons. Le nombre des particules secondaires augmente progressivement, passe par un maximum vers 18 000 - 20 000 mètres d'altitude et diminue ensuite du fait de l'absorption atmosphérique.

Les particules du rayonnement cosmique galactique sont soumises à la distribution et aux variations du champ magnétique dans le système solaire. Leur intensité varie à la surface de la terre selon le temps, la latitude et l'altitude.

Variations dans le temps

Lorsque les zones actives du soleil émettent des nuages de plasma, on observe des réductions de l'intensité du rayonnement cosmique galactique. Ce sont les réductions de FORBUSH.

Variations avec la latitude

Le champ magnétique de la terre empêche les particules chargées de franchir les lignes de forces sous une inclinaison supérieure à un angle donné.

L'intensité du rayonnement cosmique galactique est maximale au pôle magnétique et minimale à l'équateur géomagnétique.

Variations avec l'altitude

Celles-ci sont consécutives aux épaisseurs d'atmosphère traversées.

B/ Rayonnement cosmique solaire

Une éruption solaire se manifeste en fait par une augmentation de la brillance d'une certaine surface de la chromosphère. La dimension et l'intensité du phénomène définissent assez bien l'importance de l'éruption solaire. On pourrait également considérer sa durée.

La fréquence des éruptions est liée à l'activité solaire qui est un phénomène cyclique. Les deux derniers maximums ont eu lieu en 1957 et en 1968 et les minimums en 1954 et 1964.

En juillet 1970 on a observé 500 éruptions, tandis qu'il n'y en eut que 3 en 1964 qui eurent toutes lieu le même jour.

Dans certains cas, les éruptions solaires provoquent des phénomènes radioactifs qui se produisent au voisinage de la chromosphère et qui s'étendent dans la couronne solaire. Ces éruptions s'accompagnent d'émission de particules, protons et électrons, qui sont accélérées et portées à des énergies considérables (de l'ordre du GeV).

Une émission de rayons X de quelques dizaines de keV se propage alors dans le milieu interplanétaire.

Pour l'habitant de la terre, les manifestations des éruptions solaires sont rares. Les émissions de rayons X se produisent lors d'une éruption sur 200 et les arrivées de particules ont lieu pour une éruption sur 1000. Ceci est dû à l'action du champ magnétique et de l'atmosphère terrestre. Le rayonnement X est absorbé par les couches les moins denses de l'atmosphère (vers 80 000 mètres) et

il ne pénètre pas au-dessous de 30 000 mètres. Ce sont les ions créés qui perturbent les ondes radioélectriques.

Quant aux particules chargées, leur pénétration augmente avec la latitude ; seules celles qui ont une énergie très élevée (quelques GeV) peuvent atteindre la surface de la terre aux latitudes moyennes et équatoriales.

Aux altitudes de croisière des avions supersoniques, la protection n'étant pas suffisante, ces événements doivent être pris en considération.

La prévision et l'identification des événements solaires est faite en permanence par des équipes travaillant en collaboration internationale.

De 1966 à 1968, six à huit événements auraient été détectables à l'altitude de vol du Concorde soit 3 à 4 pour 10 000 éruptions.

II - Moyens dosimétriques utilisés

La complexité du problème dosimétrique posé et sa nouveauté ont conduit les responsables de la sous-commission médicale "Concorde" à faire appel aux spécialistes de différents laboratoires qui dans une large concertation scientifique et technique utilisèrent les avantages complémentaires de l'expérience acquise, du savoir-faire et des équipements disponibles.

Deux types de dosimètres ont été utilisés. Une première catégorie que nous appelons des dosimètres "passifs" qui intègrent et gardent en mémoire les événements et une autre catégorie qui s'apparente aux appareils de mesure et qui donne à chaque instant du vol les indications sur l'intensité du rayonnement.

I/ Dosimètres passifs

a/ Ensemble dosimétrique pour les particules chargées et les ions lourds

Il est constitué par des émulsions nucléaires et par des matières plastiques qui permettent de déterminer les doses dues aux particules chargées.

Le système dosimétrique maintient à l'abri de la lumière un empilement constitué d'une émulsion nucléaire Ilford K5 d'une épaisseur de 100 micromètres et 5 pellicules de nitrate de cellulose de 200 micromètres d'épaisseur.

Dans le type d'émulsion nucléaire utilisé toutes les particules chargées qui composent le rayonnement cosmique, électrons, mésons, mésons chargés, protons, particules α , ions lourds même s'ils sont au minimum d'ionisation, sont enregistrées et laissent une trace après développement.

Les cinq pellicules de nitrate de cellulose servent ensuite à déterminer le flux des ions lourds.

b/ Ensemble dosimétrique pour les neutrons

Nous avons utilisé le dosimètre photographique CEA/STEPPA contenant une émulsion nucléaire NTA pour déterminer la "dose neutron" due au rayonnement cosmique. Sept de ces dosimètres sont placés sur les sangles des parachutes des équipages.

Deux dosimètres témoins sont conservés au sol.

Le dispositif complet est renouvelé chaque mois.

c/ Ensemble dosimétrique pour les champs électromagnétiques et les particules ionisantes

Dans une première approche du problème qui nous était posé, nous avons essayé de mesurer la dose absorbée due à la composante électromagnétique du rayonnement cosmique.

Nous avons utilisé des dosimètres radiothermoluminescents au fluorure de lithium et au sulfate de calcium et nous avons également embarqué des empilements d'émulsions nucléaires à sensibilité variable afin de déterminer le spectre de transfert linéique d'énergie des particules ionisantes.

Nous utilisons les émulsions :

- K5 : pour l'enregistrement de toutes les particules ionisantes,
- K2 : pour l'enregistrement des particules dont le TLE est supérieur

à 1 keV par micromètre (dans l'eau)

KI : pour l'enregistrement des particules dont le TLE est supérieur à 6 keV par micromètre

KO : pour l'enregistrement des particules dont le TLE est supérieur à 8 keV par micromètre.

Ce système avait déjà été étudié et est utilisé au C.E.A. pour la dosimétrie auprès des grands accélérateurs (I). Il a fallu l'adapter au problème posé du fait de la faiblesse du niveau des doses enregistrées.

Nous avons dû étudier une méthode d'effacement du bruit de fond des plaques nucléaires. Cette méthode a été récemment mise au point et les essais effectués auprès de l'accélérateur Saturne ont donné pleine satisfaction.

La filtration totale correspondant aux divers emballages et supports des émulsions est de 450 mg/cm² environ.

2/ Dosimétrie par enregistrement des débits de dose

a/ Systèmes d'enregistrement du débit de dose dû aux neutrons et aux rayonnements gamma - VAMEGA

On a utilisé un matériel appelé VAMEGA déjà employé depuis plusieurs années sur les avions subsoniques longs courriers. Ce dispositif a été modifié pour assurer la mesure des neutrons. Il comprend un détecteur gamma qui est un tube Geiger-Muller, associé à un détecteur à hélium 3 entouré d'un modérateur en polyéthylène.

b/ Détecteur de bord AWRE installé sur tous les avions Concorde

Ce détecteur a été conçu par l'United Kingdom Atomic Energy Authority, Atomic Weapons Research Establishment (AWRE) à Aldermaston (2). Il indique le débit de dose en mrem/h sur une échelle logarithmique à 4 décades allant de 0 à 1 000 mrem/h. Il comporte également un affichage digital de la dose cumulée en mrem. Trois tubes Geiger-Muller permettent de mesurer la dose due aux particules chargées et aux gamma, un facteur de qualité égal à 1,5 étant automatiquement appliqué ; un compteur proportionnel au trifluorure de bore avec modérateur en polyéthylène permet de mesurer la dose due aux neutrons en utilisant un facteur de qualité égal à 10. Après traitement, les signaux des 2 systèmes de détection entrent dans une voie de comptage unique. L'échelle de l'indicateur est divisée en zones "normale" "alerte" et "action ; le niveau d'action étant relié au système d'alarme central de l'avion. La définition de ces 3 zones est en cours d'étude. Sur l'avion prototype, le signal de sortie du détecteur de bord est également enregistré sur l'équipement central d'enregistrement d'essais en vol (QS system) de façon que l'on puisse faire des lectures de durée de vol et de débits de dose à des altitudes sélectionnées.

III - Résultats et discussion

I/ Résultats obtenus avec les ensembles dosimétriques constitués par les émulsions nucléaires K5 et les plaques de nitrate de cellulose

Dès le retour au laboratoire des dispositifs d'exposition à la suite d'une expérimentation d'un mois, les émulsions nucléaires Ilford K5 de 1 000 micromètres d'épaisseur sont exposées à la lumière blanche parallèle passant à travers le négatif d'une grille millimétrique codée. La grille impressionnée sur une des faces de la pellicule d'émulsion nucléaire détermine un système d'axes de référence qui permet de faire toujours des mesures dans les mêmes conditions et dans des volumes d'un mm³. Les traces dues aux particules chargées du rayonnement cosmique, observables dans l'émulsion nucléaire, sont soit granulaires, soit continues. Ces phénomènes sont fonction de l'énergie et de la charge des particules.

En tenant compte de l'épaisseur avant développement et de la grille, on mesure, sous microscope, la longueur en projection des traces traversant un volume d'un mm³ d'émulsion nucléaire ou s'y arrêtant, les coordonnées des points d'entrée et de sortie ou d'arrêt des traces ainsi que l'épaisseur de l'émulsion

nucléaire après développement dans la zone du volume considéré. Ces mesures sont effectuées dans 10 volumes d'un mm³ présélectionnés par la grille sur chaque émulsion nucléaire contenue dans un dispositif d'exposition. Les longueurs en micromètres sont calculées à partir de ces mesures. Les traces se trouvant dans le volume d'un mm³ d'émulsion nucléaire correspondent à des particules présentant une perte spécifique d'énergie par unité de longueur variable ou non le long de leur trajectoire. En considérant des pertes spécifiques d'énergie par micromètre, la sommation de ces grandeurs sur toute la longueur de toutes les traces nous donne l'énergie totale "déposée" dans le volume d'un mm³. La perte d'énergie par unité de longueur peut être déterminée expérimentalement par des mesures de granularité ou des mesures photométriques. Ces travaux ont déjà été faits dans le cas des émulsions nucléaires exposées à bord des cabines spatiales Apollo.

Mais le temps nécessaire à ces mesures est très grand et incompatible avec un dépouillement mensuel. De plus, dès le début de cette exploitation, on a constaté que le nombre de traces continues est de l'ordre de 10 p. cent par rapport au nombre total et que le nombre des ions lourds est nul. Cette constatation a conduit à employer une méthode plus rapide.

En partant des fins de traces de protons d'accélérateurs, d'énergie initiale de 150 MeV, enregistrées dans l'émulsion nucléaire Ilford K5, on a déterminé le parcours résiduel pour lequel les traces de protons passent de l'état granulaire à l'état continu. Il est de l'ordre de 70 micromètres, ce qui correspond à une énergie du proton de 3 MeV et une perte spécifique d'énergie de 24,2 keV par micromètre. Rappelons que la perte spécifique d'énergie des protons au minimum d'ionisation est de 0,549 keV par micromètre.

En multipliant la somme des longueurs de toutes les traces contenues dans le volume d'un mm³ par la valeur 24,2 keV par micromètre, nous obtenons la limite supérieure de l'énergie totale "déposée". Un volume d'un mm³ d'émulsion nucléaire a une masse de 3,8.10⁻³g. Par définition, un rad correspond à une énergie de 6,24.10¹³eV "déposée" dans un gramme de matière. La dose en millirad est donc égale à :

$$\frac{\text{Energie totale "déposée" } \times 10^3 \times 10^3}{3,8 \times 6,24 \times 10^{13}}$$

Les doses maximales dues aux particules chargées calculées selon cette méthode sont données dans le tableau I. Ces valeurs sont les moyennes obtenues à partir des 10 volumes d'un mm³ examinés dans chaque émulsion nucléaire d'un dispositif d'exposition. Ce tableau indique le nombre d'heures de vol au-dessus de 12 000 mètres d'altitude.

Il est important de noter que sur l'ensemble des émulsions exploitées de 1970 à 1973, nous n'avons jamais repéré de trace d'ion lourd.

2/ Résultats obtenus avec les dosimètres à émulsion NTA

Par l'intermédiaire de la même grille millimétrique, on détermine le nombre moyen des traces contenues dans 10 surfaces élémentaires d'un mm².

Pour ces films et pour les neutrons d'énergie comprise approximativement entre 1 et 10 MeV, on admet d'après les résultats expérimentaux que 127 000 traces par cm² correspondent à 1 rad ; 1,27 trace/mm² correspond donc à 1 millirad. Il n'est toutefois pas possible de déterminer la dose due aux neutrons du rayonnement cosmique à l'aide de cette équivalence. En effet, les résultats expérimentaux ont été obtenus avec des flux de neutrons purs, alors que les neutrons ne constituent qu'une partie du rayonnement cosmique. L'émulsion NTA utilisée n'a que 40 micromètres d'épaisseur. L'examen détaillé de l'émulsion nucléaire K5 de 1 000 micromètres d'épaisseur a permis de déterminer que 90 p. cent des traces sont des traces de particules chargées et non des protons de recul. Il y a donc lieu de considérer que 0,127 traces par mm² correspond à 1 millirad. Le tableau (I) donne les doses en millirads dues aux neutrons dans le poste de pilotage.

3/ Résultats obtenus à l'aide de l'ensemble dosimétrique pour les champs électromagnétiques et les particules ionisantes

Trois ensembles dosimétriques sont placés à bord de l'avion :

- un à l'avant,
- un autre au centre,
- le troisième à l'arrière.

a/ Dosimètres radiothermoluminescents

Le tableau de la figure n° 5 donne des exemples de résultats de mesures effectuées à l'aide des dosimètres au fluorure de lithium (3).

On constatera que les doses mesurées sont très faibles et très proches de la limite inférieure de détection. La durée des vols à haute altitude a été insuffisante pendant nos essais ; il s'ensuit que la part de l'irradiation subie lors des vols est très faible par rapport à celle reçue au sol pendant la période d'intégration (7 semaines environ). Nous estimons que la marge d'erreur de ces mesures est de ce fait assez grande (coefficient de variation égal à 40 p. cent).

Pour diminuer celle-ci, nous avons décidé d'utiliser lors des vols du second semestre 1972, un produit nettement plus sensible, le sulfate de calcium activé au dysprosium. Ce dernier est en effet trente fois plus sensible que le fluorure de lithium et, bien que n'étant pas "équivalent aux tissus mous", il nous a donné d'excellents résultats lors des mesures effectuées sur des vols de ballons (4) pour le compte du Groupe Européen de Biophysique Spatiale. En effet, aux altitudes où sont effectuées ces expériences, la contribution des rayonnements électromagnétiques de faible énergie est tout à fait négligeable. Quant à la sensibilité aux particules ionisantes, elle ne diffère guère de celle du fluorure de lithium (5). Nous espérons obtenir des résultats plus précis à l'aide de ce matériau (le coefficient de variation des mesures obtenues est égal à environ 10 p. cent). Si l'on désire une précision encore meilleure, il est nécessaire soit de faire les mesures immédiatement après un vol prolongé, soit d'attendre que la fréquence et la durée des vols soient suffisantes.

b/ Emulsions nucléaires à sensibilité variable

Le classement des particules en fonction de leur TLE est effectué d'après les caractéristiques des émulsions (seuil de sensibilité) et l'aspect des traces. A titre d'exemple on a admis que sur une émulsion K5, les traces correspondent à un TLE inférieur ou égal à 0,5 keV par micromètre dans l'eau, les traces denses présentent un TLE supérieur. D'autre part les protons de recul provenant des interactions (n,p) engendrés dans l'émulsion, ne sont pas pris en compte.

Il est évident que ces critères sont quelque peu subjectifs et que les spectres de TLE obtenus doivent être considérés comme une simple approche du problème. Cela est d'autant plus vrai que la précision des mesures est altérée par le fait que les doses reçues au sol ne sont pas négligeables par rapport à celles reçues en vol.

Sur le tableau de la figure n° 6 nous avons reporté, à titre d'exemple, les résultats obtenus à l'aide d'un empilement d'émulsions placé à l'arrière de l'avion au mois d'avril 1972. Les valeurs mentionnées correspondent aux seules irradiations subies pendant les vols, la contribution du rayonnement cosmique et tellurique au niveau du sol a été retranchée.

Nous avons calculé la valeur de la dose absorbée à la surface du corps humain, à partir des données de ZERBY et KINNEY (6) valables dans le cas d'un faisceau isotrope de protons qui frapperait une seule face du corps humain. Il convient de préciser que dans ce cas, la dose absorbée superficielle est très voisine de la dose absorbée au point où l'ionisation est maximale (7).

On remarquera, pour cet empilement, une nette prédominance des particules à très faible TLE (TLE 0,5 keV/micromètre). Il n'en est pas toujours ainsi ; ces proportions varient selon les conditions des vols et les emplacements dans la cellule de l'avion.

Par contre, si l'on exclut les traces de protons de recul provenant des interactions des neutrons avec les matériaux hydrogénés, on ne dénombre pas de particule de TLE supérieur à 6,2 keV par micromètre. Il est vrai qu'à l'altitude de vol du Concorde il semble que les ions lourds aient déjà été absorbés dans l'atmosphère.

Il est possible à partir de ces spectres de calculer un facteur de qualité moyen pour les rayonnements ionisants. Compte tenu des données de NEUFELD, SNYDER et TURNER (7) celui-ci est légèrement inférieur à 1,3.

Ce résultat est donné à titre d'exemple, les expérimentations ne sont pas encore assez avancées pour que l'on puisse faire une synthèse valable des résultats en fonction des conditions des vols et des emplacements dans la cellule.

4/ Résultats obtenus avec les appareils de détection et d'enregistrement des débits de dose VAMEGA et AWRE

Pour le tableau n° 7 nous avons reporté les résultats maximaux moyens des mesures obtenues à l'aide de l'appareil VAMEGA lors d'un groupe de 18 vols au-dessus de l'Atlantique Nord et une série de 11 vols effectués lors de la tournée en Amérique du Sud.

On remarque que les résultats de la seconde série sont, du fait de la différence de latitude, nettement inférieurs à ceux de la première.

Les résultats obtenus à l'aide de l'appareil AWRE sont comparables. A titre d'exemple, lors de la mission du 21.7.71 effectuée au-dessus de l'Atlantique Nord, les débits de dose équivalente mesurés sont respectivement :

- Neutrons	0,20 mrem/h
- Radiations ionisantes et électromagnétiques	0,66 mrem/h
Total	0,86 mrem/h

En aucun cas on n'a observé une activité solaire anormale.

Si en cours d'exploitation normale le cas se présentait, l'altitude de vol de l'avion serait modifiée selon des critères qui sont actuellement étudiés par la Commission Médicale Franco-Britannique.

En fait, ce cas devrait se présenter assez rarement. De 1966 à 1968, huit évènements seulement auraient été enregistrés à l'altitude de croisière.

Pendant le cycle solaire le plus actif jamais observé entre 1954 et 1964, On trouve 38 évènements détectables sous forme d'irradiation à l'altitude de Concorde. Pour le cycle actuel, à peu près moitié moins actif, on arrivera sans doute à une vingtaine d'évènements, probablement tous de faible intensité. En fait, dans le cycle précédent, dix évènements seulement auraient provoqué une irradiation à un débit de dose supérieur à 1 millirem par heure. Pour le cycle actuel, probablement aucun n'atteindra cette valeur d'ici à 1974 ou 1975. Qu'en sera-t-il ensuite ? On a peu d'éléments pour effectuer une prévision : l'accélération de particules à des énergies aussi élevées et le fait que la terre se trouve sur leur trajectoire forment des conditions assez exceptionnelles observées un nombre de fois trop restreint pour qu'une statistique valable puisse être utilisée. On dispose cependant de deux indications :

- 1 - Pendant un cycle solaire, le nombre et l'énergie de ces évènements semblent en relation avec l'activité maximale atteinte au cours de ce cycle.
- 2 - On possède des indications positives qui montrent que les prochains cycles solaires vont encore, dans les cinquante années à venir, décroître en intensité.

Conclusions

Les doses reçues à l'altitude de croisière du T.S.S. Concorde sont faibles; nous n'avons jamais détecté d'ion lourd.

Le problème des éruptions solaires a retenu notre attention. Leur fréquence est relativement faible ; on en aurait détecté 8 au maximum de 1966 à 1968 à l'altitude considérée. Les appareils d'alarme placés à bord des prototypes n'en ont jamais décelé pendant les essais.

Note

Ces travaux de dosimétrie ont été réalisés sous les auspices de la Commission Médicale du Transport Supersonique du Ministère des Transports Publics, présidée par Monsieur le Médecin Général Inspecteur RABOUTET et de la Sous-Commission Médicale Française du Groupe Aéromédical CONCORDE.

Les dispositifs dosimétriques ont été installés dans l'avion avec les conseils du Commandant TURCAT et de Messieurs JOATTON et DESTARAC de la S.N.I.A.S.

Figure I

	Nbre d'heures de vol > 12000 m	Part. chargées mrad	Neutrons mrad
Janvier 1972	7h 38	2,4	0,2
Février "	8h 31	1,1	0,2
Mars "	0h 45	0,1	0,1
Avril "	0h 22	0,3	0,1
Mai "	0h 22	0,3	0,1

Résultat des doses absorbées mesurées à l'aide d'émulsions nucléaires de 1000 μ - Poste de pilotage

Figure 2

Mois	Heures de vol au-dessus de 12 000 m	Doses reçues pendant les vols (millirad)		
		Avant	Milieu	Arrière
Janvier 1972	7h 38	3	3	3
Février "	8h 31	3	3	3
Avril "	0h 22	2	2	2
Mai "	3h 38	2	2	1,5
Octobre "		0,8	0,8	0,8
Novembre "		2	2	2

Dose absorbée pendant la durée des vols mesurée à l'aide de dosimètres radiothermoluminescents

(L'irradiation reçue en dehors des vols a été retranchée)

Figure 3

Intervalle de TLE keV//m	Densité de traces - traces cm ⁻²	Dose absorbée superficielle à une fluence unitaire	Dose absorbée superficielle	Contribution relative à la dose
TLE ≤ 0,5	3950	1,6 10 ⁻⁷ rad	0,63 10 ⁻³ rad	73 p.cent
0,5 < TLE ≤ 1	450	3,1 10 ⁻⁷ rad	0,14 10 ⁻³ rad	16 p.cent
1 < TLE ≤ 2,6	80	4,3 10 ⁻⁷ rad	0,03 10 ⁻³ rad	4 p.cent
2,6 < TLE ≤ 6,2	6	10 10 ⁻⁷ rad	0,06 10 ⁻³ rad	7 p.cent
6,2 < TLE ≤ 8,1	0		0	0 p.cent
8,1 < TLE	0		0	0 p.cent
Spectre total			0,86 10 ⁻³ rad	

Résultat de l'empilement d'émulsions nucléaires du mois d'avril 1972 placé à l'arrière de la cellule

Figure 4

Série de Missions	Débit de dose équivalente dû aux neutrons	Débit de dose équivalente dû aux neutrons γ^+ particules ionisantes mrem/h	Total mrem/h
Atlantique Nord	0,31	0,58	0,89
Amérique du Sud	0,13	0,34	0,47

Débit de dose équivalente moyenne maximum mesuré à l'aide de l'appareil VAMEGA lors de deux séries de missions

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THE APPROACH TO RADON PROBLEMS IN NON-URANIUM MINES IN SWEDEN

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Abstract

In Sweden there are about 60 underground mines. These are for the mining of ferrous and sulphide ores and none are for uranium. Radon measurements were made in all of these mines during 1969 and 1970. By using an assumed equilibrium ratio of 0.5 between radon daughters and radon, the radon daughter levels were calculated to exceed 30 pCi/l (0.3 WL) in 22 mines with more than 1000 underground employees and in a few mines the levels were estimated to exceed 100 pCi/l (1 WL). Hence, non-uranium miners were the largest group of Swedish workers receiving significant radiation doses. However, these workers are not legally classified as radiological workers.

The results initiated more detailed measurements and research. The sources of radon in mines are found to be radon-rich water, ventilation air from abandoned areas with caved materials and, to a lesser extent, minerals particularly rich in radium. In some mines the radon levels vary with the season with a maximum in the summer. No simple correlation to the type of minerals or their geological properties has yet been found.

Instructions on protective measures against radon in mines were issued in March 1972. The regulations are based on a maximum of 30 pCi/l (0.3 WL) of radon daughters as the average over a year. There are also regulations on the maximum delay period for preventative action, on ventilation, on respirators, on measurements and control and on medical examinations.

Epidemiological studies on the lung cancer frequency among miners have been made. The period investigated was the years 1961-1968. A significant excess of lung cancer has been found.

Introduction

The first measurements on radon in a Swedish mine (Boliden) were made at the beginning of the 1950s. The results of these early measurements and the limited experience of radon problems in non-uranium mines at that time did not give any reason for anticipating high radon levels in Swedish mines. However, improved measuring techniques, continuous progress in radiation protection generally and internationally observed radon problems in many uranium mines resulted in new measurements being initiated at the end of the 1960s.

The first new results (from Näsliden and Långsele) indicated the possibility that radon problems might exist in Swedish mines even though they are not uranium mines. A few additional measurements (in Zinkgruvan and Danne-mora) also proved that there is no simple relation between the radon concentration in a mine and the geology of the mine; the first approach, selecting mines

of interest solely from geological considerations, had to be abandoned. In 1969 it was therefore decided to make a rough but rapid survey of the radon levels in all the mines in Sweden (about 60).

Since 1970 great efforts have been made to develop appropriate sampling and measuring techniques, and to initiate and carry out research on existing problems. As radon in mines was a new and completely unknown occupational hygiene problem for most of those concerned, much effort was put into providing appropriate information. Very shortly, it also appeared necessary to prepare special instructions for radiation protection in mines and these were issued in March 1972. Because of the relatively high radon concentrations found in many mines, it was considered necessary to consider the possibility of an excessive incidence of lung cancer among Swedish miners. An epidemiological study on lung cancer was therefore started in 1971. This report is meant to be a summary of some of the results of the work done on the problem of radon in Swedish mines.

Principles of sampling and measuring techniques

The measurements in Swedish mines have mainly been made on radon although they have been supplemented by measurements on radon daughters. Radon measurements are performed by taking air samples in evacuated 4.8 l commercial propane containers which are opened in the mine. The sample is transferred to an evacuated 18 l ionization-chamber via a drying agent. The lower limit for these chambers is about 0.5 pCi/l with a fresh 4.8 l sample. The samples are measured above ground, either in a field laboratory at the mine or at the National Institute of Radiation Protection (NIRP) in Stockholm. In the latter case, the sampling can be done by the mining staff and the containers sent to Stockholm by mail.

Radon daughters are sampled and measured in the conventional way, using glassfibre filters. In evaluation of the radon daughter concentration, the method of Kusnetz¹ is used. As there is no good Swedish expression for WL the result is expressed in equivalent pCi/l (1 WL is equivalent to 100 pCi/l). Measurements on the glassfibre filters are made above ground in general.

The principles of measurements

Three types of measurements have been made, namely: guiding measurements, basic measurements and checking measurements.

Guiding measurements

The guiding measurements were performed twice during the period 1969-1970, once during the winter and once during the summer. Three of the evacuated containers described above were sent to each mine and the mining company was asked to take one air sample in return air, one at a working place with "normal" ventilation and one sample in an unventilated drift. The samples were expected to be representative for the average radon levels in the mine, for working places and for potentially high radon levels, respectively.

In answer to questionnaires, information was given by the company about the place and time of sampling, ventilation conditions, presence of running water, adjacent minerals, number of worker etc. All results related to radon only. In estimating the corresponding radon daughter concentration, a 50 % equilibrium ratio was assumed. In accordance with the chosen Swedish terminology, this ratio is referred to as the "dose factor".

Basic measurements

The basic measurements are made by the laboratory staff during a visit to the mine. The purpose of the measurements is to find the true radon and radon daughter concentrations, the reasons for the activity levels and the best means of decreasing the concentration if necessary. The sampling and measurement are preceded by detailed discussions between the radiological team and the representatives of the employees and the employers about ventilation systems, presence or absence of working places in particular areas, suspected high radon levels, radioactive minerals etc. The choice of appropriate places for sampling is based on these discussions.

Checking measurements

Checking measurements are initiated by the company itself if there have been major changes in the ventilation, or when there is reason to expect high radon concentrations in new parts of the mine. Normally only radon measurements are necessary to guide possible extra preventative action. Periodic checking measurements are also needed as a consequence of the regulations on checking the exposure of the miners (see below). If the only measurements made are those on radon in air samples sent by mail to the laboratory in Stockholm, the radon daughter concentration is estimated by using the dose factor (or factors) found in the basic measurements if this dose factor is still believed to be adequate.

Concentrations of radon and radon daughters in the mines

After the guiding radon measurements in 1969 and 1970, the radon daughter exposure level was estimated for all mines. The numbers in parenthesis in Table 1 are the first estimated results. At the time of writing (May 1973) basic measurements have been made in 26 mines, from the guiding measurements most of these were expected to have radon daughter concentrations above 30 pCi/l. As a result of these measurements there have been a few changes in the grouping of the mines and miners as indicated in Table 1 by the 1970 numbers without parentheses. This was the situation in 1970. Since then, countermeasures have been taken in many mines, if not yet in all. A few mines have shut down (not because of radon) and a few new mines have been built and the 1973 numbers in Table 1 reflect the situation as it is believed to be in May 1973. It should be noted, however, that the grouping of the mines and miners is based on the highest representative levels found in the working areas of each mine - or of the parts of one large mine. The numbers of overexposed miners have therefore probably been somewhat overestimated.

Radon daughter concentration pCi/l	Number of mines		Number of miners	
	1970	1973	1970	1973
≤10	(25) 26	26	(1100) 1400	1400
10 - 30	(13) 12	19	(1700) 2000	2700
30 -100	(17) 16	10	(1700) 700	610
100 -300	(5) 6	0	(140) 620	0
> 300	(0) 1	0	(0) 21	0

Table 1. Numbers in parenthesis are those estimated from the guiding measurements 1969-1970, the ones without parentheses from basic measurements since 1969.

Great efforts have been made to reduce the radon daughter levels and this work is still in progress. The countermeasures mainly consist of changes in the ventilation system and often new ventilation shafts have to be built. However, it is expected that within about a year the numbers of mines and miners in the exposure group >30 pCi/l will be reduced considerably.

The measurements in 1969 and 1970 showed that many mines had higher, some considerably higher, radon levels in the summer (May-October) than in the winter. 19 mines with radon daughter levels >10 pCi/l were found to have summer values more than 50 % higher than winter values and on average for all mines the radon concentration was twice as high in the summer than in the winter. The main reason is probably reduced or changed ventilation.

Apart from the general radon and radon daughter values for the mines, it has also been of interest to find the potential high values in unventilated areas or elsewhere in the mine. Up to now 7 mines have been found to have such areas with radon levels exceeding 1000 pCi/l; radon concentrations of 1000-2000 pCi/l have been found in four mines, 6000 pCi/l in one mine, 10,000 pCi/l in one and 20,000 pCi/l in one. There are also a few mines with radon levels of 300-1000 pCi/l. In some of these areas with high radon levels, mining work has probably been carried out. The very high radon levels appeared in unventilated drifts where there was water.

The source of radon

There are three major factors to consider when seeking the causes of high radon concentrations; they may appear alone or in combination. These factors are ventilation, water and radioactive minerals. The first two predominate.

Ventilation

There are two basic ventilation principles:

- (a) The air is taken in via a shaft from the ground level down to the bottom of the mine and at each level the air is forwarded in tubes or in drifts by fans to the working places. This is illustrated in Figure 1 (left).
- (b) The air is taken in via crushed rock in abandoned upper parts of the mine or another part of the mine. This is illustrated in Figure 1 (right).

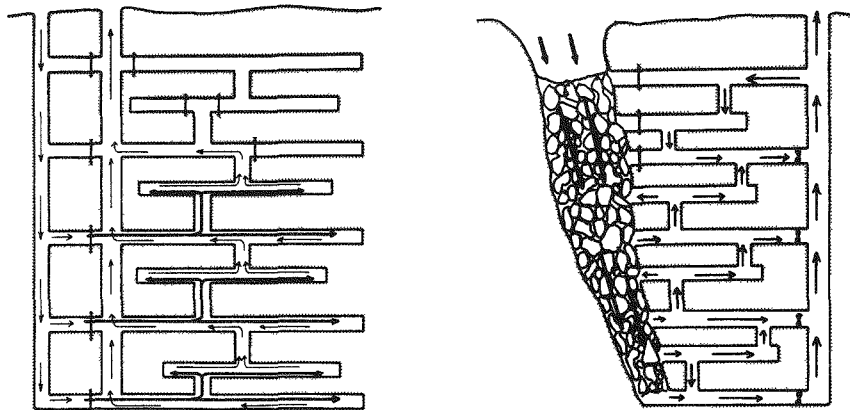


Fig. 1: Two basic ventilation principles in Swedish mines
shaft ventilation (left) and "crushed rock" ventilation (right).

There are also combinations of these principals. The advantage of ventilation principle (b) is that during the winter the inlet air becomes comfortably warm during its passage through the crushed rock and it then contains no air impurities - except radon. During the passage through crushed rock and abandoned spaces, the air is contaminated by radon which diffuses from radium in the rock and from radon-rich water in the rock. By the time it reaches the working places the ventilation air may already have too high a radon concentration. Table 1 shows that probably 23 mines had radon daughter levels higher than 30 pCi/l in 1970. The radon source has been identified for all but one of these mines and the result is as follows:

ventilation air as the predominant source - 17 mines
 water and rocks in the working areas as predominant source - 3 mines
 major contributions from both the ventilation air and sources in the working areas - 2 mines

The ventilation air is the predominant radon source. Sources in the working areas are local running water and emanation from the rock wall in drifts and other spaces. Ineffective ventilation with recirculating air results in growth of the radon concentration in the air.

Even if the fact that the air is drawn through crushed rock is the main reason for the high radon levels, it is not certain that this ventilation system always leads to very high radon levels. There are examples (Mimergruvan, Blötberget, Risbergsfältet) where the intake of air is by this principle but where the radon concentrations are not especially high (15-50 pCi/l). Contributory factors are probably the venting rate, the amount of crushed rock, its structure and radium content, presence or absence of radon-rich water, etc. As it is out of the question to investigate the condition of abandoned areas and spaces with crushed rock it is very difficult to determine the real emanating source unambiguously. However, work on this problem continues.

As mentioned above, there are seasonal variations of the radon levels in many mines. This has been studied systematically in several mines by taking an air sample once a week at the same place in the mine, normally on return air. The sample has been sent by mail to the NIRP. The result from one mine is shown in Figure 2.

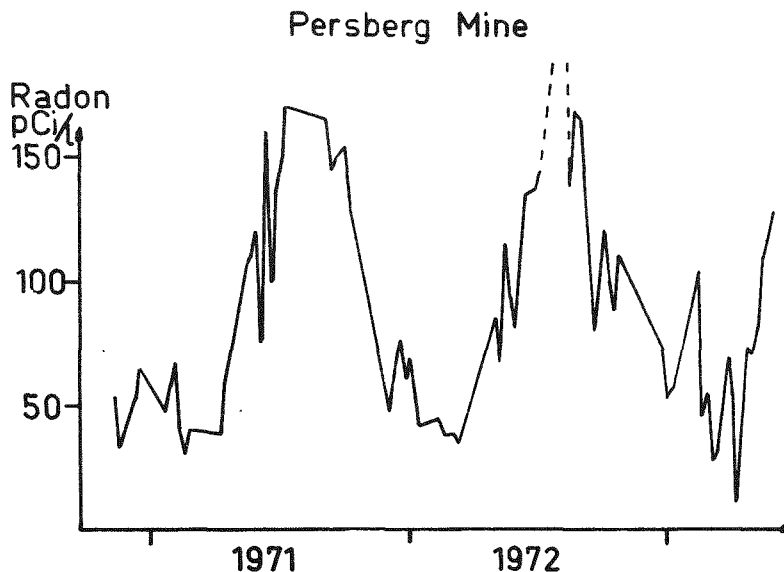


Fig. 2: Radon concentration in the return air of Persberg Mine measured once a week.

Figure 2 illustrates the seasonal variation found in some mines. The samples were taken at a depth of 150 m on return air. If the temperature outside the mine is plotted with an appropriate scale, the temperature curve and the radon curve will follow one another quite closely. It is a fact well known to mining engineers that the ventilation efficiency tends to decrease when the difference between the temperature in the mine and outside the mine decreases and the observed correlation to the temperature may reflect that phenomenon. However, one must not exclude the possibility that the variations may also be a result of different routes taken by the ventilation air or of an increase in the emanating power of the crushed rock. In a more detailed study on radon by means of a continuously running ionization chamber in a mine, diurnal variations have been observed, see Figure 3.

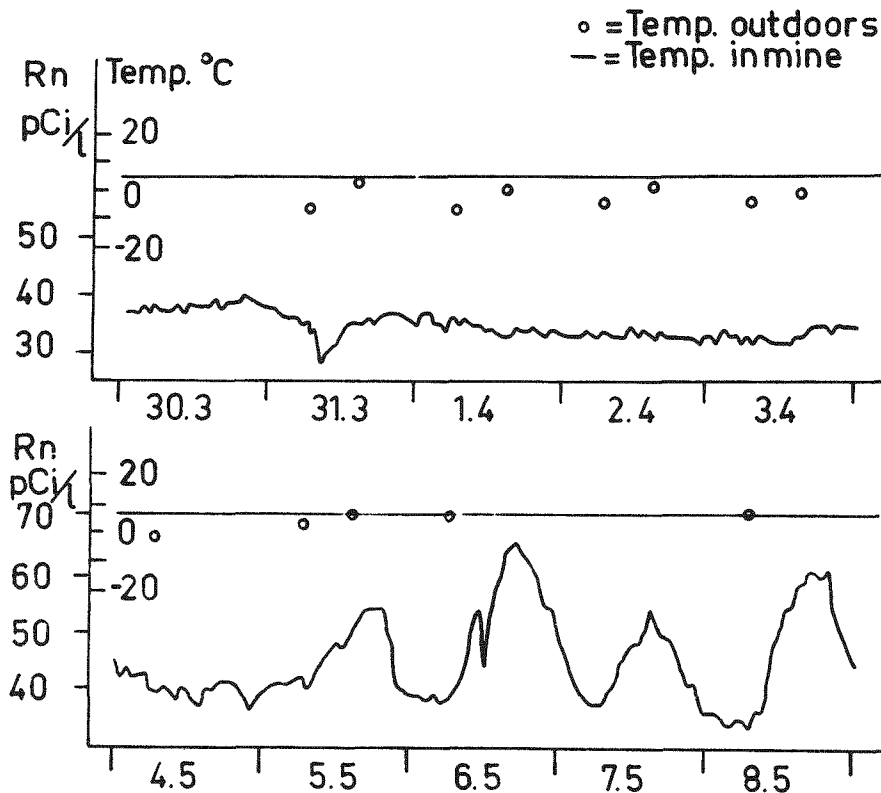


Fig. 3: Radon concentrations in the intake air at a depth of 450 m in Nygruvan Mine measured by a continuously running ionization chamber. The air has passed abandoned parts of the mine. The Figure shows March 30 through April 3 (upper curve) and May 4 through 8.

In March and April (the upper curve), the temperature outside the mine is still below the temperature in the mine at 450 m and the radon levels in the air are practically constant. In May (the lower curve) the nights are still rather cold but during the day the temperature rises above that of the mine. The radon concentration varies with a minimum during the night and a maximum during the day. This result serves to prove the influence of the temperature on the radon concentration when the temperature outside the mine is close to that inside the mine.

Water

It is difficult to prove generally the total effect of radon-rich water on the overall radon levels in a mine. Nevertheless, it has been possible to show some correlations in some mines. In one mining district (Kiruna) it has been

possible to compare two adjacent mines. They have the same geology with no exceptional radium-containing minerals in the rock or in the ore. In one mine (KUJ) the radon concentration of the water is 10-20 nCi/l. The water comes from a nearby district with minerals containing radium of the order of 100-5000 pCi radium/g. The water found in that district contains up to 60 nCi/l. The radon levels in air in the mine are 20-40 pCi/l with the exception of some unventilated parts of the mine with very high radon concentrations. In the disused adjacent mine (Loussavaara) the radon concentration of the water is very low (<0.1 nCi/l) and the radon concentration of the air in the unventilated mine is only 8 pCi/l. It is therefore reasonable to conclude that the water is a significant radon source in KUJ.

Radon-rich water may also cause very high radon levels in unventilated drifts. In KUJ a radon concentration in the air of 20,000 pCi/l was found in an unventilated drift. The water flow was about 50 l/s and contained 10 nCi radon per litre. Radon levels of that order of magnitude in the air are quite possible with the conditions which exist. The practical conclusion from this and other similar observations is that whenever there is a combination of bad ventilation and a large water flow there is reason to anticipate high radon levels in the air.

Minerals

The significance of different minerals in rock and different types of ore for the radon levels in a mine is not yet fully established.

One parameter of interest is the radium content of the rock. However, it is likely that other parameters are more relevant, as e.g. porosity, presence of cracks, humidity. Water enhances the diffusion of radon from the rock and wet mines are therefore subject to the double effect of enhancement of radon leakage and radon release directly from the water. Radioactive minerals have been found in a few mines and they have caused local radon levels of the order of 100 pCi/l. It is also possible that radioactive minerals appear in some of the crushed rock causing a high radon concentration in the ventilation air when that type of ventilation is used.

Protection instructions

The Swedish instructions on protection against radon in mines were issued in March 1972². They contain a regulative part and a descriptive and explanatory part including 5 chapters on physical and biological aspect of radon and its daughters, on geological aspects, on estimation of necessary ventilation air, on sampling methods and on estimation of annual exposure. The regulative part includes regulations on permitted radon daughter exposure, on preventative action such as ventilation etc., on measurements and checks and on medical examinations. An extract from some of the most important points is presented below.

Permitted radon daughter exposure

1. The radon daughter exposure should be as low as practicable. If the annual exposure is more than 60,000 pCi·hrs/l, there is an overexposure and preventative action is needed.

2. The radon daughter concentration in working places should be as low as practicable. For planning of preventative action an operational limit (or "basic value" or "derived working limit") on radon daughter concentration of 30 pCi/l shall be used.

Note: 30 pCi/l is by definition equivalent to 0.3 WL.

If the radon concentration has been measured instead of the radon daughter concentration and the dose factor is not known, the dose factor shall be assumed to be at least 0.5.

Note: The dose factor is defined as the relation between the radon daughter concentration and the radon concentration.

3. If the annual radon daughter exposure is expected to exceed 60,000 pCi·hrs/l, preventative action shall be taken within the times given below. However, a total overexposure of 600,000 pCi·hrs/l during an employee's whole working time in a mine shall be avoided as counted at the latest from the time when the radon daughter concentration was determined for the first time. For estimation of total overexposure, earlier exposure shall be taken into account, if possible.

Preventative action

1. Preventative action shall be planned and performed without delay and in a reliable way.

2. The maximum time within which preventative action shall be taken in continuously used working places in the mine after a basic measurement depends on the average radon daughter concentration during the year. Corresponding radon daughter concentrations and max. times are 30-60 pCi/l, 3 years; 90 pCi/l, 1.5 years; 120 pCi/l, 1 year etc. After checking measurement the corresponding max. times are shorter by a factor of three.

Other regulations

1. Any part of the mine which has higher radon daughter concentration than 1000 pCi/l shall be sealed off by a wall impermeable to radon. Those parts of the mine which are not in use and which have radon daughter concentrations of 100-1000 pCi/l or where the concentrations are unknown shall as a rule be prohibited areas.

2. Interchange of employees shall be made if necessary to prevent overexposure if other preventative action has not proved satisfactory.

3. Measurement to check the radon daughter concentration in the mine shall be made within the periods specified in Table 2.

Radon daughter concentration pCi/l	Time
<10	2 years
10 - 30	1 year
>30 -100	6 months
>100	3 "

Table 2. The time within which check measurements shall be made as a function of the radon daughter concentrations found in the last measurement.

4. Due to the radiological hazard in mines, special medical examinations should be made with respect to the functioning of the lungs and possible pulmonary diseases. Persons suffering from chronic disease in the bronchial system or the lungs and persons who have tuberculosis which has not with certainty become inactive or with healed tubercular changes of large extent should not be employed in work which involves particular risk of exposure to

radon daughters. Since there is a considerably enhanced risk of lung cancer for smokers and since it is feared that this risk is greater when a smoker is exposed to radon daughters, employees should be made aware of this fact and be advised against smoking, both at work and at other times.

Methods of calculating the exposure

As there is no good dosimeter for radon or radon daughters, the exposure is determined by indirect methods. The method proposed in the regulations is that each mine is divided into zones according to the radon daughter levels, <10, 10-30, 30-100, or 100-300 pCi/l. It is recommended that a maximum of two zones should be used. Once a year the approximate residence time in different zones is estimated for the miners and with the aid of special tables in the regulations, the exposure range is determined. The exposure ranges correspond to the same ranges as for the radon daughter level zones.

Epidemiological studies on the lung cancer rate

When the extent of the radon problems in Swedish mines was fully recognized, epidemiological studies on the lung cancer incidence among miners were started. There have been local studies in some mining districts, Zinkgruvan³, Boliden⁴, Kiruna⁵ and MalMBERGET⁶ and an overall study including all mining districts⁷. This overall study started in April 1971 and the main purpose was originally to find out whether there was any excessive incidence of lung cancer at all among the miners. The study was retrospective and covered the years 1961-1968. As there were no measurements on radon in mines at that time it was not possible, in the initial study, to make any detailed exposure-effect correlations, although such analysis is in progress.

The study involved, firstly, a comparison between expected and observed numbers of lung cancers within the mining districts and, secondly, a corresponding comparison for the miners alone. The results are shown in Table 3. Appropriate corrections have been made for the age distributions in the districts in calculating the expected number of deaths as based on the death rate in the county in which the respective districts were located. In the special study on miners, the expected number of deaths is calculated as being proportional to the number of employees and to the death rate in the district.

Group	Expected numbers of lung cancers	Observed numbers of lung cancers
All men other than miners in the mining district	125	99
Underground miners only	6	26
Above ground miners only	6	7

Table 3. Expected and observed numbers of lung cancers among men aged 20-64 years during the years 1961-1968 in mining districts for non-miners and miners. The observed numbers of lung cancers are those which relate to cancer which appeared and caused death within 5 years of cessation of employment at the mine.

It can be seen from Table 3 that there is a significant excess of lung cancers among underground miners. No excess is found among miners working above ground or among other men in the mining districts. The total number of men in the mining districts is 136,600 and the number of miners is about 13,000, of which about one half are underground workers. Of the 26 observed cases of lung cancer, 21 occurred among miners who had worked more than 10 years underground. Of the total of 48 miners belonging to this group (more than 10 years under-

ground) 21 (58 %), died within 5 years of ceasing employment and 39 persons (81 %) within 15 years.

There are obvious difficulties in estimating the true radon daughter exposures in a retrospective study without earlier measurements on radon daughters in the mines. Nevertheless, attempts are in progress to reconstruct the former ventilation conditions in some mines and more reliable values for the exposure may be given than are possible today. However, if the mines and corresponding miners are divided into two groups, one experiencing less than 30 pCi/l of radon daughters and the other more than 30 pCi/l according to the measurements since 1969, the distribution will be as shown in Table 4. The numbers in Table 4 are based on very recent follow-up studies including the years 1961-1971.

Radon daughter concentration pCi/l	Number of lung cancer deaths Age at death (years)			Number of mines	Number of miners 1966
	20-64	65-79	20-79		
<30	9	7	16	9	2,760
>30	27	23	50	11	2,099

Table 4. Observed numbers of lung cancer deaths among miners who have worked more than 10 years underground. All but 2 of the miners aged 20-64 years died within 5 years of cessation of employment at the mine. 30 pCi/l is equivalent to 0.3 WL.

It is clear from Table 4 that the relative numbers of lung cancer deaths are much greater in the mines with radon daughter levels in excess of 30 pCi/l than in the other mines.

Most of the observed lung cancer deaths among miners aged 20-64 years occurred during the last half of the period of investigation (24 of 36). The year of commencement of underground work for the deceased miners is more scattered: 8 miners during 1920-30, 12 miners during 1931-40, 12 miners during 1941-50, 3 miners during 1951-50 and 1 miner in 1961.

By assuming as a first approximation that the radon daughter levels in Swedish mines found by the measurements since the end of the 1960s are representative for earlier years, it is possible to make some qualified guesses as to the exposures. This assumption is, of course, uncertain but there is some justification for it. The ventilation has been improved with time and it is reasonable to assume that this would cause the radon levels to decrease. On the other hand the mines are continually being deepened, and that will increase the amount of crushed rock and also make the ventilation more difficult, i.e. the radon levels would increase with time if no other factors were involved. The resulting effect may well have been a relatively constant radon situation over the last few decades. On this assumption it is possible to determine an exposure-effect relationship according to Table 5. The exposure ranges in Table 5 are caused by the radon daughter ranges 1-10, 10-30, 30-100 and 100-300 pCi/l and the range of employment time for the deceased workers.

Estimated cumulative exposure, WLM		Employees underground (average)	Expected number of lung cancer deaths 1961-1971	Observed number of lung cancer deaths 1961-1971	Calculated annual mortality per 10 ⁴ miners from lung cancer	
Range	Average				Expected	Observed
2- 36	15	1001	1.25	2	1.1	1.8
13- 112	48	1852	2.31	7	1.1	3.4
48- 528	218	1488	1.85	15	1.1	9.2
170-1512	696	525	0.66	12	1.1	21

Table 5. Lung cancer deaths 1961-1971 among miners aged 20-64 years. All the deceased worked more than 10 years underground and all except two died within 5 years after cessation of employment.

The last column of Table 5 is plotted in an exposure-effect diagram in Figure 4 together with the results of the lung cancer studies in the USA among uranium miners⁸. There appears to be good agreement. Considering the considerable statistical errors and the uncertainty of the exposures it is, however, not possible to draw any conclusion about the dose-effect relation in the region of the low exposures around 100 WLM.

The average exposure is estimated to be 163 WLM (range 90-275 WLM corresponding to the radon daughter ranges mentioned above) assuming an average employment time of 30 years. If 1 WLM corresponds to 2 rads it follows that the rate of excess lung cancer is 1.7 cases per year per rad per million miners. The studies on uranium miners in the USA gave 0.9, on Newfoundland fluorspar miners 1.1 and on British underground iron miners 3.0⁸.

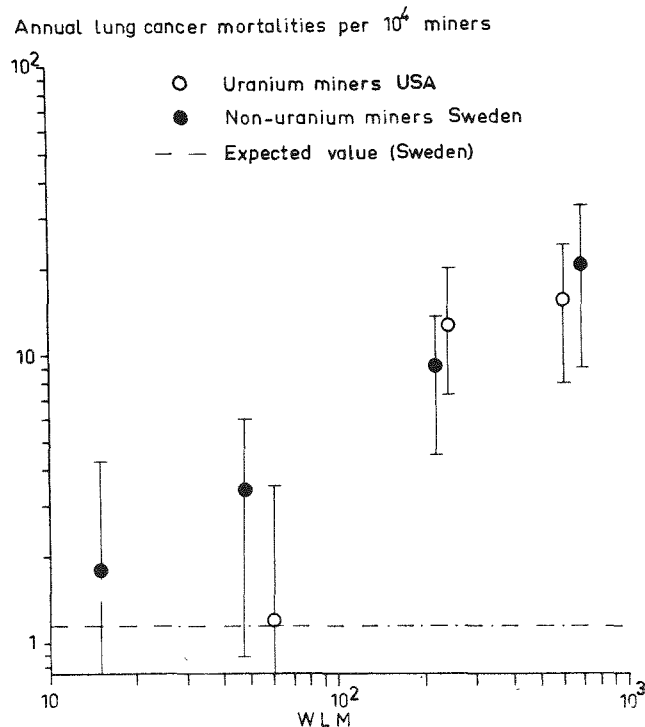


Fig. 4: Comparison between the lung cancer mortalities for American uranium miners and Swedish non-uranium miners as a function of the radon daughter exposure. 95 % confidence limits are shown for the mortality values.

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QUELQUES DONNEES NOUVELLES CONCERNANT LA PROTECTION
DES MINEURS DANS LES MINES D'URANIUM

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Abstracts

New data concerning the safety of miners
in uranium mines

Reports will be made, on the one hand, on the first results obtained regarding the action of radon and its elements in an experimental uranium mine at La Crouzille and, on the other hand, on the prospective use of a new portable individual dosimeter proportionate to the exposure of uranium miners to radon elements.

Introduction

Dans certaines mines d'uranium on a observé chez les ouvriers une augmentation de la mortalité par cancer du poumon. Cette augmentation semble d'autant plus marquée que les concentrations en radon et en descendants radioactifs sont plus élevées. Il importait donc d'intensifier les recherches dans ce domaine pour mieux connaître le comportement du radon et de ses descendants afin :

- de fixer des normes précises d'une sévérité suffisante mais non excessive étant donné les difficultés technologiques rencontrées pour abaisser les niveaux,

- d'améliorer l'efficacité des moyens de protection utilisés ou d'en développer de nouveaux,

- de mettre en place un système de surveillance du personnel précis, efficace et bien adapté aux conditions de la mine.

C'est pourquoi en 1969 nous avons décidé de créer dans une ancienne mine d'uranium, une mine laboratoire où il soit plus aisé de contrôler les différents paramètres expérimentaux que dans une mine en activité. Nous avons entrepris un programme de recherche dont nous présentons ici les principaux résultats actuellement acquis.

Nous décrivons également les résultats obtenus avec un nouveau détecteur individuel mesurant l'activité des descendants du radon.

I - Mine laboratoire

I/ Situation et implantation

La mine est située sur la division de La Crouzille à Fanay. Nous avons isolé du reste de la mine le quartier des Tenelles qui comprend deux niveaux séparés verticalement de 80 mètres. Le quartier est composé du montage d'entrée d'air M.560 reliant le jour au niveau 200 dans la galerie B.560; laquelle conduit en 800 m environ aux pieds des montages M. 500 et M. 50I raccordant le niveau 200 au niveau I20. La tête du montage 50I débouche dans la galerie B.30I du niveau I20. La B.30I communique avec le jour par le montage M.305. Les laboratoires sont installés, l'un au niveau 200, l'autre au niveau I20 en tête des montages M.50I et M.500; ils sont éclairés, chauffés, alimentés en courant électrique 220 V - 380 V, en air comprimé, et reliés par téléphone au central de la mine (fig.1).

La ventilation est assurée par deux ventilateurs de 18 chevaux chacun. Le ventilateur VI souffle l'air en tête du montage M.560; il assure l'aérage en mettant la mine en pression; V2 aspire dans la R.30I, au pied du montage M.305 et assure l'aérage en mettant la mine en dépression. Un système de trappes permet d'ajuster le débit de ces ventilateurs.

Les barrages nécessaires ont été réalisés pour isoler ce quartier des anciens travaux et limiter le circuit d'aérage à un parcours parfaitement connu.

2/ Aérage

Nous avons défini complètement les paramètres d'aérage de la mine laboratoire, en mesurant systématiquement les débits d'air dans chaque branche du circuit, et les différences de pression statique entre chaque noeud.

La mine est mise en pression ou dépression par l'action des ventilateurs VI ou V2. L'utilisation de diaphragmes permet de régler les débits de ventilation pour des valeurs comprises entre 2 et 10 m³/s environ.

La température, l'hygrométrie, la vitesse de l'air et la perte de charge entre la base et la tête des montages M.500 - 501 sont mesurées en permanence.

3/ Résultats

a/ Particules ultrafines non fixées sur des noyaux de condensation

Nous rappelons uniquement ici les résultats obtenus précédemment (I) (2) au moyen de filtres précédés ou non par des tubes de Zélény ou des batteries de diffusion : l'activité sous forme de particules ultrafines, c'est-à-dire atomes non combinés ou ayant fixé quelques molécules diverses, dont le coefficient de diffusion est compris entre $5,4 \cdot 10^{-2}$ et 10^{-2} cm²/s (et pour lesquelles il paraît plus justifié d'adopter un coefficient de diffusion moyen de $1,5 \cdot 10^{-2}$ cm²/s), représente en moyenne 3 % de l'activité du Radium A supposé à l'équilibre avec le radon; cette activité est inférieure à 10 %, sauf en cas d'arrêt de travail dans la mine où l'on peut trouver alors des valeurs de l'ordre de 25 %.

b/ Etude de l'influence de la ventilation sur la concentration en radon dans la mine

La concentration en radon est mesurée soit au moyen de prélèvements effectués dans des fioles recouvertes intérieurement de sulfure de zinc selon la technique en usage dans les mines françaises (4) soit au moyen de chambres de désintégration (5) de 15 litres fonctionnant à un débit de 10 l/mn permettant de mesurer des concentrations supérieures à $0,4 \cdot 10^{-10}$ Ci/l.

Deux chambres de désintégration ont été placées au niveau I20 (point L2) et des prélèvements ont été effectués régulièrement pour contrôler les résultats et suivre les variations rapides amorties par les chambres de désintégration. Les mesures ont été effectuées dans différentes conditions de ventilation : fonctionnement en pression en utilisant le ventilateur VI et fonctionnement en dépression en utilisant le ventilateur V2. Les différences de pression entre l'extérieur et le point L2 ont varié entre - 43 et + 55 mm d'eau. Chaque type d'aérage a été maintenu assez longtemps pour que la concentration ait atteint une valeur stable. Certains essais ont duré 4 jours (6).

Les résultats concernant les concentrations CRn, le débit de radon QRn en fonction du débit Q sont présentés dans les fig. 2 et 3.

On peut noter qu'à débit égal la concentration en radon dans l'air de la mine est plus faible quand la mine est en surpression que lorsqu'elle est en dépression; le facteur de réduction passe de 0,5 à 0,9 quand le débit augmente de 2 à 10 m³/s. Ceci confirme l'observation faite dans le cas d'une mine en activité, celle de La Chapelle Largeau, où l'on a eu une réduction de 20 % des concentrations moyennes qui passaient de $3,3 \cdot 10^{-10}$ Ci/l en 1965 la mine étant en dépression à $2,7 - 2,4 \cdot 10^{-10}$ Ci/l en 1966 et 1967, la mine étant en surpression.

On observe aussi que le débit de radon dans la ventilation au niveau du point L2, pour un débit de ventilation constant de 2 m³/s décroît régulièrement quand la pression croît en ce point (tableau I).

c/ Etude de l'influence de la ventilation sur la concentration en descendants du radon

Dans les mêmes conditions de ventilation on a effectué des mesures de l'énergie α totale suivant la méthode exposée par THOMAS (2). Les résultats exprimés en "Working Levels" WL sont également précisés dans la fig. 3.

On constate aussi qu'à débit égal, le niveau exprimé en Working Levels est plus faible lorsque la mine est en surpression; le facteur de réduction est constant et égal à 0,7 lorsque le débit est supérieur à 3 m³/s.

Ces résultats ne sont probablement pas valables dans tous les cas, mais ils confirment que la mise en surpression des mines d'uranium apporte une réduction des quantités de radon dans l'atmosphère de ces mines, cette réduction pouvant être appréciable dans certains cas.

II - Détecteur individuel pour la dosimétrie des descendants du radon

La surveillance individuelle du personnel est effectuée actuellement à partir de mesures locales, le calcul des quantités inhalées par chaque agent tenant compte du temps de travail aux différents lieux.

Un détecteur individuel capable de fournir une mesure correspondant à un prélèvement continu et représentatif de l'air inhalé par le mineur permettrait d'obtenir des valeurs plus significatives. C'est pourquoi nous avons développé un tel appareil breveté permettant d'évaluer l'énergie α totale des descendants du radon (7).

Pour évaluer cette énergie il est nécessaire de mesurer séparément l'activité du Radium A et celle du Radium C' qui se trouvent dans des proportions très variables. L'appareil (fig. 4) pesant environ 150 g comprend un dispositif de prélèvement alimenté par batterie permettant de collecter les descendants du radon sur une membrane millipore; le débit est de l'ordre de 4 litres par heure. Un détecteur constitué par du nitrate de cellulose coloré en rouge (LR II5 Kodak Pathé) est placé face au filtre à environ 20 mm. Les particules α émises au niveau du filtre doivent traverser un collimateur à deux canons et des écrans avant d'atteindre le nitrate de cellulose.

Sur le premier canon l'épaisseur de l'écran est choisie de façon à arrêter les α du Radium A et à détecter ceux du RaC' dont l'énergie résiduelle est de l'ordre de 3 MeV.

Sur le deuxième canon l'écran est tel que l'énergie résiduelle des α du RaA est de l'ordre de 3 MeV alors que celle du RaC' est de l'ordre de 5 MeV.

Les caractéristiques du nitrate utilisé sont telles que les α de 3 MeV sont détectés et ceux de 5 MeV ne le sont pas.

Le premier canon permet donc de mesurer le RaC' et le deuxième le RaA, ce qui permet d'évaluer aisément l'énergie α totale des descendants du radon accumulés sur le filtre pendant un temps très long de l'ordre de la semaine ou même du mois.

Grâce à l'utilisation des collimateurs et des écrans les traces de particules apparaissent après attaque chimique sous forme de taches blanches toutes identiques sur fond rouge (fig.5). Il est alors aisé d'évaluer le nombre de traces par la mesure du flux lumineux de longueur d'onde convenablement choisie qui traverse le nitrate (8).

Les premiers résultats obtenus avec un prototype dans la mine expérimentale ont montré une bonne concordance pour une dizaine d'essais effectués pour des états d'équilibre variés avec les valeurs fournies par la méthode de THOMAS.

Actuellement quatre appareils sont en cours d'essai de longue durée dans les conditions d'utilisation au fond de la mine.

Conclusions

La protection contre le radon dans les mines d'uranium ayant une importance considérable, des expérimentations de longue durée ont été entreprises dans une mine expérimentale. On a pu déjà constater que la fraction de particules ultrafines non fixées ayant un coefficient de diffusion moyen voisin de $1,5 \cdot 10^{-2} \text{ cm}^2/\text{s}$ et non pas égal à $5,4 \cdot 10^{-2}$ devrait être prise égale à 3 % de l'activité du Ra A à l'équilibre.

En ce qui concerne les moyens pour abaisser les niveaux de pollution, la mise en surpression des mines est d'ores et déjà à considérer comme efficace. Quant à la surveillance du personnel, l'utilisation de détecteurs individuels capables d'évaluer correctement l'énergie α totale quel que soit l'état d'équilibre est à prévoir au moins pour une partie des mineurs, les premiers résultats des essais étant satisfaisants.

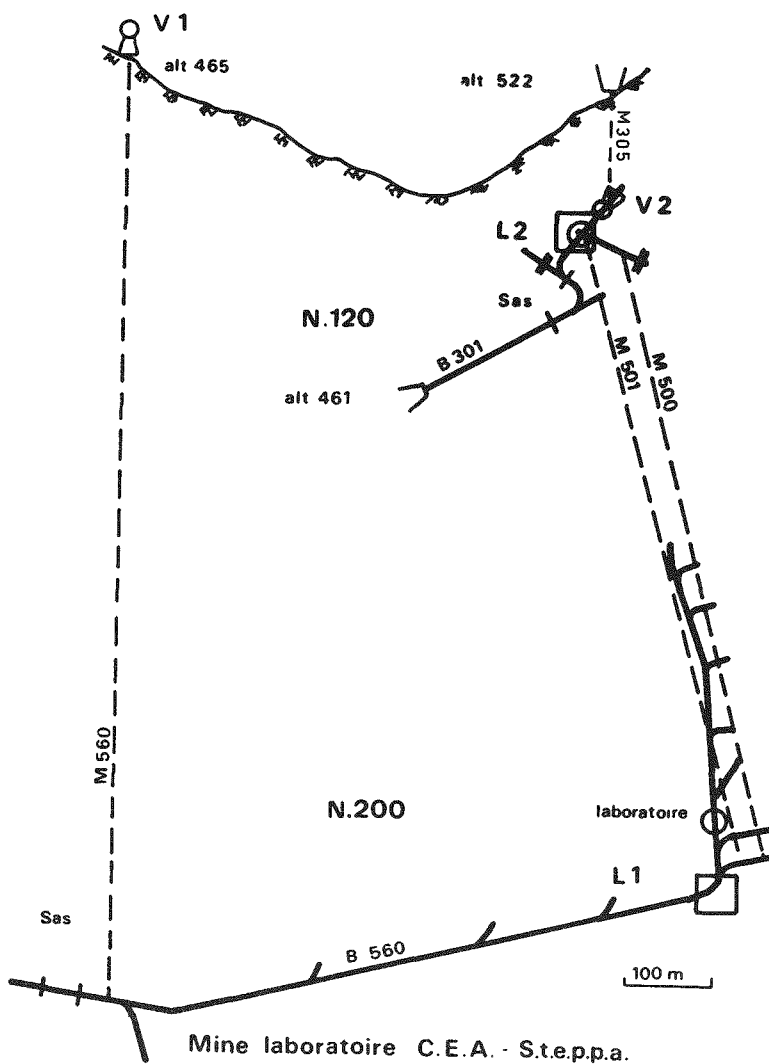


Fig. 1

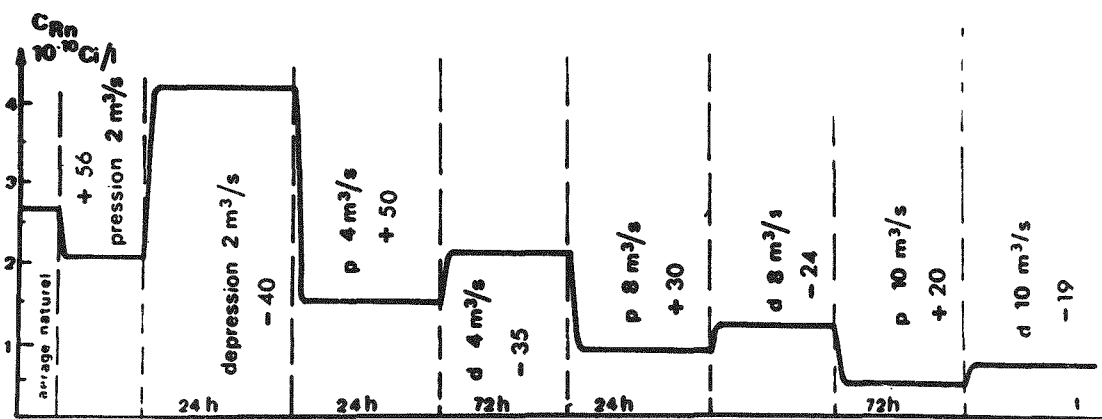
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Tableau I

Débit de radon (10^{-7} Ci/s)	Pression (mm d'eau)
9,2	- 45
7,4	- 2
7,3	+ 2
5,7	+ 56

Variation du débit de radon dans la mine
 en fonction de la pression
 pour un débit constant de $2 \text{ m}^3/\text{s}$



Influence du mode d'aerage sur la concentration du radon

Fig.2

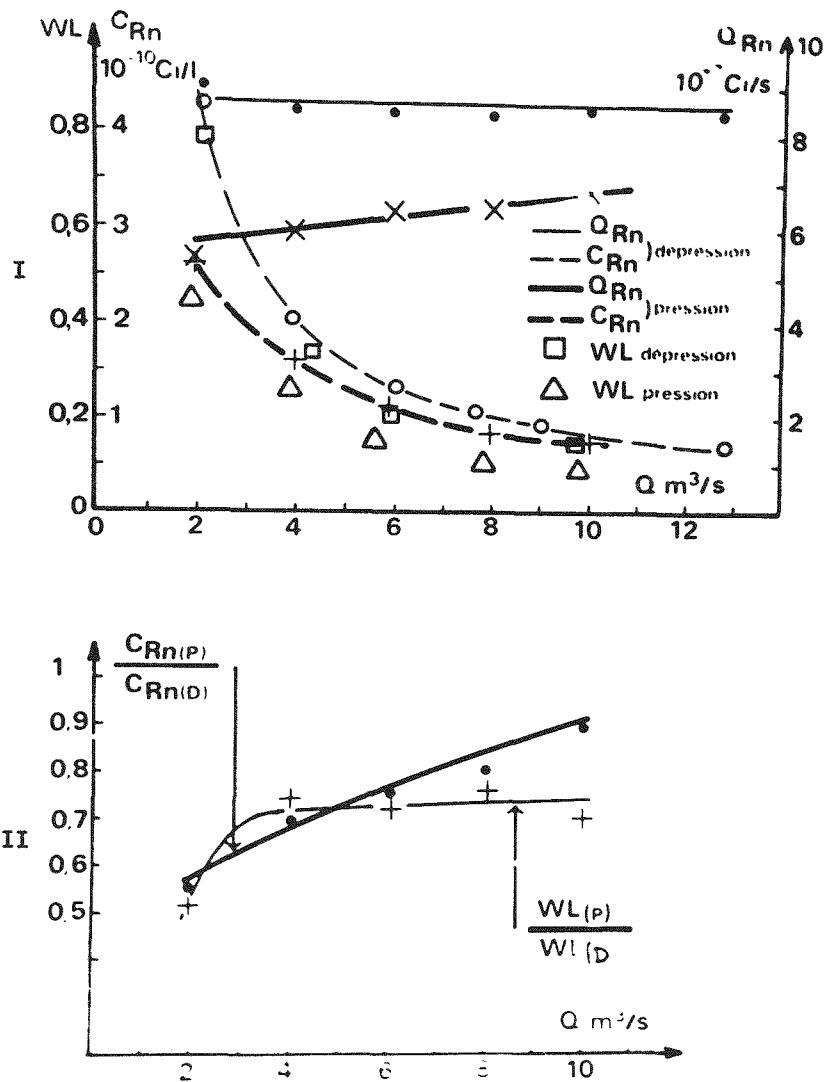
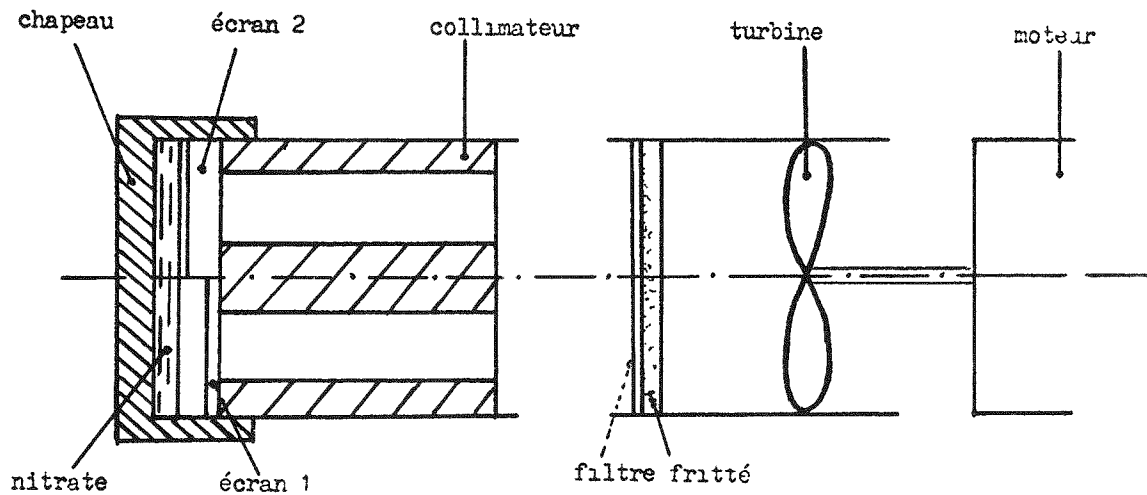


Fig. 3

- I - Concentration en radon et en descendants du radon exprimée en WL et débit de radon en fonction du débit de ventilation, la mine étant en dépression.
- II - Rapport entre la concentration en radon ou la concentration en descendants du radon pour la mine en pression et celle correspondante pour la mine en dépression, en fonction du débit de ventilation.

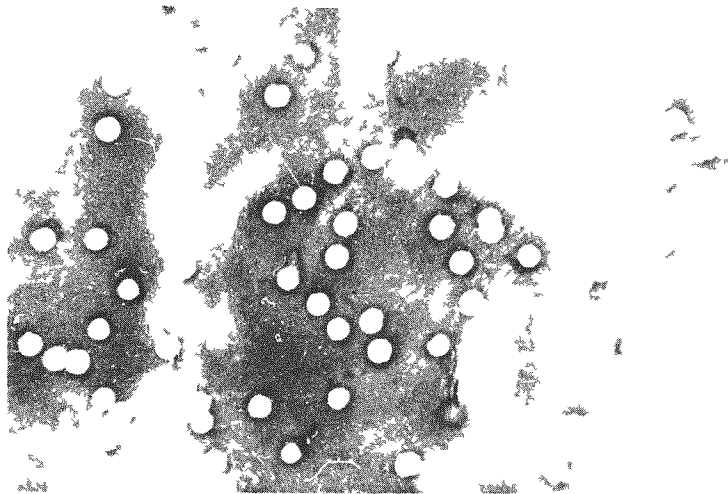


- Schéma de principe de la tête de prélèvement

Fig. 4

Traces de particules α provenant de Radium A
 dans le détecteur LRIIS Kodak-Pathé
 utilisé dans l'appareil de prélèvement individuel
 pour la dosimétrie des descendants du radon

Fig. 5



LONG-TERM MEASUREMENTS OF RADON DAUGHTER ACTIVITY IN MINES

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Abstract

A rugged, portable filter monitor for the long-term measurement of Rn-daughter activity in mines with direct indication of the cumulative exposure is described. The monitor is equipped with rechargeable batteries and enables continuous or fractionated air sampling at preset time intervals over a period of 1 - 2 weeks with one battery charge. The results of continuous measurements over a one year-period in a fluorspar mine are discussed.

Introduction

A significant excess of lung cancer mortality with increasing accumulated Radon(Rn)-exposure has been observed among uranium miners in the USA and CSSR. This fact emphasizes the suspicion that workers in mines with high Rn-content in air belong to that groups of radiation workers with the highest somatic radiation risk. Compared with this risk the surveillance of the radiation exposure of these miners is still not adequate. The main causes for this inconsistency are the complex distribution of Rn and its daughters in mine air, which varies with time and place, and the difficulties of personnel air monitoring under the abnormal working conditions in mines.

In the past air monitoring in these mines was restricted mainly on measurements of Rn or its daughters in single air probes, which were taken in more or less large intervals of time. Taking into account the varying air activity in a mine working area robust, portable and battery-operated monitors are needed which enable long-term measurements and indicate the accumulated exposure over a long time period. Several instruments of this type were proposed and tested in the last years.¹⁻⁶

With respect to their applicability in uranium and fluorspar mines we have studied different methods to determine the time-integral of the activity concentration of Rn and its daughters. For the continuous measurement of Rn-gas itself we used the electrostatic deposition of RaA-ions, formed by decay of Rn-atoms in a chamber with Rn-permeable walls. The results however have indicated, that the Rn-sensitivity of this method depends strongly on the air humidity.⁶ This method seems therefore not suitable for

Rn-monitoring in mines.

For long-term measurements of the accumulated exposure to Rn-daughters in mines we have developed on the basis of the filter method portable monitors with low-power consumption. In this paper the design and properties of this monitor and the results of test measurements in a fluorspar mine are described.

Description of the Air Monitor

Two types of surveying instruments were developed: A direct-indicating monitor with a Silicon- α -detector and a nondirect-indicating instrument using track etch foils as integrating α -detector.

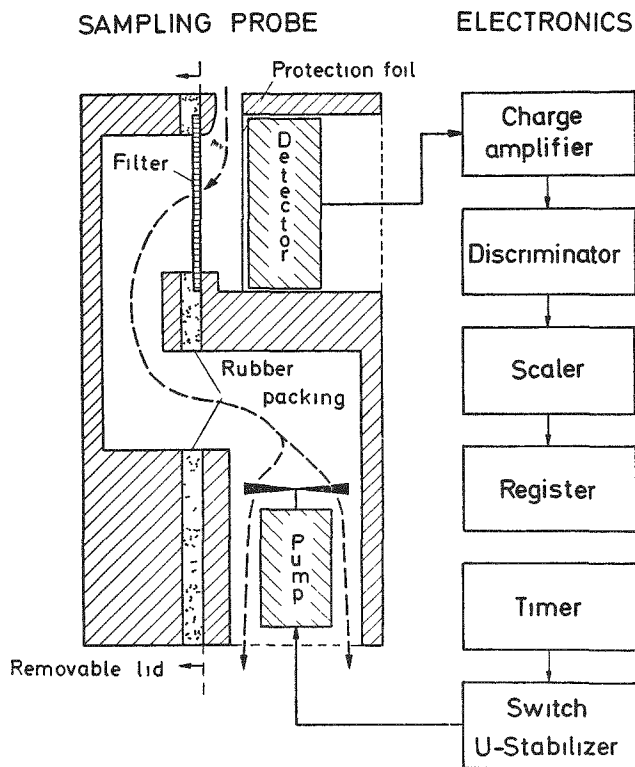


Fig. 1:

Cross section through the sampling probe of the monitor and block diagram of the electronics

Figure 1 shows a schematic cross section through the sampling probe of the direct-indicating instrument and a block diagram of its electronics. The outside air is sucked in through slits in the sampling probe, passes the fibrous filter and is exhausted on the other side of the probe. The sampling probe can be easily opened for filter replacement. The small air blower is mounted in the sampling probe; its operation voltage of 3,5 V is stabilized to assure a constant air flow-rate. With the normally used cellulose-asbestos filters (effective filter diameter 18 mm) a flow-rate of 2.5 liter/hour was adjusted. As α -detector a p-Silicium semiconductor with an active surface of 240 mm² ($\phi=17.5$ mm) is used, whose surface is protected by a thin mylar foil of 1 mg/cm² thickness. The distance between filter and detector surface is 5 mm.

After amplification and pulse height discrimination the detector pulses are counted by a combination of an electronic scaler and a 4-digit mechanical register. The electronic part includes also a timer and switching circuit for the air blower, which enables automatic, fractionated air sampling in preset time intervals. All electronic parts are designed for low power consumption. The total power consumption of the monitor is about 300 mW, from which about 200 mW are required for the air blower. For the power supply rechargeable dryfit PC-accumulators are used, which enable with one battery charge an operation time of 8 - 9 days at continuous sampling and of about 18 days at fractionated sampling.

The electronics, the counter and the accumulators are enclosed by a stable, water and dust protected metal housing, to which the sampling probe is flanged on. All operating elements and plug sockets for external connections (rate meter, pulse height analyser) are mounted behind a lid to prevent contamination by dust and undesired changes of the adjusted operation values. Only the mechanical register can be read through a window in the metal housing. The instrument dimensions are 23 cm x 12 cm x 32 cm (height). Its total weight is 7 kg, from which about 5 kg falls to the accumulators.

In a second type of this instrument the Si- α -detector is substituted by a track etch foil with the same effective diameter; in this type the detector electronics and the counter are omitted. Foil etching and α -track counting is done by the usual techniques and are described in detail elsewhere.⁶

Instrument Calibration

For the monitoring of Rn-daughter mixtures in mines and room air the concept of potential α -energy concentration and the unit 1 WL = 1.3×10^5 (pot. α -) MeV/liter air have been introduced. As described earlier the described instrument was constructed with the aspect to determine the time integral over this energy concentration over long exposure periods. The total number $Z_\alpha(T)$ of α -tracks or α -pulses, respectively, counted with the instrument during a time period T is connected with this integral or accumulated exposure E(T) by the equation⁶:

$$Z_\alpha(T) = \frac{\beta \eta f v}{p} \cdot E(T)$$

In this equation means η the counting efficiency of the used type of detector, $f > 0.99$ the deposition efficiency of the filter, $v = 2.5 \pm 0.3$ liter/hour the air flow rate and $p = 7.68$ MeV the potential α -energy of one ^{218}Pb (RaB)- or ^{214}Bi (RaC)-atom.

The correction factor β depends on the relative composition of the Rn-daughter mixture in the measured air, which varies with the ventilation rate λ_v and the rate constant λ_a for the attachment of free daughter atoms to particles in the considered mine area.⁶ It was calculated on the basis of the box model for Rn-atmospheres which was developed by one of the authors⁷ and is given in figure 2. This graph shows that the variation range of the correction factor β is rather small and a constant value $\beta = 1.05$ can be applied to most mine and room atmospheres with sufficient accuracy.

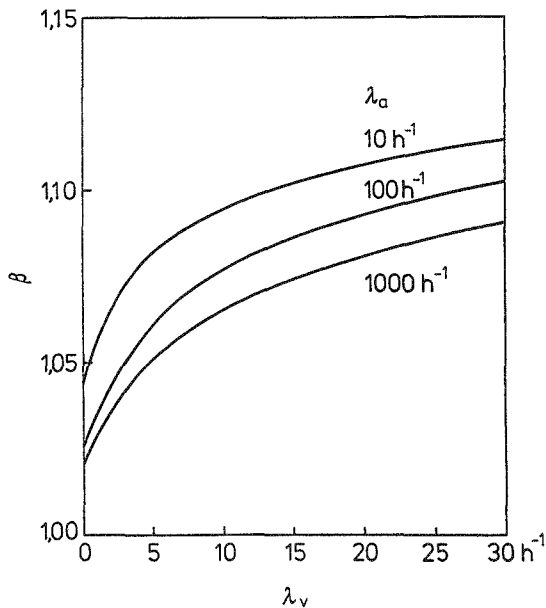


Fig. 2:
Correction factor β for
instrument sensitivity
(see text)

The counting efficiency η of the direct-indicating instrument with a Si- α -detector was determined by comparison with calibrated α -sources. At the normal operational conditions used it results $\eta_{Si} = 0.18 \pm 0.01$. With this value it follows an instrument sensitivity in the case of continuous sampling:

$$\begin{aligned}
 S &= Z_{\alpha}(T)/E(T) \\
 &= 0.061 \pm 0.011 \text{ counts per (pot.}\alpha\text{)MeV}\cdot\text{hour/liter air} \\
 &= 7900 \pm 1400 \text{ counts per WL}\cdot\text{hour}
 \end{aligned}$$

This corresponds to a sensitivity of 79 ± 14 counts per pCi \cdot hour/liter air of each daughter nuclide in the case of radioactive equilibrium in air. The built-in scaler enables a reduction of this sensitivity in steps of 1/2 to a 1/128 of this value or 62 counts per WL \cdot hour. In addition the sensitivity can be reduced by the built-in timer for fractionated air sampling. The background counting rate is about 5 counts per hour. The lower detection limit of the instrument is therefore comparable with the mean concentration of Rn-daughters in atmospheric air.

The sensitivity of the instrument with track etch foils was determined by simultaneous field measurements with both types of instruments. Depending on etching conditions and the used foil material it is about a factor 0.2 - 0.6 lower than the sensitivity for the direct-indicating instrument.

Test Measurements

Test measurements with 2 instruments of each type were performed so far in 3 fluor spar mines in East Bavaria.⁶ One direct-indicating monitor was continuously in use for one year at the same working area in a mine drift and was operated and controlled by the foreman of the miners in this area. He read off the counting register of the instrument normally at the beginning and the end of

each working shift. The resulting time variation of the potential energy concentration in the mine air during this one year-period is shown in figure 3. The annual mean value was about 1 WL, whereas the daily mean values are varying in the range from 0.4 - 3 WL. Figure 3 indicates that the short-time variations from day to day in this mine area are rather small. However, a rather strong long-term variation of the Rn-daughter level is observed.

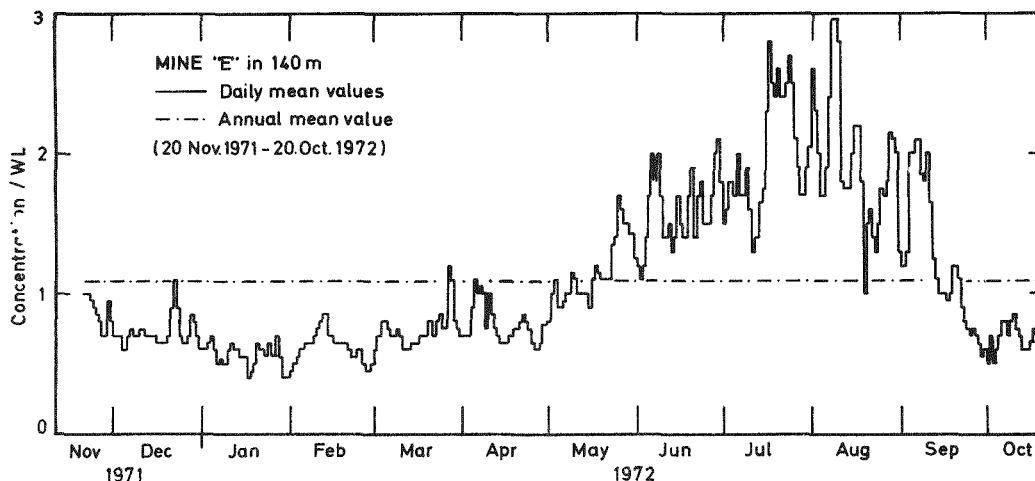


Fig. 3: Variation of the continuously measured Rn-daughter activity in a working area of a fluorspar mine in East Bavaria (Nov. 1971 - Oct. 1972).

This variation is mainly due to the change of the air ventilation during the extension of the mine gallery. During the period from November 1971 till about May 1972 a rather constant activity level of about 0.5 - 1.0 WL was observed. In the following months the fresh-air supply to the driving gallery was reduced. During this period the air-activity increased and reached rather high values of 2 - 3 WL in July - September 1972. After break-through of a new wind gate the supply with air of low Rn-content increased. Subsequently the Rn-level in the working area decreased strongly and reached a rather constant level of about 0.6 WL in October 1972.

The variation of the Rn-exhalation from the walls of the gallery due to the mining activity was probably not so significant in this fluorspar gallery, but might be certainly of more importance in other galleries, especially in uranium mines.

In either case, these test measurements indicate the necessity of long-term Rn-monitoring to get more information about the real cumulative exposure of miners.

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ОЦЕНКА РАДИОАКТИВНОГО ФАКТОРА НА ПРЕДПРИЯТИЯХ
ПРОМЫШЛЕННОСТИ РЕДКИХ И ЦВЕТНЫХ МЕТАЛЛОВ.

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Summary

When working with natural radioactive raw materials of different mineral composition and different specific activity due to the type and conditions of the particular jobs the environment in which man works can reveal different extents of activity of individual components of the factor of radiation.

Surface jobs with mineral raw materials and their processed products are extremely typical for their dusts containing long-term α -irradiators. Such dusts pollute the air environment of some production premises of ore-dressing and chemical and metallurgical enterprises.

Under examination was the state of health in workers with long-term of service. No harmful effects upon the respirative organs which could be directly related to the radiation factor have been discovered so far. However the longest term up to 20 years was only in a small group of the examined.

The amount of Th incorporated into the workers' lungs evaluated by the preliminary data taken from the entire body and the Tn content in the respirated air did not exceed 150 pCi.

The authors studied 3500 white ordinary rats intratracheally injected with dusts of various specific activity as well as chronically inhaled for long terms. It has been found out that with a combined dust-radiation action the lowest effective dose to affect the lungs is close to 200 rem.

In view of the increased fibrogenic properties of dusts containing radioactive elements the authors stress the necessity to consider the long-term natural radioactive component of mineral dusts when fixing the permissible limits of general pollution within the operation zone of the production premises. The authors regard as open the question on possible distant effects in workers with 30-50 years of service since with the length of service increased it is quite possible for the lungs to absorb doses equal to the maximum permissible ones.

Радиационная опасность на предприятиях промышленности редких металлов обусловлена содержанием в добываемых и обогащаемых породах естественно-радиоактивных элементов ряда тория и урана. Несмотря на незначительное содержание в исходном минеральном сырье естественно-радиоактивных элементов (от $1 \cdot 10^{-11}$ кюри/г до $1 \cdot 10^{-10}$ кюри/г), в ходе изучения условий труда выявлены работы, выполнение которых может сопровождаться загрязнением воздушной среды радиоактивными аэрозолями и газами.

Для наземных работ с минеральным сырьем и продуктами его переработки особенно характерно постоянное содержание в пыли, загрязняющих воздушную среду некоторых рабочих зон и производственных помещений, долгоживущих альфа-излучателей. К таким работам относятся работы на шарошечных станках в открытых карьерах, все виды работ при сухих методах обогащения концентратов редких и цветных металлов на обогатительных фабриках, многочисленные операции по доводке концентратов, шихтовке, затариванию и т.д. Поэтому, понятен наш интерес к оценке роли естественно-радиоактивного компонента промышленных пылей предприятий промышленности редких и цветных металлов, тем более, что ряд исследователей, оценивавших последствия радиационного воздействия на рабочих урановых рудников^{1,2}, высказывали предположение о том, что пыль находится среди таких факторов, которые могут обусловить возникновение рака легких при дозах облучения меньше тех, которые признаны необходимыми для индуцирования рака при "чистом" радиационном воздействии.

Сравнение уровней заболеваемости различных профессиональных групп из числа лиц, работающих на предприятиях промышленности редких металлов и изучение состояния здоровья стажированных рабочих не выявило повреждающих эффектов со стороны органов дыхания, которые можно было бы поставить в прямую связь с воздействием радиационного фактора. Но наибольший стаж - до 20 лет, был достигнут лишь в небольшой группе обследованных рабочих. Содержание тория в суточном количестве биосубстратов не превышало 20 мкг с калом, 10 мкг с мочой. Количество инкорпорированного тория в легких рабочих, оценивавшееся по результатам предварительных измерений на счетчике всего тела и по содержанию торона в выдыхаемом воздухе, не превышало - 150 пикзюри.

На 3500 белых беспородных крыс проведены экспериментальные исследования по изучению патогенных свойств труднорастворимых торийсодержащих минеральных пылей, в связи с их радиоактивностью. При этом в экспериментах с однократным интратрахеальным введением использовались пыли различных редкоземельных торийсодержащих руд и концентратов цирконовой и лопаритовой группы с присущей им удельной активностью и ее искусственным увеличением до $1 \cdot 10^{-6}$, $1 \cdot 10^{-7}$ кюри/г. Увеличение удельной активности исследуемых пылей за счет искусственных добавок ферриторита, торанита или искусственной ThO_2 нами применялось для усугубления условий эксперимента, которые бы позволили, пусть даже в расчете на нереальный "худший" случай, выявить влияние радиационного фактора на фоне пылевого.

При обычной постановке исследований, с использованием пылей только природной удельной активности, как мы уже знали, и исходного уровня удельной активности и 12-ти месячного срока наблюдения было недостаточно для того, чтобы составить представление о возможных неблагоприятных последствиях воздействия такой комбинации факторов - пылевого и радиационного³.

Эксперименты с хроническим ингаляционным осуществлялись с

помощью ингаляционных установок⁴. Запыленность воздуха в камере с учетом дисперсности пыли и коэффициента ее задержки, была выбрана равной 300-400 мг/м³ с тем, чтобы к концу затравки в легких накопилось 50-75 мг вещества - количества, достаточного для развития в легких пневмокониотического процесса, на фоне которого выявлялось влияние радиационного компонента пыли.

Таким образом, в экспериментальных условиях создавалась модель хронических пылерадиационных воздействий на фоне различных поглощенных доз на легкие - от нескольких бэр, до 750 бэр и более. Величина суммарной дозы, в каждом конкретном случае диктовалась возможностью создания аналогичной дозы в процессе 50-летнего профессионального контакта лиц, работающих с естественными радиоактивными веществами разной удельной активности, не превышающей в производственных условиях $n \cdot 10^{-8}$ кюри/г.

Экспериментальные исследования выявили, что при комбинированном пылерадиационном воздействии минимально-эффективной дозой на легкие является доза близкая к 200 бэр. Выявлению этой величины способствовали данные по изучению темпов прироста веса тела животных в динамике, функции внешнего дыхания, состояния показателей периферической крови, продолжительности жизни животных, а также результаты исследования биохимических и патоморфологических изменений тканей легкого, как критического органа.

При экспериментальном изучении сравнительной патогенности естественно-радиоактивных минеральных пылей и тех же пылей с усиленным за счет искусственных добавок радиационным компонентом многие из вышеуказанных показателей дали многочисленные свидетельства усиления фиброгенности пылей по мере увеличения их удельной активности.

Так, рис. 1 демонстрирует увеличение сухого веса легочной ткани, наблюдающееся как в сериях с рудными пылями наибольшей удельной активности, так и в аналогичных, по активности, сериях с пылью концентрата, что особенно отчетливо (достоверность >95%), проявляется к 12 месяцам от начала эксперимента.

К этому же сроку увеличивается абсолютное количество окси-пролина (рис. 2) в легочной ткани животных, подвергнутых воздействию пылей наибольшей удельной активности, суммарных и нерастворимых белков, снижается интенсивность включения меченой по C¹⁴ аминокислоты лизина и глицина. Патоморфологические исследования выявили признаки слабо прогрессирующего диффузно-очагового пневмокониоза, хронического бронхита и бронхиолита, плазматизации лимфоидных клеток лимфатических узлов. В эксперименте отмечено некоторое усиление патогенных свойств пылей к 15-18 месяцам от начала воздействия за счет общей дозы, близкой к 200 бэр, что выражалось в более очаговом характере пневмосклероза (рис. 3) и увеличении лимфоретикулосарком легких животных.

При исследовании усугубленного влияния смесей циркония с двуокисью тория и чистой двуокиси тория отмечено снижение продолжительности жизни животных (рис. 4,5). Рисунки демонстрируют сложность происходящих в организме животных процессов, так как при дозе большей 200 бэр на легкие в сериях 2 и 3, в одном случае, у животных этих серий ET₅₀ было существенно ниже, чем таковое у животных возрастного физиологического контроля (К) и животных 1 серии (рис. 4). В другом, несмотря на дозу превышающую 200 бэр (2 серия), ET₅₀ было аналогично таковому у контрольных животных (рис. 5). Основные статистические параметры, приведенные на рис. 4-5, получены при обработке данных по методу Литчфильда и Блисса.

На основании экстраполяции результатов экспериментальных исследований было установлено также, что период полувыведения тория,

входящего в состав пыли природных соединений-минералов циркона и лопарита, попавших в легкие, для человека находится в пределах от 4,8 до 5,2 лет. Тогда при соблюдении допустимых уровней запыленности воздуха рабочей зоны расчетным путем получено, что доза, поглощенная легкими рабочих, занятых на производстве цирконового концентрата, может достигать 300 бэр, а при производстве лопаритового концентрата 1300 бэр в течение 50-летнего профессионального контакта.

Вместе с тем, ПДД для персонала за 50 лет равна 750 бэрб. В связи с этим установленная нами в эксперименте минимально эффективная доза, равная 200 бэр, может быть свидетельством необходимости учета содержания в пылях радиоактивного компонента при гигиеническом нормировании пылей в воздухе производственных помещений.

Таким образом на основании проведенных исследований можно сделать следующие выводы:

1. Минимальная эффективная доза на легкие белых беспородных крыс при комбинированном пылерадиационном воздействии равна 200 бэр.

2. Некоторые виды работ с естественно-радиоактивными веществами, такими как лопаритовый концентрат, должны относиться к радиационно-опасным, так как в производственных условиях возможно получение легкими рабочих за 50 лет профессионального контакта доз, больших 200 бэр.

3. При работах с веществами малой удельной активности (не более $1 \cdot 10^{-9}$ юри/г) радиационная безопасность может быть достигнута соблюдением общегигиенических требований, касающихся допустимых уровней общей запыленности воздуха рабочей зоны, которые должны устанавливаться с учетом наличия в пыли долгоживущего альфа-активного компонента.

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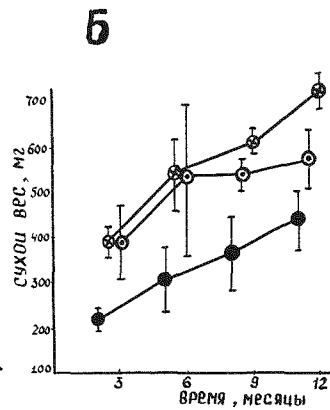
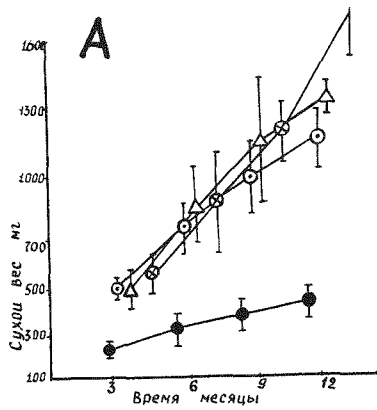


Рис. 1. Сухой вес легочной ткани при однократном интратрахеальном введении пылей цирконовых руд - А и цирконовых концентратов - Б

Обозначения

А
 - 10^{-11} кюри/г
 Δ - 10^{-9} - " -
 - 10^{-7} - " -

Б
 - 10^{-10} кюри/г
 - 10^{-8} - " -
 - контроль

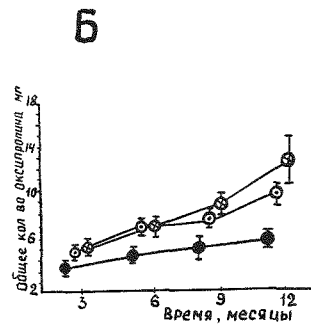
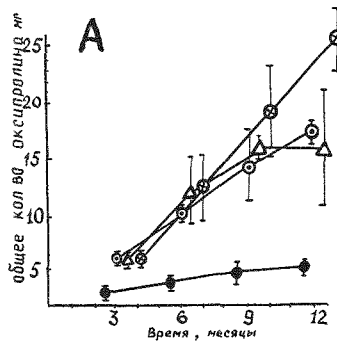


Рис. 2. Оксипролин легочной ткани.

Обозначения те же.

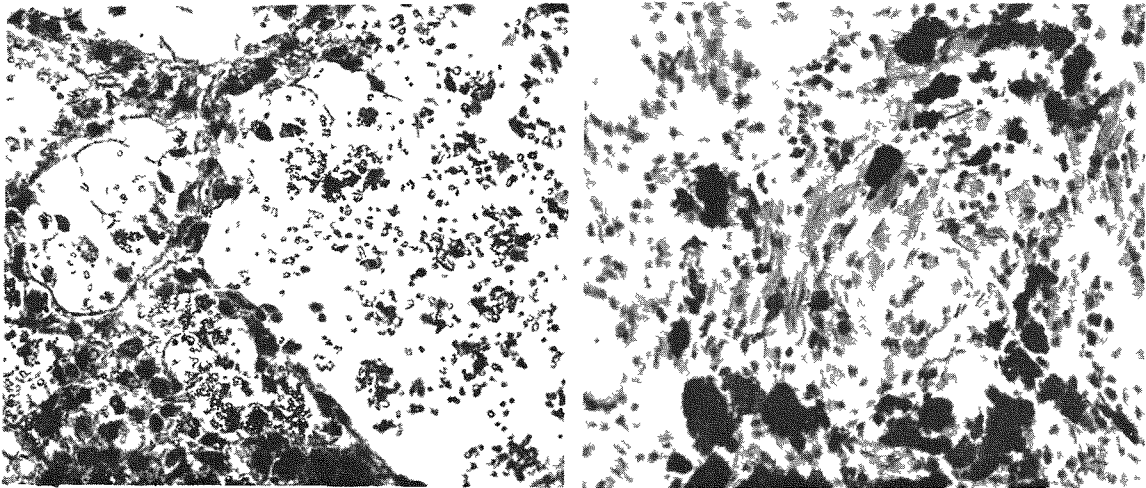


Рис. 3. Легкие крысы при ингаляции пыли: а) лопаритового концентрата через 6 мес. Кониофаги с большим количеством пылинок не гибнут, пролиферация гистиоцитов альвеолярных перегородок; б) чистой двуокиси титана через 15 мес., резко выраженный фиброз в зоне отложения пыли. Гематоксилин-эозин, $\times 350$.

обознач серии	ET, сутки, нрч			K O		1 O		2 O	
	16%	50%	84%	ET ₅₀	f _{TR}	TR	f _{TR}	TR	f _{TR}
K O	400	500(498-570)	625	1,14					
1 O	353	415(375-457)	483	1,06	1,2	1,175			
2 O	460	530(485-577)	650	1,09	1,06	1,167	1,27	1,14	
3 O	220	304(268-340)	425	1,145	1,65	1,205	1,36	1,18	1,74
обознач серии	N	S	f s	K O		1 O		2 O	
				SR	f _{SR}	SR	f _{SR}	SR	f _{SR}
K O	12	1,24	1,12						
1 O	8	1,16	1,077	1,07	1,14				
2 O	19	1,18	1,08	1,05	1,142	1,02	1,115		
3 O	21	1,375	1,145	1,06	1,19	1,185	1,165	1,164	1,166

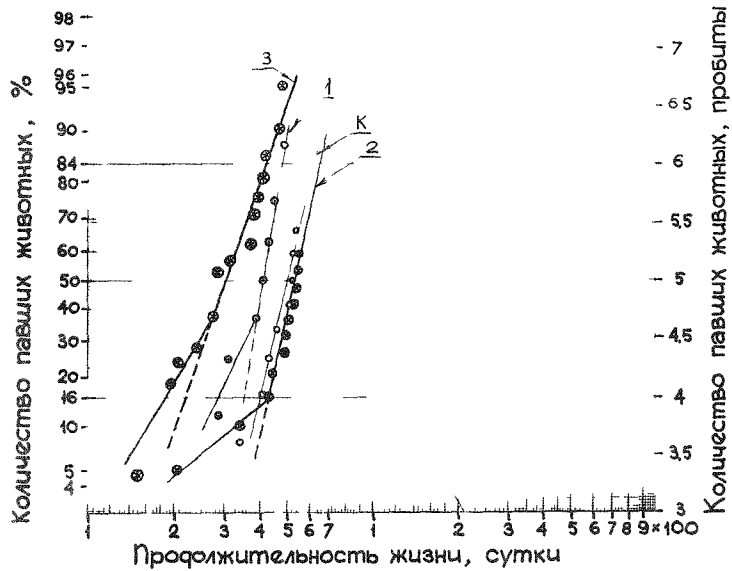


Рис 5 Выживаемость животных при хроническом ингаляционном воздействии аэрозолей лопаритового концентрата / I серия - 0,65% Th; II - 10,16% Th; III - 46,2% Th; K - контроль.

обознач серии	ET, сутки, нрч			K O		1 O		2 O		3 O	
	16%	50%	84%	ET ₅₀	f _{TR}	TR	f _{TR}	TR	f _{TR}	TR	f _{TR}
K O	396	545(480-620)	750	1,135							
1 O	352	460(394-545)	600	1,17	1,19	1,22					
2 O	330	395(330-470)	480	1,195	1,18	1,24	1,16	1,27			
3 O	264	370(280-480)	520	1,33	1,47	1,36	1,24	1,38	1,07	1,395	
4 O	284	370(325-420)	490	1,195	1,47	1,19	1,22	1,07	1,24	1,0	1,36
обознач серии	N	S	f s	K O		1 O		2 O		3 O	
				SR	f _{SR}	SR	f _{SR}	SR	f _{SR}	SR	f _{SR}
K O	17	1,37	1,12								
1 O	9	1,295	1,18	1,18	1,175						
2 O	8	1,295	1,14	1,18	1,0	1,19					
3 O	5	1,39	1,24	1,27	1,07	1,27				1,28	
4 O	16	1,3	1,095	1,15	1,0	1,16	1,17	1,26	1,07		

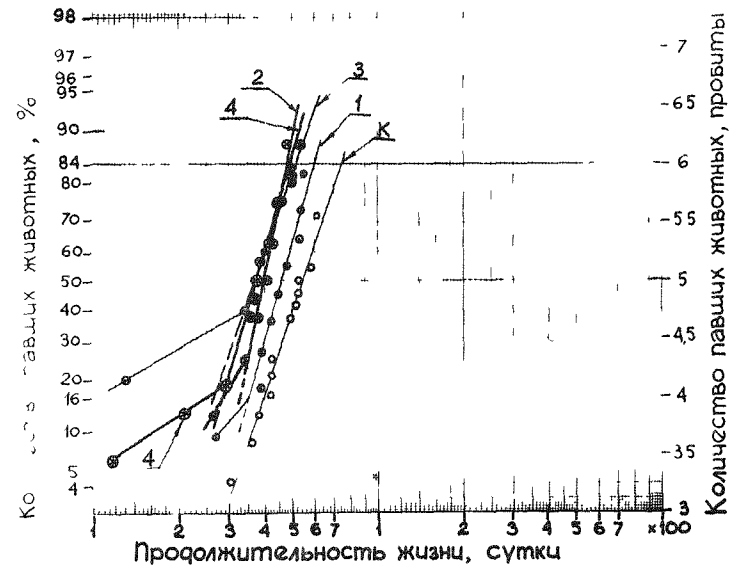


Рис 4 Выживаемость крыс при ингаляционном воздействии аэрозолей цирконового концентрата разной удельной активности / 1 - 0,025% Th, 2 - 4% Th / чистой двуокиси тория / 4 / и цирконового концентрата в сочетании с торием / 3 / ET - эффективное время / продолжительность жизни для 16, 50 и 84% павших животных /.

A RADON DAUGHTER MONITOR FOR USE IN MINES

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Abstract

A self-contained monitor for the measurement of airborne RaA, RaC and the working level, WL, in the shortest practicable time and with a minimum of calculation is described. The ratio of two gross alpha-counts gives, directly, the ratio of airborne alpha-emitter concentrations, RaC':RaA, after collection on a filter. This is achieved because the contribution from airborne RaB to the measured alpha-activity is minimised by choosing short sampling and counting times. The RaA concentration is derived from the first alpha-count, during air sampling. WL is derived from the second alpha-count. No back-ground correction is required. Systematic errors in estimated quantities are small. With a total measuring time of eleven minutes, the limits of detection are approximately 1 pCi/litre for RaA and 10^{-4} for WL.

Introduction

A radiation hazard from the short-lived radon daughters in uranium and other metal mine atmospheres has been convincingly shown.¹ Most of the epidemiological studies made in this context have used the working level (WL) as the unit of exposure. 1 WL represents the concentration of radon daughter activity in an atmosphere, and is defined as any combination of the daughters (RaA, RaB, RaC and RaC') in one litre of air that results in the emission of 1.3×10^5 MeV of potential alpha energy in decaying to RaD. The unit is numerically equal to the total alpha-energy arising from the decay of 100 pCi/litre each of RaA, RaB and RaC, but does not depend on the state of daughter disequilibrium. Nevertheless, it is often useful to know the RaA concentration and state of daughter disequilibrium as well as the WL. The concentration of the first daughter, RaA, because of its short half-life, responds rapidly to the radon gas concentration. This information can be valuable, for example, in locating a point of injection of fresh radon and/or daughters into an airstream. With the location of such a point, corrective action can be considered. In this case, a quick, on-the-spot assessment is very helpful, as additional measurements may be indicated. For this purpose, the RaA and RaC concentrations adequately describe the state of daughter equilibrium, whilst a measurement of RaB adds only little useful information.

A prototype radon daughter monitor (RDM) has been developed with the above points in mind. We have chosen to measure only the RaA and RaC concentrations, hence the counting procedure and the calculation of results have been greatly simplified. Both sampling and measurement normally take only 11 minutes and results can be calculated using a slide rule. An important feature of the method is that electronic requirements are simple, amounting only to the scaling of gross alpha-activity.

The Radon Daughter Monitor

Fig. 1 is a photograph of the prototype monitor. The instrument is portable and weighs 5 kg.

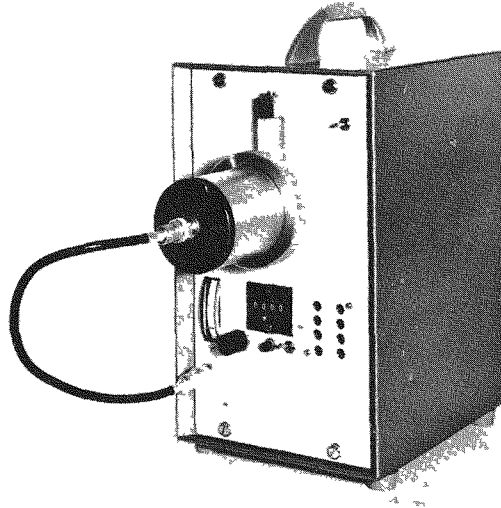


Figure 1: The Radon Daughter Monitor

Mechanical details

Air is drawn through a glass fibre filter (GF/A, Whatman UK) at 10 litres/min. The carbon vane pump (L10, Rotheroe & Mitchell UK) is battery powered. Sampling rate is monitored and all components are mounted in a sealed case.

Alpha-counting

Alpha activity on the filter is detected by a 450 mm² diameter surface barrier diode (ORTEC) mounted in the filter holder. The front electrode of the detector has been specially thickened to 0.5 mg cm⁻² of gold. This reduces sensitivity to chemical contaminants that might be picked up from mine air.

Scaling

A charge sensitive pre-amplifier and an amplifier with an adjustable threshold are used. The discrimination level is set for a particle energy of 800 keV in Silicon. This effectively rejects beta-pulses from RaB and noise picked up from the pump motor. Integrated circuits and a binary display are used for two decades of scaling logic. Four higher decades are counted and displayed by a mechanical register (Landis & Gyr UK). A maximum count rate of 1000/sec is attained by this arrangement. Power for the circuitry is supplied by a battery of mercury cells, with a separate dry battery for the display bulbs.

Operation of the monitor

Operation is controlled by a single switch, the sampling and counting times being measured by a watch.

The Sampling and Counting Scheme

We have chosen a procedure that is simple to use underground. Thus, sampling times are restricted to 2, 5 or 10 minutes. Equal sampling and counting times are used. A fixed, one minute, delay between the 2 alpha-counts reduces the likelihood of timing error.

Conversion factors

Mercer's general formulation of radon daughter decay² was used for calculations on a digital computer. Computations were checked against published data^{3,4}.

Fig. 2 shows the build up of alpha-activity for equal sampling and counting times when equal airborne concentrations of radon daughters are collected on a

filter. RaA contributes almost half of the total alpha-activity during sampling. At short time intervals, both during and after sampling, the contribution from RaB is small.

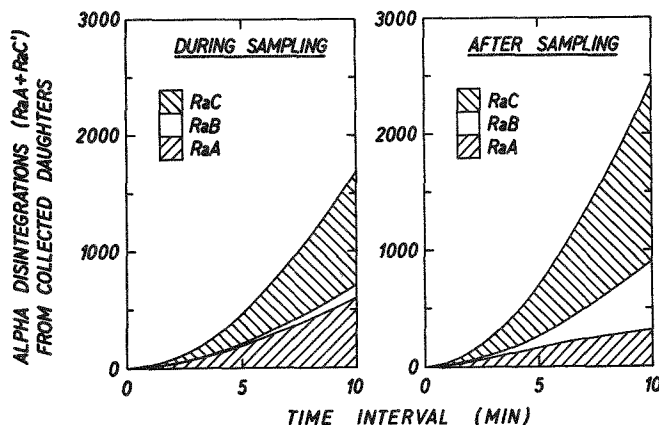


Figure 2: Alpha disintegrations from 1pCi/l. each of RaA, RaB and RaC, sampled at 10 l./min and collected on a filter. Disintegrations after sampling refer to equal sampling and counting times with a 1-minute delay in between. The envelopes of the curves give total alpha disintegrations.

In order to estimate the desired unknown concentrations, RaA and RaC, from only 2 gross alpha-counts, some assumption must be made about the RaB concentration. We have assumed that the ratio RaB/RaA is a uniquely defined function of the ratio RaC/RaA.

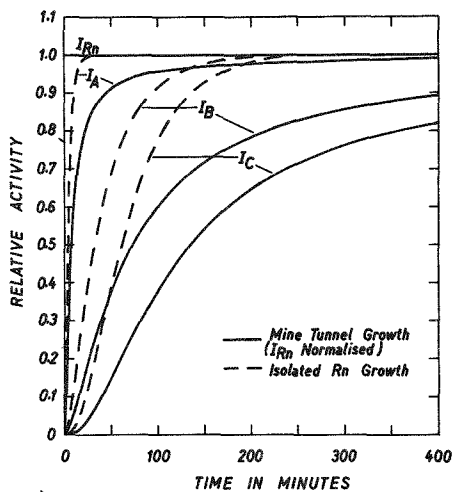


Figure 3: Relative activities of daughters and parent radon as a function of growth time, according to two theoretical models of growth.

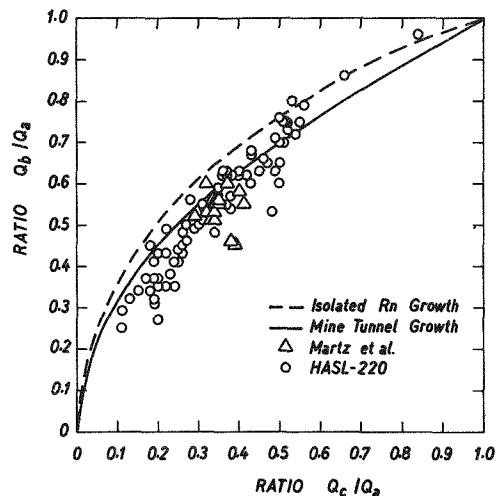


Figure 4: Ratios of radon daughter activities for two theoretical growth models. Plotted points are ratios measured in U.S. uranium mines.

The RaB approximation. Fig. 3 shows the growth of radon daughters from parent radon under 2 different conditions. The 'Mine Tunnel' model⁵ describes growth during de-emanation of radon at a uniform rate from the walls of a mine tunnel. Air is moving through the tunnel, thus growth time is equated to transit time. The 'Isolated Radon' model³ describes growth in still air. Fig. 4 shows that a different function relates RaB/RaA to RaC/RaA in the 2 models. Measured values of these ratios, from U.S. uranium mines^{6,7}, are also plotted in the figure. The measured values are better represented by the 'Mine Tunnel' model, but they do tend to fall below this (solid) curve. Rolle⁸ found that

similar, relatively small, departures from the simple model are predicted when 'young' and older air mix in a mine, e.g. downstream of a junction. Plate-out of unattached daughters on tunnel walls can also disturb the daughter equilibrium. However, the measurements plotted in Fig. 4 indicate that daughter equilibrium is adequately described by the solid curve. We have assumed this to be generally true. The curve is a good fit of the relationship⁸
 $RaC/RaA = (RaB/RaA)^2$.

Calculated factors. Figs. 5-7 show the factors calculated to relate equilibrium ratio, Q_C/Q_A , RaA concentration, Q_A , and WL to alpha-counts recorded by the RDM. Curves are given for both growth models. Full computer tabulations are available from the authors. These allow for the slightly lower counting efficiency observed for RaA than for RaC with GF/A filters⁹. The calculated factors have been verified by comparing laboratory measurements with the RDM and simultaneous measurements by the modified Tsivoglou method¹⁰.

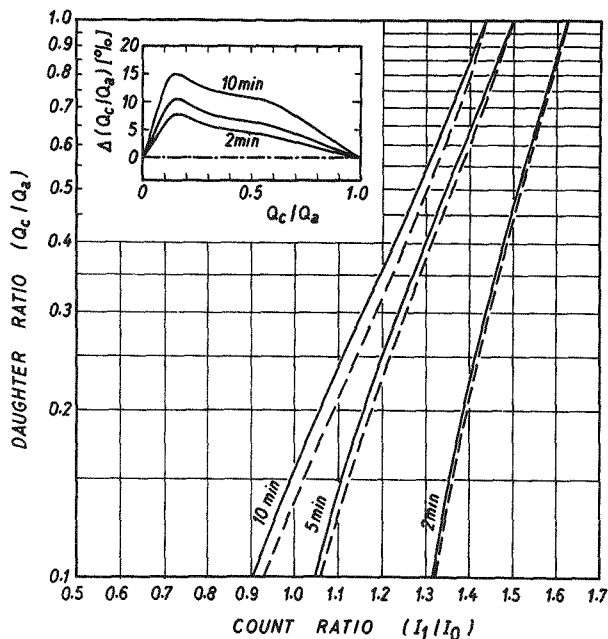


Figure 5: Curves relating the measured ratio of two alpha-counts to radon daughter ratio, Q_C/Q_A , for different sampling times. Equal counting efficiencies for RaA and RaC are assumed. Solid curves assume 'Mine Tunnel' model, dashed curves 'Isolated Rn'. Inset shows percentage systematic error in estimate of Q_C/Q_A if the wrong model chosen.

Systematic errors. The possible errors caused by departures from an assumed model are smallest for short sampling times. In Figs. 5-7, the inset figures show the magnitude of systematic errors that would arise when sampling in still air, if the 'Mine Tunnel' model were assumed to hold. These are a function of the true Q_C/Q_A . Reference back to Fig. 4 shows that similarly small errors would have been recorded for the values measured in uranium mines.

Statistical precision. Fig. 8 (a, b & c) shows the calculated coefficients of variation in estimated quantities for a range of airborne daughter concentrations. Fig. 8(a) also shows that a realistic background alpha-count of 1/min has a negligible effect on the precision of estimating Q_A . Therefore, neglecting background, the precision in an expected value, say Q_A , can be calculated as $S(Q_A = 1)/\sqrt{Q_A}$. Similarly, the minimum detectable concentrations, corresponding to a coefficient of variation of 0.5, can be calculated from these curves. For high radon daughter concentrations, the maximum counting rate of 1000/sec limits the sampling time. A two-minute sample is advised for concentrations in excess of 3 WL.

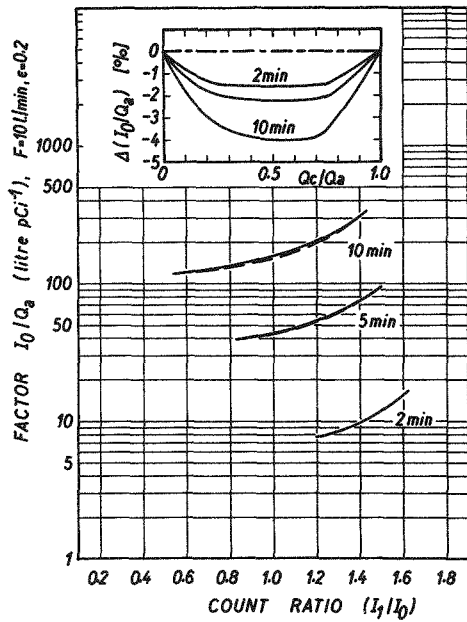


Figure 6: Factors relating count during sampling, I_0 , to RaA concentration, Q_A , for different sampling times (counting efficiency 0.2). Solid and dashed curves refer to different models as in Fig. 5. Inset shows percentage systematic error arising from choice of the wrong model.

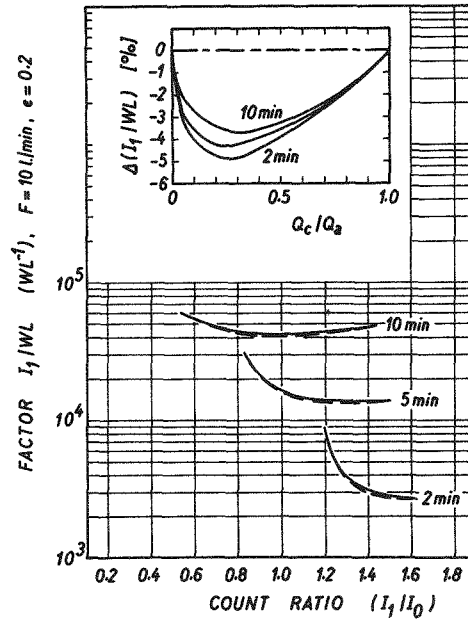


Figure 7: As Fig. 6, but showing factors relating count after sampling, I_1 , to working level, WL. With a 5-minute sample and $Q_C/Q_A \geq 0.3$, the factor is almost constant.

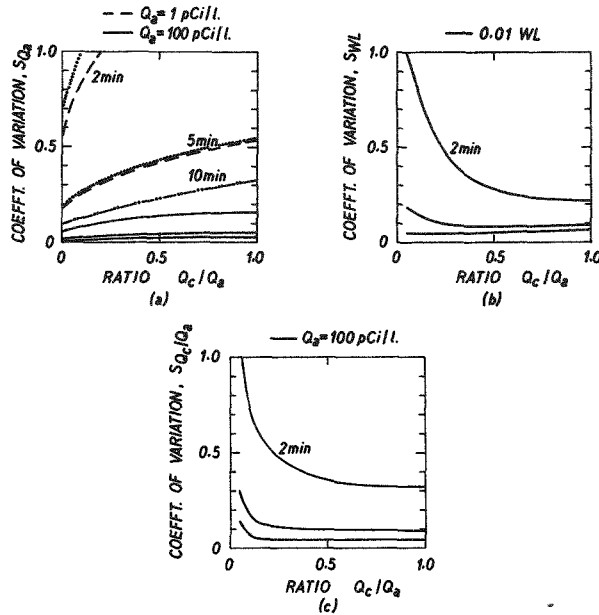


Figure 8: Statistical precision in estimates of (a) Q_A , (b) WL and (c) Q_C/Q_A as a function of daughter equilibrium for 2, 5 and 10 minute sampling times. In (a) two sets of data are given; for $Q_A = 1$ and 100 pCi/l. Dotted curves show the effect of 1 count/min background when $Q_A = 1$ pCi/l.

Comparison with Other Methods

The table displays for comparison statistical precision, number of counts required for a measurement, total time and the subsequent computation for our method (RDM) and 4 established methods. Two of these methods^{6,10} give individual daughter concentrations. The other two are presented as rapid methods, giving WL only: The Single Gross Alpha Measurement Procedure¹¹ (SGAMP) and the Instant Working Level Meter¹² (IWLM). Statistical precision for the RDM compares favourably with other methods, whilst the monitor offers significant advantages in field use. Under conditions of disequilibrium, both the RDM and SGAMP are subject to systematic errors. These are of the order of 10% for the SGAMP¹¹.

TABLE 1
COMPARISON OF METHODS FOR MEASURING RADON DAUGHTER CONCENTRATIONS*

	Spectrometry	Modified Tsivoglou	SGAMP (Kusnetz)	IWLM	RDM
RaA	$\pm 5\%$	$\pm 12\%$	-	-	$\pm 6\%$
RaC/RaA	$\pm 6\%$	$\pm 12\%$	-	-	$\pm 9\%$
WL	$\pm 3\%$	$\pm 3\%$	$\pm 1\%$	$\pm 11\%$	$\pm 1\%$
No. of counts	2	3	1	2	2
Time	35 min	35 min	16 min	4 min	11 min
Computation	Simultaneous equations		Slide rule	Direct Readout	Slide rule

*Calculated coefficients of variation for 100 pCi/l. each of RaA, RaB, RaC. Sampling flow rate 10 l./min, except IWLM (3 l./min). Counter efficiency 0.2.

Field experience with the RDM

The monitor has undergone continuous development based on underground and laboratory comparisons with established methods. Development has reached the stage where the pump and counting system function reliably underground, even in very humid conditions. Good correlation has been obtained between routine measurements of WL with the RDM and the standard Kusnetz method¹³.

Conclusion

The radon daughter monitor described gives a rapid, comprehensive and sensitive measurement of radon daughter activity. Ease of measurement is achieved with only small and acceptable systematic errors. The complexity of the instrument has been reduced to a minimum.

Acknowledgements

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RADON EMANATION STUDIES IN JADUGUDA
URANIUM MINE

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Abstract

Radon gas emanating from the rock surfaces is the major source of radon in uranium mines and ventilation is the only effective means of keeping the radon levels within the acceptable limits. Quantitative estimation of the radon emanation rates is therefore essential for calculating the fresh air requirements of a working face.

Measurements of radon emanation rates have been carried out in laboratory and in the underground mine. In laboratory, the uranium ore sample is enclosed in a large air-tight glass container. Radon from the sample diffuses out and accumulates in the air volume of the container. Samples are periodically drawn from the container and radon activities are determined. The radon emanation rate is then computed from the activities obtained at different sampling intervals.

This paper describes in detail the studies conducted in the laboratory and in the uranium mines. Results obtained are presented and compared with the emanation rates published elsewhere in literature.

INTRODUCTION

The major radiation hazard in a uranium mine originates from the short-lived radon - progeny, concentrations of which mainly depend on that of the parent, radon-222. Formed within the ore body, radon enters mine atmosphere by diffusion through the rock surface. The rate of emanation is characterised by the ore grade and porosity of the rock^{1,2}. Suction effect caused by lowering of the atmospheric pressure is another additive factor³. Although underground water⁴ and broken ore piles also contribute substantially to the radon content of a mine drift, the main supply comes from the continuous diffusion through the ore body. Ventilation plays the most effective role in reducing the airborne radiation levels underground. Knowledge of radon emanation rate is therefore essential for an efficient and economic design of a uranium mine ventilation system.

This paper describes the efforts being made at Jaduguda to estimate the radon emanation rate in the uranium mine. The present investigations were confined to study the dependence of emanation rate on the grade of ore. Simple laboratory and field methods have been described. Emanation rates obtained in Jaduguda Uranium Mine have been compared with those reported in literature from elsewhere.

METHOD OF EMANATION RATE MEASUREMENT

Laboratory Experiments

Small pieces of uranium ore were placed in glass jars of approximately one litre capacity, and were covered with air tight lids provided with a stop cock. The gas emanating from the ore samples was allowed to accumulate in the free-air-space of the emanation jars. Radon levels within the jars were usually found to reach measurable concentrations in 5 to 6 hours after sealing. The radon build-up in the jar follows an exponential pattern, and reaches a constant concentration value after a period of about a month. In order to establish that our measurements were done during the near linear region of the build-up curve, the build-up was followed and was found to be fairly linear within the first 5 days. All our sampling for emanation studies were done within the first two days. Samples of air were drawn from the jar at desired intervals directly into evacuated scintillation flasks. Air drawn from the emanation jar during sampling was replaced by introducing radon free air. Sampling from each jar was repeated at suitable intervals and the concentration of radon at each sampling instant was estimated by measuring the activity in the scintillation flask after a lapse of about 200 minutes.

The rate of diffusion of radon through unit surface area of the ore piece is termed as the Emanation Rate, 'J'. It can be calculated using the following formula suggested by Thompkins et al¹.

$$J = \frac{K(C_2 - C_1 e^{-\lambda t})}{3600} \times \frac{V}{A} \dots\dots(1)$$

Where $K = \frac{\lambda}{(1 - e^{-\lambda t})} =$ depletion factor,

$C_1 =$ radon concentration in the accumulation volume at instant t_1 , Ci/l,

$C_2 =$ radon concentration in the accumulation volume at instant t_2 , Ci/l,

$\lambda =$ decay constant of ^{222}Rn , h^{-1}

$A =$ emanating area of rock surface, cm^2

$V =$ Radon accumulation volume, l.

Field Experiments

For field measurements underground, holes of 34 mm dia and 1 to 3 metre length were drilled in the ore body. Holes were thoroughly washed to remove loose particles of ore dust. After flushing with compressed air the holes were sealed with rubber stoppers provided with a stop cock. Radon emanating from the inner surface of the rock was allowed to accumulate in the drill hole. Samples of radon were directly collected in pre-evacuated scintillation flasks at known time intervals. Volume and surface area of the drill hole were measured. The concentrations of radon obtained within the holes at different sampling instants were used to estimate the radon emanation rate using the relation shown in equation (1).

EMANATION RATE DATA

Radon emanation rates of twenty one ore samples from different areas of the mine were measured in the laboratory. Grade of ore in respect of each sample was estimated radiometrically. The ore grades thus obtained were grouped into discrete classes for simplicity. For instance, all values from 0.070 to 0.090 were considered as 0.08 and those from 0.090 to 0.110 as 0.10 and so on.

The mean of emanation rates of the samples corresponding to the different groups of ore grade are presented in Table-1.

Table-1

Radon Emanation Rates of Uranium Ore Samples

Grade of ore (% U_3O_8)	Radon Emanation Rates(J) $\times 10^{-16}$ Ci/cm ² sec.
0.02	0.10
0.04	0.41
0.06	0.14
0.08	0.25
0.10	0.67
0.12	0.35
0.14	0.32
0.16	0.85
0.20	0.12
0.24	0.61
Range of J: 0.10×10^{-16} to 0.85×10^{-16} Ci/cm ² .sec.	

For field measurements, drill holes at 15 locations in the mine were chosen and the emanation rates were estimated. Rock samples were chipped off from around the individual holes and the ore grades were estimated. The values of J obtained for different groups of ore grades around the drill holes are given in Table-2.

Table-2

Radon Emanation Rate in the Mine

Grade of ore (% U_3O_8)	Radon Emanation Rate(J) $\times 10^{-16}$ Ci/cm ² .sec.
0.02	0.13
0.06	0.37
0.08	0.36
0.10	0.19
0.12	1.20
0.16	1.18
0.18	1.35
0.20	1.69
Range of J: 0.13×10^{-16} to 1.69×10^{-16} Ci/cm ² sec.	

DISCUSSION

The radon emanation rates obtained in laboratory experiments are generally lower than those obtained under actual mining conditions. The deviations in the two ranges vary from a factor of 1.3 to 2. This variation may be attributed to a variety of reasons. One is that the ore pieces used in the laboratory studies were very small as compared to the massive ore body inside the mine. Secondly, since the mine employs exhaust type of ventilation, the barometric pressure under ground is depressed in comparison to that outside. The difference in the pressure is of the order of 32 mm of water gauge. It has been reported that reduction of pressure increases the emanation of radon³.

Contrary to expectations, the attempts made to establish a precise relation between the ore grade and emanation rate did not yield any conclusive result in respect of laboratory experiments. Although no definite explanation could be given for this anomaly, it is likely that the violent forces at work during blasting might have altered the rock characteristics to varying degrees and hence the inconsistency. In case of the underground experiments, however, the emanation rate did appear to follow the ore grade, as may be seen from Figure-1.

As the atmospheric concentrations of radon in our mines are generally found to be well within the permissible limits, a comparison of emanation rate in Jaduguda mines with those elsewhere may be of interest. For this reason, emanation rates prevalent in American mines and in some soils have been compiled in Table-3.

Table-3
Comparative Values of Emanation Rates

Area/Countries	Radon Emanation Rate J, $\times 10^{-16}$ Ci/cm ² .sec.
Jaduguda Mines, India	0.13 to 1.69
New Mexico, USA, (Mines) ²	500.00
Southern Utah, USA, (Mines) ⁵	150.00
Socorro, New Mexico (Soil) ⁶	0.90
Pelindaba, South Africa (Soil) ⁷	0.14
Tailings used as back fill in Jaduguda	14.20

This comparison shows that radon emanation rate in Jaduguda mine is much lower than those obtained in American mines. Though the ore grade in American mines are presumably higher than that in our mine (0.07% U₃O₈), their emanation rates are too high to be accounted for by the ore grade alone. It is known that in New Mexico uranium occurs in sand stone and in Utah in shales. These rocks are highly porous as compared to our densely packed archaean and metamorphic rocks. The soils of socorro and Pelindaba, though having lower uranium and radium concentrations, have radon emanation rates comparable with that of Jaduguda mine. Thus, the porosity appears to affect the emanation rates of radon substantially.

Rate of radon emanation was determined by the authors from coarse uranium mill tailings, used as backfill in the mine. The rate was 14.2×10^{-16} Ci/cm².sec. The radium content of these sands was of the order of 60 pCi/g while that of the ore of grade 0.07% U₃O₈ is about 200 pCi/g. The emanation rate of this ore as seen from Figure-1 is 0.41×10^{-16} Ci/cm².sec. The emanation rate from the sand is thus about 35 times that of the ore despite the radium content being only about one third. The emanation rate from the tailings would therefore be about a hundred time greater than that from the ore when normalised to equal radium content. The porosity of the sand was about 50% while the porosity of the ore was about 0.5%. The porosity ratio of the sands to the ore is therefore the same as the emanation ratios, indicating that porosity plays a far more important role in radon emanation than the ore grade alone.

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RADON EMANATION IN JADUGUDA URANIUM MINE

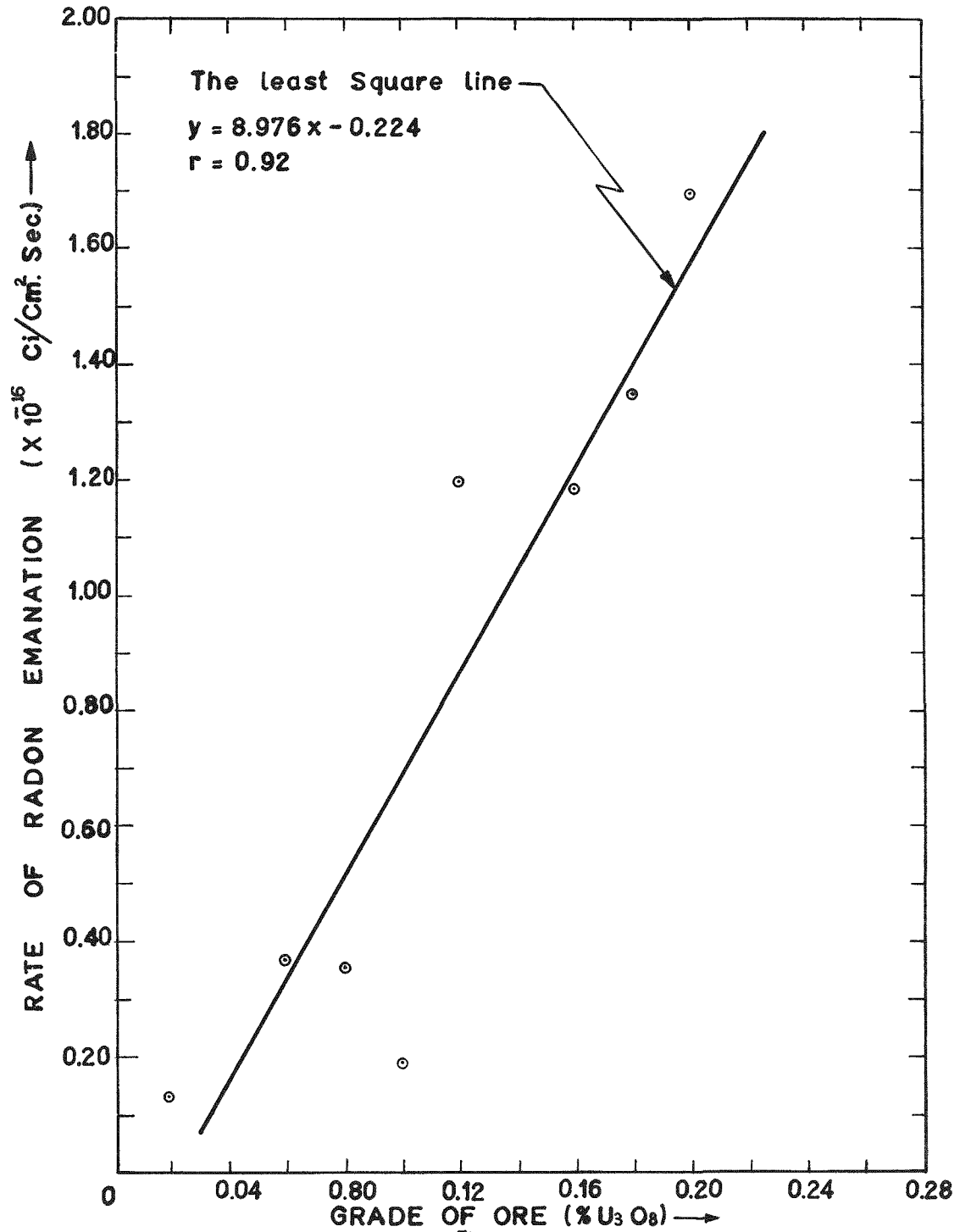


Fig 1

DISTRIBUTION OF AIRBORNE ACTIVITY IN
A URANIUM MILL USING COBWES

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ABSTRACT

Cobwebs found in the different sections of the uranium mill at Jaduguda were observed to trap dust particles in the respirable size range (about 60% respirable). As such these were taken to represent the dust particles inhaled by the workers. The dust collected from the cobwebs were analysed for U(nat), ionium (^{230}Th), radium (^{226}Ra) and polonium (^{210}Po). The results indicate that equilibrium in the airborne activity exists only in the initial stages of operations, viz during crushing and screening. In other sections of the mill the equilibrium is found to be disturbed to different degrees.

INTRODUCTION

For a realistic estimation of inhalation hazard associated with uranium ore milling, the activity fraction of the constituent long-lived alpha emitters in respirable dust must be known. This may be accomplished by sampling and analysing the activity content in the respirable fraction of the airborne dust.

It was observed that cobwebs present at the plant site trap very fine dust particles. The purpose of the present study is to determine the size distribution of the particles thus trapped and to estimate the respirable fraction to decide whether the dust on cobweb can be taken to represent the dust inhaled by the workers. Further cobweb samples collected from different operation sites have been analysed chemically for the individual long-lived alpha emitters to study the distribution of alpha activities in the airborne dust.

EXPERIMENTAL DETAILS

The Cobweb

The cobweb found at the plant are mostly woven by an orb weaving spider "Microthema Gracilis". The cobwebs have the following properties:

Diameter of the net	- 25 - 30 cm
Spiral spacing	- 3 - 5 cm
Number of support thread	- 22 - 28
Diameter of support thread	- 1.25 - 1.75 μm
Diameter of spiral thread	- 1 - 1.5 μm
Shape of the frame	- hexagon

The cobwebs were collected from different units of the plant like Crushing and Screening, Grinding, Dewatering, Filtration and Clarification, Precipitation and Recovery and Tailings Treatment plant. Five samples were collected in each area at intervals of three months.

Particle Size Measurements

The dust adhering to the cobweb samples was stripped using distilled water to which a few drops of 0.01 N sodium hydroxide was added. An aliquot of the dust laden liquid was transferred to an A.O. Spencer Bright Line Counting Chamber. After about thirty minutes delay (for complete settling of the suspended dust) the dust particles were sized using May's type graticules. On an average more than two hundred particles were sized in each case.

Estimation of Respirable Fraction

In order to make the necessary calculation to estimate the respirable fraction of the dust collected on the cobweb we have followed the method suggested by Schulte¹. The respirable fraction is obtained as a product of the mass frequency and the corresponding lung deposition factor obtained from the LASL curve.

Chemical Analysis

The sample was dried and weighed. Organic material was oxidised by repeated treatment with conc. HNO₃, leached and taken up in 4 N nitric acid. Estimation of U (nat), ²³⁰Th, ²²⁶Ra and ²¹⁰Po were carried out taking suitable aliquots of the prepared solution.

Percentage distribution of the long-lived alpha activities were determined by taking the ratio of the counts due to the individual isotope to that of the total.

RESULTS

Size Distribution and Respirable Fraction

The dust particles obtained from cobwebs were found to follow log normal distribution. Representative data on the size distribution and respirable fraction are given in Table 1 and 2.

Table-1
Size Distribution and Respirable Fraction

Diameter		Frequency		Lung Deposition % (LASL Curve)	Respirable Fraction	Remarks
Projected area D _p (μm)	Aerodynamic D _{ae} (μm)	No.	Mass %			
0.68	0.75	56	1.2	100	1.2	
0.10	1.18	61	5.2	100	5.2	
1.56	1.67	54	13.0	100	13.0	
2.20	2.36	27	18.3	80	14.6	CMAD=1.07
3.10	3.33	12	23.1	55	12.7	σ _g = 1.77
4.40	4.73	4	22.1	30	6.7	MMAD=2.85
6.25	6.70	1	16.8	10	1.7	

CMAD = Count Median Aerodynamic Diameter

MMAD = Mass Median Aerodynamic Diameter

σ_g = Geometric standard deviation

Table-2

Statistical Parameters and Respirable Fraction
Of Cobweb Dust at Different Operational Stages

Operation	Bulk Density of the dust	CMAD	Geometric Standard Deviation	MMAD	% respirable Schulte Method ¹ Regression graph ²	
Crushing and Screening	2.7	1.08	1.79	2.99	51.4	58
Grinding	2.7	0.99	1.95	3.75	44.0	48
Dewatering	2.7	1.12	1.79	3.00	59.9	58
Filtration and Clarification	2.7	1.18	1.69	2.70	57.6	65
Precipitation and Recovery	5.4	1.67	1.94	6.24	14.5	25
Tailings treatment	2.7	1.22	1.72	2.95	55.0	60

Distribution of Long-lived Alpha Emitters

Percentage contribution of long-lived alpha activities in air during different operations are given in Table-3 and shown in Figure-1.

Table-3

Activity Distribution of Long-lived Alpha Emitters

Operation	Percentage of gross long-lived alpha emitters			
	U(nat)	²³⁰ Th	²²⁶ Ra	²¹⁰ Po
Crushing and screening	39.50	20.50	20.30	19.70
Grinding	45.50	16.50	20.30	17.70
Dewatering	58.00	15.00	12.00	15.00
Filtration and clarification	72.00	11.30	6.40	10.30
Precipitation and recovery Sec.	91.00	6.90	1.10	0.80
Tailings treatment	30.50	21.70	23.90	23.90

DISCUSSION OF RESULTS

For calculating the respirable fraction of dust trapped in cobweb, the schulte method¹ has been followed and the results have also been verified by an alternative method², following a theoretical curve between MMAD and respirable fraction for given standard geometric deviation(Table-2).

Chemical analyses of cobweb dust collected from different stages of operation indicate that during the initial stages i.e. crushing and screening radioactive equilibrium exists in the airborne activities(Table-3, Fig.1). Airborne uranium activity in this section is about the same as the rest of the radionuclides of interest. But during subsequent operations, uranium activity is predominant as compared to the rest.

Higher values of uranium in the airborne dust during grinding and dewatering stages is probably due to the addition of recycled iron cake and slurry containing significant amounts of uranium.

After leaching of the crushed ore most of the activities except uranium remain with the waste cake which is separated during filtration and sent for tailings treatment. One would expect airborne uranium in this section to be less than the other three alpha emitters considered. But in practice this was found not to be the case.

The predominance of uranium at the stages coming after filtration is easily explained, since the solutions and solids handled are rich in uranium as compared to other alpha emitters. But it is seen that the airborne uranium is higher also in the filtration section contrary to expectations. This is possibly due to cross contamination from precipitation and recovery areas; since both the sections are housed in the same building without partition between them. During tailings treatment which is separately housed, as expected uranium values are less than radium, polonium and ionium.

CONCLUSION

Cobweb dust sampling provides a long-term air sample of particles that are most likely to be inhaled by the workers. This has an edge over the conventional air sampling techniques with cyclone or other size discriminating devices, which are suitable only for grab sampling and hence are inadequate as far as average conditions are concerned. Analyses of cobweb samples for long-lived alpha emitters give fairly good idea about the activity distribution during different stages of operation.

Since the $(MPC)_a$ for gross long-lived alpha activities depends on the state of radioactive equilibrium of the constituent nuclides in air, we propose to revise the existing $(MPC)_a$ value on the basis of the above findings.

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AN INSTANT WORKING LEVEL METER WITH AUTOMATIC INDIVIDUAL
RADON DAUGHTER READOUT FOR URANIUM MINES[‡]

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Abstract

The Instant Working Level Meter (IWLM) evaluates the Working Level and the individual Rn-daughter concentrations in an uranium mine atmosphere within five minutes. The instrument is portable and fully automatic. The WL and the RaA, RaB and RaC concentrations (pCi/liter) are displayed in digital form. Calculation of these quantities is performed by a pre-programmed CMOS calculator chip using the counts observed in the instruments three channels (RaA, RaB + C, RaC'). The Rn-daughters are collected on a membrane filter at a flowrate of 12 liter/min. α -spectroscopy is performed with a silicon surface barrier detector, the $\beta + \gamma$ - counts are detected with a plastic scintillator plus PM tube. No assumptions about Rn-daughter equilibrium are made. Only constancy of the Rn-daughter concentrations during the time of sampling (2 minutes) is assumed. The unit is entirely solid state with exception of the photomultiplier. The range of the instrument is 0.01 - 100 WL.

Introduction

The commonly used methods¹⁻³ to determine the WL (Working Level) and the short-lived Rn-daughter concentrations in uranium mine atmospheres suffer from several shortcomings. It takes a minimum of 17 minutes to complete the measurements using the fastest of these methods and in the most frequently used procedure¹ the Rn-daughter equilibrium and the influence of the build-up time of the activity on the resulting WL is neglected. The first attempt to solve these problems⁴ produced an IWLM (Instant Working Level Meter) which was capable of automatic WL determination but was limited by the low air sampling rate and the high γ -sensitivity of its β -detector. The pseudo-WL due to γ -background has been reduced by about a factor of fifty in the instrument described in this paper. This was achieved by increasing the sampling rate to 12 liter/minute, use of a thinner scintillator (0.003 in.) and shielding of the β -detector.

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Description

Mechanical Assembly

A schematic drawing of the air sampling system is given in Fig. 1.

After a one minute background counting period and a two minute sampling period, the filter paper tape (Gelman, Acropor, pore size 0.8μ) is moved from the air intake by spring tension to position the active area between the α -detector (ORTEC, silicon surface barrier detector) and the β -scintillator (NE 102). After a delay of three seconds, the two minute counting period starts automatically. After this period, the WL and the Ra A, Ra B and Ra C concentrations in pCi/liter are read out on command. After completion of the measurement the active spot is moved from the counting position and discarded. We found a self-absorption of 0.4% in the filter paper (Gelman, Acropor) using the method of J. Shapiro⁵. A carbon vane pump (GAST) driven by a printed circuit motor delivers a sampling rate of about 12 liter/minute. A tachometer senses the revolutions per minute and is part of a feedback system ensuring constant flow-rate.

Electronic System

The electronic circuitry consists of three major subsystems: the detection subsystem, a control-computer subsystem and a power subsystem. (See functional block diagram.) The detection subsystem is further broken down into the α -detection channel and β -detection channel, the pump motor regulator and drive circuit, the high voltage regulator and the solenoid actuator for the paper drive system.

Detection Subsystem. The α -detection channel consists of a surface barrier detector, a high gain charge sensitive preamplifier, a pulse amplifier, and a single channel analyzer to separate Ra A from Ra C'. The overall α -detection gain is 5 volts per pC.

The β -detection channel consists of a 10 stage low noise high gain photomultiplier, a NE 102 scintillator, a high gain charge sensitive preamplifier and a discriminator.

Power Subsystem. The power subsystem consists of 13 rechargeable Gel cell batteries and a connector which selects either individual cells for the external charging circuit or combines the batteries into the power pack which consists of an 18 V, 3 Ah battery for operation of the pump, a 6 V, 3 Ah battery for the digital components and ± 12 V, 1 Ah batteries for the linear components.

Control-Computer Subsystem. The control section of the control-computer subsystem consists of a CMOS driven sequential control circuit which applies the timing pulses for the fully automatic operation.

Also included in the control section are three burst generators, one for each output (Ra A, Ra (B+C) and Ra C'), used to generate double pulses for each single input pulse during the background counting period. These doubled background counts are subtracted from the normal sample count through the use of

up-down counters (accumulators) to compensate for the γ -background. Gamma shielding is achieved through the use of a lead shield (see Fig. 1) and by positioning the batteries around the detector section. The control section also controls the input gating, signal routing and digital resets of the computer section to provide the proper compensated two minute sample count to the accumulators.

The computing subsystem is essentially a pre-programmed buss oriented digital processor. This processor accepts digital data from the accumulators, combines it with various stored constants and from this data calculates Working Level and the individual Rn-daughter concentrations. The processor consists of a MOS calculator circuit, a program memory, a constant memory, a system clock generator, accumulators, calculator driver, display driver, and display as shown in Fig. 2.

The entire circuit except for the memories is assembled from CMOS digital integrated circuits which offer very low power consumption and high immunity to electrical noise.

In order to understand the operation of the circuit, a brief explanation of it's component parts follows:

- 1) The program control section consists of two programmable read only memories (PROM's) which store the program steps, a program counter which advances once for each program step, and a 4 line to 16 line decoder which translates part of the digital word from the memory into individual commands such as add, enter accumulator A, enter constant 5, multiply, etc. The data lines from the PROM's are also used to select the location of a particular constant in the constant memory.
- 2) The constant memory consists of two PROM's which hold the 12 constants required for the calculations; a memory address register which locates and clocks out the constant requested by the program; a set of clock controls; and a set of transmission gates which tie the memories to the data buss. The constant consists of six information words which may include a decimal point. The use of PROM's allows a field change of constants should changes in counting efficiency or flowrate alter the equations.

The information is entered onto the data-buss in a bit-parallel, digit serial fashion. The system clock provides the timing information required to serialize the data. In a particular timing sequence, C0 through C15, the clock pulse C1 steps the program memory register to the next location. If the command at this location calls for entering data from an accumulator, the constant memory or the flowrate correction switch, then clock pulses C2 through C15 enter the data into the calculator. If the new program step called for an operation such as multiply or add, then this operation would be entered into the calculator at time C1. When C1 appears for the second time, the program again advances and new data is entered or a new operation performed.

The clock generator consists of an oscillator driving a binary scaler

whose output is decoded in a 4 line - 16 line decoder to generate clock lines $C_0 - C_{15}$. These lines are then routed to the proper locations in the system.

The data buss is terminated in the calculator driver card which shifts the voltage levels from those required by the PROM's to those required by the calculator circuit. After the level shifting, the data buss is decoded in a 4 line to 10 line converter to obtain digit information for the calculator while a separate conversion is performed to obtain the decimal point information. These decoded data are then used to control information inputs to transmission gates which enable the calculator inputs. The calculator is interfaced to a Light Emitting Diode (LED) display for the presentation of data.

Theory and Calibration of the IWLM

The WL is a linear combination of the short-lived Rn-daughter concentrations as shown in the following equation:

$$WL = 1.052 \times 10^{-4} N_A + 5.908 \times 10^{-5} (N_B + N_C) \quad (1)$$

Therefore, the three unknowns N_A , N_B , N_C (atoms/liter) have to be determined to evaluate the WL. This is done by relating these quantities to the counts observed in the three channels of the instrument, as shown below:

$$\begin{aligned} A &= 0.580386 E_A V N_A \\ B + C &= (0.036204 E_B + 0.001584 E_C) V N_A + \\ &+ (0.098134 E_B + 0.006941 E_C) V N_B + \\ &+ 0.131000 E_C V N_C \\ C' &= (0.001584 N_A + 0.006941 N_B + 0.131000 N_C) E_A V \end{aligned} \quad (2)$$

A = α -counts in Ra A - channel
 B+C = $\beta + \gamma$ -counts from Ra B and Ra C
 C' = α -counts in Ra C' - channel
 V = flowrate (liter/minute)
 E_A = detection efficiency for Ra A and Ra C'
 E_B = detection efficiency for Ra B
 E_C = detection efficiency for Ra C

The numerical coefficients in (2) follow from the laws of radioactive series decay. The half-lives used are:

Ra A: $T_{1/2} = 3.05$ min.
 Ra B: $T_{1/2} = 26.8$ min.
 Ra C: $T_{1/2} = 19.7$ min.

For example, the numerical coefficient (0.580386) in the equation for A follows from:

$$(1 - \exp(-\lambda_A t_B)) \exp(-\lambda_A/20) (1 - \exp(-\lambda_A t_D)) / \lambda_A \quad (3)$$

with:

λ_A = decay constant of Ra A = 0.227621
 t_B = 2 min. sampling time
 t_D = 2 min. counting time

The first term in (3) describes the build-up, the second the decay of the RaA activity during the 3 sec. delay and the third the accumulation of counts during the counting period. The analogous coefficients for daughter and granddaughter products are more complex and are not given here. E_A is determined by comparison with a calibrated hemispherical gas-flow proportional counter. E_B and E_C are calculated in the following manner. First N_A , N_B and N_C are determined from the α -counts observed using the following equations:

$$\begin{aligned}
 N_A &= 0.926838 E_A V A(5) \\
 N_B &= (-0.879403 A(5) - 11.12606 C'(5) + 2.752840 C'(30)) E_A V \quad (4) \\
 N_C &= (0.049957 A(5) + 4.232080 C'(5) - 0.251541 C'(30)) E_A V \\
 A(5) &= \text{RaA counts observed during five minutes starting three} \\
 &\quad \text{seconds after the end of the two minute sampling time.} \\
 C'(5) &= \text{RaC' counts observed during the same time interval as above.} \\
 C'(30) &= \text{RaC' counts observed during thirty minutes starting three} \\
 &\quad \text{seconds after the end of sampling.}
 \end{aligned}$$

The numerical coefficients in (4) follow again from the laws of radioactive series decay. Since their derivation is straightforward but lengthy, it is not given here. With N_A , N_B and N_C known E_B and E_C can be determined from the following equations:

$$\begin{aligned}
 BC(5) &= (0.127907 N_A + 0.236138 N_B) V E_B + \\
 &\quad + (0.010200 N_A + 0.028418 N_B + 0.311000 N_C) V E_C \\
 BC(30) &= (0.981722 N_A + 1.050628 N_B) V E_B + \\
 &\quad + (0.385895 N_A + 0.478116 N_B + 1.256959 N_C) V E_C \quad (5) \\
 BC(5) &= \text{total } \beta \text{-counts observed during five minutes starting three} \\
 &\quad \text{seconds after the end of sampling.} \\
 BC(30) &= \text{total } \beta \text{-counts observed during thirty minutes starting} \\
 &\quad \text{three seconds after the end of sampling.}
 \end{aligned}$$

With E_A , E_B and E_C so determined, equations (2) can be inverted. Properly scaled the inverted equations give the Rn-daughter concentrations in pCi/liter and the WL as a linear combination of A, B + C and C'. These inverted equations are programmed in the calculator subsystem of the IWLM. It is clear from this description of the calibration that the IWLM determines the Rn-daughter concentrations and the WL without any assumptions about Rn-daughter equilibrium. Since all weighing coefficients are strictly proportional to $1/V$, a flow-rate variation can be easily corrected for. This correction is accomplished by setting the ratio (calibration flowrate/observed flowrate) on a thumb wheel switch indicated in Fig. 2. Recalibration of the IWLM, if used at different elevations or with different flowrates, is therefore unnecessary.

Tests

We tested a prototype IWLM in the experimental Dakota Mine in New Mexico. The results are shown below:

Test	IWLM				α -Spectroscopic Method (See Eq. (3))				Kusnetz Method
	WL	RaA (pCi/litre)	RaB (pCi/litre)	RaC (pCi/litre)	WL	RaA (pCi/litre)	RaB (pCi/litre)	RaC (pCi/litre)	WL
1	0.88	173	85	68	0.83	171	80	66	0.85
2	1.44	231	129	140	1.51	233	150	137	1.58
3	1.11	219	104	92	1.12	217	109	91	1.12
4	1.47	292	149	105	1.44	296	146	107	1.43
5	0.71	110	69	63	0.76	142	73	64	0.71
6	0.32	74	33	21	0.31	76	30	20	0.31
7	0.52	89	62	28	0.40	90	38	29	0.44

WL, RaA and RaC concentrations determined by the IWLM and the spectroscopic method are in good agreement. The same is true for the RaB concentrations in most cases. The reasons for deviations like in Test 2 and 7 are not clear. More tests are needed to clarify these discrepancies.

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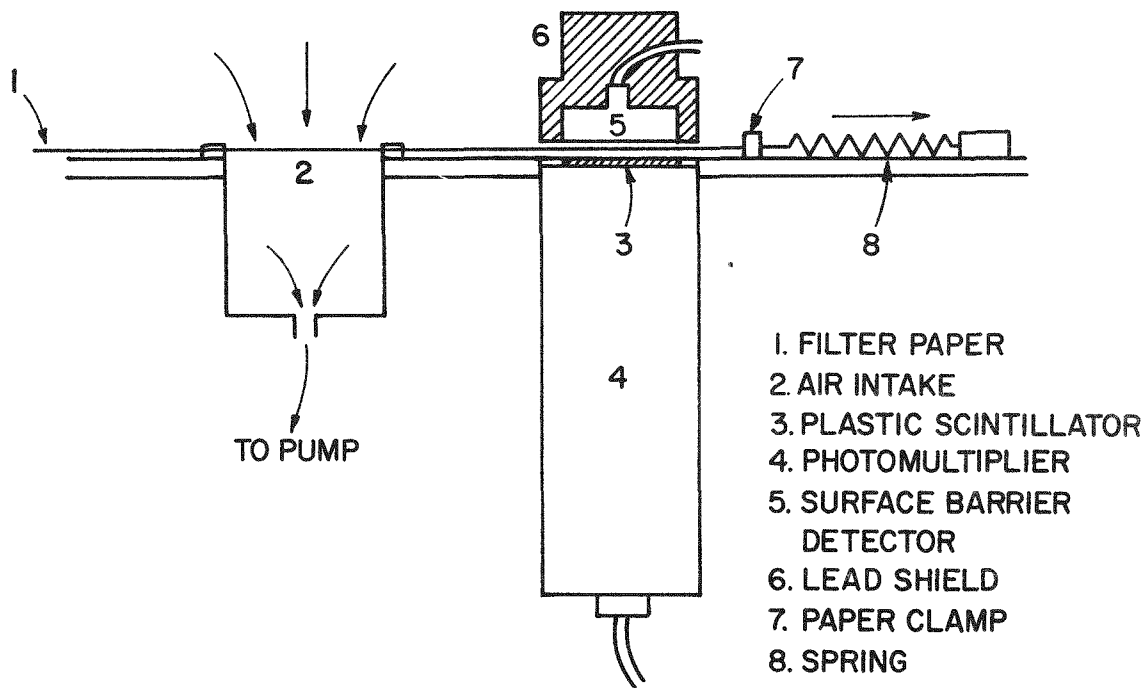


Fig. 1 Detection, air sampling and paper transport mechanisms of the IWLM.

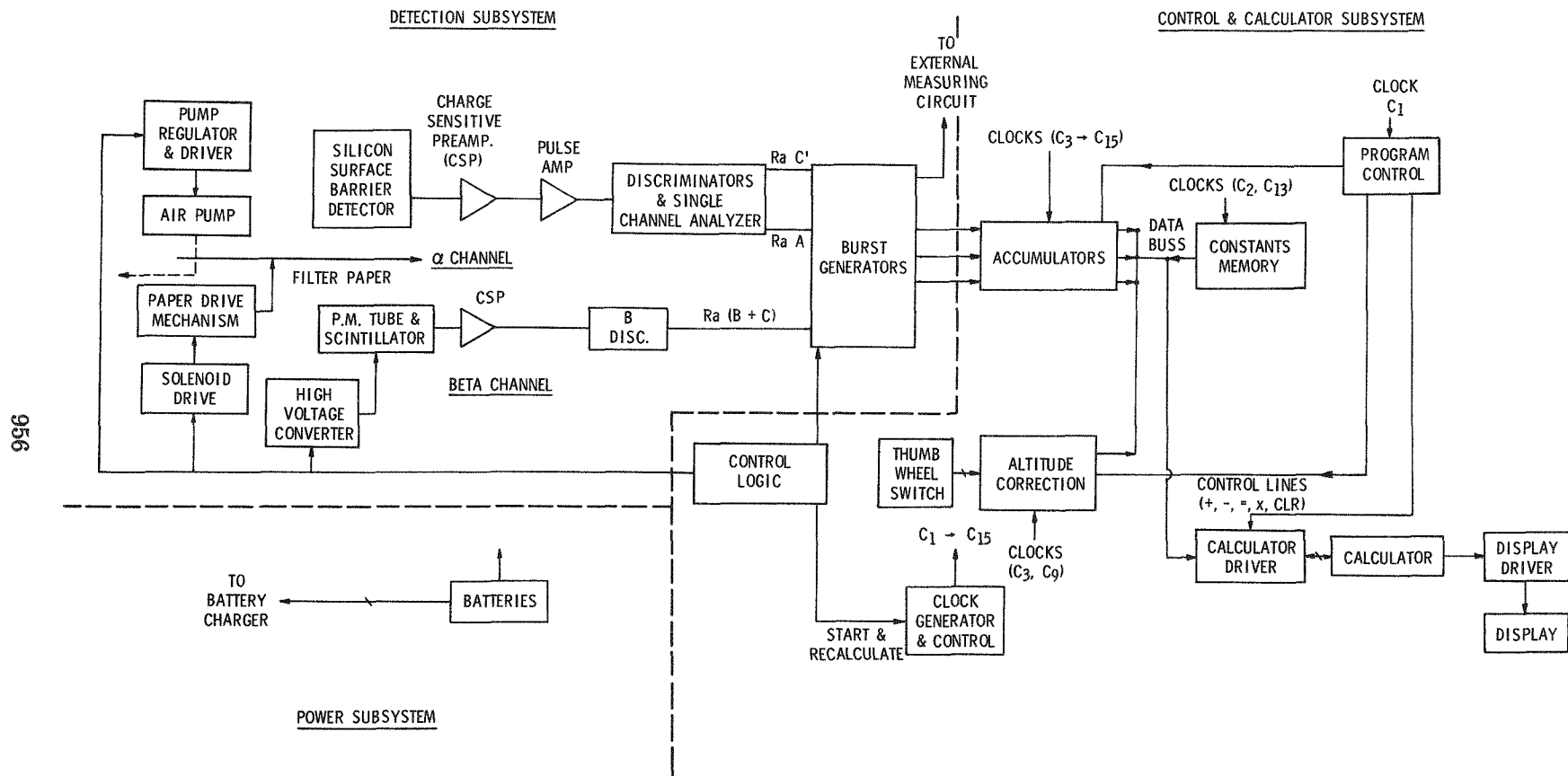


Fig. 2 Functional block diagram of the IWLM.

IN VIVO MEASUREMENTS

MONITORING OF LOW-ENERGY X-RAY RADIONUCLIDE CONTENT IN HUMAN BODY

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Abstract

A 15 cm -dia 0.1 cm thick NaI detector and an Ar-CH₄ proportional counter with a 300 cm² window are used for in vivo measurements of ²¹⁰Pb, ²³⁹Pu and ²⁴¹Am content in human body. The subject's background was determined from the correlation of the counting rates in two channels, e.g. for ²¹⁰Pb: 30-55 and 100-150 keV, respectively. The detectors were calibrated on an anthropomorphic phantom with a thickness of the tissue absorption layer varying from zero to 4 cm.

Calculation methods for nuclide content in critical organs and cases illustrating different types of radionuclide distribution in human body are given.

Introduction

The amount of internally deposited radionuclides in personnel is accepted by present-day international and national standards as one of the basic criteria used in the sphere of radiation protection. However, the evaluation of human body burdens of incorporated ²¹⁰Pb, ²³⁹Pu and ²⁴¹Am is materially complicated by the difficulties involved in obtaining data on the distribution of these radionuclides in individual critical organs. The difficulties involved in detection are due to a number of factors and particularly, to the low-energy radiation of radionuclides assessed. The possibility of obtaining such information is determined to a large extent, by proper choice of techniques of in vivo and calibration measurements which depend on the pattern of radionuclide distribution among and in the subject's organs.

There are three main types of distribution of the above radionuclides in human organism: lung, osteohepatic and a combined one. The specific pattern of distribution is determined by the rhythm and route of the nuclide administration, the chemical form of the deposited compound and some other factors. The combined type of radionuclide distribution is the most general and complicated one, as far as direct measurements are concerned.

Many important aspects of in vivo assessment of low-energy radionuclide content in human body have been discussed in the papers¹⁻⁶. This work deals with the methods and illustrations of measurement procedures for ²¹⁰Pb, ²³⁹Pu and ²⁴¹Am for different

patterns of their distribution in the body.

Equipment

The unit comprises a detection system and a measurement panel with channels for registering energies in the ranges of 10-25, 30-55, 35-70 and 100-150 keV. The detection system consists of two proportional 7.5 cm Ar-CH₄ gas filled counters with a 300 cm² window each and two scintillation counters with 15 cm-dia 0.1 cm thick crystals of NaI(Tl). The counter window panes are made of 0.03 cm-thick beryllium plates. The counters are mounted in an iron chamber 2x1.5x1.5 m with a wall thickness of 15 cm. The scintillation counter background is 20 cpm (10-25 keV) and 65 cpm (35-70 keV). The proportional counter background is 3 cpm (10-25 keV). The counter sensitivity is characterized by the magnitude of calibration coefficients (see Fig.1).

Prediction of the Subject's Background

The sensitivity and accuracy of in vivo measurements of low-energy internally-deposited nuclides are determined, to a great extent, by the individual subject's background level and precision of its measurement. It is impossible to measure directly the specific background in a given contaminated subject because the continuous distribution of pulses of the diffused radiation of high-energy radionuclides ⁴⁰K, ¹³⁷Cs, etc. and the photopeak of the low-energy radionuclide measured, e.g. ²¹⁰Pb, ²³⁹Pu, ²⁴¹Am are registered in the same energy interval. We suggested a method of prediction of the subject's background which is based on the probable correlation between the background counting rate in the main channel where the photopeak of the nuclide measured is registered, and the background counting rate in the reference channel, where this peak does not occur. The correlation between the background counting rates in these two channels was derived from our measurements carried out in a group of people of the different build and with different body contents of high-energy radionuclides. The experimental data processed by the method of least squares were approximated by the following equation:

$$N^M = \alpha + \beta N,$$

where N^M - expected background counting rate, cpm, in the main channel in the energy interval of E_1-E_2 ; N - registered counting rate in the reference channel in the energy interval of $E_3 - E_4 = 100 - 150$ keV; α and β - parameters whose numerical values for different energy intervals are given in the Table:

Radionuclide	E_1-E_2 keV	Position of Detector	α	β
²¹⁰ Pb	30 - 55	Over chest	42.5	1.37
		Over skull	14.80	0.50
²³⁹ Pu	10 - 25	Over chest	9.7	0.19
		Over skull	0.19	0.33
²⁴¹ Am	35 - 70	Over chest	18.6	1.33
		Over skull	11.5	0.88

Methods

Since the radionuclides of lead, plutonium and americium are

generally deposited in the lungs, liver and skeleton. The detectors were placed over the lungs and the liver in front of the subject's chest and over the frontal bone of the skull, a location most convenient for skeletal measurements.

The body contents of ^{210}Pb and ^{241}Am were measured by scintillation counters, while that of ^{239}Pu - by both scintillation and proportional counters.

The set of equations for the values of the activity of the radionuclide deposited in each of the critical organs is:

$$J_i = \sum_{j=1}^4 q_j \varepsilon_{ij} \quad , \quad (i=1,2,3,4) \quad (\text{Eq.1})$$

where J_i - the counting rate of the detector over the i -organ, which is determined by the measured nuclide radiation; q_j - the desired activity of the radionuclide deposited in the j - organ; ε_{ij} - calibration coefficient which is numerically equal to the counting rate measured by the detector over the i -organ for unit activity of the radionuclide in the j -organ of the phantom. The indices used of the equation set (Eq.1) are : 1 - the right lung 2 - the lung; 3 - the liver and 4 - the skeleton.

The value J_4 includes the counting rate measured over the skull, while ε_{44} - the same for unit activity of the radionuclide content in the entire skeleton.

Radionuclide quantum radiation was measured in the intervals: 10 - 25 keV for ^{239}Pu ; 10-25 keV and 35-70 keV for ^{241}Am and 30-55 keV for ^{210}Pb . The calibration coefficients ε_{ij} (Fig.1) were obtained by means of an anthropomorphous phantom which comprised the skull, trunk and arms and legs.

Since the actual distribution of deposited radionuclides involves the values of ε_{41} , ε_{42} and ε_{43} close to zero, these values were disregarded in the calculations, too.

Solutions to the equation set (Eq.1) were found from $q_j = \Delta_j / \Delta$ where Δ is the determinant of the system and Δ_j is the determinant of the undetermined value q_j .

The fourth equation was excluded from the set of equations (Eq.1) and the terms containing ε_{14} , ε_{24} and ε_{34} - from the three first equations, when ^{239}Pu content was calculated, on the assumption that the X-ray radiation of ^{239}Pu is nearly completely absorbed by skeletal bones.

In assessing ^{210}Pb or ^{241}Am , the counting rate J_4 determined by the measured radionuclide radiation was found to be equal to that registered by the detector less the background value. In the measurements of ^{239}Pu content this equation may be upset due to the presence of the impurities of ^{241}Am , the energies of the X-ray lines of which lie within the range of ^{239}Pu radiation. In such case, the values of J_i were obtained from the expression:

$$J_i = I_i - \sum_{j=1}^3 q_j^{Am} \varepsilon_{ij(10-25)} \quad (\text{Eq.2})$$

where I_i - the counting rate detected over the i -organ less the background value; q_j^{Am} - the amount of the activity of ^{241}Am deposited in the j -organ; $\varepsilon_{ij}^{Am}(10-25)$ - calibration coefficients in the interval of 10-25 keV obtained experimentally as a result of insertion of ^{241}Am -emitters into the phantom organs. Values for q_j^{Am} were computed from the data of measurements of the gamma-radiation of ^{241}Am in the energy interval of 35-70 keV.

In Vivo Measurements Examples

Case of Lung Distribution of ^{241}Am

Four people were exposed to the radiation from an americium

source as a result of a failure of containment during experiments. The nuclide was inhaled in the form of insoluble americium dioxide. 0.4 to 2.2 nCi²⁴¹Am was found in the lungs. Fig.2 shows the spectrum of ²⁴¹Am radiation recorded over the lungs of one of the subjects three months after the exposure. Distinct gamma-spectra recorded over the subject's head were detected at the beginning of measurements only (Fig.3). As they were not detected after repeated decontamination measures had been taken, it was an indication of the surface contamination of the skin and hair. Therefore, it provides evidence that it was a case of lung distribution of ²⁴¹Am.

Case of Osteohepatic Distribution of ²¹⁰Pb

The measurements were carried out in a group of volunteers who took a hydrochloric acid solution of ²¹⁰Pb, at pH = 3. The gamma-spectra recorded over the subjects' head and liver are given in Fig.4 and 5. The activity Q deposited in the whole skeleton was assessed with due regard to the equation: $Q = q \cdot k(t)$, where q is the activity deposited in the skull; k(t) is the coefficient of conversion of the skull activity to that of the skeleton which allows for both the ratio of the masses of the skull and the whole skeleton and the nuclide distribution in the compact and trabeculate tissue portions of skeletal bones. The value k(t) varies with time from 16.5 on the first day of radionuclide administration to 12.3 on the 100th day, owing to the continuous re-distribution of bone-seeking elements.

The measurement results showed the skeleton/liver ratio to be 3 to 4 in the average of four subjects on the 100-th day of ²¹⁰Pb administration, which points to the skeletal distribution of lead chloride in human body.

Case of Combined Lung-Osteo-Hepatic Distribution of ²³⁹Pu and ²⁴¹Am

Measurements in man usually fail to produce statistically significant spectra of plutonium and americium radiation. Such spectra, however, may be obtained by long-term measurements, if the thickness of the muscular, fat and skin tissues of the subject is not great. Fig.6 shows the spectra recorded by a scintillation counter placed over the lungs, liver and skull of the subject. The peaks of the pulse amplitude distribution corresponds to the energies of 17 keV and 60 keV, which indicates the incorporated plutonium and americium. Fig.7 shows the spectra obtained by means of a proportional counter placed over the skull and the right lung of the same subject. It also shows three characteristic peaks which make it possible to identify the X-ray radiation of plutonium with energies of 13.6 keV, 17.4 keV and 20.4 keV respectively. The said spectra provided evidence that this is a typical case of the lung-osteo-hepatic type of radio-nuclide distribution.

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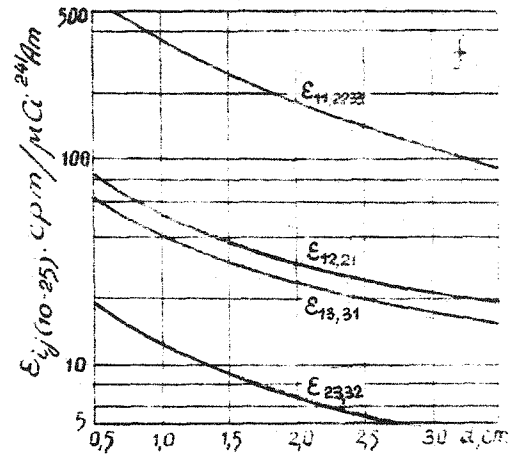
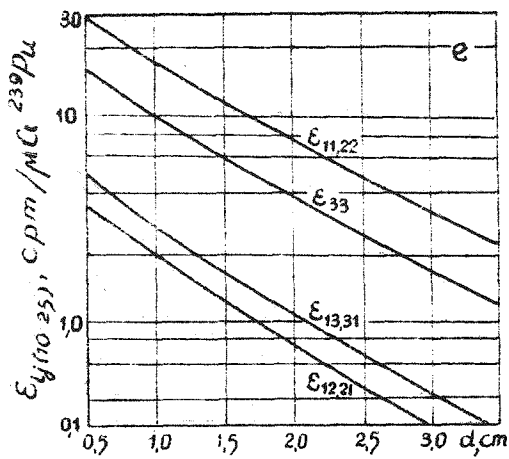
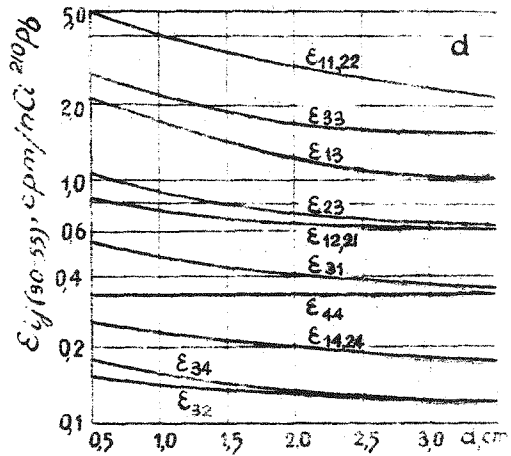
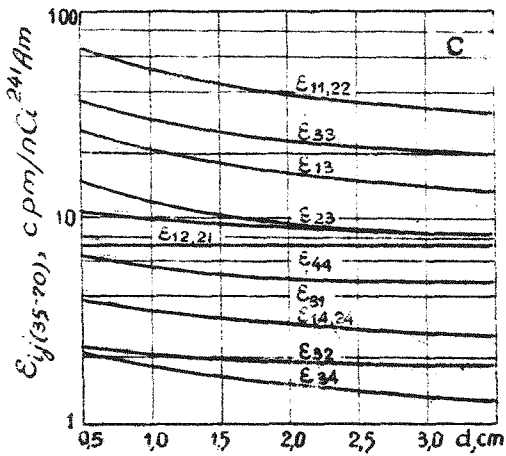
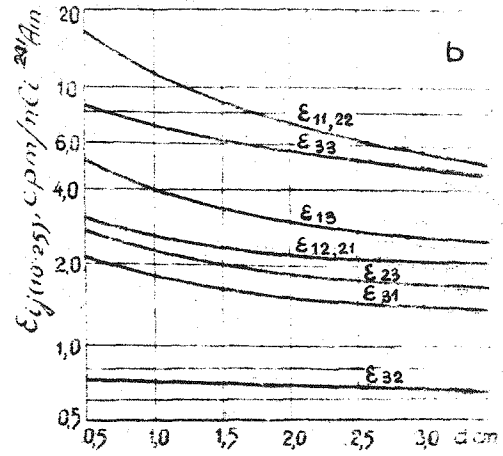
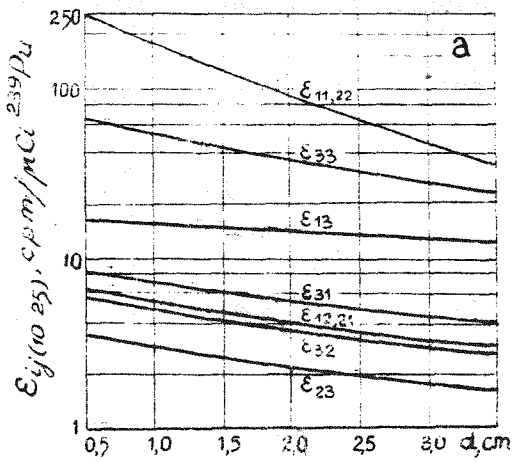


Fig. I. Calibration coefficients ϵ_{ij} vs soft tissue thickness for scintillation counter in the energy range: a) 10-25 keV ^{239}Pu , b) 10-25 keV ^{241}Am , c) 35-70 keV ^{241}Am , d) 30-55 keV ^{210}Pb and for proportional counter in the energy range 10-25 keV: e) ^{239}Pu , f) ^{241}Am .

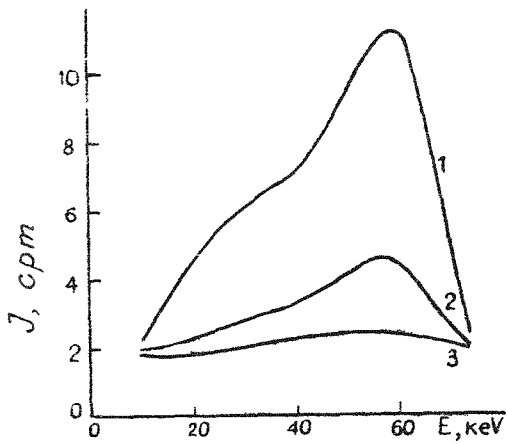


Fig. 2. Spectra ^{241}Am : 1-fantom (IOnCi), 2-contaminated subject (2nCi), 3-control subject.

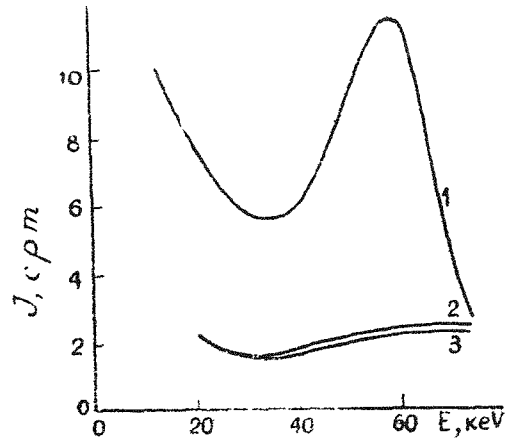


Fig. 3. Spectra ^{241}Am : 1-contaminated 3.10.72; 2-6.22.72. 3-control subject.

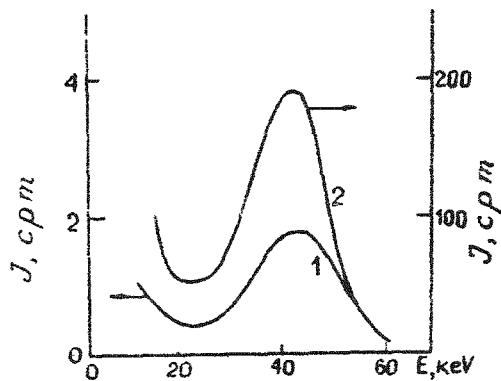


Fig. 4. Net spectra ^{210}Pb : 1-over head contaminated subject; 2-over skull of fantom.

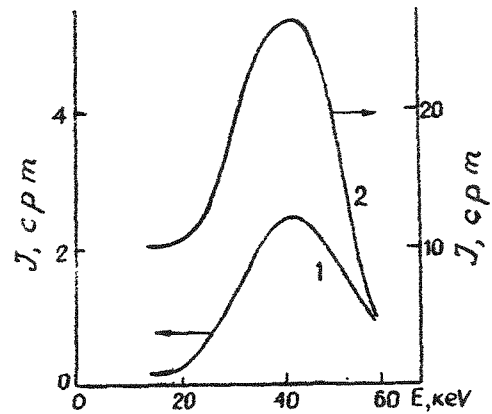


Fig. 5. Net spectra ^{210}Pb over liver: 1-contaminated subject, 2-fantom.

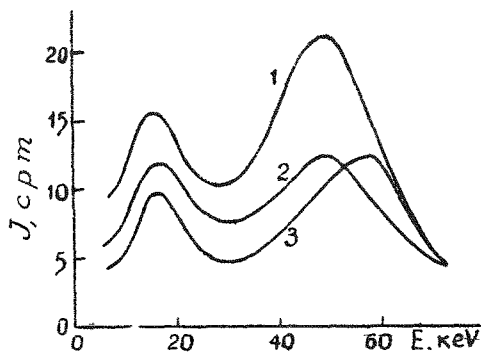


Fig. 6. Spectra Pu+Am over: 1-right lung, 2-liver, 3-Head of contaminated subject.

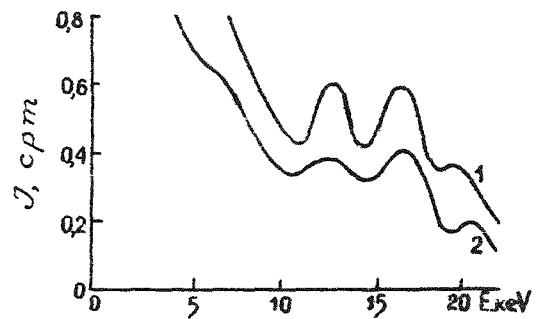


Fig. 7. Spectra Pu+Am over: 1-right lung, 2-head contaminated subject.

ОПТИМИЗАЦИЯ УСЛОВИЙ ГАММА-СПЕКТРОМЕТРИЧЕСКИХ ИЗМЕРЕНИЙ
В РАДИАЦИОННОЙ ЗАЩИТЕ

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Аннотация

На основе нового критерия радиометрического качества и анализа параметров около 100 спектрометров гамма-излучения человека рассматриваются вопросы оптимизации измерения радиоактивности препаратов и организма человека.

Abstract

Some problems of sample and human body radioactivity measurement optimization are discussed on the basis of new radiometric quality criteria and analysis of about 100 whole-body counters' data.

Определение оптимальных условий спектрометрических измерений малых активностей производится с помощью критерия радиометрического качества (КРК), максимальное значение которого соответствует параметрам, обеспечивающим наивысшую чувствительность методики. Ранее было показано^{1;2}, что используемые на практике КРК в виде комбинаций скорости счета препарата n_0 и фона n_φ ($n_0^2/n_\varphi; n_0$) дают неточный оптимум режима, т.к. действительны только в экстремальных случаях $n_0 \ll n_\varphi$ и $n_0 \gg n_\varphi$ ^{3;4}. Истинный оптимум при любом соотношении n_0/n_φ указывает КРК $Q = I/\varepsilon^2 T$, где ε - относительная ошибка и T - продолжительность измерения препарата. Так, например, для исходного $n_0/n_\varphi = 5$ КРК Q указывает, что без ухудшения чувствительности методики допустимо увеличение фона в $n_{\varphi 2}/n_{\varphi 1} = 20$ раз, если при этом эффективность возрастает в $n_{0 2}/n_{0 1} = 3$ раза, (см.рис.1), тогда как КРК n_0^2/n_φ допускает возрастание фона только в 9 раз. Вычисление Q производится с помощью набора кривых $Q = \text{const}$, в координатах $n_0; n_0/n_\varphi$ или по выра-

жению Q через эти параметры. Кривые зависимости Q от активности A препарата дают сопоставление приборов по чувствительности с учетом их эффективности и фона, а значение Q указывает степень трудоемкости радиометрии определенных уровней активности. Соотношение Q , A , E_γ , ширины канала регистрации определяет оптимальный режим и реально измеряемую активность. Энергетическая зависимость Q близка к обратной пропорциональности в диапазоне $0,3 \pm 0,6$ Мэв для A от I до 50 пккюри, имеет плоский минимум в области $0,7 \pm 0,8$ Мэв с нарастанием при $E_\gamma = I$ Мэв, существенный при $A \ll I$ пккюри; при A от $0,05$ до I пккюри Q уменьшается вплоть до $E_\gamma = I,5$ Мэв. Для спектрометров с кристаллами NaJ $\varnothing 40 \times 40$ мм и $\varnothing 70 \times 70$ мм Q изменяется в диапазоне $E_\gamma (0,06 \pm 2,5$ Мэв), соответственно, в 25 и 10 раз. При этом отличие Q от упрощенного КРК $- n^2/n_\phi$ достигает $15-30$ уже при активности препаратов $0,1 \pm 1$ нкюри. Для оценки трудоемкости радиометрии препаратов различной активности в диапазоне $E = 0,1 \pm 1,5$ Мэв были рассчитаны значения Q для различных вариантов геометрии измерения. На рис.2 приведена номограмма для кристалла NaJ $\varnothing 70 \times 70$ мм в чугунной защите толщиной 10 см и препаратов объемом от 5 до 2000 см³, позволяющая оценить степень трудоемкости радиометрии препаратов активностью от $0,01$ до 10 нкюри. Результаты анализа показывают, что обобщенный КРК Q уточняет до $2-7$ раз оценки детектируемых уровней активности, которые можно получить из упрощенных КРК. С применением КРК Q построена номограмма (рис.3), связывающая значения A , n_ϕ , эффективности (Θ %), полной продолжительности измерения T и относительной ошибки (ε %) в диапазоне $A = 0,5$ пккюри $\pm 0,1$ мккюри; $n_\phi = 0,01 \pm 1000$ имп/мин; $\Theta = 0,001 \pm 100$ %; $T = 1 \pm 1000$ мин.; $\varepsilon = 2 \pm 50$ %. Номограмма весьма удобна при планировании экспериментов с радиометрическими измерениями.⁶

Оптимизация конструкционных параметров аппаратуры приобретает особое значение при разработке спектрометров гамма-излучения человека (СИЧ), для которых характерна высокая стоимость защитной камеры и детекторов большого объема. В данном случае из-за трудности учета всех факторов расчетные методы или моделирование не позволяют определить достаточно достоверно зависимость радиометрических параметров СИЧ от конструктивных параметров, характеризующих геометрию измерений, материал и толщину защиты и пр. Нами был предпринят анализ параметров более 100 СИЧ, опубликованных в сводке⁷. Рассматривались дифференциальные и куму-

лятивные распределения параметров, исследовалась связь между параметрами, определялись значения для каждого СИЧ. Несмотря на существенные различия конструкции СИЧ, анализ выявил отдельные закономерности.

Ниже приводятся результаты относящиеся к оценке толщины d защиты и объема кристаллов детектора. Ранее было показано ⁸, что для сравнения качества защит следует пользоваться плотностью фона (отношением скорости счета фона N_p к интервалу энергий ΔE и полной поверхности S кристалла), которая в широком диапазоне энергий гамма-излучения не зависит от размеров кристалла. Распределение СИЧ по величине плотности фона в области 0,66 и 1,46 Мэв показано на рис.4. На рис.5 представлена зависимость от толщины d средней плотности фона для 4 групп стальных защит в диапазонах толщин $d=10+12,5$; $14+16$; $16-19$ и 20 см. Из рис.5 видно, что увеличение толщины защиты от 15 до 20 см сопровождается уменьшением фона порядка 30%, т.е. снижением минимальной обнаруживаемой активности лишь на 15-20%. Поэтому 15 см следует считать оптимальной толщиной стальной защиты. При такой толщине больший эффект дают дополнительные затраты, направленные не на усиление защиты, а на увеличение объема кристалла детектора. По средним значениям плотностей фона в области 0,66 и 1,46 Мэв защиты из свинца толщиной 10-12 см оказались эквивалентны 20 см стали, а толщиной 5-6 см - 15 см стали и на 30-35% эффективнее стальных защит толщиной 10-12 см. Поэтому облегченные защиты транспортных СИЧ целесообразно изготавливать из свинца.

На рис.6 показаны кумулятивные распределения СИЧ по величине относительной статистической ошибки $\Delta K/K$ определения содержания калия (150 г) в организме человека за 20 минут (нижняя кривая) и по величине активности A_0 инкорпорированного цезия-137, определяемой с точностью 20% за 5 минут (без учета вклада K-40) - верхняя кривая. Распределения показывают, что 90% СИЧ способны измерять за 5 минут менее 10 нкюри цезия-137 и определять за 20 минут среднее содержание калия с точностью лучше 8%. Зависимость от суммарного объема V кристаллов СИЧ средних значений A_0 и $\Delta K/K$, полученных для отдельных групп СИЧ с типичными значениями V , представлена на рис.7 (здесь опущены несколько СИЧ с малым объемом кристаллов, для которых A_0 и $\Delta K/K > 20$). Видно, что радиометрические качества СИЧ резко ухудшаются при $V < 1500$ см³. Однако, с увеличением суммарного объема

кристаллов от 2000 до 6800 см³ чувствительность возрастает в среднем лишь в 1,7 раза. Поэтому с точки зрения радиометрического качества оптимальным является применение одного кристалла $\emptyset 15 \times 10$ см ($V \sim 1800$ см³) или $\emptyset 20 \times 10$ ($V \sim 3200$ см³). Увеличение суммарного объема кристаллов свыше 3500 см³ целесообразно лишь в том, случае, если применяют несколько детекторов для улучшения изочувствительности геометрии измерений.

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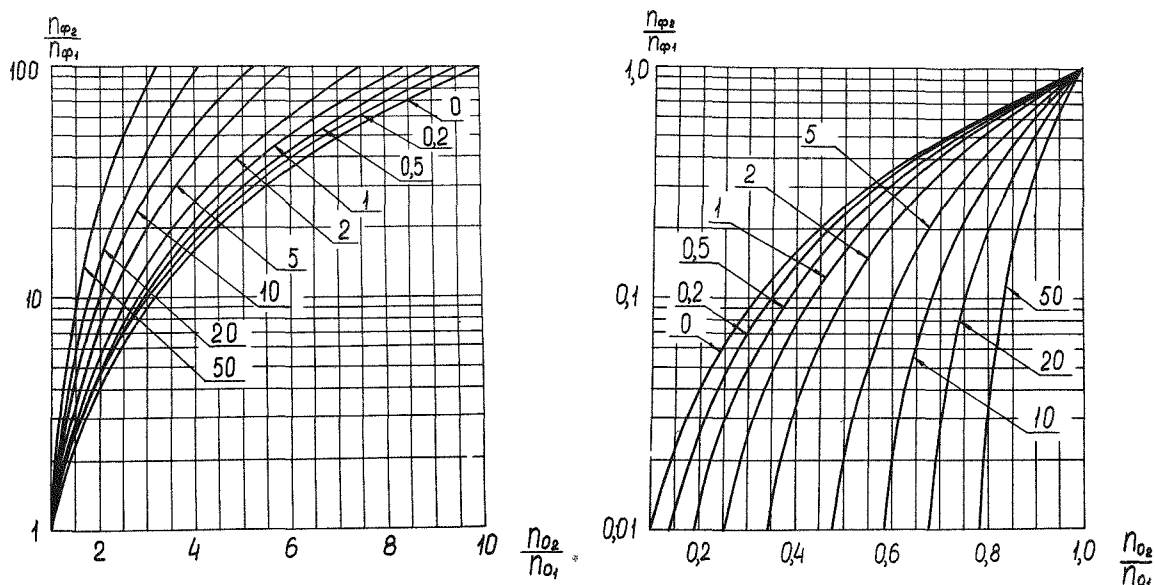


Рис. 1.

ПОДПИСИ К РИСУНКАМ.

- Рис.1. Допустимая кратность изменения фона $n_{\phi 2}/n_{\phi 1}$ при изменении эффективности регистрации в n_{o2}/n_{o1} раз, не вызывающая ухудшения чувствительности методики. Параметр у кривых соответствует исходному значению n_o/n_{ϕ} .
- Рис.2. Степень трудоемкости радиометрии проб объемом от 5 до 2000 см³ с помощью детектора с кристаллом NaJ \emptyset 70x70 мм в чугунной защите толщиной 10 см.
 I зона: $\epsilon \leq 10\%$ T=1 мин II зона: $\epsilon \leq 10\%$ T=15 мин.
 III зона: $\epsilon \leq 20\%$ T=1 час IV зона: $\epsilon \leq 30\%$ T=4 часа.
- Рис.3. Номограмма для определения статистических характеристик режима радиометрии.
- Рис.4. Распределение СИЧ по плотности фона в области 0,66 и 1,46 Мэв.
- Рис.5. Зависимость средней плотности фона в области 0,66 и 1,46 Мэв от толщины α стальной защиты.
- Рис.6. Распределения СИЧ по величине относительной ошибки $\Delta K/K$ определения 150 г калия в организме человека за 20 мин. (нижняя кривая) и по величине A_o активности цезия-137, измеряемой за 5 минут с точностью 20% (верхняя кривая).
- Рис.7. Зависимость средних значений $\Delta K/K$ и A_o от суммарного объема V кристаллов СИЧ.

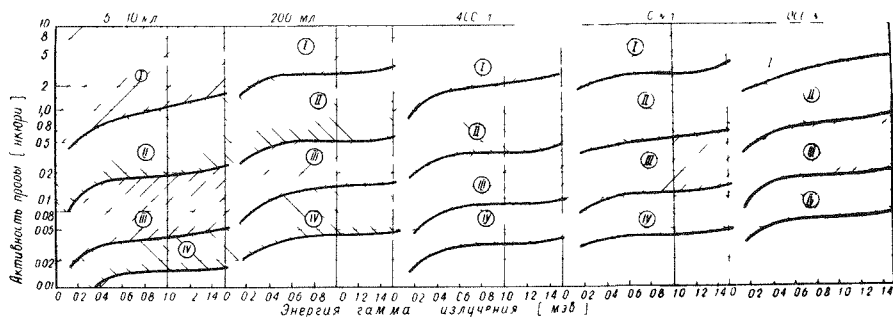


Рис. 2.

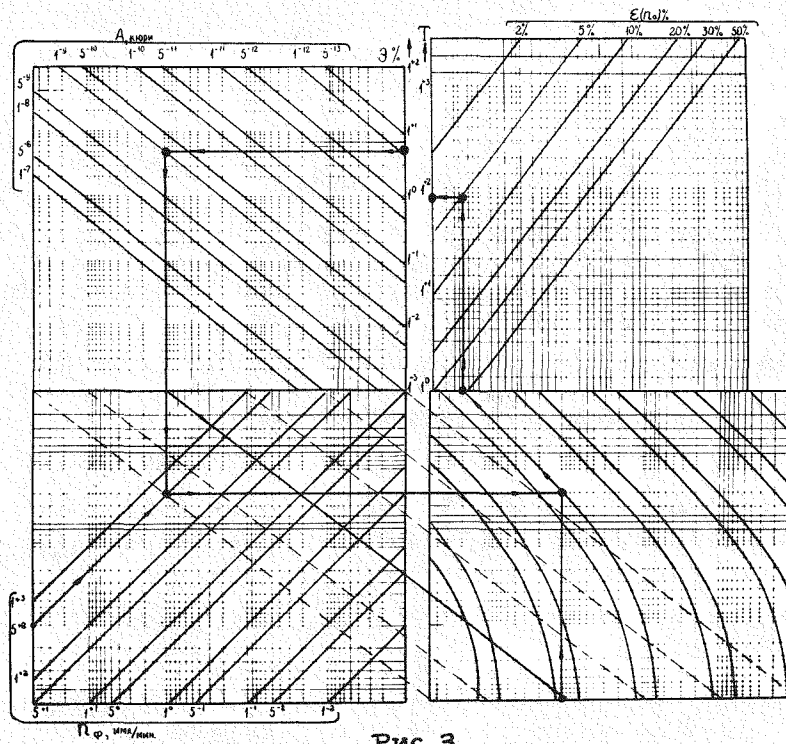


Рис. 3.

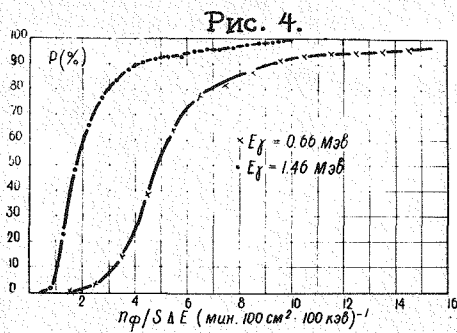


Рис. 4.

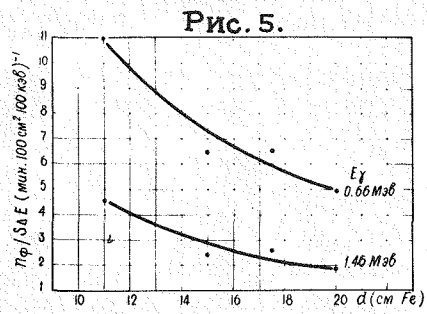


Рис. 5.

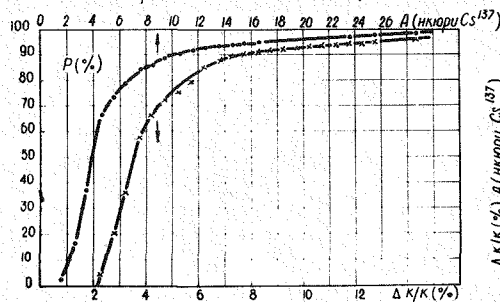


Рис. 6.

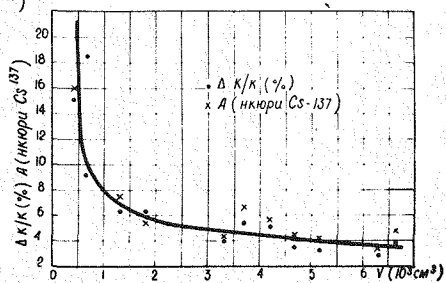


Рис. 7.

THE METHODS OF ABSOLUTE CALIBRATION OF EQUIPMENT FOR
MEASUREMENTS OF Pb-210, Pu-239 AND Am-241 IN HUMAN BODY

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Abstract

Calibration of detectors designed for in vivo measurements of ^{210}Pb , ^{239}Pu and ^{241}Am in human body was made by means of an anthropomorphic phantom. Absorption of low energy photons (13 - 60 keV) in materials simulating various biological tissues was studied. The calibration coefficients for the fat tissue ($Z=5.9$) were 3 times as high as those for the muscular one ($Z=7.4$), when the tissue thickness was 2 cm. In this connection, the distribution and ratio of the said tissues in human body were studied, using a group of 30 men. In calibration phantom measurements, a tissue equivalent material simulating the biological tissue with 22% of fat tissue and 78% of muscular one was used. Detectors have been calibrated for measurements in the energy bands: 10-25 keV ^{239}Pu , 30-55 keV, ^{210}Pb , 10-25 keV and 35-70 keV ^{241}Am and tissue absorber thicknesses in the range from 0.5 to 4 cm. The effect of such factors as variations in the shape and location of body organs, radiations of isotopes deposited in the skeleton and some others was taken into account.

Introduction

The interpretation of the results of in vivo measurements of the body ^{210}Pb , ^{239}Pu and ^{241}Am depends on the choice of calibration coefficients appropriate to a specific subject. Detectors may be calibrated, using (a) volunteers who take a safe dose of the assayed isotope ^{1,2}, or (b) anthropomorphic phantoms. The calibration made on volunteers may provide sufficiently realistic coefficient values which allow for both the complex structure of the bone and soft tissue shield and the actual distribution of inhaled aerosol throughout the lungs.

However, such factors as the availability of volunteers, the choice of isotopes to simulate a given radionuclide as well as the possibility of modelling of nothing more than a particular case of the radionuclide distribution in the lungs limit the scope of this direct method of calibration.

This paper deals with the further development and improvement of the phantom calibration technique which can provide numerical calibration coefficients for the deposited radionuclide distribu-

tion of the lungs-liver-skeleton pattern.

Phantom Design and Tissue Equivalent Materials

The basic criteria used in the phantom development were its tissue equivalence in the 10-100 keV radiation band, modelling of different types of radionuclide distribution and simulation of different thicknesses of the tissue absorber in the region of the chest.

The complete phantom assembly consists of component phantoms, of the skull, chest and arms and legs. A natural human skull is used as the skull phantom and hollow polyethylene cylinders simulate the extremities.

The chest phantom assembly comprises a thin (0.2 cm) plexi-glass shell, containing a human thoracic skeleton and man-made organs (the lungs and liver). The free space of the shell is filled with the tissue equivalent material. The shell front wall is movable and it is provided with an attachment for setting it at a desired distance from the phantom sternal ribs. The front wall and the phantom base are supplied with portholes for filling the tissue equivalent material and artificial organ replacement.

Phantom Filling Materials

The muscular and fat tissue and the skin are the main constituents of the human tissue covering the thoracic cage. The calculated values of the mass absorption coefficients for these tissues (cm²/g) at 10-100 keV shown in Table 1 (Columns 2, 3 and 4) suggest that the fat component should be taken into account in the selection of the tissue equivalent material. (The skin, as far as its absorption properties are concerned, may be assumed to be similar to the muscular tissue).

Table 1

Tissue Energy band	Pectoral muscle	Fat	Skin	Mean tissue shield	Tissue equivalent material
10	5.249	2.406	4.854	4.579	4.520
15	1.633	0.815	1.499	1.356	1.357
20	0.789	0.409	0.722	0.684	0.673
40	0.259	0.217	0.237	0.247	0.243
50	0.221	0.195	0.199	0.212	0.209
60	0.201	0.184	0.181	0.195	0.195
100	0.170	0.161	0.151	0.166	0.163

The experimental testing of the fat and muscular tissues for their absorption properties were carried on the phantom, which was alternately filled with materials simulating each of these tissues. Attenuation curves for the X-ray radiation of ²³⁹Pu emitters uniformly distributed throughout the lung model were obtained. The experimental results given in Table 2 are consistent with the data in Table 1 and support our suggestion.

Table 2

Tissue thickness, cm	1	2	3	4
The ratio of photons which passed through the "fat" to those which passed through the "muscles"	1.4	3	6	10

The ratio of the thicknesses of the fat and muscular tissues covering the thoracic cage in the region of the lungs was measured in 18 male corpses and was found to vary from 34/66 to 10/90, the mean value being 22/78.

Table 1 (columns 5 and 6) gives values of the mass absorption coefficients for the experimental mean tissue consisting of 22% fat and 78% muscles and the tissue equivalent sugar and magnesium oxide based material. The electronic densities of this material and the simulated tissue are $3.24 \cdot 10^{23}$ and $3.32 \cdot 10^{23}$ electron/gram, respectively.

The models of organs, i.e. the lungs and liver, were made of the tissue equivalent material to conform to the average size, shape and density of their human prototypes. The model shell was capron. The lung model shell was filled with a cotton fabric soaked in a NaCl solution to reproduce the desired values of $Z=7.4$ and $\rho=0.27$ g/cm³. The same material was used for filling the liver and chest shells. The experimental coefficients of self-absorption of ²³⁹Pu radiation in the lungs and liver were found to be 3.4 and 5.1, respectively.

Measurement of Soft Tissue Thickness

Owing to a considerable attenuation of low-energy X-ray radiation in the tissues, the accuracy of calibration coefficients selected for the measurements in the monitored subject is materially dependent on the precision of measurements of the thicknesses of soft tissues covering the thoracic cage.

To develop techniques for measurement of soft tissue thicknesses, radiograms of tissues in a special sagittal plane were made. Previously, the ratio of the average thickness across this section d_s to the mean thickness of these soft tissues across the whole area covered by the detector d_g was obtained in corpse measurements.

The equation $d_s/d_g = 1.05 \pm 0.12$ holds for this ratio in a wide range of W/H variations of 0.37 - 0.52, where W - weight, kg; H - height, cm.

The subject's posture proved to influence the thickness of the tissue shield in the chest region. The smallest thickness of the soft tissues over the lungs was found in in vivo measurements to be, when arms are raised and placed behind the back of the subject's head. It was shown experimentally that, for this posture, the absorber thickness decreases by 0.48 cm, as compared with a pose with arms at sides, which is consistent with the value of 0.5 cm, given in This posture was accepted as standard in all subsequent measurements.

The measurement data for soft tissue thicknesses in 26 subjects were related to different parameters of the body and approximated by means of functions, such as $d = \varphi(W, H, C, C_1, C_2)$, where C, C_1, C_2 - circumferences of the chest, waist and hips, res-

pectively, cm; W - weight, kg, H - height, cm. These parameters were measured simultaneously with radiograms being taken. The dependence of d on the selected parameters is best approximated by the equation:

$$d = 118 W/H \cdot k_1 \cdot k_2 - 39.2, \text{ where } k_1 = C/C_2, k_2 = C/C_2$$

The graph showing this relationship is given in Fig.1.

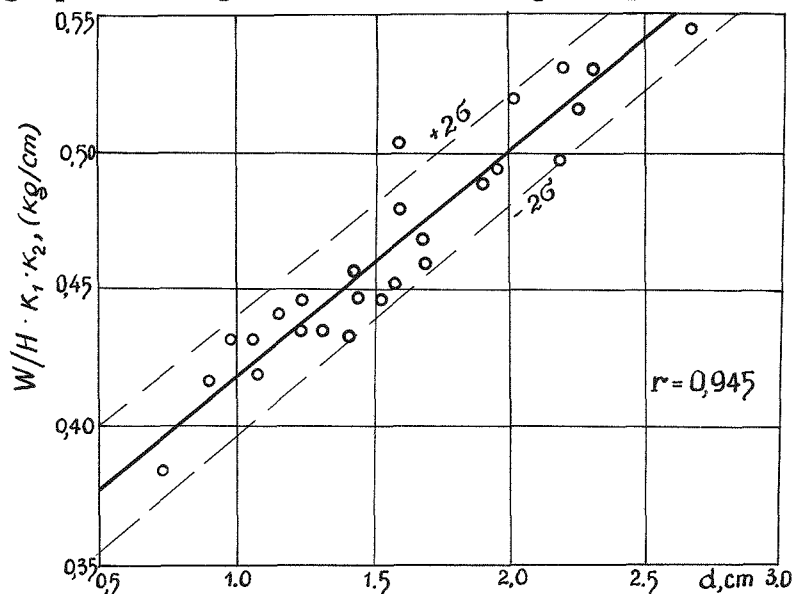


Fig.1. The correlation of soft tissue thickness d and parameters W/H, k_1 and k_2 .

Thus, simple measurements of the body parameters make it possible both to assess the thickness of the soft tissue layer covering the thoracic cage and to choose suitable calibration coefficients.

Calibration Procedure

The lungs, liver and skeleton are the chief sites of ^{210}Pb , ^{239}Pu and ^{241}Am deposition in human body. Suitable sets of calibration coefficients are required for the assessment of burdens of these radionuclides in the said organs on the basis of in vivo measurements.

100 point emitters of known activity were placed uniformly alternately throughout the models of the lungs and liver. The distribution of ^{210}Pb or ^{241}Am emitters in skeletal bones is given in Table 3. Measurements in the four points, i.e. the left lung, the right lung, the liver and the skull, were carried out for each insertion of the emitters into one of these models. The pulse count rate was registered at 10-25 keV for calibration with respect to ^{239}Pu , 10-25 keV and 25-70 keV (^{241}Am) and 30-35 keV (^{210}Pb).

The data of the standard calibration of NaI scintillation counter ($S=177 \text{ cm}^2$ and $h=0.1 \text{ cm}$) for a 1.7 cm - thick soft tissue are given in Table 4.

Table 3.

Distribution of activity in the skeleton

Bones of skeleton	Weight %	Quantity of sources		Activity nCi
		11 nCi	2nCi	
Skull	14.1	50		550
Chest Cage	15.3		210	588
1. Sternum	1.2		16	44.8
2. Ribs	9.1		126	352.8
3. Clavicles	1.2		16	44.8
4. Scapulae	3.8		52	145.6
Vertebral Column	9.0	38		418
1. Cervical	1.4	5		55
2. Thoracic	4.2	14		154
3. Lumbar	3.4	12		132
4. Sacrum	2.0	7		77
Pelvis	7.8	27		297
Arms	16.2	56		616
1. Radii	5.9	20		220
2. Ulnae	4.4	16		176
3. Hands	5.9	20		220
Feet	35.6	126		1386
Femora	15.7	56		616
Tibiae	14.2	50		550
Feet	5.7	20		220
Total	100	297	210	3855

The sets of calibration coefficients for soft tissue thicknesses from 0.5 to 3.5 cm for scintillation and proportional counters are given in our paper "Dosimetric Monitoring of Content of the Radionuclides with a Low Radiation Energy in the Human Organism"

The total error for calibration coefficients (Table 4) due to the variations in the organ shape, the different location of the organs in the phantom the detector displacement with respect to the phantom front wall, the counting statistics and the precision of emitters' calibration is 16% for ^{239}Pu ; 8% - for ^{210}Pb and 6% - for ^{241}Am .

The error due to the assumption of the uniform distribution of the assayed radionuclide throughout the organ was determined separately. The values of calibration coefficients for different patterns of radionuclide distribution in the lung model are shown in Tables 5 and 6.

Table 4

Calibration Factors for Homogeneous Distribution cpm/nCi/d=1.7cm

Source position	Counter position	Radionuclides and energy band			
		²³⁹ Pu	²¹⁰ Pb	²⁴¹ Am	
		10-25	30-55	10-25	35-70
Left lung	left lung	0.120	3.1	8.5	40.0
	right lung	0.004	0.7	2.5	8.4
	liver	0.04	0.2	0.7	2.0
Right lung	left lung	0.004	0.7	2.5	8.4
	right lung	0.120	3.1	8.5	40.0
	liver	0.006	0.4	1.6	5.0
Liver	left lung	0.003	0.8	2.1	10.0
	right lung	0.016	1.4	3.3	18.0
	liver	0.040	1.8	5.9	22.6
Skeleton	left lung		0.22		3.6
	right lung		0.22		3.6
	liver		0.15		1.7
	skull		0.40		7.5

Table 5

Calibration Factors (cpm/nCi, d=17 cm) for ²⁴¹Am 35-70 keV

Position		Emitter distribution in the lung			
Emitters	Detectors	Uniform	Upper part	Lower part	60% in lymphatic nodes; 40%-uniform
Left lung	Left lung	40.0	35.0	43.0	42.0
	Right lung	5.4	7.9	8.7	8.3
	Liver	2.0	1.2	2.3	1.9

Calibration Factors (cpm/nCi, d=1.7 cm) for ²³⁹Pu/10-25 keV

Position		Emitter distribution in the lung			
Emitters	Detectors	Uniform	Upper part	Lower part	60% in lymphatic nodes; 40%-uniform
Left lung	Left lung	0.120	0.085	0.130	0.105
	Right lung	0.004	0.003	0.003	0.003
	liver	0.004	0.001	0.005	0.003

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ЧУВСТВИТЕЛЬНЫЙ МЕТОД РАДИОМЕТРИИ
ИНКОРПОРИРОВАННЫХ НУКЛИДОВ ТОРИЕВОГО РЯДА

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The complex of the apparatus and methods for the assessment of Th-228 body burden in man was developed. The correlation is found between content of thoron ($\text{Q}_{\text{Th}}\text{A}$) in the breath, which was measured by a special scintillation apparatus (0.05 pCu/l sensitivity), and Ra-224 in the excretion ($\text{Q}_{\text{R2}}\text{X}$), which was measured by the method of the high emanating samples preparation (0.05 pCu sensitivity), with the activity of Th-228-body burden in man.

Расширение использования в промышленности тория и различных неметаллических руд, в которых торий присутствует как примесь ведет к увеличению контингента людей, работающих в условиях контакта с радионуклидами ториевого ряда. В связи с этим понятен интерес к проблеме контроля поступления в организм и накопления в нем тория и продуктов его распада /1 ; 2 ; 3 ; 4 /.

В соответствии с указаниями МКРЗ (ICRP-10) и НКРЗ СССР (НРБ-69) такой контроль должен строиться в основном на определении индивидуального содержания в организме изотопов ториевого ряда, среди которых большое значение имеет радиоторий, ответственный вместе с дочерними продуктами за 90% энергии альфа-распада ториевого ряда.

Наиболее точную оценку содержания радиотория в организме (в пределах своей чувствительности) дает метод гамма-спектрометрии человеческого тела с помощью СИЧ, однако, как показывает практика, его чувствительность (1-5 нкюри) в большинстве случаев недостаточная для проведения массовых обследований. В принципе большую чувствительность могут обеспечить методы оценки содержания радиотория в организме по активности торона в выдыхаемом воздухе и радия-224 в выделениях. Для их реализации нужно знать величины коэффициента эманирования торона и перехода эндогенного радия-224 в выделения, которые для наиболее важного в условиях профессионального контакта ингаляционного поступления тория в организм изучены недостаточно.

Чтобы получить необходимые соотношения, мы предприняли попытку с помощью аппарата камерных моделей проанализировать имеющуюся в работах / 6 ; 7 ; 8 ; 9 ; 10 и других / экспериментальную информацию о метаболизме тория и продуктов его распада в организме при их внутривенном и аэрогенном поступлении.

Анализ показал, что в случае аэрогенного поступления радиотория в растворимой, ионной форме ежедневно с экскретами выводится доля равновесного радия-224, равная $4 \pm 5\%$ от содержания радиотория в организме - λ_{T_2} , причем перераспределение со временем депонированного радиотория существенно не сказывается на этой величине; чего нельзя сказать об активности торона в выдыхаемом воздухе, которая за время $2T$ (где $T \sim 60$ дн - период полувыведения торона из легких) изменяется от 13 до 6% от величины λ_{T_2} . Отметим, что скорость выведения радиотория не превышает $0,2\% \lambda_{T_2}$ в день.

При поступлении радиотория в нерастворимой форме ежедневная экскреция равновесного радия-224 зависит от дисперсности пыли, с которой радиоторий поступает в легкие: для пыли двуокиси тория с $СМД \sim 1 \mu$ ежедневно выводится $\sim 1\% \lambda_{T_2}$, а для пыли двуокиси с $СМД = 0,2 \mu$ - $2\% \lambda_{T_2}$; в то же время эманирование торона зависит от дисперсности слабее и для указанных пылей коэффициент эманирования $\sim 6\% \lambda_{T_2}$ и также как и экскреция радия-224 существенно от времени не зависит.

Для определения содержания в выделениях радия-224 нами используется методика, основанная на эманомерии сухих пэпаратов, коэффициент эманирования торона из которых специальным приготовлением доведен до величины большей 0,8.

Чувствительность метода 0,05 пкюри радия-224 в пробе /12/. Этот метод определения радия-224 проще альфа-спектрометрии / 13 / или метода "смешивания" / 14 /, с его помощью произведена оценка величины ежедневной экскреции радия-224 с мочой - F у людей, не имевших контактов с торием, $F \sim 1$ пкюри/с.

Весьма чувствительный метод измерения содержания торона в выдыхаемом воздухе был предложен I. Hursh и A. Lovaas / II /. Однако его реализация наталкивается на трудности поддержания высокой и стабильной сорбционной способности угольной ловушки, с помощью которой из выдыхаемого воздуха извлекается торон.

Для определения концентрации торона в выдыхаемом воздухе у людей в данной работе сконструирован удобный в эксплуатации, автономный сцинтилляционный эманомер, в котором регистрируется альфа-распад торона и тория-А в проходящей через камеру прибора струе воздуха.

Объем и конструкция камеры выбраны так, чтобы обеспечить оптимальную величину коэффициента использования активности эманиции $S = 200 \frac{1/\text{час}}{\text{пкюри/л}}$ при средней скорости продувки 8 л/мин.

Этот прибор, когда последний соединен с человеком, легкие которого в течение 20' очищаются безрадоновым воздухом, как показали специальные измерения не превышает 10-12 1/час.

Это позволяет измерять концентрацию торона в выдыхаемом воздухе $\sim 0,05$ пкюри/л.

С помощью этого прибора произведена оценка естественного содержания торона в выдыхаемом воздухе у некурящих ($0,14 \pm 0,02 \frac{\text{пкюри}}{\text{л}}$ для группы 10 чел.). У курящих концентрация торона оказалась вдвое большей ($0,28 \pm 0,03 \frac{\text{пкюри}}{\text{л}}$ для группы 10 чел.), что подтверждается данными G. Jovet / 15 / о возможности накопления тория в легких при курении.

Приведенные выше соотношения, связывающие активность тория-228 в организме с активностью радия-224 в выделениях и торона в выдыхаемом воздухе, а также данные о чувствительности использованных радиометрических методик позволяют сделать заключение, что в случае ингаляционного поступления радиотория в растворимой форме целесообразно определять его содержание в организме по активности радия-224 и выделениях; предел обнаружения при этом ~ 20 пкюри

в организме. Если радиоторий поступает в нерастворимой форме, то лучше, по-видимому, воспользоваться эманометрическим методом (предел обнаружения ~ 10 пкюри радиотория в легких).

В обоих случаях эти методики позволяют определять радиоторий на уровне естественного его содержания в организме.

Разработанные методы применяются для обследования на содержание радиотория у рабочих рудников и горнообогатительных фабрик промышленности редких металлов. У некоторых из них обнаружено повышение содержания торона в выдыхаемом воздухе в пределах 0,3 - 0,6 пкюри/л; поскольку торий в этом случае поступал в форме нерастворимой пыли, можно на основании вышеизложенного заключить, что в легких у них имеется депо радиотория с активностью 60-120 пикоюри.

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ASSESSMENT OF PLUTONIUM IN HUMAN LUNGS
WITH THIN NaI(Tl) DETECTOR SYSTEMS

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Abstract

The development of detector systems for assessment of inhaled Plutonium-Americium dust deposited in human lungs by direct detection of externally emitted low-energy photons, is a continuing programme at Trombay. This paper describes the work done with thin NaI(Tl) scintillation detector systems. The background data inside Trombay steel room in different low-energy bands for crystals of thicknesses 1, 2 and 5 mm are presented. To study the capabilities of three detector systems, each consisting of a set of crystals of the same thickness, a realistic chest phantom of an Indian adult was designed and employed. The chest phantom was constructed from a rib cage of an Indian adult enclosed in a hard polythene cover provided to simulate the chest profile. Measurements on absorption and scattering of low-energy photon (17, 22 and 60 keV) by four constructional materials were made to verify their degree of equivalence to human tissue and granular sugar was used as tissue equivalent material in phantom construction. The counting efficiencies and limits of detection of three detection systems for point sources of plutonium distributed in the central plane of each simulated lung of the designed phantom are reported.

A few normal subjects were counted with one detector system and the increase in background in low-energy region was investigated. The natural radioactivity of the subjects was monitored with a (20.32 cm dia. x 10.16 cm thick) NaI(Tl) crystal in a 50 cm Arc Chair. Finally, the effect of body build of a subject on the counting efficiency of plutonium is commented and our future programme is briefly indicated.

Introduction

At the Bhabha Atomic Research Centre, Trombay, we have an on-going programme of design and development of systems for in vivo assessment of plutonium and other transactinide elements deposited in the lungs. This paper (1) presents the results of our studies on the suitability of Trombay-produced thin NaI(Tl) crystals for this application, (2) describes the design and construction of a chest phantom and its use for calibration of the Pu lung monitor, and (3) compares our results with those of other laboratories using NaI(Tl) detectors, as reported in the literature.

Evaluation of Trombay-Produced NaI(Tl) Crystals

The Trombay-produced NaI(Tl) crystals, used in the present study, are of 7.6 cm diameter and 2 and 1 mm thick. They are canned in Al and

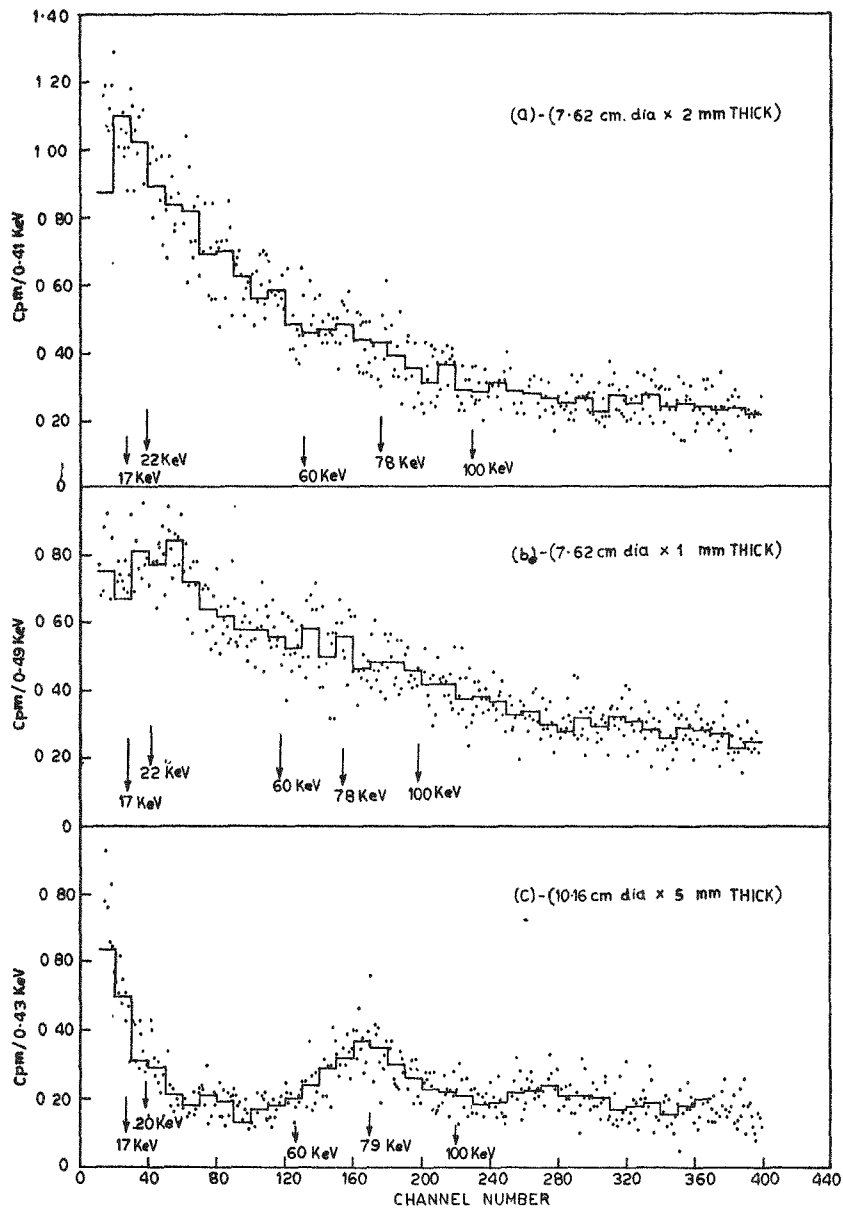


FIG. 1 - A COMPARISON OF THE BACKGROUND SPECTRA OF LOW ENERGY RADIATION INSIDE TROMBAY STEEL ROOM FOR THREE DIFFERENT THIN CRYSTAL DETECTORS. HISTOGRAMS REPRESENT TEN POINT AVERAGES.

are provided with 1 mil thick Al foil radiation entrance window and 3 mm thick glass optical window. They were coupled to selected low-noise Dumont 6363 phototubes and the output from the phototube was fed through an FET pre-amplifier to a 400-channel pulse height analyser fabricated in our laboratory.

A typical pulse height spectrum of a Trombay-produced Pu source, obtained with these crystals, showed that the L X-rays of U are not resolved and exhibited a peak at an average energy of 17 keV. A small peak at 60 keV pointed to the presence of ^{241}Am in the source, the ^{241}Am concentration being about 0.3% of the total Pu alpha activity. The energy resolution at 17 keV was estimated to be about 50%, compared to 46% obtained with a Harshaw integral assembly Model No. 16 MBS 5M/5A Q X (10.16 cm dia x 5 mm thick NaI(Tl) crystal, having 1 mm Be radiation entrance window and 2.54 cm thick quartz optical window) Fig.1 shows the background spectra of low-energy radiations inside the Trombay steel room¹, observed with the three detectors. The histogram depicted represents ten point averages. The Harshaw assembly showed the lowest background rates in the energy bands of interest. These spectral measurements have indicated the aspects in which Trombay-produced crystals require improvement to be suitable for Pu detection².

Construction of Chest Phantom

Speight et al³ had suggested some tissue-equivalent materials for construction of chest phantoms. Due to non-availability of Lincolnshire bolus and Mix D and the handling problems of water, it was decided to test three probable constructional materials, viz. masonite, perspex and sugar, for their degree of tissue-equivalence. The effective atomic number (Z) and electron density (n) for tissue are quoted as 7.33 and 3.32×10^{23} respectively and for water as 7.42 and 3.36×10^{23} respectively. Perspex and masonite have much higher values of Z and n than tissue or water. The half-value thicknesses for water, sugar, perspex and masonite were found as 6.0, 6.0, 7.0 and 5.25 mm respectively at 17 keV, and 14.0, 14.0, 16.75 and 12 mm respectively at 22 keV². The effects of forward scattering by almost equal thicknesses of the four materials as measured by energy shift, degradation of spectral resolution and variation of the ratio of count-rate in the 24-43 keV energy band to that in the 60 keV peak (43.5 - 76 keV) were all found to indicate a similarity of behaviour between water and sugar². Sugar was, therefore, chosen in the construction of the phantom. No attempt was made to simulate the presence of fat in the chest region.

The chest phantom was constructed from a thoracic cage with clavicles, scapulae and shoulder blades taken out of the cadaver of an Indian adult. The whole rib cage fixed at the bottom to a perspex sheet was first enclosed in a frame of thin perspex strips. A hard polyethylene sheet was used to cover the frame to get the chest profile. All measurements were matched to a subject. The vertebrae contained a copper rod to keep them firmly attached. This, however, would not affect the calibration for Pu, since the bone is essentially a dark body for low-energy photons. Two symmetric polyethylene bags were filled with saw dust (density 0.3 g/cm^3), each weighing about 450 g. These were shaped to a human lung and inserted inside the rib cage. Prior to this insertion, point sources of Pu deposited on perspex (1 mm thick both sides) were stuck to a filter paper which was spread longitudinally in the central plane of each simulated lung. The

presence of other parts of the respiratory tract and the heart were not simulated. Granular sugar was filled inside the polyethylene enclosure surrounding the rib cage, to provide simulation of tissue. The chest phantom thus constructed had a circumference of 83 cm. The total activity of Pu incorporated in the simulated lungs was 2.5 uCi.

Multi-Crystal Arrays

The phantom described above was employed to obtain the counting efficiencies and the minimum detectable activities for Pu in lungs for a multi-crystal array geometry simulated with a single detector. The multi-crystal array was selected with a view to achieve a large coverage of the frontal area of the chest phantom. The six crystal array chosen is shown in Fig. 2 together with the phantom outline. Each crystal array consisted of detectors of the same thickness. Thus, three multi-crystal arrays were studied to assess the capabilities of these systems and to evolve an array system of Trombay-produced crystals to count suspected cases of internal contamination by Pu and Am.

Table 1 gives the results with our systems and compares them with data of various Pu-lung monitors employing thin NaI(Tl) scintillation detectors as reported in the literature. The systems compared are those of Swinth & Griffins⁴, Ishihara et al⁵. The first column lists the various monitor parameters. In the net spectrum of radiation from point sources of plutonium distributed in the central plane of each lung longitudinally of the chest phantom we observed that attenuation of 13.6 and 16.9 keV results in shifting of the 17 keV peak to about 20 keV. The presence of ²⁴¹Am was indicated by peaks at 29 keV (26 keV + escape) and then at about 57.5 keV. A major conclusion drawn from Table 1 is that Pu lung monitors employing thin NaI(Tl) crystals in different configurations are not capable of achieving the MDA for plutonium desirable for routine monitoring. We achieved the best results when ²⁴¹Am is used as a tracer for Pu. The use of ²⁴¹Am is certainly not valid for soluble Pu. Nevertheless an immediate estimate of lung burden of Pu for a subject involved in an accident may be derived on this basis if isotopic composition of the contaminant is known. The MDA quoted for our systems are for very low ²⁴¹Am (0.5% of total alpha activity) content in Pu.

The MDA quoted for the multi-crystal arrays are all based on the statistical criterion only, i.e. three times the standard deviation in background rate for a given time of counting. Each subject would increase the background in the low energy region of the array, depending upon the level of internal contamination by other radionuclides and the thickness of the detectors employed. For the 5 mm crystal array, we observed that a subject with 125 gm K and 1 nCi ¹³⁷Cs increased the background by 20 cpm (12-25 keV) region and by 120 cpm in (43.5 - 76 keV) band. ¹³⁷Cs and K contents of the subjects are determined by whole body counting in 50 cm arc chair using a 20.32 cm x 10.16 cm NaI(Tl) detector. Collection of these data is continuing with a view to derive some useful correlations.

The counting efficiencies and MDA for Pu for the three six crystal arrays reported in Table 1 are thought to be valid for humans having body build and distribution of Pu in lungs similar to that of phantom. Several methods to correct the calibration factor for differing body builds have been proposed^{6,7}. We have found it advantageous to use the concept of

Table 1

COMPARATIVE DATA OF VARIOUS Pu-LUNG MONITORS EMPLOYING THIN NaI(Tl) SCINTILLATION DETECTORS

Sl.No.	Monitor Parameters	Swinth and Griffin	Ishihara et al	Our Multi-Crystal Array
1.	Detector size employed and geometry used	Array of 52 NaI(Tl) (2" dia x 1 mm each) four boxes of 13 detectors each. Two boxes positioned on the chest of the reclining subject. Each box edge 2" below the top of the chest. The other one on each side of the armpits	Single (8" x 4 mm) NaI(Tl) on the centre axis of the phantom and at a distance of 1.4 cm from the top plate to the crystal face	(10.16 cm x 5 mm); (7.6 cm x 2 mm); (7.6 cm x 1 mm) Three multi-crystal arrays studied each consisting of a set of six crystals of the same thickness, simulated with a single detector. Detector arrangement relative to phantom is shown in Fig.2. Crystal face in every position remains horizontal with the minimum distance from phantom surface being 1 cm.
2.	Phototubes coupled	Low noise 2" diameter crystal separately	Single 7" dia VMP/II/170 20th Century Electronics without light guide	(10.16 cm x 5 mm) NaI(Tl) : RCA-8055 through 2.54 cm of quartz pipe (7.6 cm x 2 mm) NaI(Tl) : Dumont-6365 through 0.50 cm of glass (7.6 cm x 1 mm) NaI(Tl) : Dumont-6365 through 0.50 cm of glass
3.	Phototube Noise predominance	Below 10 KeV	Below 13 KeV with no light guide	Below 12 KeV for all
4.	Total Sensitive Detector Area	15771 cm ²	103.271 cm ²	154.871 cm ² for 5 mm crystal 57.1 cm ² for the other two.
5.	Energy Bands used to cover (a) Pu- X-ray 17 KeV photopeak (b) Am-241 gamma	(a) (13.6 to 25.6 KeV) (b) (13.6 to 66.5 KeV) both X-ray and gamma covered.	(a) (16 to 31 KeV) (b) (35 to 73 KeV)	(a) (12 to 25 KeV) (b) (43.5 to 75 KeV)
6.	Energy Resolution at 17 KeV level	-	67.5% with no light guide	*46%, 50%, 48%
7.	Background counts per min With distilled water phantom	(a) Pu Ch.- 56.2±2.9 (b) Pu+Am Ch.- 204.8±2.8 (In 4" thick Pb shield lined with Cd and Cu)	(a) Pu Ch.- 51.2 (b) Am " - 135.0 (With no light guide) (In shielded chamber 20 Iron + 3 mm of Pb)	(a) Pu Ch.- 84.0, 204.0, 132.0 (b) Am Ch.- 105.0, 246.0, 234.0 For six crystal array In shielded chamber 20 cm Iron + 3 mm of Pb.
8.	Calibration phantom and source position	Alderson Remab phantom filled with water. Plutonium (720 ppm Am) mixed with lung equivalent material kept in lung cavities	Phantom of Lucite sheets cluster of pt. sources placed in the model of human lungs	Chest phantom from human rib cage. Tissue equivalent material used is granular sugar. Point sources distributed longitudinally in the central plane of each simulated lung.
9.	Counting efficiency for Pu-239 in lungs counts per sec/nCi	(a) 0.641x10 ⁻³ (Pu ch) (b) 4.38x10 ⁻² (X-ray and Am gammas)	(a) 1.34x10 ⁻² (b) -	* (a) Pu ch. 3.83x10 ⁻³ ; 3.46x10 ⁻³ ; 1.65x10 ⁻³ For six crystal array each.
10.	Minimum Detectable amount of Pu	(a) 49 nCi 20 min counting time (b) 67 nCi	(a) 6.2 nCi of Pu-239 100 min (b) -	* (a) Pu Ch. 12 nCi; 28 nCi; 34 nCi and 2.44, 10, 8 nCi if Am is used as a tracer. (For count- Trombay Pu) ing time. For six crystal array each and counting time of 100 min.
11.	Contribution from degraded Cs-137 and K-40 gammas	Human subjects add in (a) Pu Ch.- 63.2 to 130.2 cpm (b) Am Ch.- 458 to 798 cpm	A subject with 130 gK and 4 nCi of Cs-137 adds in (a) Pu Ch.- 12.6 cpm (b) Am Ch.- 75.6 cpm	For 5 mm detector array only; a subject with 125 gK and 1 nCi of Cs-137 contributes: (a) Pu Ch.- 20.0 cpm (b) Am Ch.- 120.0 cpm.

*The three successive values are for 5 mm, 2 mm and 1 mm thick detectors respectively.

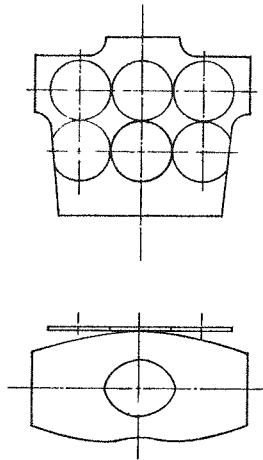


FIG 2-THE MULTI-CRYSTAL ARRAY AND THE PHANTOM
OUTLINE

effective tissue thickness (ETT) which is defined as the thickness of a tissue equivalent material that produces for a point source of Pu, the same overall attenuation of X-rays as would occur in a subject having Pu deposited in lungs. ETT takes account of self-absorption of Pu X-rays in the lungs and the attenuation in tissue overlying the ribs. Since our Pu contains traces of ^{241}Am , we determined the variations in the ratio of count-rates in Am and Pu peaks with different thicknesses of overlying water for a point source on the axis of a thin NaI(Tl) crystal at a fixed distance of 10 cm. Calculating the same ratio for the phantom, we found ETT for the phantom as 6.3 cm. This value was also confirmed by the observed shift of the 60 KeV peak of ^{241}Am .

Our future programme of work in this field will include development of phoswich detectors and improvements in the methods of calibration.

Acknowledgement

We wish to express our sincere thanks to Shri U.R. Marwah, Technical Physics Division, BARC for the supply of thin NaI(Tl) crystals and to Ss B.J. Vaidya, G. Krishnamachari and J.M. Vidhani for the use of the 400-channel pulse height analyser and instrumentation support. The secretarial help rendered by Ss M. Idicula and G.D. Mistry is deeply appreciated.

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IN VIVO MEASUREMENTS FOLLOWING EXPOSURE TO ^{133}Xe
AND ASSOCIATED DOSE ASSESSMENT PROCEDURES

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ABSTRACT

Information is presented on the retention and distribution of ^{133}Xe in the human body as determined by in-vivo counting. Calculations by other workers have shown that for exposure to ^{133}Xe gas the critical dose is that to the skin. It is shown here, with reference to three cases, how measurements of the body content of ^{133}Xe made by in-vivo counting can be used to estimate skin doses.

By reference to actual recent cases attention is drawn to problems caused by ^{133}Xe intakes in the interpretation of external contamination and plutonium-in-lung measurements.

INTRODUCTION

^{133}Xe is produced in the fission of ^{235}U with a total yield (direct and by chain) of about 7%. It decays with a half life of 5.27 days to stable ^{133}Cs by the emission of beta particles of maximum energy 0.34 MeV, gamma rays of energy 81 KeV are emitted in 35.5% of the disintegrations and caesium K X-rays of 30 KeV energy are emitted following 56% of disintegrations. Xenon, a noble gas, which is present in the atmosphere with a partial pressure of about 8 mN/m², is in general chemically inert but appears to combine specifically with haemoglobin (1).

^{133}Xe has been used extensively in medical science for the investigation of lung function; information on the procedures and computations of the retention of ^{133}Xe when used for this purpose have been presented by Matthews et al (2). Measurements of the distribution and retention of ^{133}Xe in the body following both experimental and accidental inhalation in a laboratory manufacturing ^{133}Xe for medical application have been reported by Venner and Devell (3). Guillot (4) has reported on retention experiments with ^{133}Xe , ^{131m}Xe , ^{125}Xe and also stable xenon isotopes. The International Commission on Radiological Protection (5) state that the primary hazard from ^{133}Xe is from external radiation from submersion in a cloud and calculations of internal doses arising from inhalation of radioactive noble gases presented by Whitton (6) confirm this.

Because it is a gas it is possible for ^{133}Xe to leak from nuclear reactor fuel elements into operating areas. Whilst the reactor is operating any ^{133}Xe will usually be accompanied by other fission product noble gases notably ^{88}Kr ($t_{1/2}$ 2.8h) whose daughter product ^{88}Rb is a solid emitting energetic beta particles; the leak will therefore usually be rapidly detected by conventional filter paper air samplers. Because of its longer half life however ^{133}Xe may be released a day or so following reactor shut down virtually without the ^{88}Kr . If the release is a slow one which leads to more or less uniform contamination of the air in the reactor containment conventional external radiation monitor-

ing will adequately detect and assess the hazard. Sometimes however the release may be very localised and although when the activity is dispersed throughout the operating area the resultant radiation level is very low, significant doses may be received by individuals in the immediate vicinity of the release as we will show later.

THE UPTAKE AND RETENTION OF ^{133}Xe IN THE HUMAN BODY FOLLOWING INHALATION

As stated by Matthews et al (2) the uptake of ^{133}Xe into the body is a function of its solubility in blood. However this apparently straightforward situation is complicated by the presence in the atmosphere of naturally occurring stable xenon. The solubility of a gas in a liquid is a function of the partial pressure of the gas and since the normal atmosphere contains xenon at a partial pressure of about 8 mN/m^2 the body will be saturated with xenon at this partial pressure.

We have made measurements of the retention of ^{133}Xe in persons who have been exposed to ^{133}Xe in a reactor environment. Some had been exposed to low level uniform concentrations during reactor operating periods and some to small localised clouds of high concentration following reactor shut down. Measurements made from 1 to a few hours following cessation of exposure to low level uniform concentrations showed the half life of ^{133}Xe excretion to be about 2 hours. Measurements on persons exposed to small high concentration clouds were made over a longer period up to about 80 hours after exposure and showed retention curves very similar to those reported by Venner and Devell (3), viz an initial rapid excretion phase lasting about 4 hours during which the half life is less than 1 hour followed by a slower elimination rate with half life of about 6 hours for the remainder of the period.

DISTRIBUTION OF ^{133}Xe IN THE BODY FOLLOWING INHALATION

We have made measurements of the distribution of xenon-133 in the body (1) after exposure for several hours to low-level contamination and (2) after exposure to a small cloud of high concentration. Profile curves, obtained one day after exposure, are shown in figure 1. Some differences between the curves may be attributed to differences in the scanning techniques. Curve (1) was obtained by scanning with a collimated detector above the supine subject whereas a more finely collimated detector, located under the body, was used for scan (2). The 'depressions' in counting-rate in scan (2) could be due to absorption of the 80 KeV gamma-rays in bone. The 'depressions' in the counting-rate from the chest could be due to attenuation in the ribs and the decrease in counting rate at about 110 cm could be attributed to absorption in the pelvis. These effects would not be so marked with the wider-angle collimator particularly as it was used above the subject. The maxima of the distributions occur at 60-80 cm from the top of the head and could indicate accumulation of xenon-133 in liver. Neither plot shows the large depression in the chest region noted by Venner and Devell (3) in a scan made some hours after an accident inhalation.

CALCULATION OF SKIN DOSE FROM IN-VIVO MEASUREMENTS

The critical tissue dose from exposure to ^{133}Xe is that to the skin. If however the ^{133}Xe is released in the form of a small cloud normal external radiation monitoring devices will often not give a correct indication. Under those circumstances however it is possible to calculate a skin dose from

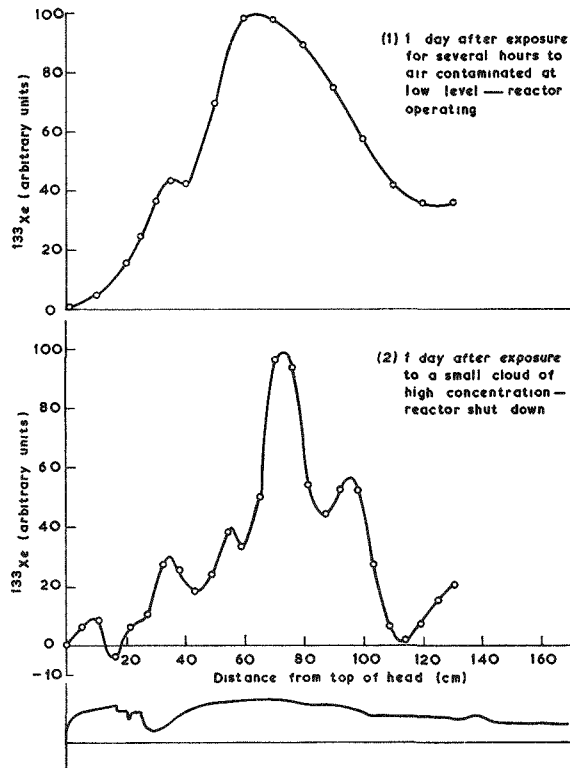


FIG 1 DISTRIBUTION OF ^{133}Xe IN TWO SUBJECTS

in-vivo measurements. Although the calculation will be subject to considerable error it should be sufficiently accurate to decide whether or not an over exposure had occurred. This is illustrated by the following cases.

Case 1

A technician L, removing a thermocouple from a shut down reactor, was working at a glove box containing a mixture of reactor blanket gas and purging argon at a pressure just above atmospheric when a glove was snagged and torn releasing some of the gas mixture. In vivo measurements were carried out on the technician and his assistant who was standing about 2m away at the time of the incident. These both showed ^{133}Xe but the intake by the assistant was only 1% of that of the technician, confirming that only a small cloud of gas had been involved. In vivo measurements were made on the technician over the period 4-75 hrs after the exposure showed a retention pattern similar to that obtained by Venner and Devell (3) after controlled inhalations; for the period 4-10 hrs the fall was more rapid by about a factor of 2. By extrapolation and interpolation of the retention curve we estimated a body content of about 9 mCi of ^{133}Xe at 1 hr and 300 μCi at 10 hours after exposure. The Venner and Devell plot (3) shows that for controlled inhalations the retained amounts of the inhaled activity at those times are 4% and 0.5% respectively. Applying these factors to our estimated body content we obtained estimates of 200 mCi and 60 mCi respectively for the initial inhaled amount. In view of the more rapid fall from 4 hours to 10 hours in our case we chose the higher figure as being a more correct estimate (perhaps even a little on the low side). If the activity was breathed in during some unknown but short period of time, t hours (it is not necessary to know this time), whilst the subject was breathing at the standard man rate of 1 m^3 per hour, the activity concentration in the cloud would have been $200/t \text{ mCi/m}^3$. On the assumption that the radius

of the small cloud of gas was about equal to the range of the beta particles (80 cm) conventional calculation showed the beta dose rate to a plane in the centre of the cloud to be 20/t rem/hr.

The gamma dose rate from a cloud of this size would be several orders of magnitude lower and was ignored. When we eliminate our unknown exposure time (t) we obtain a skin dose of 20 rems. We were therefore able to say that, despite a film badge recorded dose of only 0.5 rem gamma and 0.4 rem beta, the technician probably received a dose to the skin of his face of more than the 13 week permitted dose (15 rem) but less than 1 year's permitted dose (30 rem) and appropriate administrative action was taken. The most significant internal organ dose from this inhaled quantity was calculated from the information given by Whitton (6) as 1.3 rem to the tracheal mucosa, which is much less significant than the skin dose.

Case 2

Several men became internally contaminated with mixed fission products during removal of a fuel element two days after reactor shut-down.

The most highly contaminated man, subject G, had a body content of 12.8 μ Ci of ^{133}Xe 21 $\frac{1}{2}$ hours after the release. Measurements made during the following three days indicated that ^{133}Xe was being removed from the body with an effective half-life of 8.7 hours (biological half-life 9.3 hours): this is in agreement with Venner and Devell's retention curve at this time after inhalation (3).

By extrapolation of the retention curve we estimated a body content at 10 hours of 32 μ Ci ^{133}Xe . According to Venner and Devell's data (3), retention of ^{133}Xe at 10 hours is 0.5% and we therefore estimated the initial body burden as 6.4 mCi. It is probable that most of the intake occurred in a short period of time since the measured general air levels were high (>3 nCi/ml) for about an hour and then dropped to 0.6 nCi/ml and it is also probable that a cloud of much higher concentration existed close to the source for a shorter time. As before we assumed that subject G's initial body burden was acquired in a short period while breathing at 1 m³/hr, then calculation of the β -dose rate at the centre of a cloud of gas, as above, showed that subject G may have received a skin dose of 0.6 rem. This is less than the 13-week permitted dose, but greater than the dose to any internal organ. The most significant internal dose was 40 mrem to the tracheal mucosa and the corresponding lung dose was 8 mrem (6).

Case 3

^{133}Xe was also identified in subject M, 2 $\frac{1}{2}$ hours after a release of mixed fission products which occurred during removal of a rig from an operating reactor. The total body content of ^{133}Xe at this time was 0.5 μ Ci. Several measurements made during the next few hours indicated that xenon was being removed from the body with a biological half-life of 2 hours. However, a further measurement made three days later suggested that there was some long-term retention greater than that predicted by the Venner and Devell retention curve. The faster clearance may correspond to removal of xenon from the water-containing tissues and the longer-term clearance may represent elimination from the less well-perfused fatty regions of the body.

The initial intake was estimated by extrapolating the measured value at 2 $\frac{1}{2}$ hours to 1 hour after intake, with the observed half-life of 2 hours, and

then applying Venner and Devell's figure of 4% retention at one hour. This suggests that subject M's initial intake was 20 μ Ci. The β -dose to the skin of the face was estimated to be 2 mrem and internal doses to body organs were negligible.

INTERFERENCE FROM INTERNALLY INCORPORATED ^{133}Xe WITH HEALTH PHYSICS MEASUREMENTS

Internally incorporated ^{133}Xe gives rise to relatively small internal dose commitments as indicated in Case 1 above where the highest internal organ dose from an initial intake of 200 mCi was calculated as 1.3 rem. However incorporated ^{133}Xe in much smaller amounts can lead to misleading results being obtained from health physics measurements as is illustrated by the following two occurrences.

i. The technician referred to in Case 1 above monitored his body immediately after the incident and believed himself to be highly contaminated (about 200 x dwl). Several unsuccessful attempts at decontamination by showering were made before a health physicist was consulted who suggested that internally incorporated ^{133}Xe was the most likely cause; gamma spectrometry of a blood sample confirmed this. Later concurrent in-vivo counting and monitoring with a contamination probe showed that an internal content of 300 μ Ci gave rise to a counting rate at the surface of the body (using a thin walled, 30 mg/cm², Geiger Muller tube of dimensions 14 cm long and 1.5 cm diameter) of 10 cps equivalent to approximately 2 dwl of skin contamination.

ii. A laboratory worker who normally worked in a laboratory handling ^{239}Pu went into a reactor operating area to view an experiment on his way to keep an appointment for a ^{239}Pu -in-lung measurement. This measurement was made

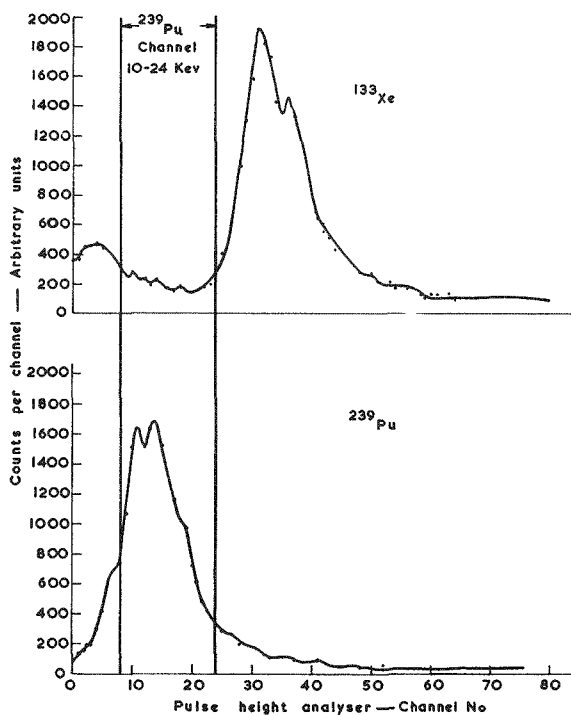


FIG. 2. PROPORTIONAL COUNTER X-RAY SPECTRA

using a gas filled proportional counter of the type described by Taylor (7). The initial result, which was assessed by the total counting rate in the 17 KeV ^{239}Pu channel, caused some concern as it was equivalent to more than 100 times the maximum permissible lung burden. Examination of the spectrum from the counter however showed peaks at about 30 KeV and 5 KeV and ^{133}Xe was suspected.

A further count using a scintillation detector indicated a body content of about 0.6 μCi of ^{133}Xe and a repeat measurement with the proportional counter a few days later showed no activity in the plutonium channel. A plot of the spectrum from ^{133}Xe in the body as given by the proportional counter together with a spectrum of ^{239}Pu for comparison is shown in Figure 2.

CONCLUSIONS

In-vivo measurements made on men exposed to air contaminated with xenon-133 in nuclear reactor environments show retention patterns similar to those reported (2). Elimination of xenon-133 is a complicated function of time, indicating that many body compartments are involved in the uptake and retention. Uptake, retention and also distribution within the body may vary depending upon the partial pressure of the inhaled xenon. Profile scanning measurements suggest that the distribution within the body is different for the two modes of uptake discussed. The reasons for this are not readily apparent, but some of the differences between the two profile curves may be due to different scanning techniques.

The critical dose from exposure to ^{133}Xe is that to the skin but, as we have shown, the results of in vivo measurements of the body content and elimination rates may be used to calculate this dose.

^{133}Xe incorporated in the body in amounts which give rise to trivial doses of radiation can interfere with health physics measurements and health physicists for reactor areas should be aware of the possibilities.

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In-vivo monitoring of nuclear fuel workers

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Introduction

As well as being a major source of uranium, Canada possesses a sizable uranium processing industry. Several plants located in Southern Ontario fabricate fuel rods for use in the Canadian nuclear power industry and also for export.

Working conditions in these plants are specified by the Atomic Energy Control Board, and monitoring of workers by means of criticality badges and urinalysis is performed by the Radiation Protection Bureau and the Ontario Department of Health.

Urinalysis results frequently show evidence of uranium ingestion, but the relation of this to total body burden is uncertain and a direct, in-vivo method of body burden measurement is required.

Cofield (1) made such measurements using a standard whole body counter in a steel room, and Quastel et al (2) of this Bureau made similar measurements, together with extensive urinalysis and other biological measurements, on 15 uranium workers. This study confirmed that body burden could not be accurately estimated from daily urinary excretion.

Unfortunately, the use of a conventional whole body counter means that the subjects have to travel to Ottawa, a distance of 250 miles from the major uranium processing plants, and while this can be arranged for small sample populations it is obviously impractical for the plants' total exposed work force. It was therefore necessary to move the equipment to the plants, and the present study was undertaken to see whether a portable system would have sufficient sensitivity to be useful.

Theory

The usable radiations from U-Nat. are as follows:

U-238	-	none	U-235	-	185 keV (54%)
Th-234	-	63 keV (3.5%)	Th-231	-	84 keV (10%)
		93 keV (4%)			

Since the subjects to be monitored had long exposures to uranium it was considered reasonable to assume secular equilibrium of the ingested material, which being of refined reactor grade had a very low content of radium or radium daughters. It was therefore decided to use the low energy radiations

from the thorium daughter, for which a 20 cm diameter x 3 mm thick NaI crystal, already available, would have a high efficiency. The response of this detector to a U-Nat. phantom is shown in fig. 1 as a broad peak extending from around 50 to 90 keV. The detector has negligible sensitivity to the 185 keV gammas from U-235.

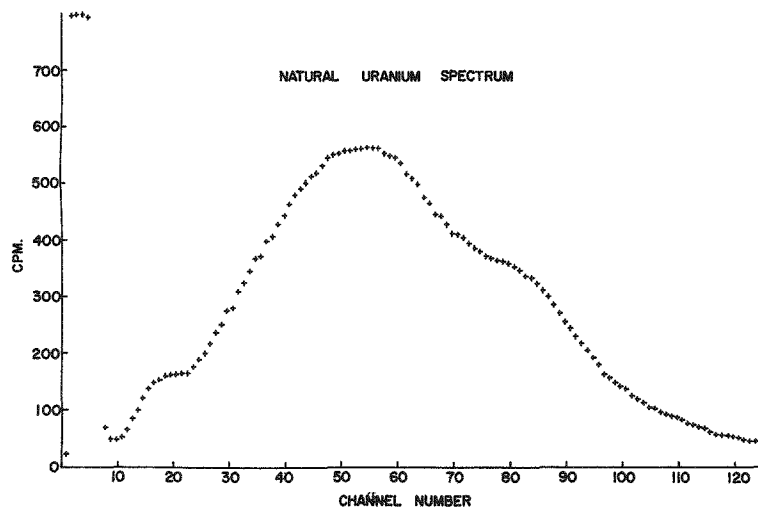


Fig. 1 Response of 20 cm x 3 mm NaI crystal to U-Nat. phantom

The phantom used for calibration purposes was a Remcal (Alderson Research Laboratories Inc.) phantom, with 9.2 g of uranium oxide dust in the lung cavities. The dust was sprayed onto adhesive coated paper tissues which were then made to adhere to the inside of four polythene bags. One bag was introduced into each of the four lung cavities and inflated with air. The basal lobes contained about four times as much uranium as the apical lobes.

The remainder of the phantom was filled with water and the net count rated observed was 3.86 cpm/mg U-Nat., which with a background of 3000 cpm gives an S^2/B ratio of 0.0049.

Using the formula of Altshuler and Pasternak (3), the minimum significant measured activity is 12.4 mg and the minimum detectable true activity is 24.8 mg, these figures being for a single 10 min. count. The crystal is also sensitive to scattered radiation from Cs-137 and K-40 to the extent of 8 cpm/nCi Cs-137 and 0.5 cpm/g K. For a normal 70 kg adult with a body burden of 5 nCi Cs-137 and containing 180 g K this would amount to a count rate of 130 cpm. Since the thin crystal is not sensitive to the primary radiation from either Cs-137 or K-40 it is not possible to measure each subject's content of these isotopes directly. The count rate from them has instead been considered part of the subject background, which is discussed further below.

Description of apparatus

The detector is a 3 mm thick by 20 cm diameter NaI (Tl activated) crystal coupled to 3 low-noise PM tubes. Each PM tube has its own H.V. supply, but the signals are collected by a common cable and fed into a single preamplifier and thence to the amplifier which is an integral part of the

Technical Measurement Corporation pulse height analyser. The date is recorded by either a TMC parallel printer or a Tally paper tape punch.

The settings of the H.V. Supplies are adjusted by placing a small uranium source on the crystal axis at a distance of about 40 cms. and switching on one H.V. supply at a time, and adjusting them so that the three spectra overlap as precisely as possible.

The detector is housed in a stainless steel cylinder which is lined with 1/8" lead and supported on a counterbalanced stand. See fig. 2.



Fig. 2 Detector, counterbalanced stand and lead tent with near-side panels removed.

The shielding is based on a design by Eisenbud et al (4) and consists of 1/8" lead sheet fastened to plywood and placed over a tent shaped framework. A sheet of 1/8" lead is placed under the mattress on which the subject lies.

The effect of this shielding is to reduce the subject background in the 30-100 keV region from 9,500 cpm to 3,000 cpm.

This equipment is portable, the total weight being about 600 lbs. Two men can load it into a station wagon in about 30 mins. and on arrival at the counting location, the system can be unloaded and assembled in about an hour. A room about 10 ft square is sufficient to house the equipment and the counting bed, located away from any active area. So far no unduly high background rates have been encountered, despite the fact that the plants visited process large quantities of radioactive material.

Subject Background

Preliminary work in the laboratory indicated that the background count

on a subject inside the tent varied considerably, depending on his size and shape. This variation was much more than would be expected just from differences in Cs-137 and K-40 content. Therefore forty Radiation Protection Bureau staff members were studied and their backgrounds were measured together with their height, weight and "chest thickness", i.e. the front to back measurement of chest thickness made at inspiration. The latter measurement was chosen as the simplest one to give some index of the bulk of tissue under the detector.

A series of empirically chosen expressions combining the three anthropomorphic parameters were analysed using a least squares method, for their correlation with the observed count rate. The expressions were of the type $X = (W/H) \log C$ or $X = \exp(W/H)$ etc. and the correlation coefficients were all quite similar at around 0.84. The highest coefficient, 0.851, was obtained with the expression $X = (W/H)\sqrt{C}$ which yielded an equation for Y, the subject background; $Y = K(3517.4 + 259.46X)$ where K = correction factor to allow for differences in ambient background between our laboratory and other counting locations. A plot of Y vs X is shown in fig. 3, which includes the 95% confidence limits for a single estimate of Y given a value of X.

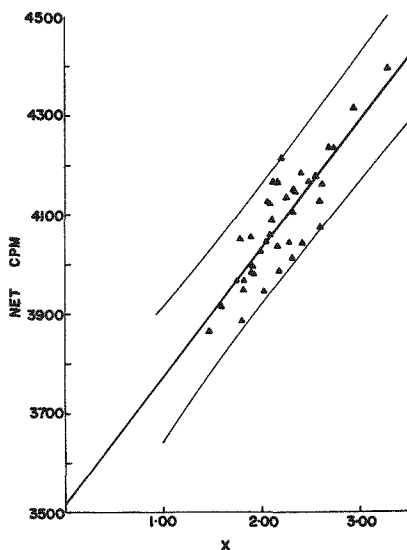


Fig. 3 Plot of observed subject background count-rate vs X where $X = \text{weight/height} \times \sqrt{\text{chest thickness}}$. (The 95% confidence limits are shown)

In terms of cpm these confidence limits represent ± 120 cpm on a typical subject background of 3000 cpm (K being significantly less than one at the plant where the bulk of the subjects were counted), whereas counting error for 10 min. count = 17.3 cpm. This error is more or less equal to the count rate expected from 30 mg U-Nat., the maximum permissible body burden. This means that only burdens in excess of the maximum permissible can be confidently detected. In an attempt to improve this situation, a more detailed multiple regression test was run on the data. This analysis was carried to the point where an equation containing six terms was derived, but the correlation coefficient associated with this equation was 0.8631, which was not significantly greater than with the much simpler equation originally chosen.

Other corrections

In dealing with radiations at 100 keV or below, tissue absorption must be taken into account. On the assumption that the uranium is located in the lungs or pulmonary lymph nodes, it is necessary to make some estimate of the thickness of tissue overlying these organs. Deane (5) has measured this thickness ultrasonically and correlated it with the weight/height ratio of each subject. His formula is as follows:

$$T \text{ (thickness (mm))} = 0.071 + 0.512 \frac{W \text{ (weight (kg))}}{H \text{ (height (m))}}$$

Ramsden et al (6) did a similar study, and obtained a different formula, which included a measurement of the chest circumference (C)

$$T \text{ (cm)} = 15.3 \frac{W \text{ (kg)}}{H \text{ (cm)}} - 0.01 C \text{ (cm)} - 3.55$$

We were not able to obtain an ultrasonic device and make actual measurements, and so T was calculated both ways and the mean taken, for each subject. As there were differences between the thicknesses obtained with the two formulae, in some cases as much as 25%, a mean value was taken. This was used to read off the appropriate absorption correction from fig. 4, the error associated with this procedure being estimated as $\pm 4\%$, due to the relatively small slope of fig. 4.

Newton et al (7) in their work on the measurement of plutonium in the lungs, took into consideration the self-absorption of the lung tissue itself, but for the present work, in view of the absorption curve in fig. 4 this has been ignored.

Collection of data

In the fall of 1971 a brief visit was made to a uranium processing plant in Southern Ontario. This was the first trip with the portable equipment and the main object was to see whether it could be transported and set up easily, and would be stable in operation. The time from arrival at the plant to counting the calibration standard was about $1\frac{1}{2}$ hours, and frequent counting of the calibration standard indicated that the response of the system was very stable.

The plant management made a small room 10' x 10' available to us in the administration area of the building and the subjects arrived after a shower and change of clothing. The subjects were 3 workers who had been studied five years earlier by this laboratory (2).

The subjects were counted supine with the 8" detector in contact with the chest surface. The counting time was 10 min.

The results from this preliminary study, while showing some counts above the background of a normal subject, were calculated without the benefit of the subject background equation subsequently developed, and were therefore considered merely an indication that the system could work and that a further field trip to study a larger sample of workers would be justified.

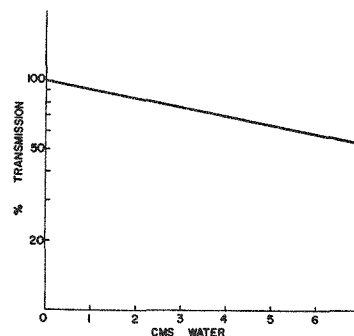


Fig. 4 Relative transmission of U-Nat. radiations in 60-90 keV region in water.

Such a trip was undertaken some time later, and involved a larger nuclear fuel processing plant. This plant contained a large stock-pile of uranium and there were some misgivings as to what the background count would be. It turned out however to be not significantly different from that at the other plant.

A total of twenty-four subjects were counted, with results as shown in table 1, calculated on the assumption that only U-Nat., in equilibrium, was present.

Table 1
Results of Measurements in Nuclear Fuel Workers

Subject #	X $(\frac{W}{H} \sqrt{C})$	Estimated background c.p.m. ¹	Observed count rate c.p.m. ²	Net count rate c.p.m.	Absorption correction ^{*3}	mg U-Nat.
1	2.47	3056 ± 125	3298 ± 18	242 ± 126	0.78	62 ± 25
2	1.79	2927 ± 121	3401 ± 18	474 ± 122	0.87	109 ± 24
3	2.15	2995 ± 122	3005 ± 17	11 ± 123	0.82	3 ± 24
4	1.93	2953 ± 121	2975 ± 17	22 ± 122	0.85	5 ± 24
5	1.87	2941 ± 120	2981 ± 17	40 ± 121	0.87	9 ± 24
6	2.36	3035 ± 120	3047 ± 17	12 ± 121	0.81	3 ± 24
7	2.42	3046 ± 123	3011 ± 17	-35 ± 124	0.80	-
8	2.09	2985 ± 123	3244 ± 18	259 ± 124	0.83	62 ± 25
9	1.76	2920 ± 120	3090 ± 17	170 ± 121	0.88	38 ± 24
10	1.71	2912 ± 122	3018 ± 17	106 ± 123	0.87	24 ± 24
11	2.17	2999 ± 123	3217 ± 18	218 ± 124	0.82	53 ± 25
12	1.75	2919 ± 122	3204 ± 18	285 ± 123	0.88	64 ± 24
13	1.57	3075 ± 121	3041 ± 17	-34 ± 122	0.77	-
14	2.20	3005 ± 123	3291 ± 18	286 ± 124	0.86	66 ± 25
15	1.74	2917 ± 120	2984 ± 17	67 ± 121	0.89	15 ± 24
16	1.87	2941 ± 120	2968 ± 17	29 ± 121	0.87	6 ± 24
17	2.68	3097 ± 123	3326 ± 18	229 ± 124	0.77	59 ± 25
18	2.36	3035 ± 120	3349 ± 18	314 ± 121	0.80	78 ± 24
19	2.97	3151 ± 128	3282 ± 18	131 ± 129	0.75	35 ± 26
20	1.89	2945 ± 120	3211 ± 18	266 ± 121	0.86	62 ± 24
21	2.36	3035 ± 120	3374 ± 18	239 ± 121	0.80	84 ± 24
22	2.07	2980 ± 122	3047 ± 17	67 ± 123	0.83	16 ± 24
23	1.90	2948 ± 120	4908 ± 22	1960 ± 121	0.85	459 ± 24
24	2.07	2983 ± 123	2947 ± 17	-36 ± 124	0.83	-

¹ Errors estimated from fig. 3

² Counting error only

³ Correction factor read from fig. 4 using tissue thickness calculated as described in text.

As will be noted some had very high values, but subsequent discussion with the plant health physicist revealed that these had worked in the UF₆ plant, and during the processing of this material, the thorium does not enter the gaseous phase and is left behind in the "ash". It was therefore assumed that the high readings were due, at least in part, to the inhalation of thorium 234, during the handling of this "ash", and measurements taken on some of the UF₆ workers six months later were down to much lower levels, which tended to confirm this assumption.

Discussion

The results from these field trips indicate that a significant proportion of the workers examined were contaminated with uranium or its daughters. The errors, mainly due to uncertainties in the background count estimate for each subject, were such that only lung burdens greater than 30 mg could be detected with any confidence, and in some cases the material detected could have been mainly 24 day Th-234 rather than U-Nat.

The problem of identification is in fact two-fold, first to identify the degree of enrichment of the uranium, and second, to differentiate thorium from uranium. A recently acquired dual crystal (Phoswich*) detector has been put to use in some preliminary experiments to look into these problems.

The detector is a 12.5 cm diameter, 1 mm thick NaI (Tl activated) crystal backed by a 12.5 cm diameter, 5 cm thick CsI (Tl activated) crystal, and a pulse shape discrimination circuit (Harshaw NC-25) is used to allow only those photons that are completely absorbed in the thin crystal to be recorded. The sensitivity of this detector is 1.25 cpm/mg U-Nat. (50 keV - 110 keV) for an average subject background of 600 cpm. If these figures are corrected to compensate for the smaller area of the 12.5 cm detector compared to the 20 cm detector, an S^2/B ratio of .0067 is obtained. This is better than the figure of .0049 for the 20 cm detector, but could probably be improved further by the use of a thicker front (NaI) crystal, of say, 3 mm.

The natural uranium source gives a spectrum with well-defined peaks at about 60 keV and 90 keV, and the change in relative heights of the peaks with changing enrichment was investigated, on the grounds that the Th-231 from U-235, with its gammas at around 84 keV would affect the 90 keV peak more than that at 60 keV. The peak ratio being defined as $\frac{\text{ch. 103-162}}{\text{ch. 163-216}}$ at a gain of 0.48 keV/ch.

The plot of peak ratio vs enrichment is shown in fig. 5, and there is a marked drop in the 60 keV/90 keV ratio with increasing enrichment up to about 20%. Beyond that the curve levels out but since the material used in the plants under investigation rarely exceeds 20% enrichment, this should not detract from the utility of the curve in estimating the enrichment of the uranium present.

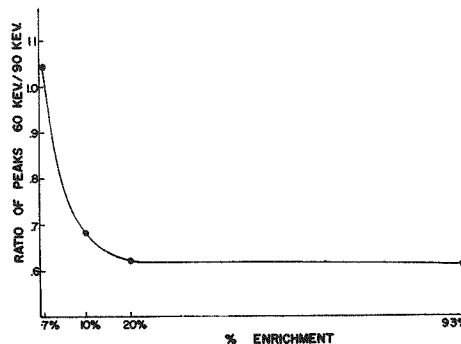


Fig. 5 Ratio of 60 keV/90 keV peaks vs enrichment

The above measurements were made with sources of high activity and so the net count rates were high. With actual subjects the net count rates would normally be much lower, and prone to the errors involved in estimating subject body background discussed already.

(* Harshaw Chemical Co. Ltd.)

Future work with the dual crystal will include an experiment to determine the relationship between the 60 keV/90 keV peak ratio and tissue thickness, and another to explore any correlation between the body background of a normal subject in the 60 keV-90 keV region and that in the 200 keV and above region, using the 5 cm thick CsI crystal as a detector and counting all the events occurring within it. The range from 200 keV upwards would include contributions from Cs-137 and K-40 but not from the 185 keV line of U-235.

It would also be very useful if the 185 keV line of U-235 could be detected by the CsI crystal in order to distinguish between uranium and separated thorium but early results indicate that the background counts in that region are too high for sufficient sensitivity.

Conclusions

The study shows that given certain conditions, the 20 cm x 3 mm single crystal with portable shielding can detect lung burdens in the region of 30 mg U-Nat. The conditions are that only U-Nat. is present, in equilibrium with its thorium daughters, and that no separated thorium is present.

In the major uranium processing plant studied these conditions hold for a substantial portion of the personnel, and those for which they do not hold can be identified. The system can, therefore, give useful information, and is suitable for routine monitoring within the stated limits.

A dual crystal (Phoswich) system, however, offers considerable promise for refining the method to determine enrichment and to improve the subject background estimates, and hence the sensitivity. The problem of separated Th-234 will require either a much more substantial counter of the shadow-shield type, or a method of repeated counting to detect the decay of the thorium.

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APPLICATION OF PHOSWICH DETECTORS FOR LUNG COUNTING PLUTONIUM-238

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Abstract

Mound Laboratory's Whole Body Counter was designed and calibrated for the detection of ^{238}Pu in the lungs. This paper summarizes the basic counting program since December 1969. The primary discussion is centered around the phoswich detection system. A unique triple coincidence pulse shape discrimination technique was used to reduce the background more than one order of magnitude as compared to a standard NaI(Tl) detector. Detection limits are given as a function of the subject's tissue thickness between the lungs and detectors. For a typical subject with an effective tissue thickness of 2.3 cm over the lungs, the system has a detection limit of 4 nCi.

Introduction

Numerous laboratories have employees who work daily with plutonium. At Mound Laboratory about 450 employees are routinely monitored for ^{238}Pu as part of the overall radiological health protection program. An important part of the program is the routine and special lung counting which gives a direct assessment of the most common mode of uptake - inhalation.

This paper briefly summarizes the development of lung counting capabilities since 1969 with the primary emphasis on the phoswich detector system. The sensitivity of this system lies in the pulse shape discrimination instrumentation which is used to lower the background by more than one order of magnitude compared to a standard NaI(Tl) detector. The major problems of implementing the detector/pulse shape discrimination system are also discussed. Also included is a discussion of detection limits as a function of the chestwall tissue thickness where the chestwall is the primary absorber of low energy photons emanating from the lung.

Historical Development

Detectors The radiation safety program at Mound Laboratory was upgraded in 1969 with the completion of the Body Counting Facility. The design of the facility was reasonably standard with a Packard Instrument Company¹ steel room and a semi-aged air supply from the crawl space in the adjacent administration building. In 1969, two standard NaI(Tl) detectors, 10.2 cm diam by 0.4 cm thick, were coupled through amplifiers to a multichannel analyzer. Room background in the 6-27 keV band was about 0.45 count/min/cm². A typical count on an unexposed individual was 0.542 count/min/cm² with a minimum detectable activity of about 11 nCi of ^{238}Pu (approximately 3/4 m.p.l.) using a 4000-sec count.

*Mound Laboratory is operated by Monsanto Research Corporation for the U.S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

A diagram of the phoswich detectors purchased from Harshaw Chemical Company² in July 1969 is shown in Fig. 1. By May 1970 the two phoswich detectors were in use with a significant improvement in sensitivity over the standard NaI(Tl) detectors. Room background was about 0.0185 count/min/cm² in the 14-25 keV band; however, the pulse shape discrimination system resulted in a loss of about 6% of the detected '17 keV' photons. Even so, a typical background of an exposed individual was only 0.0346 count/min/cm² (14-25 keV) which allowed a minimum detectable activity³ (3 σ above background) of 5.1 nCi of ²³⁸Pu.

In October 1972, two additional phoswich detectors, 12.7 cm diam, were purchased; the only difference was the larger diameter of the crystals. With the same pulse shape discrimination system, the room background decreased to 0.0178 count/min/cm² and the minimum detectable activity was slightly improved at 4.0 nCi of ²³⁸Pu.

Calibration Procedures The most prominent photons for counting ²³⁸Pu in-vivo are the ²³⁴U L x-rays with an average energy of 17 keV. The total counts in the complete 17 keV region, i.e., from 5-28 keV, were originally integrated for analysis. However, it was soon discovered that 14-25 keV was the optimum area of integration. This was determined by maximizing S²/B, where S is the net count rate and B is the background. Originally, a simple two point calibration curve was used for lung deposition assessments as shown in Fig. 2.

The two points used for the exponential fit were obtained by counting the Remab phantom⁴ full and then one-half full of liquid. This gave two chestwall tissue thicknesses, i.e., two different thickness absorbers, at which the counting rate per unit activity was measured. This was knowingly in slight error for thin chestwalls because of the lung-to-detector distance with the phantom one-half

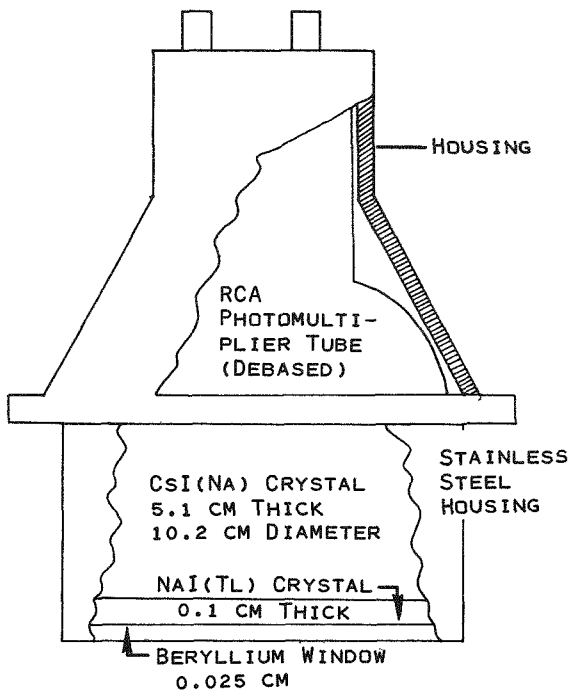


Fig. 1 Phoswich detector diagram.

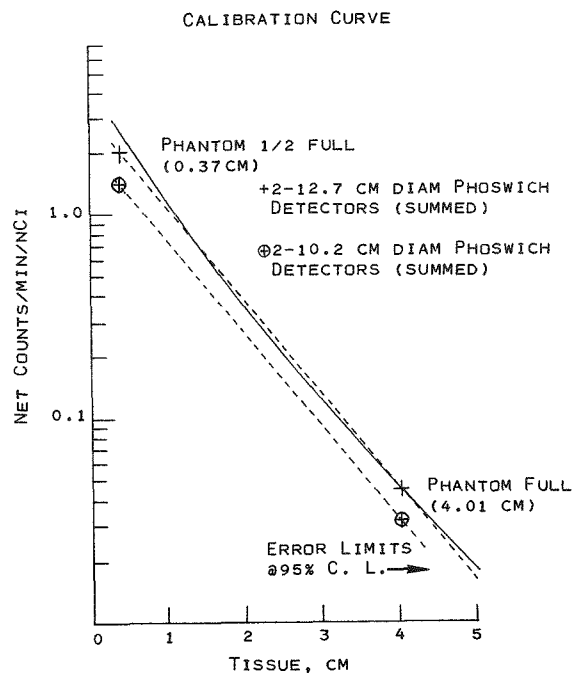


Fig. 2 Calibration curve.

full of liquid. This error was later corrected by developing the proper shaped curve using ^{238}Pu doped lungs from the phantom and beefsteak absorbers.⁵ Once the correctly formed calibration curve existed, it was overlaid and correlated to the one suitable phantom measurement where the phantom was full of liquid and had an equivalent chestwall thickness of 4.04 cm (See Fig. 2).

Counting Procedures As part of the overall radiological health program, all plutonium workers are routinely counted on a quarterly basis. Any employe is promptly scheduled for a special lung count upon discovering 200 dis/min or more on a nasal swab. More than 50% of those needing special counts have had at least a small amount of surface contamination on various parts of their bodies. One major problem encountered in special counting is determining whether the plutonium was detected from within the lungs or from contamination of the chest surface. As little as 50 dis/min on the skin's surface can cause a false reporting of a Type B Incident. Procedures used at Mound Laboratory to eliminate "false" interpretation of data are outlined below:

1. Carrying out extremely thorough surface decontamination, including "washing" the subjects chest with ethylenediaminetetrachloroacetic acid.
2. Placing lead loaded gloves on the subjects hands and arms.
3. Counting the subject with a lead shot filled curtain around each detector for shielding.
4. Requiring a confirming lung count taken from the subjects back.

Instrumentation

The two phoswich detectors currently used at Mound Laboratory are summed together into the pulse shape discrimination system as shown in Fig. 3. Because of the two dissimilar crystals in a phoswich detector, each output pulse will be characteristically shaped by the crystal in which the absorbed photon lost its energy. Since the pulse shape discrimination system is aligned to accept only those low energy (17 keV) [NaI(Tl)] pulses, noise and most high energy background pulses [CsI(Na)] are rejected. About a 5-8% loss of detector efficiency has been experienced using the pulse shape discrimination system.

Discussion

The operation of the pulse shape discrimination system is rather unique. Initially the system was set up to discriminate by using only rise time and crossover times as shown outside the dotted lines in Fig. 3. Because the room background of 0.053 count/min/cm² (14-25 keV) was not as low as expected, the system was thoroughly reinvestigated. Extraneous background counts were caused by cosmic radiation. Typical amplifier output pulses appeared similar to those shown by solid lines in Fig. 4.

Careful examination of the cosmic ray overload pulses resulted in the discovery of a preamplifier bleed-off pulse following the initial saturated pulse by 40-80 μsec . This bleed-off pulse was

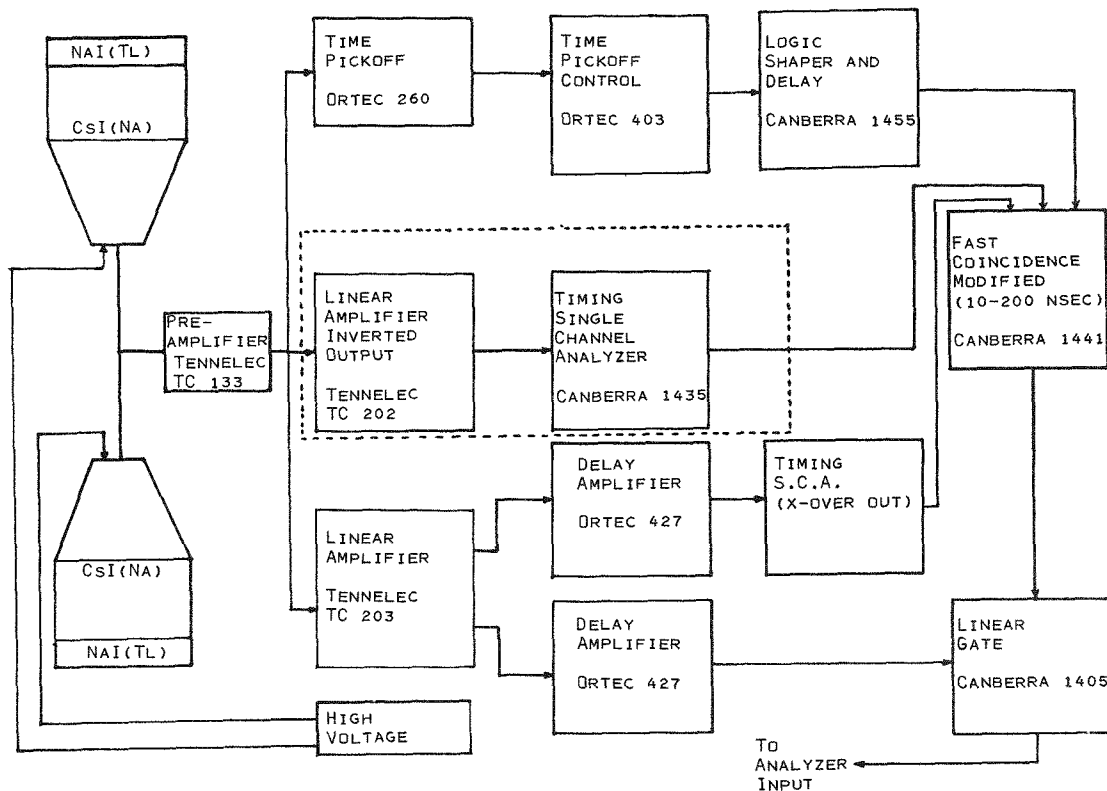


Fig. 3 Pulse shape discrimination system.

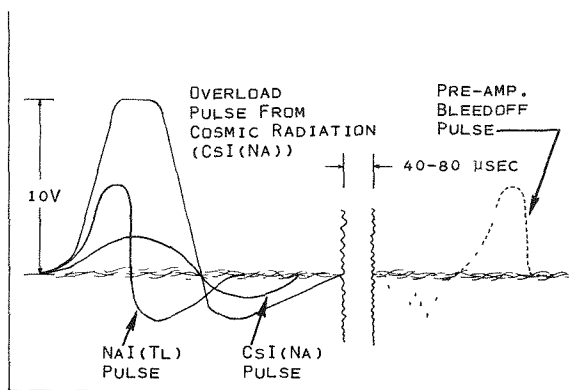


Fig. 4 Amplifier output pulses.

similar to the '17 keV' pulses from ^{238}Pu . A portion of these bleed-off pulses would pass the discrimination system and cause extraneous background counts in the 17 keV region. Two solutions existed for this problem. The first and simplest was to use an integral discriminator to inhibit the system output for about 100 μsec after the detection of any saturating pulse. The dead time is insignificant at the count rate of interest, but can be determined. The second method (in current use) to eliminate the extraneous bleed-off pulses made use of the bleed-off pulse shape. By using an inverted bipolar pulse input to a timing single channel analyzer in the leading edge mode, only those bipolar pulses with a second lobe were accepted. (See Fig. 3).

In November 1969, efficiency measurements using the internal gate of the multichannel analyzer indicated an electronic problem in the system. The efficiency for the

system decreased when the distance between the source and the detector was increased. Numerous efficiency measurements were made using the analyzer gate and then compared to results obtained under the same counting conditions with an external linear gate. The efficiency of the system using either gate was approximately 90% at counting rates of 2000 counts/min. At counting rates less than 500 counts/min, the efficiency of the system dropped to less than 50% when the analyzer gate was used, but remained at about 90% with the external gate. The unusual behavior of the analyzer gate was examined further by letting the coincidence logic pulse trigger an oscilloscope simultaneously with the linear gate in the analyzer. Observation of both the oscilloscope and the analyzer's visual display revealed that many pulses which triggered the oscilloscope did not register on the analyzer's display. Such pulses were not being stored in the analyzer's memory. It was later confirmed by the manufacturer that indeed there was a design error in the analyzer's gate. Prior to resolving the problem, abnormally low background counting rates were observed.

Results

The comparison of a standard NaI(Tl) detector system versus a phoswich detector system of the same active area, shows an unquestionable improvement for 17 keV photons in ^{238}Pu lung counting. A direct comparison is shown in Table 1.

Table 1

Standard NaI(Tl) Detectors Compared with Phoswich Detectors

<u>Detector</u>	<u>Steel Room Background from 14-25 keV (counts/min/cm²)</u>	<u>Unexposed Person Count from 14-25 keV (counts/min/cm²)</u>	<u>Minimum Detectable Activity* (nCi)</u>
Std. NaI(Tl) 10.2 cm diameter 0.4 cm thick	0.245	0.276	11
Phoswich Detector 12.7 cm diameter 0.1 cm thick NaI(Tl) 12.7 cm diameter 5.1 cm thick CsI(Na)	0.0178	0.0274	4

* 3σ above background (2.31 cm chestwall thickness).

The minimum detectable activity³ for ^{238}Pu lung counting has been improved by about a factor of three by changing to the phoswich detector system. The minimum detectable activity as a function of chestwall tissue thickness and unexposed subject counting rate is shown in Fig. 5.

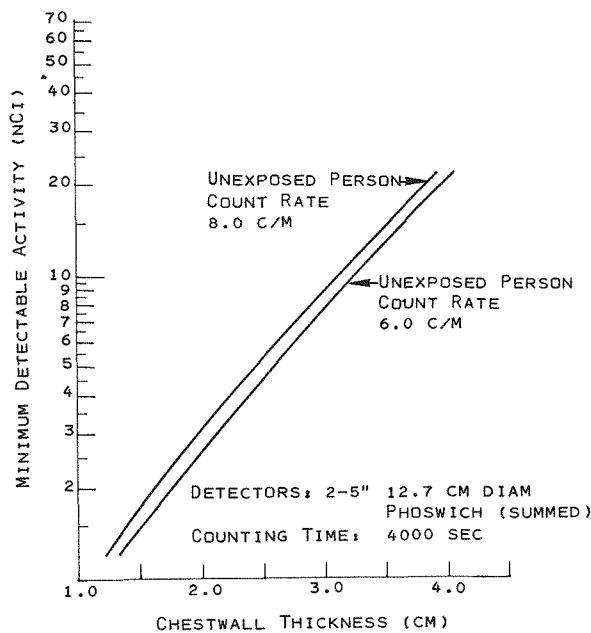


Fig. 5 Minimum detectable activity curve.

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CHESTWALL TISSUE MEASUREMENTS FOR LUNG COUNTING APPLICATIONS

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Abstract

Mound Laboratory's Whole Body Counter was designed and calibrated for the detection of ^{238}Pu in the lungs.

Quantitative measurements depend upon the detection of the 17 keV (average) x-ray associated with the decay of ^{238}Pu . Because the half-value layer for 17 keV x-rays in tissue is only 6-7 mm, the effective thickness of the tissue overlaying the lungs must be accurately determined for proper interpretation of the counting data.

The tissue thickness over the lungs is determined by ultrasonic measurements over the second, third, and fourth rib in the manner suggested by Ramsden, Peabody and Speight.

This paper presents a review of the instrumentation and technique used at Mound Laboratory to obtain the tissue thickness measurement based on our experience in making these measurements on more than 700 different persons.

Introduction

During the last several years, various ultrasonic instruments have been used for determining human chestwall thickness. The chestwall thickness is extremely important where low energy photons are counted for lung burden assessments.

Even though reproducibility of the chestwall measurement error itself can be maintained at $\pm 5\%$, it can propagate very significant errors in lung burden assessments. For the case of ^{238}Pu assessments where 17 keV (average) photons are counted, an error of 6-7 mm would cause the final lung count to be misinterpreted by as much as 100%. It is therefore necessary to maintain the best possible accuracy and precision in making chestwall tissue measurements for lung deposition assessments.

This paper discusses the major problems of making chestwall tissue measurements and the effect they have on making ^{238}Pu lung deposition assessments. The instrumentation and techniques used at Mound Laboratory during the last four years are reviewed. A discussion of different methods of deriving the chestwall thickness that have been investigated is also included.

Calibration

The effect of the chestwall as an absorber when lung counting for ^{238}Pu must be considered during calibration. Mound Laboratory

*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

calibrates for lung counting using a Remab Hybrid phantom that has known quantities of ^{238}Pu distributed uniformly throughout the phantom lungs.¹

For ^{238}Pu , the most prevalent photons available for counting are the 17 keV (average) uranium L x-rays. The three ^{234}U L x-rays (13.6, 17.2, and 20.4 keV) are not attenuated by tissue at the same rate and therefore give a transmission curve similar to the beef-steak curve shown in Fig. 1.

Two 12.7 cm diam phoswich detectors were used in a "normal" counting geometry to obtain the data in Fig. 1, and thus the significant geometry effects are included.

When the statistical errors that would normally be considered in calibration procedures are disregarded, the error in relative transmission caused by an error in the chestwall tissue thickness can be readily observed from the dotted lines in Fig. 1. An error of 6 mm results in a transmission error of about 100%. The chest-wall effect on minimum detectable activity (3σ greater than background) is shown in Fig. 2. A typical subject with no exposure and a counting rate of 7.5 counts/min for 4000 sec is used in the example.

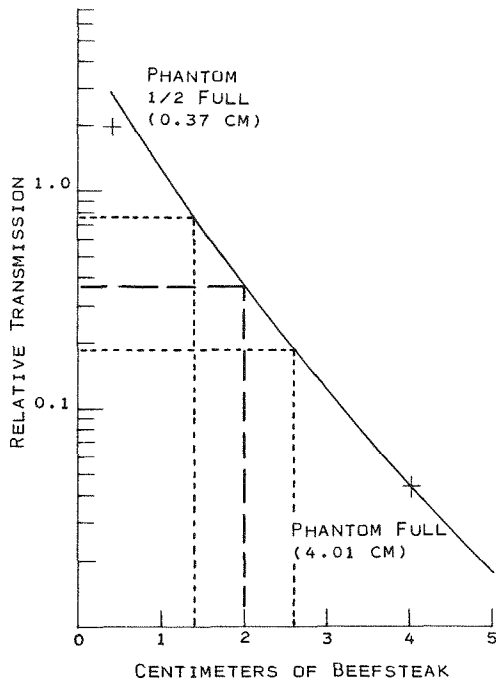


Fig. 1 17 keV (average) x-ray attenuation. [Absorber: beefsteak, ground and frozen. Source: phantom lungs (^{238}Pu spiked). Detectors: 2 each 5 in. NaI(Tl)-CsI(Na) Phoswich.]

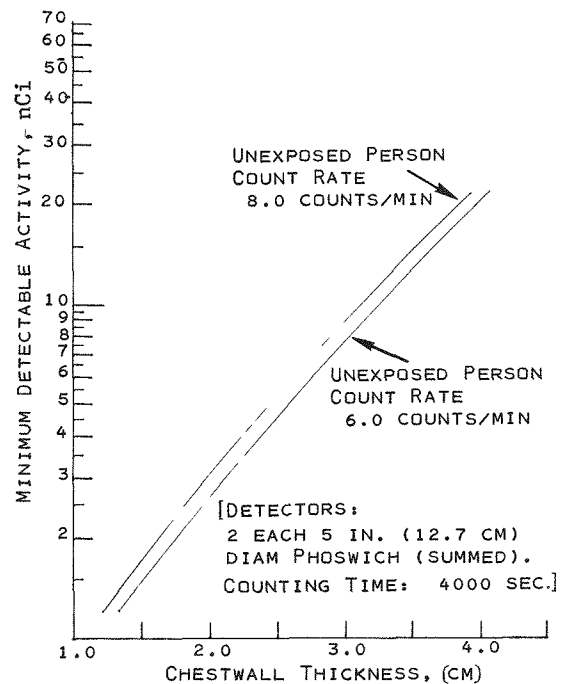


Fig. 2 Minimum detectable activity as a function of chestwall thickness

Instrumentation

In 1968 the only known investigation into making chestwall tissue measurements had been made by Ramsden et al.² Since lung counting of employees was to be routine, it was also desirable to determine the chestwall tissue thickness rapidly and accurately.

An ultrasonic sounding instrument was purchased from Hewlett Packard.³ Instruments of this type are used in the medical profession for brain and heart studies. The transducer (0.64 cm diam) is electrically pulsed and produces an acoustic frequency of 2.5 MHz which is transmitted through a coupling medium into the subject to be measured. The acoustic pulses are partially reflected at any interface where the acoustic impedance changes. The reflected portion of the pulse is then detected by the transducer and the time delay from transmission is measured. This time difference is displayed on the horizontal axis of the cathode ray tube which is calibrated in centimeters of tissue. The position of any acoustic interface is a linear function of the velocity of sound in that medium and is converted to a depth measurement from the crystal transducer. The intensity of the reflected pulse is a function of the depth and acoustic impedance mismatch and is displayed on the vertical axis of the cathode ray tube. A typical trace on the instrument is shown in Fig. 3.

This instrument has an adjustable distance marker that is adjusted via a 10-turn potentiometer, and once it is aligned with the echo of interest, it reads the distance from the transducer to the interface producing the echo. This mode of operation is commonly referred to as the "A-scan mode." A calibration block of plastic is supplied with the instrument.

Methods and Procedures

Ultrasonic chestwall measurements were made on cadavers using various approaches followed by sectioning of the chestwall and physical measurements made with calipers.⁴ Rather than measure directly to the lung interface between the ribs, it was advantageous to use the tissue to rib interface since the intensity of the echo was more sharply defined. However, to do this, additional rib thickness must be added to the tissue thickness overlying the ribs. After numerous rib thickness measurements on skeletons, this rib thickness was found to range from 0.3 to 0.5 centimeters⁴ depending on general bone structure of the skeleton.

An average thickness of the chestwall was found for the area between the detectors and the lungs. The nine measured points are located as shown in Fig. 4 and on the right side of the chest.

The photon attenuation is a function $e^{-\mu x}$ where μ is about 1.15 cm^{-1} for tissue and x is the absorber thickness in centimeters. Therefore, the average thickness is not a simple mean of several measured thicknesses, but instead, is an "exponential average." The true average thickness was found by measuring nine points to the rib, averaging the values of $e^{-1.15x}$, equating this average to $e^{-1.15x}$, solving for x , and then adding the rib thickness.

Asymmetry between the right and left halves of the chestwall thickness could not be defined within the precision of measurement.

A highly developed technique was necessary in order to align the transducer to achieve a well defined structure as shown in Fig. 3. The most obvious difficulties occur on subjects with appreciable amounts of fatty or muscle tissue in the chest area.

A second method of measuring the chestwall thickness between the ribs in the same chest area was also briefly investigated.

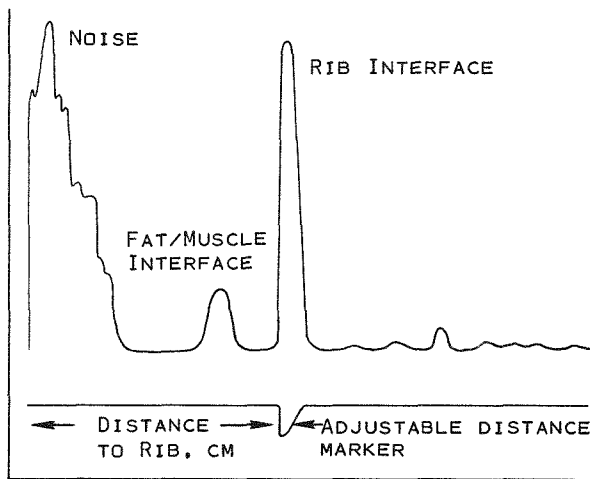


Fig. 3 Ultrasonic display of chest-wall measurement

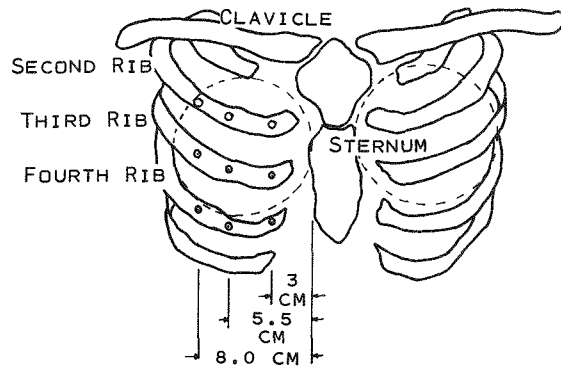


Fig. 4 Ultrasonic measurement points. (Approximate area covered by detectors shown in dotted lines.)

The same basic principles of measurement apply; however, the cathode ray tube display is slightly different.

Results and Discussion

After more than 1,000 chestwall determinations had been made, the possibility of discarding the procedure in lieu of a prediction method was investigated.

The average chestwall measurement for 741 different subjects was found to be 2.28 ± 0.748 cm (2σ). Therefore, in the simplest form, it is possible to use this value for all subjects but with an extremely large resultant error in plutonium lung assessments. The error could be in excess of 100% at the 95% confidence limit if this thickness is related back to Fig. 1.

Another prediction method used various physical body parameters, which can be quickly measured, to predict the chestwall thickness as investigated by Ramsden et al.³ and Dean.⁵ A stepwise multiple regression analysis was used to investigate different prediction equations for chestwall tissue thickness. The data used were limited to 644 different subjects for which all physical parameters were measured.

The stepwise multiple regression used is a statistical technique for analyzing a relationship between a dependent variable (chestwall thickness-T) and a set of independent variables (see Table I) in order of their importance. The criteria of importance is based upon a reduction of the total variation in the dependent variable. In each given step the independent variable most important in this reduction is entered in the regression. Unless the percentage of the total variation accounted for by an independent variable was greater than 1%, the variable was eliminated.

The results of these analyses for three major groups of independent variables are shown in Table I.

TABLE I Regression Analyses

1. Independent Variables: Weight (W), Height (H), Chest Circumference (CC), Waist Circumference (WC), Chest Thickness (CT) and Age (A)

<u>Step No.</u>	<u>Variable</u>	<u>Regression Equation</u>	<u>Total Variation Accounted for by Regression</u>	<u>Multiple Correlation Coefficient</u>	<u>Standard Error of Estimate (cm)</u>
1	W	$T = a+bW$	40.3%	0.634	0.292
2	H	$T = a+bW+cH$	46.1%	0.678	0.270
3	A	$T = a+bW+cH+dA$	51.7%	0.718	0.264

Example of Step 3 Regression Line: $T = 4.0185 + 0.0107 W - 0.0475 H - 0.0088A$
 Standard Error of Estimate (σ) = 0.264 cm

6001

2. Independent Variables: W/H, CC, WC, CT, A

1	W/H	$T = a+b(W/H)$	43.1%	0.657	0.285
2	A	$T = a+b(W/H)+cA$	48.2%	0.694	0.273

Example of Step 3 Regression Line: $T = 0.8388 + 0.6880 W/H - 0.0082A$
 Standard Error of Estimate (σ) = 0.273 cm

3. Independent Variables $(W/H)^{\frac{1}{2}}$, CC, WC, CT, A

1	$(W/H)^{\frac{1}{2}}$	$T = a+b(W/H)^{\frac{1}{2}}$	43.8%	0.661	0.284
2	A	$T = a+b(W/H)^{\frac{1}{2}}+cA$	48.3%	0.694	0.273

Example of Step 3 Regression Line: $T = 0.9690 + 2.2249 (W/H)^{\frac{1}{2}} - 0.0077A$
 Standard Error of Estimate (σ) = 0.273 cm

Of the two ultrasonic methods described, the first was the more objective and easier to use. The method of measuring between the ribs to the lung interface is more difficult to use because of the lesser intensity echo from that interface which therefore could introduce significant error in accuracy.

Because of errors involved in predicting the chestwall thickness in the upper chest area, the precision from ultrasonic measurement was investigated on 45 subjects using the first method described previously. Over a one year period, all plutonium operating personnel that were scheduled for lung counting and found to have had two or more chestwall measurements were considered. Only the data from those subjects whose weight and thickness (front to back) had not varied more than 4% were used. The results of applying these constraints left 45 suitable subjects and two standard deviations were found to be 0.112 cm.

One error that is generally not considered in discussion of chestwall thickness measurements is that resulting from the ratio of fat to muscle. Although a small error is introduced from the direct ultrasonic measurement, the propagated error in a plutonium lung burden assessment can be of the order of 10%.

Even though several methods of predicting the chestwall tissue thickness were investigated, the results indicate that the most precise technique of determining this value is by ultrasonic measurement. It is, however, possible to conserve time used for routine counting by making only four to six routine chestwall measurements weekly to maintain the technique needed in actual lung burden assessment cases.

Acknowledgements

The authors would like to thank Ernest Arnett, M.D., for his invaluable work with the cadaver measurements and E. L. Saenger, M.D., and Dr. J. Kerieakes for the use of the medical facilities at the University of Cincinnati Medical College as well as their advice on techniques. Grateful thanks are also given to Mr. Anthony Grandillo for performing the regression analyses of the data.

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In vivo MEASUREMENT OF URANIUM CONTAMINATION IN THE LUNG

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Abstract - The commonly used method for detection of internal contamination of uranium, as well as of all other radionuclides - is urinalysis. In cases of nontransportable natural uranium contamination by inhalation, experience has shown that urinalysis gives erratic results which do not enable accurate calculation of the inhaled amount. A method for direct *in vivo* measurement of natural uranium, utilizing the 186 keV line of uranium-235, has been developed utilizing a Whole Body Counter by placing the detector directly over the lung area (1, 2). This procedure requires however corrections due to the presence of caesium-137 and potassium-40 in the body and the parallel measurement of a "double" as a control. A newly developed method gives direct *in vivo* determination of uranium contamination by counting the soft L X - rays (10 - 20 keV) contribution of uranium-238 and uranium-234. Counting is done inside the low background Whole Body Chamber with two proportional detectors (30 x 15 cm) flushed with argon and methane. Quantitative calibration was done with sponges saturated with UO_2 and placed in a chest cavity of a male goat weighting 75 kg. Counts due to uranium are much higher than those contributed by the normally existing amounts of caesium-137 and potassium-40 thus enabling a detection threshold of at least $1/3$ of the maximum permissible lung burden. Work is in progress for improvement of apparatus and methodology.

With the increased availability and use of radioactivity, knowledge of the behavior in the human body of inhaled or ingested radioactive material has acquired a new significance.

Since most chemical elements which enter the body are eventually excreted, analysis of the urine became the most commonly used method for detection of radioactive internal contamination.

Any activity above zero of radioactivity in the urine is an indication of the presence of a contaminant, and rather elaborate methods of determination of the true amount of this radioactivity have been developed and are in practice in all bioassay laboratories in nuclear centers.

Accurate counting of radioactivity in urine is an easy task, at least for β and γ emitting radioisotopes. Calculating from the result the actual amount of radioactive material which exists in the body is another matter and is rather complicated and inaccurate. The rate of excretion of an element from the body depends on its physical state, chemical formula, particle size, the metabolic activity of the body, and other such factors. These become even more complicated when one deals with insoluble and α emitting elements.

Our experience in one case of inhalation of insoluble natural uranium dust strengthened the notion that it is difficult to rely solely on urinalysis for quantitative determination for the amount of uranium inhaled and retained in the human body (1, 2). As has been found in this case - the urine of the contaminated subject showed great variations in uranium concentration from day to day (Fig.1), ranging from a high of about 2000 μ /liter to a low of about 100 μ /liter.

After analysis and reconstructing the case and concluding that we are dealing with a case of inhalation of insoluble natural uranium, it became clear that we have to develop a direct *in vivo* determination of the uranium in the lungs. The

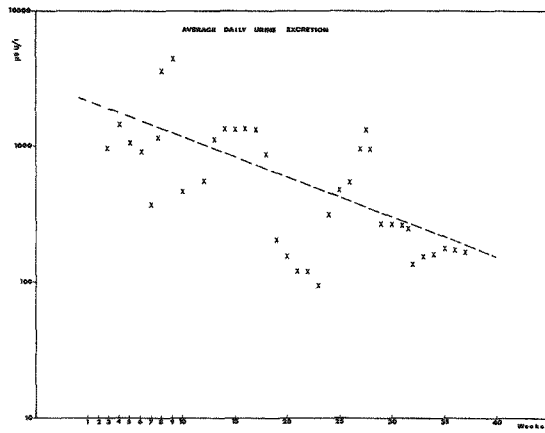


Fig. 1

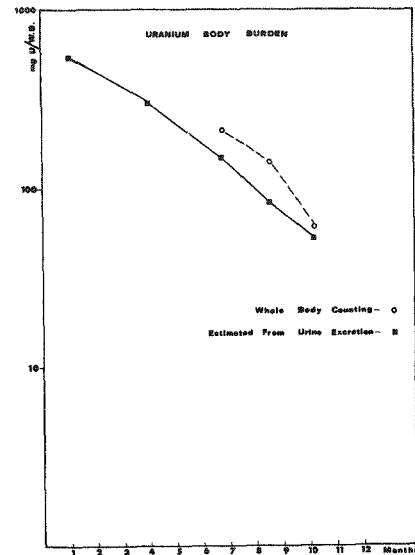


Fig. 2

availability of a Whole Body Counter was therefore useful in developing a direct counting method based on Coffield's method (3) which measures the counts at 186 KeV and 90 KeV, with the 5x9 inch crystal placed directly over the chest at a distance of 1 cm and with corrections made by counting of an unexposed "double", equal in weight and height to the exposed subject. Details of these calculations are given in Table I. Uranium standard was counted at a distance from the crystal of 22 and 11

Table I : Body Burden obtained by Whole Body Counting at 186 keV

months after exposure	cpm					μcm^{-1}	cpm		mg burden U/UR1
	U x=22cm	U x=11cm	URo	U*URo	UR1		N(UR1)	NoUR1	
7	68	130	174	179	260	0.13	86	350	207
9	74.5	144.9	180	187.8	245.7	0.113	65.7	266.4	141.6
11	78.6	149.9	147.3	155.5	175.5	0.113	20.2	114.5	61

U = Uranium-graphite standard containing 77 mg uranium. URo = control subject. UR1 = Subject under study. N(UR1) = counts obtained from subject. No(UR1) = Counts obtained from subject after correction for chest absorption.

Example of calculation:

$$N(\text{UR1}) = \text{No}(\text{UR1}) \cdot e^{-ux} ; \quad \text{No}(\text{UR1}) = \frac{N(\text{UR1})}{e^{-ux}} = \frac{86}{e^{-1.4}} = 350 \text{ cpm} ;$$

$$\text{Body Burden} = \text{mg U/UR1} = \frac{\text{No}(\text{UR1}) \cdot \text{mgU}}{\text{cpm U}} = \frac{350 \times 77}{130} = 203 \text{ mg} ;$$

cm, and under the back of an unexposed person serving as a double. Thus the chest absorption μ was determined.

Subtracting the counts obtained in the double (URO) from the counts obtained from subject (URI) gives the counts contributed at 186 KeV from the uranium in the lungs. When correction is made for chest absorption in accordance with the simplified formula $N_0(\text{UR } 1) = \frac{N(\text{UR } 1)}{e^{-\mu x}}$, the actual counts of uranium are obtained, and when the amount of uranium retained in the lung is determined.

The results of employing this method enabled us to assess the amount of natural uranium still present in the lungs of the exposed subject, to follow the rate of elimination from the body and (Fig. 2) calculate by extrapolation the amount which was inhaled at the time of exposure.

This method of direct counting of retained uranium in the lungs is certainly more accurate than urinalysis, as it counts retained rather than excreted uranium and is thus not dependant on physiological and other factors which influence the rate of excretion of radionuclides from the body. It is not, however, an easy method as it requires bringing both the subject and his "double" to the whole body counter at frequent intervals, measurements are time consuming and moreover, not all laboratories possess a Whole Body Counter which is rather expensive equipment.

Other methods for accurate direct determination of uranium contamination are therefore under investigation.

External counting of the low intensity uranium L X - rays (energies 13.6, 16.9, and 20.2 keV) seemed to us a promising technique for the estimation of insoluble natural uranium in the human body.

There are several publications on development of instruments and methods for the estimation of plutonium in the lungs by counting the L X-rays of uranium derived from it (4,5,6,7,8), but none about possibility of estimation of uranium as such.

Clearly, there are difficulties due to the fact that these low energy radiations are very easily absorbed in the tissues of the chest wall. The published half value thickness is about 0.7 cm of soft tissue and only 0.03 cm of bone. It seemed however possible that with this technique uranium in the lungs can be counted with a proportional counter without much interference of counts derived from caesium-137 (660 keV) which exists now in every human being.

We have used detector system comprised of two commercially made proportional detectors (30 cm x 15 cm) flushed with argon - methane gas and connected to a suitable single channel analyzer. The detectors were placed inside the low background

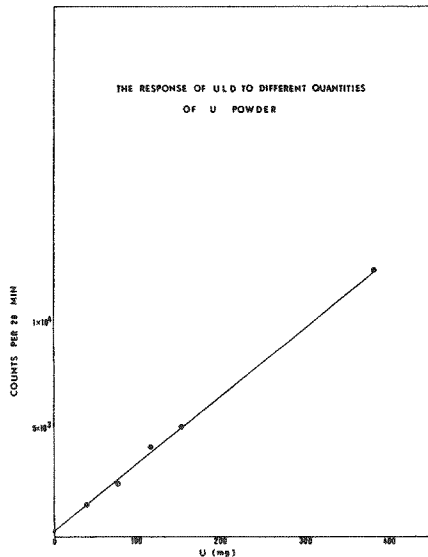
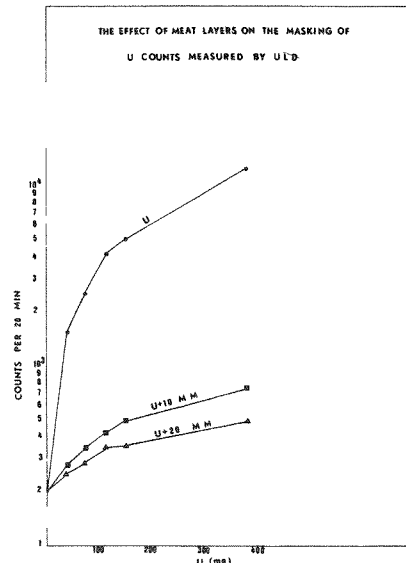


Fig. 3 Fig. 4



Whole Body Chamber. We call the system ULD = Uranium Lung Detector.

In one set of measurements the response of the ULD was checked with different quantities of UF_4 powder directly exposed to the detector or covered with meat layers, each 10 mm thick. The results are shown in figures 3 and 4 respectively.

In another set of measurements sponges saturated with suspension of UO_3 in different quantities, were placed in the intact chest cavity of a male goat, (Fig. 5) simulating contaminated lungs in the chest. The results are shown in Fig. 6.

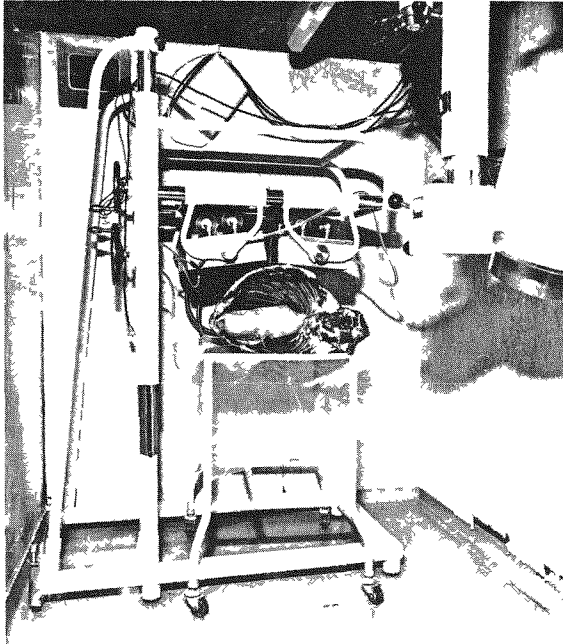


Fig. 5

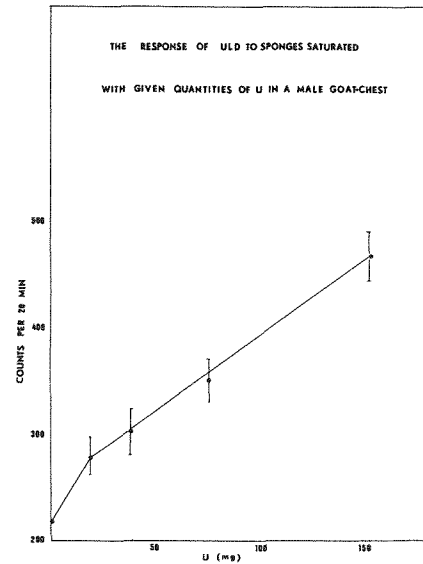


Fig. 6

From these preliminary results it seems that we shall be able to estimate without difficulties about a $1/3$ of MPBB for natural uranium.

It soon became clear that a more specific and narrow spectral region should be used in order to get specific counting response to uranium.

These spectral regions were analyzed (by connecting the system to a multichannel analyzer), in the range 0 to 25 keV.

Although more counts were obtained from uranium in the low energy region I and less in II as compared to region III, the analysis suggests that uranium is best counted in region III (and the instrument was therefore set up for this region) as in the former regions there is also high count of caesium-137 and other sources (Table II). The ratio of uranium count to caesium-137 count is highest in region III. It should be noted however that the amount of caesium-137 used for these calibrations was very high and normally existing quantities in the body (about 6 nCi) do not increase the count to any significant extent.

A more detailed study of the counts obtained from varying quantities of uranium, in the presence of 8 nCi of caesium-137 and 245 g of KCl (equal to normal body content of these isotopes in standard man), is summarized in Table II. The results confirm earlier observations and show that about $1/3$ MPLB of uranium can easily be detected in the lung when measurements are made in this system in the energy spectral region III (10 - 20 keV).

Table II : Counts Obtained in U L D

Isotope and amount counted		Regions			
		I 3-16	keV II 2-6	III 10-24	
		Counts per 20 minutes			
^{137}Cs	$8 \times 10^{-9}\text{Ci}$	153	40	38	
	$100 \times 10^{-9}\text{Ci}$	908	152	230	
	$200 \times 10^{-9}\text{Ci}$	1883	290	431	
KCl	245 mg	450	112	148	
UF_4	50 mg	2013	325	1219	
	25 mg	1065	184	753	
	15 mg	697	132	419	
	7.5 mg	400	90	203	
$^{137}\text{Cs} + \text{KCl} + \text{UF}_4$					
$8 \times 10^{-9}\text{Ci}$	245 mg	0 mg	571	107	141
"	"	50 mg	2410	423	1437
"	"	25 mg	1645	311	910
"	"	15 mg	1337	412	566
"	"	7.5 mg	885	278	280
Calculated net	50 mg		1839	316	1296
UF_4 counts	25 mg		1074	204	769
	15 mg		766	305	425
	7.5 mg		314	171	139
Calculated ratios	50 mg/8 nCi		13.1	8.2	32.1
U/Cs	7.5 mg/8 nCi		2.6	2.2	5.3
Background			100	18	24

Work is now in progress with tissue equivalent phantom. It is hoped that when all measurements are repeated in the correct spectral region under controlled conditions, it will give us another method suitable for accurate estimation of the amount of uranium contamination in the human lung.

Acknowledgements

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EXPOSURE FROM NUCLEAR POWER

"AS LOW AS PRACTICABLE" IN THEORY AND PRACTICE

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Abstract

The ICRP introduced the phrase "as low as practicable" into the language of radiation protection some years ago. Since that time, the phrase has served as a qualitative admonition to prudent judgment. The record of the nuclear power industry, in particular, bears witness to the care with which engineers have applied this concept to the design and operation of nuclear power plants.

Recently the U.S. Atomic Energy Commission has proposed to elevate "as low as practicable" to a regulatory standard with numerical limits. This proposal has the effect of reducing the ICRP limit for public exposure by a factor of 100 and essentially wipes out the opportunity for judgment, sound or otherwise.

Such a reduction in an environmental quality standard is probably unique in the field of public health. An action as dramatic as the proposed reduction is properly taken in circumstances of near-epidemic proportions. However, there is no evidence that radiation exposure at the ICRP limits has any demonstrable effects; there is in fact considerable evidence that it does not. Further, the nuclear industry has conducted its operations in such a way that public exposures are far below the ICRP limits.

The justification offered for setting a legal, numerical limit to "as low as practicable" is that if it can be done, it must be done. This is a philosophy that demands careful scrutiny. The costs in effort and money are considerable; the benefits, if any, are miniscule. Further, it is a philosophy without reason and without stopping place. Finally, it is an action which seriously undermines confidence in the ICRP standards.

Introduction

The ICRP introduced the phrase "as low as practicable" into the language of radiation protection in 1958.¹ This phrase, I believe, was intended as an admonition to prudent judgment in the face of the possibility that "any exposure may involve some degree of risk."²

The record of radiation exposure, both occupational and public, during the last several decades bears evidence to the care with which designers, engineers and operators have applied this concept. The public exposures which have resulted from the operation of nuclear power plants are particularly striking in this respect. In all but unusual instances, the public exposures from these plants have been less than a few percent of the limits recommended by the ICRP.

One can only believe that those who labored on the ICRP committee have been gratified with the result of their carefully worded recommendation.

U.S. A.E.C. Proposed Appendix I to 10 CFR Part 50

Comes now, however, the United States Atomic Energy Commission.

About two years ago this body issued a draft of a new proposed regulation.³ The issuance of regulations is, of course, a responsibility of governmental agencies, and the US AEC has met this responsibility fully. The new proposed regulation of concern to us is known as "Appendix I to 10 CFR Part 50." For those of you who are not familiar with this proposed regulation, a terse summary may be useful.

The proposed Appendix I to 10 CFR Part 50 elevates "as low as practicable" to a regulatory standard with numerical limits. These limits are, for all practical purposes, 1/100 of the ICRP public limit; i.e., 5 mrem per year to any organ of the body. There are also, in this proposed regulation, annual release limits for certain isotopes, and some talk of the need for flexibility to allow nuclear power plants to produce electricity now and then. The essence of the regulation is, however, the public organ limit of 5 mrem per year.

The reduction of an environmental quality standard by a factor of 100 is probably unique in the field of public health. What circumstances, one wonders, have called for an action as dramatic as this?

Possible Need for the Proposed Regulation

Is it that the present limits of radiation exposure are producing unacceptably high rates of injury and death?

Is it that the nuclear power industry is abusing the present limits and that the proposed rule is needed to enforce them?

Is it that the margin of safety in the present limits has been found to be smaller than was intended?

Taking each of these possible explanations in turn, we find that (1) there is no evidence that radiation exposure at the present ICRP limits has any demonstrable effects, and considerable evidence that it does not, (2) the nuclear power industry has conducted its operations in such a way that maximum public exposures have seldom exceeded 1/100 of the ICRP limits, and (3) the U.S. National Council of Radiation Protection completed in 1971 an extensive review of the bases for radiation exposure limits and concluded that there is no need to reduce them.⁴

The recent BEIR Report⁵ encourages consideration of quantifying the "as low as practicable" concept, but states that "there should not be attempted the reduction of small risks even further at the cost of large sums of money that spent otherwise, would clearly produce greater benefit."

As far as public health is concerned, there is no demonstrated need for the proposed regulation. Why, then, is this dramatic reduction of exposure limits necessary?

Justification

If one searches through the voluminous material associated with the proposed Appendix I, he finds that the justification given by the AEC is this: since the nuclear industry has shown that it can operate at about 1/100 of the ICRP limits, the industry should be required to operate this way. In other words, if it can be done, it must be done. This is a new concept in public health and deserves thoughtful scrutiny. It is a concept which is beginning to appear in public health areas other than radiation and leads to the ultimate goal where all environmental contaminants are maintained forever at zero, whatever that may be.

The concept that if it can be done, it must be done has no stopping place. For example, suppose that the nuclear industry responds to the proposed Appendix I by designing nuclear plants which operate at 1/10,000 of the ICRP limit; it follows by the concept that they must be operated at this limit. The engineer who designs a safety factor into his plant is rewarded by having it eaten away by the next set of regulations. Such a drive to perfection (if zero is indeed perfection) has certain attractions to some people who do not count the cost, but the cost must be counted and it must be paid.

The Costs

The additional costs which will be imposed on the electric energy produced in nuclear power stations by the proposed Appendix I may be examined in two ways: (1) in terms of the benefit-cost ratio, or (2) in terms of cost per person-rem avoided. (Women are being liberated by elimination of the unit man-rem from the U.S. vocabulary). Application of the benefit-cost ratio to the proposed Appendix I is a simple matter: the benefit to public health is zero; the cost will be appreciable; the value of the ratio is zero. So much for the benefit-cost analysis.

At the previous meeting of this Association, Hedgran and Lindell gave a charming paper which lead to an estimate of the value of a man-rem as about 1,000 Swedish crowns.⁶ Currency fluctuations in the intervening years have introduced more uncertainty in this figure than the uncertainty of their estimate. Estimates of the costs of the person-rem to be saved by the enactment of Appendix I range from less than one hundred "early 1973 dollars" to four million dollars per person-rem.⁷ Whatever the value of the dollar in terms of the crown, the Appendix I person-rem appear likely to be expensive.

There is another aspect to these Appendix I person-rem, quite apart from their cost. Hedgran, Lindell and the others who have speculated on the proper expenditure to spare a man-rem were considering actual exposures. Appendix I, on the other hand, is concerned largely with fictitious person-rem received by an imaginary child, drinking imaginary milk produced by an imaginary cow which is grazing on an imaginary pasture at the boundary of the plant site. Thus, real dollars are to be spent to spare make-believe doses. Estimation of the value of a make-believe person-rem may belong in the realm of fables, but surely not in the realm of radiation protection.

The Critics of Nuclear Power

You will see that our search for some justification of the numerical interpretation of "as low as practicable" has so far been in vain. Perhaps looking at the matter from the point of view of the U.S. Atomic Energy Commission may suggest an answer.

Some years ago the late Andre Cipriani said that one should remind himself that government bureaucrats, in their inner hearts, are not purposely trying to make life impossible for the rest of us. (This, as friends of Dr. Cipriani will realize, is a sanitized version of his original statement).

Critics of nuclear power have predicted an assortment of disasters which make Dante's Inferno look like a summer afternoon in the park should the development of nuclear power continue. These critics have pleaded for reduction of radiation limits by factors of ten or more, reduction of these limits to zero (whatever that may be), a moratorium on nuclear power plants until it can be shown that they are absolutely safe, and the elimination of these plants altogether and forever. Still mindful of Dr. Cipriani's comment, I realize that to some extent a governmental agency should respond to the wishes of the public it serves. Perhaps it is here that we may find the explanation for Appendix I.

Nothing on this aspect of the case appears in the pages of reports, testimony and response to which Appendix I has given rise. There is no way of knowing how near to the truth is the suggestion that the purpose of this proposed regulation is to buy off the critics, but it is the only plausible explanation I have found. If this is in fact the explanation, it is ironic indeed: the critics of nuclear power have been trying for years to discredit the AEC; now, by proposing this rule, the AEC discredits itself.

There are circumstances in which one could forgive a frightened bureaucracy for proposing a regulation as unfortunate as Appendix I. Imagine, if you will, a world where radiation is the only cause of illness and death, where nuclear power is the only source of radiation, where alternate sources of power are in ample supply, and where the production of electricity by means other than nuclear power has no ill effects on public health and the environment. In such a world, Appendix I would make good sense. It is such a world as this that many of the critics of nuclear power envisage, but our world is nothing like this. The proposed regulation makes no sense whatever for today's world.

The public has become somewhat confused and suspicious by the controversy over radiation standards. The proposed Appendix I destroys confidence in the ICRP standards, which are surely the most firmly based and carefully conceived the world has ever known.

Summary and Conclusion

In theory, the principle of "as low as practicable" is an appeal to cautious judgment. The very low public doses and the complete lack of any ill effects on public health attest to the excellence of the theory and the scrupulous observation of it by the nuclear power industry.

In practice, as in the proposed Appendix I, the principle of "as low as practicable" becomes a stringent numerical standard, which is unnecessarily low and essentially unmeasurable. The proposal has already done considerable harm. It has increased the costs of designing, constructing and operating nuclear power plants. It has introduced bewildering confusion into the planning for electric power generation. The proposed Appendix I may increase cost and public opposition to the point where fission and even fusion are eliminated as sources of energy. This, in my opinion, would be a tragic development. I urge that the Appendix I interpretation of "as low as practicable" be buried as quickly and as quietly as possible.

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РАДИАЦИОННАЯ БЕЗОПАСНОСТЬ НАСЕЛЕНИЯ ПРИ ЭКСПЛУАТАЦИИ АЭС СССР

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Abstract

The report considers some problems of radiation safety of a population due to airborne releases from the atomic power station. The basic national standards for radioactive releases (actual or predicted) from a PWR APS are given. The problem of the possible scope of the accident of APS which may cause an environmental emergency radiation dose and the problem of the population doses are discussed.

В настоящее время (к 1 января 1973 г.) в СССР эксплуатируются АЭС различного типа с общей электрической мощностью 2500 Мвт. В их числе /I-6/:

- четыре блока Нововоронежской АЭС (НВАЭС) мощностью около 1500 Мвт;
- два блока Белоярской АЭС (БАЭС) мощностью 300 Мвт;
- Сибирская АЭС мощностью 600 Мвт;
- Ульяновская АЭС с реактором кипящего типа мощностью 75 Мвт.

В июне 1973 г. состоялся энергопуск Кольской АЭС с серийным реактором ВВЭР-440, а в июле - реактора-размножителя на быстрых нейтронах с натриевым охлаждением БН-350 в г. Шевченко (реактор двухцелевого назначения). В ближайшее время предполагается пуск I блока нового серийного реактора канального типа РБМК-1000 на Ленинградской АЭС.

Планируется к 1975 г. довести мощность АЭС до 6000-8000 Мвт, а к 1980 г. - до 28000-30000 Мвт. Такая широкая программа строительства АЭС выдвигает на первый план проблему радиационной безопасности населения, проживающего в районе их размещения.

В докладе основное внимание будет уделено водо-водяным реакторам корпусного типа (ВВЭР), так как для этого типа реакторов в СССР накоплен наибольший опыт. Следует, однако, отметить, что многие положения рассматриваемых вопросов радиационной безопасности реакторов типа ВВЭР являются общими и для другого типа реакторов.

I. Основные регламенты по радиационной безопасности

Действующие в СССР официальные документы, регламентирующие допустимые уровни облучения для персонала АЭС и населения, в основном исходят из рекомендаций Международной комиссии по радиологической защите и Международного агентства по атомной энергии. Они отражены в национальных документах /7-10/, которые в нашей стране являются законодательными.

Так, согласно /7/ предельно допустимые выбросы (ПДВ) при нормальной эксплуатации АЭС и при любых погодных условиях не должны превосходить, кюри/сутки: I-131- 0,1, Sr-89, Sr-90- 0,001, суммы аэрозолей с периодом полураспада больше 1 суток (кроме I-131 и Sr-90-Sr-89)- 0,5 и смеси инертных радиоактивных газов (ИРТ) - 3500.

В настоящее время составляются программы для расчетов ПДВ конкретных АЭС. В целях прогнозирования радиационной обстановки на местности считается целесообразным на АЭС типа ВВЭР производить расчет ПДВ для изотопов H-3, C-14, Ar-41, Mn-54, Mn-56, Co-60, Kr-85, Sr-90,89, Zr-95, Nb-95, Ru-103,106, I-129,131, Xe-133, Cs-137, Ce-141,144, смеси изотопов Kr и Xe, а также I-131- I-135. Уместно заметить, что выброс таких социально значимых изотопов, как H-3, C-14, I-129, Sr-90, Cs-137 и некоторых других, нужно регламентировать не в национальных и даже не в региональных масштабах, а с учетом перспективы развития атомной энергетики (включая заводы по регенерации топлива) во всем мире.

На случай аварийных ситуаций на АЭС в СССР установлены следующие временные регламенты (см. табл. I).

Таблица I

Допустимые дозы аварийного облучения для населения D, бэр /9/

Меры защиты	Внешнее облучение	Облучение щитовидной железы	
		Дети	Взрослые
A. Дозы, которые не требуют специальных мер по защите	≤ 25	≤ 75	≤ 150
B. Дозы, требующие временных ограничений: укрытия в помещениях, ограничения потребления молока и других продуктов,	25-75	75-225	150-450
C. Дозы, требующие серьезных мер защиты вплоть до эвакуации	> 75	> 225	> 450

В настоящее время документы /7/ и /9/ пересматриваются.

2. Радиационная обстановка на АЭС типа ВВЭР

Многолетний опыт эксплуатации в СССР АЭС всех типов подтверждает, что за весь период их эксплуатации не было зарегистрировано ни единого случая, когда газоаэрозольные отходы достигли бы установленных среднесуточных величин (не говоря уже о годовых).

Рассмотрим в качестве примера одну из крупнейших в СССР Нововоронежскую АЭС (НВАЭС) с суммарной электрической мощностью ~ 1500 Мвт.

На НВАЭС работают 4 блока с реакторами типа ВВЭР: I блок (сентябрь 1964 г.) - 210 Мвт, II блок (декабрь 1969 г.) - 365 Мвт; III блок (декабрь 1971 г.) - 440 Мвт и IV блок (декабрь

1972 г.) - 440 Мвт. Реакторы ВВЭР-440 являются головными в этой серии. Сейчас на площадке НВАЭС проектируется У блок, который будет головным в серии ВВЭР-1000. НВАЭС расположена в густонаселенном районе европейской территории СССР. Именно по этой причине, а также учитывая масштабы и перспективу развития АЭС, на НВАЭС существует значительной по объему информации контроль внешней среды. Служба внешней дозиметрии производит контроль следующих сред: выброса из вентиляционных труб радиоактивных аэрозолей и газов; концентрации аэрозолей в атмосферном воздухе; скорости осаждения радиоактивных веществ на почву; воды и донных отложений реки Дон и окружающих озер; питьевой воды, грунтовых вод вблизи хранилищ твердых и жидких отходов; сбросной воды, охлаждающей конденсаторы турбин; воды с полей фильтрации, куда сбрасываются хозяйственно-фекальные отходы; фауны и флоры рек и озер; почвы, растительности, атмосферных осадков. Кроме того, на специально оборудованной автомашине измеряются уровни β - γ -загрязненности почвы, а с помощью интегрирующих дозиметров - годовые дозы гамма-излучения. Контроль радиоактивности охватывает площадь с радиусом 50 км. Кроме суммарной β -активности, анализируются концентрации Co-60, Sr-89, Sr-90, Zr-95, Nb-95, Ru-103, Ru-106, I-131, Ce-141, Ce-144, Cs-137. Исследовательскими институтами производится также измерение H-3 и C-14. Достаточно подробные данные об объеме внешнего дозиметрического контроля, величинах выбросов и радиационной обстановки на местности до 1970 г. приведены в работах /II-12/. Поэтому приведенные ниже данные относятся к 1971, 1972 и первой половине 1973 г., когда работали все блоки (см. табл. 2 и 3).

Таблица 2

Фактические выбросы аэрозолей на НВАЭС

Годы	1971 г.,	1972 г.,			1973 г. (за 5 месяцев)		
	мкюри/год	мкюри/год			мкюри		
Блоки	(I) ^х + II	I	II	III	I	II	III + IV
I-131	16,6	162,8	82,9	1,65	17,0	0,83	7,3
Cs-137	7,9	36,4	3,4	0,16	5,8	0,57	1,8
Sr-90	0,94	12,7	0,64	0,022	2,1	0,14	0,06
Ce-141, 144	5,4	27,5	2,45	0,13	5,1	0,46	0,26
Co-60	6,4	17,5	3,4	0,41	4,9	1,5	-

^х/В 1971 г. I блок находился на модернизации.

Таблица 3

Фактические выбросы ИРГ (Kr , Xe) на НВАЭС

Годы	1971 г.,	1972 г.,		1973 г. (за 6 месяцев),	
	кюри/год	кюри/год		кюри	
Блоки	I + II	I+II	III	I+II	III+IV
Выброс	380	20000	400	16000	750
% от ПДВ	0,03	1,6		2,6	

Из табл. 2 и 3 видно, что газоаэрозольные выбросы по всем четырем блокам НВАЭС очень низки. Так, выбросы по аэрозолям составляют менее 1% от ПДВ, а от двух серийных блоков ВВЭР-440 они еще меньше. По ИРГ суммарные выбросы в 1971-1973 гг. составляли от 0,03 до 2,6%, а для III и IV блоков - от 0,03 до 0,12% от ПДВ. Необходимо отметить, что основной вклад в выбросы дает I (несе-

рийный) блок. Удельная активность воды теплоносителя III и IV блоков примерно на 2 порядка меньше, чем для I блока, и определяется в основном изотопами наведенной активности F-18, Na-24, Ar-41 и K-42.

В табл. 4 и 5 приведены некоторые данные о концентрациях радиоактивных веществ в атмосферном воздухе и скорости осаждения на почву.

Таблица 4

Концентрации радиоактивных аэрозолей в атмосферном воздухе γ , IO-17 кюри / л

R, км	1971 г.			1972 г.			1973 г.		
	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144
0,5	0,53	0,89	13,5	0,27	0,57	3,8	0,12	0,30	1,40
2-3	0,43	0,65	9,2	0,22	0,51	2,8	0,06	0,11	2,74
4-5	0,58	0,92	15,4	0,25	0,23	3,5	0,10	0,16	1,52
8	0,40	0,73	11,2	0,17	0,27	2,7	0,056	0,32	1,06
15	0,46	0,72	10,8	0,17	0,25	2,4	0,048	0,10	1,56
50x	0,41	0,63	11,4	0,23	0,32	3,0	0,026	0,44	1,59

x/ г. Воронеж.

Таблица 5

Выпадение радиоактивных веществ из атмосферного воздуха μ , мкюри/км²год (в 1973 г. мкюри за I квартал)

R, км	1971 г.			1972 г.			1973 г. (I кв.)		
	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144
1,5-2	1,43	2,28	38,4	0,70	1,13	10,6	0,15	0,21	0,51
4-6	1,32	2,16	26,5	0,54	0,99	7,8	0,078	0,21	0,68
9-12	1,58	2,22	27,6	0,59	1,33	7,6	0,072	0,38	0,45
50x	1,37	2,32	35,5	0,69	0,95	12,3	0,189	0,42	1,17

x/ г. Воронеж.

Анализ табл. 4 и 5 показывает, что как γ , кюри/л, так и F, мкюри/км²год практически не изменяются с расстоянием от АЭС, а по абсолютной величине не отличаются от соответствующих величин для изотопов глобального происхождения. Заметим, что короткоживущих изотопов (в том числе I-131) ни в атмосферном воздухе, ни на почве не наблюдалось.

Анализ радиоактивности других сред (воды, рыбы и т. д.) подтверждает, что выбросы НВАЭС совершенно не влияют на радиационную обстановку, и существующие уровни загрязнения внешней среды определяются изотопами глобального происхождения, которые характерны для этого периода для центральных районов СССР.

Экспериментальные данные, приведенные в табл. 4 и 5, а также аналогичные данные за 1965-1970 гг. дают возможность определить среднегодовые скорости осаждения изотопов на почву V_g . Она оказывается равной $V_g \approx 1,0$ см/сек.

Интегрирующие γ -дозиметры, расставляемые ежегодно в радиусе до 50 км, не показывают различия внешних полей гамма-излучения от уровней естественного фона. Расчеты показывают, что в 1972 г., когда работали три блока, максимальная доза от внешнего γ -излучения, обусловленная выбросом ИРГ, была меньше 0,2 мбэр/год, а от I-131 на щитовидную железу детей (с учетом пищевой цепочки в мае-октябре) менее 1,5 мбэр/год при среднем естественном фоне

90±4 мбэр/год. Следовательно, расчетные величины дозовых нагрузок на население от АЭС меньше колебаний естественного фона и в тысячи раз меньше ПДД для отдельных лиц из населения.

Анализ газоаэрозольных выбросов на НВАЭС за весь период ее эксплуатации, а также анализ выбросов других АЭС СССР дает возможность сделать заключение, что расчетные пределы доз на отдельные лица из населения, обусловленные этими выбросами, не достигали 5 мбэр/год, а усредненные дозы по большим группам не достигали 1 мбэр/год. Жидкие и твердые отходы НВАЭС (которые в данном докладе не рассматриваются) практически не изменяют радиационной обстановки на местности.

3. Проектируемые газоаэрозольные отходы при нормальной эксплуатации АЭС

При нормальной эксплуатации радиационную обстановку как на АЭС, так и во внешней среде определяет степень герметичности твэлов и оборудования теплоносителя. Вследствие процессов коррозионно-усталостного типа оболочки тепловыделяющих элементов (твэлов) могут иметь микротрещины, а затем и более крупные дефекты. При решении балансных уравнений для расчета активности теплоносителя в проектируемых АЭС допускаются предельные условия, а именно: наличие 1% оболочек твэлов с газовой негерметичностью (микротрещины) и 0,1% более крупных дефектов в оболочках, допускающих прямой контакт топлива с водой теплоносителя /13/. Постоянная скорости утечки летучих изотопов из-под оболочек (через микротрещины) в I контур принимается равной $1 \cdot 10^{-5}$ сек⁻¹. Тогда в теплоносителе реактора ВВЭР-440 (полный объем которого равен 200 м³) удельная активность изотопов будет достигать (мкюри/л): по сумме изотопов - I39, по I-131 - 1,9, по сумме изотопов йода - 12,2, по сумме изотопов Kr и Xe - 112. Загрязненность воздуха в помещениях рассчитывается из условия допущения неорганизованных протечек теплоносителя 0,2 т/час. При этом предполагается, что из воды в паровоздушную фазу изотопы переходят в соотношениях: ИРГ - 100%, изотопы йода - 1%. Загрязненный воздух из помещений и технологические сдувки из оборудования поступают на системы подавления активности. Они включают в себя (в различных вариантах): газгольдеры выдержки (для снижения активности технологических сдувок); фильтры ФПП из тонковолокнистой ткани, дающие коэффициент улавливания по аэрозолям до 99,9%; фильтры с угольной насадкой для улавливания газообразной фазы йода. В последнее время на некоторых АЭС для задержки Kr и Xe проектируются также установки подавления активности на фильтрах с активированным углем. В табл. 6 в качестве примера приведены расчетные выбросы одной из проектируемых АЭС типа ВВЭР-440. Сравнимые (или несколько меньшие) выбросы принимаются и для других типов проектируемых АЭС. Необходимо отметить, что эти величины получены при наиболее пессимистических предположениях, о которых сказано выше. В действительности, как видно из предыдущего раздела, выбросы на реакторах ВВЭР-440 значительно меньше проектных величин.

При расчете ПДВ I-131 в атмосферу учитывается пищевой путь поступления по цепочке воздух-трава-молоко. По многолетним экспериментальным данным авторами получены следующие соотношения для I-131, которые используются при нормировании выбросов I-131:

$\frac{\text{мкюри/л молока}}{\text{мкюри/м}^3 \text{ воздуха}} = 700;$ $\frac{\text{мкюри/кг травы}}{\text{мкюри/м}^3 \text{ воздуха}} = 5000;$ $\frac{\text{мкюри/л молока}}{\text{мкюри/кг травы}} = 0,12$

Рассеяние газоаэрозольных отходов после их очистки производится через трубы высотой 120-150 м. Важным фактором защиты населения

является установление вокруг АЭС санитарно-защитной зоны. В каждом конкретном случае размеры этой зоны определяются органами Министерства здравоохранения СССР.

Таблица 6

Расчетные выбросы изотопов проектируемой АЭС типа ВВЭР-440, кюри/сутки

Изотоп	Выброс, кюри/сутки	Изотоп	Выброс, кюри/сутки	Изотоп	Выброс, кюри/сутки
Sr-89	$1,5 \cdot 10^{-5}$	Ba-140	$2,6 \cdot 10^{-4}$	Fe-59	$1,6 \cdot 10^{-5}$
Sr-90	$2,4 \cdot 10^{-6}$	La-140	$1,4 \cdot 10^{-4}$	Co-60	$2,6 \cdot 10^{-4}$
I-131	$2,2 \cdot 10^{-2}$	Na-24	$6,0 \cdot 10^{-4}$	Xe-133	250
ΣI	$5,6 \cdot 10^{-2}$	Cr-51	$1,2 \cdot 10^{-3}$	$\Sigma ИРГ$	350
Cs-137	$2,2 \cdot 10^{-4}$	Mn-56	$1,3 \cdot 10^{-3}$	$\Sigma аэрозоли$	0,18

4. О масштабе повреждения активной зоны, при котором создаются дозы аварийного облучения на местности

В работах /13, 14/ советскими специалистами высказаны соображения по поводу характера развития крупной аварии на реакторах типа ВВЭР, масштабов возможного аварийного выброса во внешнюю среду и технических средств подавления активности. Подчеркивается, что вероятность возникновения аварии может быть сведена к минимуму за счет высокого качества оборудования, периодического и непрерывного контроля за его состоянием; своевременным срабатыванием систем защиты ядерной установки, предотвращающих возникновение и развитие аварии; разработки системы локализации распространения радиоактивных веществ за пределы АЭС и надежных систем аварийного охлаждения. Наиболее опасной считается крупная невосполнимая течь теплоносителя I контура с последующим нарушением теплового режима активной зоны реактора, разгерметизацией оболочек твэлов и далее частичным оплавлением ядерного топлива. Согласно этой модели предполагается, что развитие крупной аварии на реакторах типа ВВЭР может происходить в три последовательные этапа:

I этап - разрыв I контура и невосполнимая штатными средствами течь теплоносителя в герметичное помещение, где расположено оборудование;

II этап - частичное или полное разрушение оболочек твэлов;

III этап - частичное оплавление ядерного горючего.

Основная тактика обеспечения безопасности по модели работы /14/ сводится к тому, чтобы на I этапе аварии беспрепятственно с помощью взрывных клапанов выбросить в атмосферный воздух паровоздушную смесь теплоносителя и максимально ограничить развитие II и III этапов аварии путем использования специальной системы аварийного охлаждения и конденсации пара и локализации активности в пределах герметичного помещения.

Назовем условно масштабом аварии (f_2) долю оплавленного ядерного топлива.

Используя экспериментальные данные работы /13/, ниже рассчитан масштаб аварий, при котором создаются на местности дозы аварийного облучения, приведенные в табл. I для группы А.

Расчет сделан для двух групп критических изотопов: смеси ИРГ (т. е. изотопов Kr и Xe) и смеси изотопов йода. Критическими группами населения выбраны дети в возрасте до I года. Обозначим A_n и A_c активность изотопа в активной зоне и в теплоноси-

теле I контура соответственно, кюри. Тогда полная активность аварийного выброса Q будет равна

$$Q = A_c f_3 f_4 f_5 f_6 \quad \text{I этап аварии} \quad (1)$$

$$Q = A_n f_1 f_5 f_6 \quad \text{II этап аварии} \quad (2)$$

$$Q = A_n f_1 f_2 f_3 f_6 \quad \text{III этап аварии} \quad (3)$$

Коэффициенты f_i приведены в табл. 7.

Таблица 7
Коэффициенты для расчета аварийных выбросов

Название коэффициента	ИРГ	Йод-131	Этап аварии
f_1 - доля активности, выходящей из твэла	$(0,12-2)10^{-2}$ 1,0	$(0,12-2)10^{-2}$ 0,8	II III
f_2 - доля оплавленного топлива-масштаб аварии	Подлежит	определению	III
f_3 - доля активности, вышедшая из технологического оборудования (с учетом адсорбции на внутренних поверхностях оборудования и трубопроводов)	1,0	0,5-1,0	I
f_4 - доля от содержащегося в теплоносителе газа, вышедшего при разрыве I контура	1,0	0,4	I
f_5 - доля активности теплоносителя, переходящая в парогазовую форму	1,0	$1 \cdot 10^{-2}$	I
f_6 - доля активности, вышедшая из технологического помещения (уменьшение обусловлено адсорбцией на стенах помещений и оборудования, а также радиоактивным распадом)	0,25	0,1	I-III

Поскольку в нашей работе ставится задача определения масштаба аварии, т. е. коэффициента f_2 на III стадии аварии, мы не приводим здесь результатов расчета выбросов для первого этапа аварии. Упомянем лишь, что по расчетам /13/ они не создают чрезвычайно опасных доз на местности. Методически масштаб III этапа определяется следующим образом: вначале рассчитывается аварийный выброс (Q), при котором создаются дозы аварийного облучения, приведенные в табл. I. Затем из соотношения (3) определяется соответствующая этому выбросу процентная доля поврежденной активной зоны f_2 , условно названная масштабом аварии, т. е.

$$f_2 = (Q \cdot 100\%) / (A_n f_1 f_3 f_6). \quad (4)$$

Соотношение между индивидуальной дозой D и кратковременным выбросом Q имеет следующий вид:

$$D = P \frac{0,69 \cdot M}{\text{сек.кюри}} C_i \frac{\text{сек}}{M^3} Q \text{ кюри.} \quad (5)$$

Здесь C_i - так называемый метеорологический фактор разбавления

(или фактор "вытяжки"), определяемый известной формулой Пасквилла-Гиффорда, для короткого выброса. Графическая зависимость C_1 от расстояния, высоты выброса, скорости осаждения и категорий погоды по Пасквиллу даны, например, в работах /15, 16/. \bar{P} - нормализованный множитель, численно равный мощности дозы бэр/сек при концентрации изотопа в воздухе λ , кюри/м³.

Для ряда задач более удобно соотношение

$$D \text{ бэр} = \bar{P} \frac{\text{бэр.м}^3}{\text{сек.кюри}} \left(\lambda \frac{\text{кюри}}{\text{м}^3} t \text{ сек} \right) \quad (6)$$

Здесь t - продолжительность экспозиции. Величину (λt) при постоянной во времени концентрации и $\int \lambda(t) dt$ при зависящей от времени концентрации называют интегралом концентрации.

Таким образом, из формулы (6) может быть определен интеграл концентрации (λt) или $\int \lambda(t) dt$ соответствующий заданной дозе D . Рассмотрим более подробно значения нормализованного множителя \bar{P} . Для расчета поля γ -излучения от факела радиоактивных газов ИРГ обычно допускается предположение о полубесконечном раз- мере облака. Как известно, в этом случае

$$\bar{P} = 0,25 E (\text{бэр.м}^3) / (\text{кюри.сек}), \quad (7)$$

где E - энергия γ -квантов, Мэв/распад.

Для этих условий γ -излучение на местности (с учетом многократного рассеяния в воздухе) может быть рассчитано также по формуле:

$$D, \text{ рад} = 2\pi 0,87 (\lambda t) \sum_i \frac{K_{\gamma,i}(E_i)}{\gamma_i(E_i)}, \quad (8)$$

где $K_{\gamma,i}(E_i)$ - гамма-постоянная i -ой энергии, $\frac{(\text{р.м}^2)}{(\text{сек.кюри})}$; $\gamma_i(E_i)$ - коэффициент истинного поглощения γ -квантов в атмосферном воздухе, 1/м; 0,87 - переводной множитель от единиц рентген к рад.

В табл. 8 приведены уточненные радиационные характеристики для смеси ИРГ и изотопов йода, относящиеся к реактору на тепловых нейтронах типа ВВЭР-440 с тепловой мощностью 1400 Мвт, продолжительность кампании $T = 3$ года и выдержка - от 0 до 6 часов.

Для расчета дозы γ -излучения использовались нормализованные множители \bar{P} , приведенные в формуле (7) или табл. 8. Для расчета дозы на щитовидную железу детей нормализованные множители получены, исходя из стандартов МКРЗ и модели, предложенной в работе /17/. При ингаляционном пути поступления I-131 $\bar{P} = 800$, I-131 в сопровождении I-132-I-135 $\bar{P} = 1440$ и при поступлении I-131 через молоко $\bar{P} = 1,5 \cdot 10^5$ (бэр.м³)/(кюри.сек). Заметим, что интегралы концентрации (λt) , соответствующие заданной дозе D , могут быть получены с помощью формулы (6) из соотношения

$$(\lambda t) (\text{кюри.сек}) / \text{м}^3 = D \text{ бэр} / \bar{P} (\text{бэр.м}^3) / (\text{кюри.сек}) \quad (9)$$

При разовом загрязнении пастбищ используются такие соотношения: $\bar{P} = 10,7$ (бэр.л молока)/(мкюри.сутки). Для детей до 1 года доза D на щитовидную железу равна: 11,5 бэр/мкюри I-131, 20,5 бэр/мкюри I-131 в присутствии I-132 - I-135 при ингаляции и 15,5 бэр/мкюри I-131 при поступлении с молоком. Расчет масштаба аварии произведен для дозы аварийного облучения 25 бэр от внешнего γ -излучения и 75 бэр на щитовидную железу детей (т. е. дозы, не требующие принятия специальных защитных мероприятий). За основу принят серийный реактор типа ВВЭР-440. Расстояния, где расположены населенные пункты - рецепторы, выбраны равными:

Таблица 8

Радиационные характеристики для смесей ИРГ
и изотопов йода реактора ВВЭР-440

Величина	Время выдержки, мин						
	0	1	10	30	60	180	360
Полное содержание в активной зоне, 10^6 кюри							
Kr, Xe	590	490	341	286	256	218	188
I-131 - I-135	380	360	330	320	300	240	200
I-131	34,8	34,8	34,8	34,8	34,8	34,6	34,4
Энергия E, МэВ/распад	0,74	0,57	0,52	0,51	0,47	0,35	0,25
K_{γ} , р.м ² /сек.кюри, 10^{-5}	7,90	6,4	5,7	6,1	6,0	4,9	3,8
P , бэр.м ³ /сек.кюри	0,18	0,14	0,13	0,13	0,12	0,081	0,061

$R_1 = 50$ км и $R_2 = 4$ км. Скорость осаждения на почву V_g для ИРГ принята равной нулю, а для изотопов йода $V_g = 1$ см/сек. Категория погоды принята по классификации Пасквилла. За $f_2 = 100\%$ принято полное оплавление ядерного горючего активной зоны. При этом сделано допущение, что специальных мер по подавлению и локализации активности не сделано. Результаты расчета масштаба аварии приведены в табл. 9.

Таблица 9

Масштаб аварии $f_2\%$, при котором может достигнуть доза внешнего облучения на все тело 25 бэр и доза на щитовидную железу у детей 75 бэр, $h_{эфф} = 30$ м (для реактора ВВЭР-440)

ПДД, источник	Категории погоды по Пасквиллу					
	A	B	C	D	E	F
Расстояние $R_1 = 50$ км						
D = 25 бэр, ИРГ	>100	>100	>100	>100	>100	>100
D = 75 бэр, I-131, вдыхание	>100	>100	>100	>100	23	62
D = 75 бэр, I-131-I-135, вдыхание	>100	>100	>100	78	13	35
D = 75 бэр, I-131, молоко	>100	5,6	1,8	0,7	0,12	0,32
Расстояние $R_2 = 4$ км						
D = 25 бэр, ИРГ	>100	>100	>100	>100	32	14
D = 75 бэр, I-131, вдыхание	65	5,6	3,7	1,5	0,34	0,15
D = 75 бэр, I-131-I-135, вдыхание	36	3,2	2,1	0,81	0,19	0,08
D = 75 бэр, I-131, молоко	0,35	0,03	0,02	0,008	0,0019	0,0008

Из табл. 9 видно, что для населенного пункта, находящегося на $R_1 = 50$ км, допустимые дозы аварийного облучения 25 бэр внешнего γ -излучения и 75 бэр на щитовидную железу детей при вдыхании изотопов йода или совсем не достигаются даже при стопроцентном оплавлении активной зоны, или достигаются при очень большом (и маловероятном) повреждении активной зоны. Исключение составляет пищевой путь поступления йода: если загрязняются пастбища, то достаточно сравнительно небольшого масштаба аварии ($f_2 = 0,12-1,8\%$ для категорий C - F), чтобы появилась необходимость принятия решения - прежде всего ограничения потребления молока.

На расстоянии 4 км радиационная обстановка ухудшается и при меньших масштабах аварии. Тем не менее поле γ -излучения от облака ИРГ (кроме двух последних категорий E и F) все же оказывается сравнительно небольшим даже при крупных авариях.

Однако опасность облучения изотопами йода может быть значительной даже при небольших масштабах аварии, когда оплавляются лишь доли процента ядерного горючего. Естественно, что с помощью рассмотренного метода может быть решена и другая задача - расчет дозы на население при заданном масштабе аварии.

Подобные расчеты необходимы для разработки инженерных мероприятий по подавлению активности при авариях.

Заметим, что изложенная методика определения масштаба аварии (f_2) с помощью соотношения (4) не зависит от выбранной модели подавления активности. (Здесь мы использовали одну из возможных моделей /14/.) Коэффициенты "естественной фильтрации" активности f_1 (оболочками твэлов), f_2 (стенками трубопроводов и оборудования) и f_6 (стенами помещений АЭС) характерны для любого типа аварии.

5. О методике расчета популяционной дозы

В настоящее время при оценке степени риска для населения от применения атомной энергии рассчитывают популяционную дозу. При этом обычно используется единица размерности "человек.бэр". Если N_j - число лиц, получивших дозу от определенного вида радиации D_j бэр, то популяционная доза может быть определена из соотношения

$$D_p = \sum_j N_j D_j. \quad (10)$$

Расчет популяционной дозы по формуле (10) может быть сравнительно легко осуществим от непосредственных прямых путей воздействия радиации на человека, например: на профессиональных работников атомных производств, на население от внешнего потока γ -квантов радиоактивного облака или осадков на почве; от вдыхания радиоактивных аэрозолей и т. д.

Но применение формулы (10) становится затруднительным, а иногда даже невозможным, когда нужно определить вклад в популяционную дозу от загрязненных продуктов питания, которые проходят длительный путь от места их производства (и радиоактивного загрязнения) до потребителя. Это в первую очередь относится к таким долгоживущим изотопам как $H-3$, $C-14$, $Sr-89, 90$, $I-129$, $Cs-137$ и ряду других. При определенных условиях это относится и к $I-131$.

Хотя степень загрязнения пищевых продуктов вблизи атомного объекта может быть известна, но, в какой местности, кем и когда они потребляются, часто является неопределенным. Для такого пути воздействия нами предлагается упрощенная схема, которая, однако, требует дискуссионного обсуждения.

Определим вклад в популяционную дозу от определенного изотопа в форме

$$D_p = \sum_j D \left(\frac{\text{бэр}}{\text{кюри}} \right) N_j (\text{чел}). I_{m,j} \left(\frac{\text{кюри}}{\text{чел}} \right) = D I_m. \quad (11)$$

Здесь, бэр/кюри, - нормированная доза при поступлении в организм единичной активности $I_m = \sum_j N_j I_{m,j}$ - кюри, суммарное содержание изотопа в продуктах питания в момент заглатывания пищи, - поступление в организм данного изотопа с пищей количеству лиц N_j (индекс j относится к уровню загрязненности пищи). Дозовый коэффициент D зависит от возраста человека, пути поступления в организм и радиобиологических констант и с помощью стандартов МКРЗ может быть рассчитан. Обратим внимание прежде всего на то, что в формуле (11) популяционная доза выражается в единицах "бэр", а не "человек.бэр". Собственно эта размерность для D_p вытекает и из формулы (10), если в ней доза на индивидуум D выражается в "бэр/человек". Рассмотрим теперь более подробно множитель I_m .

Если исходить из концепции беспороговости действия радиации,

то при расчете популяционной дозы D_p , а следовательно, степени риска, нет необходимости знать, как и где распределились загрязненные продукты в данной популяции. Достаточно знать, сколько радиоактивного вещества находилось в данных продуктах питания в момент их потребления населением. В простейшем случае (если пренебречь радиоактивным распадом, изменением концентрации при переработке и т. д.) суммарное поступление I_m в организм людей с продуктами питания можно заменить суммарным содержанием активности данного изотопа в продуктах питания A_m , т. е.

$$I_m, \text{ кюри в организм людей} \cong A_m \text{ кюри в продуктах питания} \quad (12)$$

Суммарную величину активности в продуктах питания, употребляемых человеком или животными (например, коровами) определить значительно легче, чем индивидуальную дозу на отдельное лицо. Эту характеристику можно получить путем непосредственных измерений уровней загрязнения продуктов питания или путем расчета, применяя, например, методику Пасквилла-Гиффорда /18-20/ с заданной скоростью осадения на почву. В последнем случае для оценки популяционной дозы требуется знание нормализованной дозы \bar{d} , бэр/кюри, и выброса в вентиляционную трубу Q , кюри.

Формула (5) дает возможность рассчитать дозу от выбросов АЭС на отдельное лицо из населения, т. е. индивидуальную дозу, и входящий в эту формулу множитель C_i представляет собой "индивидуальный" коэффициент разбавления. Если пользоваться моделью Гиффорда, то при разовом выбросе C_i определяется в виде

$$C_i = \frac{F(x)}{\pi \sigma_y \sigma_z u} \exp \left\{ -\frac{1}{2} \left(\frac{y^2}{\sigma_y^2} + \frac{z^2}{\sigma_z^2} \right) \right\}, \quad (13)$$

где $F(x)$ - безразмерная функция, учитывающая истощение радиоактивного факела за счет радиоактивного распада и осадения изотопов на почву. Остальные величины общеизвестны /19, 20/.

Расчет популяционной дозы может быть сделан по формуле, аналогичной (5), с той лишь разницей, что в нее входит популяционный фактор разбавления C_p :

$$C_p = \int \rho C_i dS = \bar{\rho} / C_i dS = \bar{\rho} \bar{C}_p, \quad (14)$$

где ρ - распределение по площади населения, животных или урожайности культур на 1 м², а \bar{C}_p - нормированный популяционный фактор разбавления, сек/м.

Поскольку угол расширения струи выбросов обычно меньше 90°, то удовлетворительным приближением можно сделать замену $y = \theta x$ и $\sigma_y = \sigma_\theta x$, где θ - азимутальный угол между направлением на рецептор и осью струи, σ_θ - его дисперсия. Подставляя в (14) значение C_i из формулы (13) и производя интегрирование по окружности, получим

$$\bar{C}_p = \sqrt{\frac{2}{\pi}} \frac{F(x)}{u} \int_0^x \frac{1}{\sigma_z} \exp \left(-\frac{z^2}{\sigma_z^2} \right) dz. \quad (15)$$

Тогда по аналогии с формулой (5) при прямом пути воздействия радиации на человека популяционная доза D_p будет равна

$$D_p = \bar{P} \left(\frac{\text{бэр}}{\text{человек}} \right) \left(\frac{\text{м}^3}{\text{кюри сек}} \right) \bar{\rho} \left(\frac{\text{человек}}{\text{м}^2} \right) \bar{C}_p \left(\frac{\text{сек}}{\text{м}} \right) Q \text{ (кюри)}. \quad (16)$$

При учете более сложного пути воздействия, когда загрязненные ра-

диоактивными веществами продукты питания поступают в торгово-заготовительную сеть, по аналогии с формулой (II) имеем

$$D_p = \bar{D} \left(\frac{\text{бэр}}{\text{кюри}} \right) I_m (\text{кюри}) = \bar{D} \left(\frac{\text{бэр}}{\text{кюри}} \right) k \left(\frac{\text{м}}{\text{сек}} \right) \bar{C}_p \left(\frac{\text{сек}}{\text{м}} \right) Q (\text{кюри}). \quad (I7)$$

В формулах (I6, I7) \bar{C}_p определяется выражением (I5). Коэффициент k зависит от типа пищевой цепочки для данного изотопа. Например, при поступлении изотопа в организм через цепочку воздух-пастбище-молоко-человек коэффициент k будет равен

$$k = k_1 V_g \left(\frac{\text{м}}{\text{сек}} \right) \bar{\rho} \left(\frac{\text{коров}}{\text{м}^2} \right) \zeta S \frac{\text{м}^2}{\text{корова.сутки}} \frac{T_{\text{эфф}} \text{сутки}}{0,693} \quad (I8)$$

Здесь k_1 - доля от поедаемого коровой с кормом изотопа, перешедшая в молоко; V_g - скорость оседания изотопа на почву; $\bar{\rho}$ - средняя плотность коров на загрязненной территории; ζ - доля изотопа, задерживающаяся на траве; S - площадь, с которой корова поедает траву в течение одних суток; $T_{\text{эфф}}$ - эффективный период полувыведения изотопа из съедобной части травы.

Для других пищевых цепочек, например, воздух-зерновые культуры-человек коэффициент будет равен

$$k = k_2 \left(\frac{\text{кюри/кг}}{\text{кюри/м}^2} \right) V_g \frac{\text{м}}{\text{сек}} \bar{\rho} \frac{\text{кг}}{\text{м}^2} \quad (I9)$$

Здесь k_2 - активность изотопа в 1 кг зерна, полученного с территории, загрязненной интегральным выпадением 1 кюри/м²; $\bar{\rho}$ - урожайность, кг/м², V_g - скорость оседания, м/сек.

Фактор \bar{C}_p - весьма удобная для оценочных расчетов величина, зависящая только от условий истечения и разбавления примеси. Как видно из формулы (I5), он не зависит от горизонтальной дисперсии σ_y . Рост вертикальной дисперсии (σ_z) с расстоянием ограничен толщиной слоя перемешивания. Поэтому единственным фактором, ограничивающим величину \bar{C}_p , является истощение струи за счет радиоактивного распада во время движения и выпадения на почву. Для долгоживущих изотопов расстояние, для которого необходимо рассчитать \bar{C}_p , достигает сотен и даже тысяч километров. Время движения выбросов со средним ветром на столь большие расстояния превосходит сутки. Поэтому при расчете \bar{C}_p существенным оказывается суточный ход категории стабильности. Фактор \bar{C}_p является численной величиной, не зависящей от расстояния в отличие от индивидуального фактора C_1 , который является функцией расстояния.

В заключение отметим, что термин "бэр", в свете вышеуказанного, является более правильным критерием оценки популяционной дозы, чем широко используемый термин "человек.бэр".

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AVERAGE EFFLUENT RELEASES FROM U.S. NUCLEAR POWER REACTORS,
 COMPARED WITH THOSE FROM FOSSIL-FUELED PLANTS,
 IN TERMS OF CURRENTLY APPLICABLE ENVIRONMENTAL STANDARDS*

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Abstract†

Between 1967 and 1972, eighteen "second generation" light-water-cooled nuclear power plants, with capacities in the range of 500-800 MW(e) have been put into operation in the United States. These were in addition to ten smaller demonstration plants and one high-temperature gas-cooled nuclear power plant in operation at the start of this period. The reported yearly air effluent releases of radioactive gases, halogens and particulates, and liquid effluent fission and activation products and of tritium from these plants are evaluated on a Ci/10³ MW(e) basis, and the overall yearly averages for the various types of reactors [boiling water (BWR), pressurized water (PWR) and high temperature gas-cooled (HTGR)] are compared.

The complete first and second generation data are used to project the effluent releases for a "reference" 1,000 MW(e) BWR, PWR and HTGR. The yearly effluent releases for such reference reactors, at an 80% availability, would be:

	<u>AIRBORNE</u>		<u>LIQUID</u>	
	<u>Gaseous</u>	Halogens & <u>Particulates</u>	<u>Fission & Acti- vation Products</u>	<u>Tritium</u>
BWR	1.66x10 ⁶ Ci	5.31 Ci	49.6 Ci	104 Ci
PWR	9,650 Ci	0.17 Ci	30.2 Ci	5,750 Ci
HTGR	2,760 Ci	< 0.02 Ci	0.27 Ci	835 Ci

These and the amounts of effluents released from reference 1,000 MW(e) fossil-fueled plants are compared in terms of relative environmental concentrations and their relationship to the applicable U.S. environmental standards for the principal constituents in their respective plant air-effluent streams. The largest required annual dilution volume for the most restrictive constituent is as follows:

<u>Plant Type</u>	<u>Pollutant</u>	<u>EPA & AEC Standard</u>	<u>Annual Discharge Quantity**</u>	<u>Air Dilution Volume (m³)</u>
Coal (3.5% S)	SO ₂	0.03 ppm	3.66x10 ⁸ lb.	2.14x10 ¹⁵
Oil (1.5% S)	SO ₂	0.03 ppm	1.23x10 ⁸ lb.	6.95x10 ¹⁴
Gas	NO ₂	0.05 ppm	2.71x10 ⁷ lb.	1.23x10 ¹⁴
BWR	Short-lived radio-gases	3x10 ⁻⁸ Ci/m ³	1.66x10 ⁶ Ci	5.54x10 ¹³
PWR	⁸⁵ Kr & ¹³³ Xe	3x10 ⁻⁷ Ci/m ³	9,650 Ci	3.32x10 ¹⁰
HTGR	Short-lived radio-gases	3x10 ⁻⁸ Ci/m ³	2,760 Ci	9.22x10 ¹⁰

**Based on 80% availability

* Research carried out at Brookhaven National Laboratory under contract with the U. S. Atomic Energy Commission.

† Revised August, 1973 to incorporate 1972 data.

Introduction

During the late 1960's a widespread concern for the overall quality of the environment evolved in the United States. This led to the enactment of a variety of legislative and administrative measures intended to decrease the quantities of pollutants released to the air and to waters.

This concern also extended to the radioactivity released to the environment in nuclear power plant effluent streams. It was heightened by the widespread dissemination by the media of claims by Sternglass¹ of a causal connection between power reactor effluents and infant mortality, as well as on the arguments made by Gofman and Tamplin² in the context of power reactors, that a U.S. population exposure of 170 millirems per year would lead to large increases in prevailing U.S. cancer mortality rates.

In what seemed to at least some observers³ a response to the popular concern so evoked, rather than to the scientific evidence, the U. S. Atomic Energy Commission (AEC) formally adopted "as low as practicable" into its regulations governing radioactive effluents, and proposed related numerical guidance for light-water-cooled nuclear power reactors.

Up to recently, much of this concern about power reactor effluents has been narrowly focused on their absolute amounts, and on the degree of risk occasioned thereby. Among the first reviews of past experience to establish patterns and trends of effluent releases was one by Blomeke and Harrington⁴ covering the years up to 1967, and one covering the same period by Brinck and Kahn⁵. Starting in 1969, the Division of Compliance of the AEC (now the Division of Regulatory Operations) published yearly summaries of power reactor effluents, itemized by the categories of noble and activation gases, halogens and particulates (with a half-life greater than 8 days), mixed fission and corrosion products (MFP) and tritium in liquid effluents. It was qualitatively apparent from these data that the overall amounts of power reactor effluents were increasing; that the gaseous and halogen releases from the boiling water reactor (BWR) type were larger than those from the pressurized water reactor (PWR) type; that their fission and corrosion product releases were rather comparable, and that tritium in liquid effluents from PWR's exceeded that from BWR's.

Since it is related to the meeting of projected electrical energy demand, a reasonable debate about the risks occasioned by nuclear power plant effluents should also include a comparative consideration of that occasioned by conventional fossil-fueled plants. One of the first such comparisons was made in 1967 by Terrill et al.⁶. They set forth discharge quantities per year - MW(e) of SO₂, NO₂, radioactive ²²⁶Ra and ²²⁸Ra in fly ash from coal, oil and gas-fueled plants, and also of radioactive noble gases and ¹³¹I from nuclear plants. Using these data with AEC concentration standards for radioactivity or recommended concentration standards for conventional agents, they calculated a yearly volume of air required for dilution [m³/MW(e)]. They utilized only a limited amount of PWR effluent release data available to them at that time, as well as concentration standards for conventional pollutants that have since been superseded by much lower ones promulgated⁷ by the U. S. Environmental Protection Agency (EPA). However, they showed that the amounts of air required for dilution of the yearly amounts of radioactivity, as well as the conventional pollutants emitted from fossil-fueled power plants, were much greater than those required to dilute the radioactivity emitted from PWR's.

That these conclusions might not apply to the same degree to BWR effluents was suggested in 1969 by Fish⁸, who also suggested that the air quality criteria for SO₂ utilized by Terrill et al. were higher than more recently

adopted ones. In a further consideration along these same lines, Hull⁹ utilized the average of all releases from nuclear power plants for 1969, as well as more conservative air quality criteria for non-radioactive agents. His conclusions were in essential agreement with the earlier comparisons, but also showed that both coal and oil plants required larger dilution volumes than BWR's. A similar, but somewhat more sophisticated comparison, which considered the residence time of airborne pollutants, was made in 1970 for coal and nuclear-fueled plants in West Germany by Jansen et al.¹⁰, who arrived at a similar conclusion.

In the absence of a much needed biological effect related unit for conventional pollutants analogous to the man-rem, these comparisons to concentration standards appear to offer the most objective basis available for weighing the relative risks of the presently available choices for producing electrical power. However, in a recent comparison along this same line, Starr et al.¹¹ observed that the air quality standards for conventional pollutants are much closer to concentrations at which prompt medical effects are perceivable, than are those for radioactivity. In the absence of well controlled studies of the possible effects of long-term exposures to low levels of conventional pollutants, if a linear dose-effect relationship is applied their effects would be greater than those suggested from considerations based on air quality standards per se.

In what follows, the previous comparisons⁹ are updated to include a consideration of how the trends in effluent release rates have been affected by the larger "second generation" light-water-cooled nuclear power plants, with capacities in the range of 500-800 MW(e), some eighteen of which have been put into operation from 1967 on, in addition to those operating at that time. It also incorporates recently adopted EPA Air Quality Standards as the basis for comparison.

Reactor Effluent Releases

In order to provide as consistent as possible a basis for evaluating the trend of releases from nuclear power reactors, it is desirable that they be separated by type, and that these releases be normalized to the integrated quantity of electricity generated, rather than on plant capacity. Since the reactor effluent release data summaries by the AEC have not until 1971 included the latter data, it is not clear that previous comparisons have been made on this basis. For 1967 and 1968 the AEC reactor effluent data utilized herein was published in 1969 by the Joint Committee on Atomic Energy¹². For the years previous to 1971, the amounts of electricity generated by nuclear reactors was obtained from a 1971 AEC report¹³ on the operating history of U.S. nuclear power reactors. Since it was available, the data for one relatively small high temperature gas-cooled reactor was also included.

The effluent release data reported by the AEC have been set forth in their four major categories, as indicated above. Starting in 1972, supplementary information on individual nuclides within these categories was also included in the AEC report. Although such data would permit a more precise evaluation of the radiological significance of reactor effluent releases, since it has not been available over the period of interest, it has not been included in this study.

Yearly overall average amounts of gaseous, halogen and particulate, liquid fission and corrosion products, and tritium activity from 1967 to 1972 have been calculated in curies per 10^3 megawatt hours(e). These have been obtained by dividing the total of each reported reactor effluent category by the total electrical power generated by the various reactor types.

The nuclear power reactors operated between 1967 and 1972, with their net electrical power capacities, are shown in Table I. Although omitted from the detailed presentations, the releases from the two reactors which ceased operation during the study period have been included. Shippingport was not included since release data from it was not included in the AEC reports.

Table I
U.S. Nuclear Power Reactors Operated Between 1967 and 1972⁽¹⁴⁾

<u>Name</u>	<u>Type</u>	<u>Power</u>		
		<u>Net MW(e)</u>	<u>Start Up</u>	<u>Shut Down</u>
Shippingport	PWR	90.0	1957	-
Dresden I	BWR	200	1959	-
Yankee	PWR	175	1960	-
Big Rock	BWR	70.3	1962	-
Elk River	BWR	22.0	1962	1968
Indian Point I	PWR	265	1962	-
Saxton	PWR	3.0	1962	1972
Humboldt	BWR	68.5	1963	-
Peach Bottom	HTGR	40.0	1966	-
San Onofre	PWR	430	1967	-
La Crosse	BWR	53.2	1967	-
Connecticut Yankee	PWR	575	1967	-
Oyster Creek	BWR	640	1969	-
Nine Mile	BWR	625	1969	-
R. E. Ginna	PWR	420	1969	-
Dresden II	BWR	800	1970	-
Millstone I	BWR	652	1970	-
H. B. Robinson	PWR	700	1970	-
Monticello	BWR	545	1970	-
Point Beach	PWR	497	1970	-
Palisades	PWR	700	1971	-
Dresden III	BWR	800	1971	-
Quad Cities I	BWR	800	1971	-
Vermont Yankee	BWR	514	1972	-
Quad Cities II	BWR	800	1972	-
Point Beach	PWR	497	1972	-
Surry I	PWR	788	1972	-

The trend of the gaseous effluent release rate is shown in Figure 1. It is apparent that the average release rate from BWR's has continued to exceed that of PWR's, but that factor has decreased from about 1,000 to closer to 100 between 1967 and 1972. An examination of the individual data, shown in Figures 1A for BWR's and 1B for PWR's, discloses that this is attributable both to the lower "second generation" BWR release rates, which have averaged about 1/10 of the pre-1967 BWR's, and to an increasing trend with time from several of the older PWR's.

A similar pattern for halogens and particulates with a half-life greater than 8 days is evident from Figure 2, except that the average release rates from BWR's have also increased over the period from 1967 to 1972. The release data for individual reactors, shown in Figures 2A and 2B, indicate that although the spread is not large, this is principally due to increases over this period from the older BWR's and that most of the "second generation" BWR release rates have been somewhat less than the overall average. The pattern for individual PWR's is less obvious, but most of the increase in the overall average is again attributable to older plants.

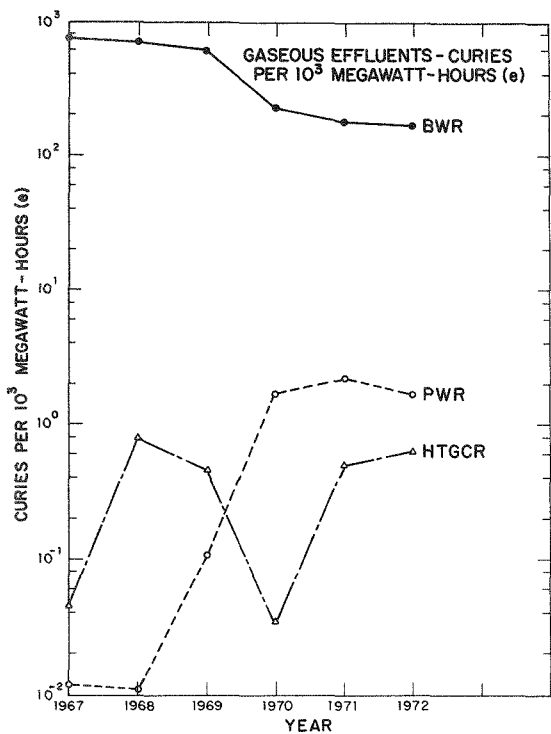


Fig. 1

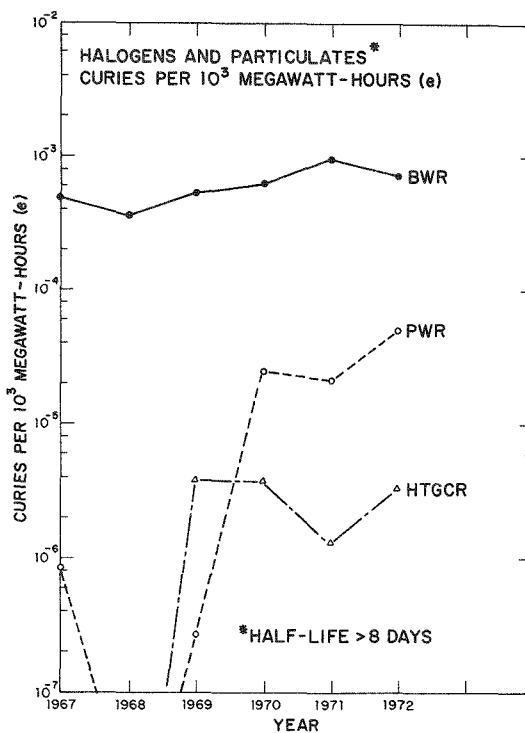


Fig. 2

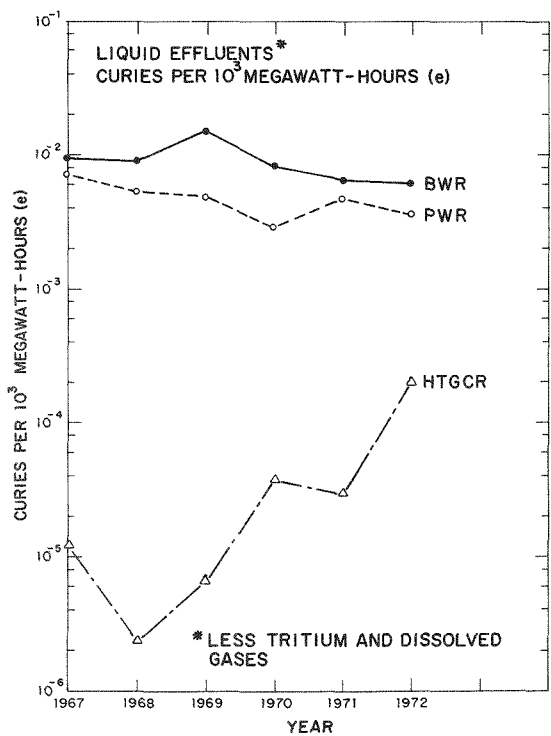


Fig. 3

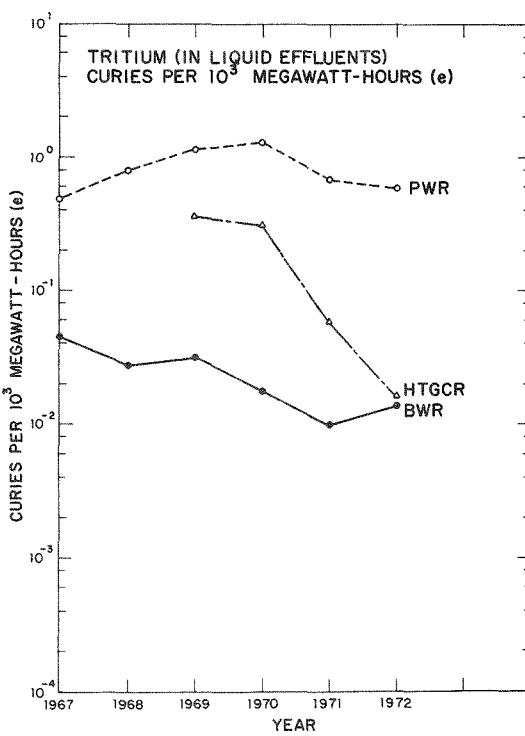


Fig. 4

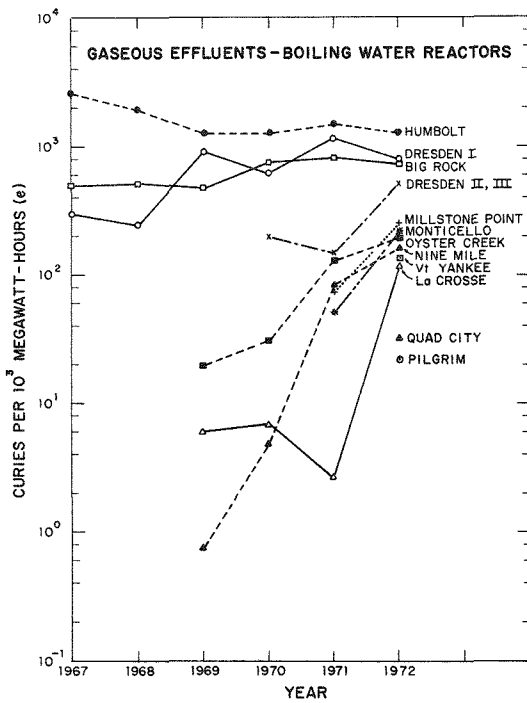


Fig. 1a

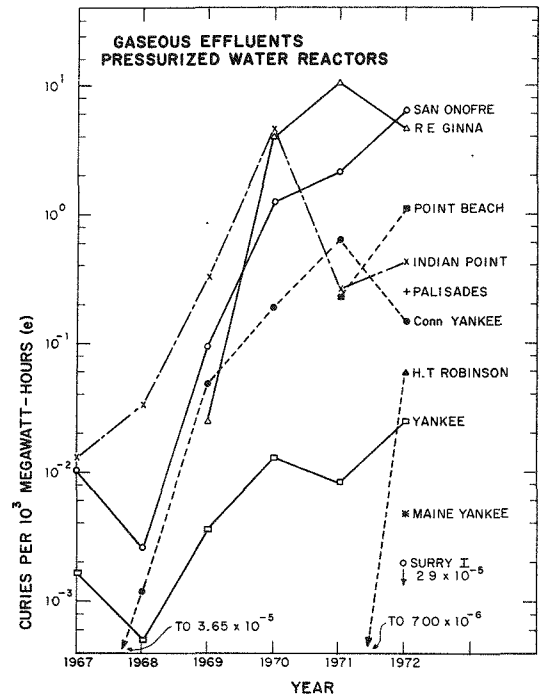


Fig. 1b

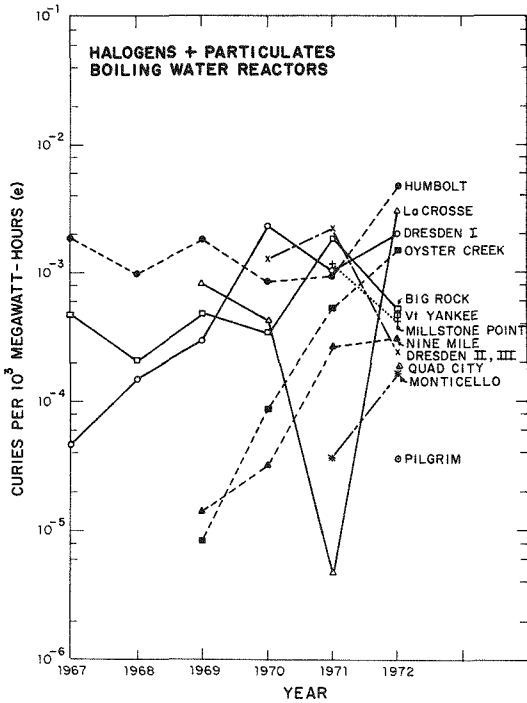


Fig. 2a

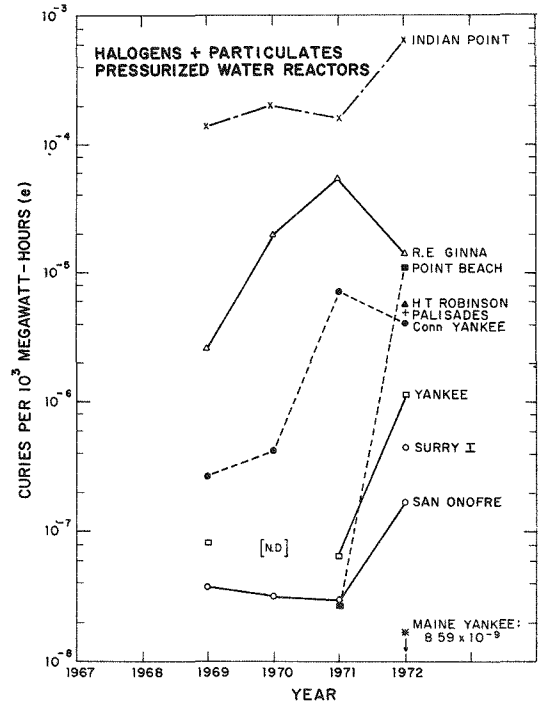


Fig. 2b

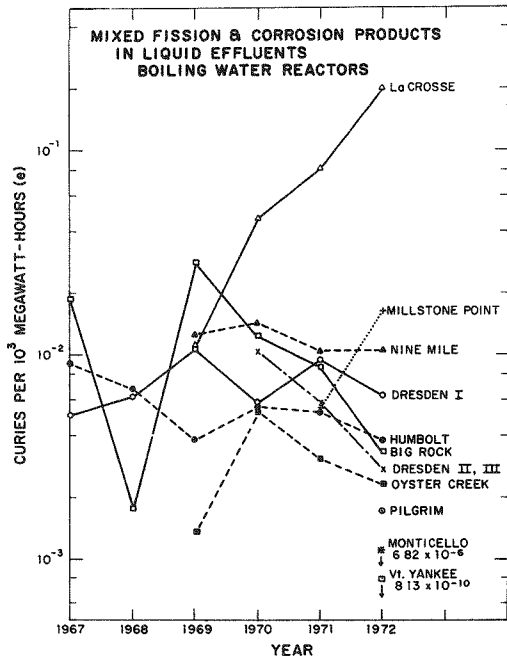


Fig. 3a

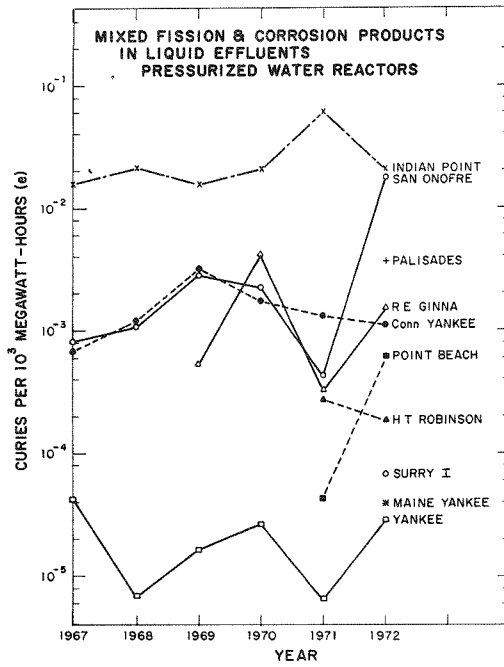


Fig. 3b

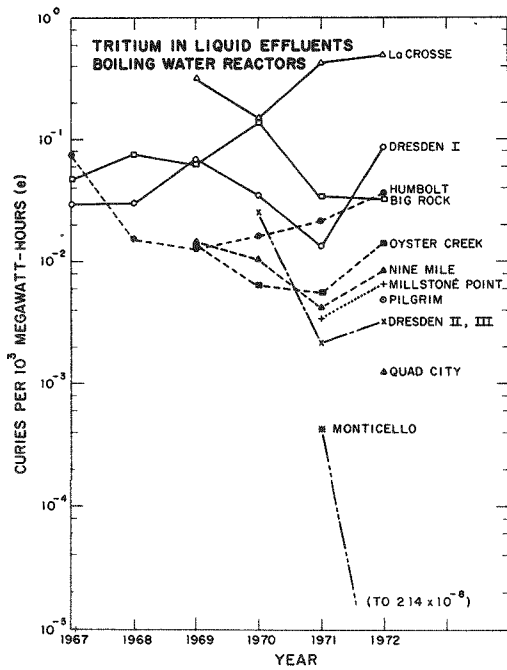


Fig. 4a

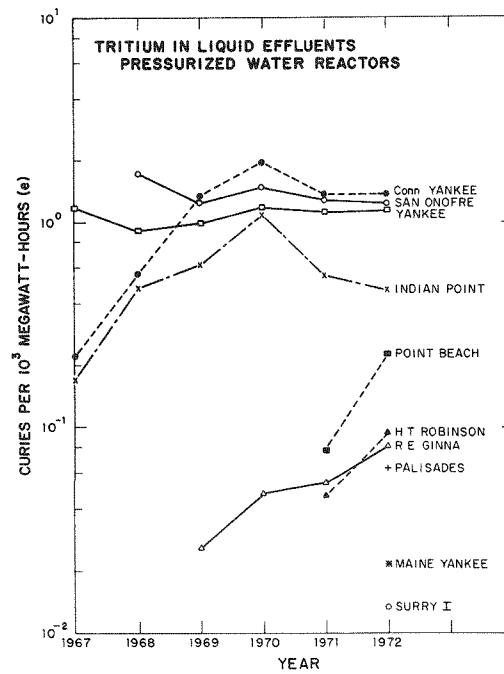


Fig. 4b

As shown in Figure 3, the average mixed fission and corrosion product activity release rates from BWR's have consistently been somewhat larger than those from PWR's. Both have declined somewhat over the period under consideration. While the relative influences of older and "second generation" reactors are not altogether consistent, the individual reactor release rate data shown in Figures 3A and 3B suggest that for both the BWR's and PWR's the release rates from the "second generation" reactors have generally been somewhat less than from the older plants.

From Figure 4, it is obvious that the relative overall release rates of tritium in liquid effluents from BWR's and PWR's has been the reverse of the situation for gaseous effluents. Although rates for both reactor types have declined in recent years, those from PWR's have been about one hundred times the rates from BWR's. In both cases the individual release data shown in Figures 4A and 4B indicate that most of the reduction in the overall averages is attributable to the lower release rates from "second generation" plants.

With appropriate caution that, as suggested by the increasing trend of many of the release rates from older plants, the performance of the newer "second generation" plants might be expected to deteriorate somewhat in this regard over future years, these data appear useful as a means of projecting the anticipated yearly effluent releases from a "reference" 1,000 MW(e) capacity nuclear power reactor, available for 80% of the time as a base load plant. These projections are:

Table II

Projected Effluent Releases of Radioactivity in the Effluents of "Reference" 1,000 MW(e) Power Reactors

	AIRBORNE EFFLUENTS (Curies/year)		LIQUID EFFLUENTS (Curies/year)	
	<u>Gaseous</u>	<u>Halogens & Particulates</u>	<u>Fission and Corrosion Products</u>	<u>Tritium</u>
BWR	1.66x10 ⁶	5.31	49.6	104
PWR	9,650	0.17	30.2	5,750
HTGR	2,760	< 0.02	0.27	835

The concern of this paper is with the relationship of effluents from nuclear power reactors and fossil-fueled plants to environmental standards in their immediate vicinities. However, it should be noted that the operation of a nuclear power reactor also results in the release of radioactivity to the environment when its fuel is reprocessed. From data given in Ref. 2, Vol. I, pp. 1711-31, for effluents released between 1966 and 1971 from the one commercial fuel reprocessing plant in the United States, the amounts so released from the reprocessed fuel of the above indicated 1,000 MW(e) capacity power reactor would be as follows:

Table III

Projected Effluent Releases of Radioactivity in the Effluents of a Plant Reprocessing Fuel with a Total Exposure of 8.72 x 10⁵ Mwd

AIRBORNE EFFLUENTS (Ci/yr)		LIQUID EFFLUENTS (Ci/yr)	
<u>Gaseous</u>	<u>Halogens & Particulates</u>	<u>Fission and Corrosion Products</u>	<u>Tritium</u>
2.68x10 ⁵ *	0.40**	53.5***	4,830

* ⁸⁵Kr - 100%; ** ¹³¹I - < 0.07 Ci; *** ⁹⁰Sr - 8.3 Ci.

It is also possible to project the release rates from a nuclear power reactor from theoretical considerations, starting with assumed fuel leakage rates, transfer coefficients from the primary to other systems, and the eventual release from the reactor to the environment. Such an approach has been utilized by the AEC in its Environmental Statement¹⁵ concerning the rulemaking action in connection with its proposed numerical guidance for light-water-cooled nuclear power reactors. For 3,500 MW(t) plants with effluent control equipment similar to that employed in currently operating power reactors, their "base case" projections are as follows:

Table IV

AEC "Source Term" Projections of Radioactivity in the Effluents from Base Case 3,500 MW(t) Light-Water-Cooled Nuclear Power Reactors

	AIRBORNE EFFLUENTS (Ci/yr)		LIQUID EFFLUENTS (Ci/yr)	
	<u>Gaseous</u>	<u>¹³¹I</u>	<u>Fission and Corrosion Products</u>	<u>Tritium</u>
BWR	2.9x10 ⁶	15	1,800	20
PWR	4.6x10 ⁴	1.2	210	350

It appears that for all categories but tritium, the AEC assumptions and calculational method lead to projected releases in excess of those predicated on experience to date. Other aspects of the overall reactor-environmental model utilized by the AEC in framing its proposals to limit radioactivity in light-water-cooled reactor effluents have been discussed elsewhere³. The influence of these proposals, whether or not they are finally adopted in their present form, seems likely to considerably diminish the release rates of most categories of nuclear power reactor effluents in the United States, thus making projections based on experience to date upper limits of what may be anticipated.

Comparison With Fossil-Fueled Plant Effluents

When the relationship of conventional pollutants from fossil-fueled power plants to air quality standards is compared to that between airborne radioactive effluents from nuclear power stations and radiation concentration guides, the much greater public concern which the latter has evoked in recent years seems difficult to comprehend. The nature of these relationships is suggested by a straightforward method of calculating the volume of air required to dilute the yearly amounts of various kinds of pollutants or radioactivity emitted from a "reference" 1,000 MW(e) capacity plant to the currently applicable EPA air quality standards or AEC radiation concentration guides. If it is further assumed that the same meteorological considerations, whatever these may be, are applicable, then these need not be specified in making such a comparison.

In the current calculation, the following heat values of fuels have been utilized:

- Bituminous Coal - 26,200 BTU/ton
- Crude Oil - 5,800,000 BTU/barrel
- Natural Gas - 1,035 BTU/ft³

Emission factors for various fossil fuels have been obtained from a U.S. Office of Science and Technology report¹⁶. The amounts of radioactivity in coal and oil originally indicated by Eisenbud and Petrow¹⁷ have been slightly modified, considering more recent data reported¹⁸ in 1968. The results are as follows:

Table V

Volume of Air Required to Meet Concentration Standards
and Average Site Boundary Concentrations for Yearly Emission
from a 1,000 Megawatt(e) Power Station
(Operated as a Base-Load Plant, with an 80% Availability)

Type Plant	Pollutant	Standard (A)	Discharge Quantity (B)	Dilution Volume (10 ⁹ m ³)	Site Bndry (C) Concentration
COAL	SO ₂ (3.5% S)	0.03 ppm	3.66x10 ⁸ lb.	2.14x10 ⁶	0.20 ppm
	NO ₂	0.05 ppm	5.50x10 ⁷ lb.	2.49x10 ⁵	0.04 ppm
	CO	9.0 ppm*	1.38x10 ⁹ lb.	63.5	0.02 ppm
	Hydrocarbons	0.24 ppm**	5.50x10 ⁵ lb.	156	0.001 ppm
	Particulates (97.5% Removal)	75 µg/m ³	1.25x10 ⁷ lb.	75,500	18 µg/m ³
	²²⁶ Ra	2x10 ⁻¹² Ci/m ³	0.0170 Ci	8.5	4.2x10 ⁻¹⁶ Ci/m ³
	²²⁸ Th	2x10 ⁻¹³ Ci/m ³	0.108 Ci	708	2.6x10 ⁻¹⁶ Ci/m ³
OIL	SO ₂ (1.5% S)	0.03 ppm	1.23x10 ⁸ lb.	6.95x10 ⁵	0.07 ppm
	NO ₂	0.05 ppm	5.42x10 ⁷ lb.	2.45x10 ⁵	0.04 ppm
	CO	9.0 ppm*	2.08x10 ⁴ lb.	0.95	2.61x10 ⁻⁴ ppm
	Hydrocarbons	0.24 ppm**	1.17x10 ⁶ lb.	4,720	0.004 ppm
	Particulates (97.5% Removal)	75 µg/m ³	5.88x10 ⁶ lb.	35,400	8.4 µg/m ³
	²²⁶ Ra	2x10 ⁻¹² Ci/m ³	6.0x10 ⁻⁴ Ci	0.3	1.5x10 ⁻¹⁸ pCi/m ³
	²²⁸ Th	2x10 ⁻¹³ Ci/m ³	1.3x10 ⁻³ Ci	6.7	3.2x10 ⁻¹⁸ pCi/m ³
GAS	SO ₂	0.03 ppm	2.78x10 ⁴ lb.	157	1.5x10 ⁻⁵ ppm
	NO ₂	0.05 ppm	2.71x10 ⁷ lb.	1.23x10 ⁵	0.02 ppm
	Particulates (97.5% Removal)	75 µg/m ³	1.04x10 ⁶ lb.	6,290	1.5 µg/m ³
NUCLEAR	⁸⁵ Kr & ¹³³ Xe	3x10 ⁻⁷ Ci/m ³	9,650 Ci	33.2 PWR	2.3x10 ⁻¹¹ Ci/m ³
	Short-lived radioactive gases	3x10 ⁻⁸ Ci/m ³	1.66x10 ⁶ Ci	5.54x10 ⁴ BWR	4.0x10 ⁻⁹ Ci/m ³
	¹³¹ I (Inhalation)	1.0x10 ⁻¹⁰ „	{ 0.2 Ci 5.3 Ci	{ 2.0 PWR 53.0 BWR	{ 4.2x10 ⁻¹⁶ Ci/m ³ 1.3x10 ⁻¹⁴ Ci/m ³
	¹³¹ I (Air-Grass-Milk)	1.4x10 ⁻¹³ „ ***	{ 0.2 Ci 5.3 Ci	{ 1,430 PWR 37,800 BWR	{ 4.2x10 ⁻¹⁶ Ci/m ³ 1.3x10 ⁻¹⁴ Ci/m ³

* Maximum 8-hour concentration, once per year. Yearly average not specified.

** Maximum 3-hour concentration (6-9 A.M.) once per year.

*** "Concentration factor" of 700 applied to inhalation standard for ¹³¹I.

(A) EPA "National Primary and Secondary Air Standards", (Federal Register, Vol. 36 No. 84, Part II, pp. 8186-87, 4/30/71), and AEC "Standards for Protection Against Radiation", 10CFR20.

(B) Discharges from PWR and BWR are derived from weighted average 1967-1972 release data as summarized by the Directorate of Regulatory Operations, USAEC.

(C) Based on average X/Q at 500 m (for release height of 100 m) of 6.2x10⁻⁸ sec/m³, for 25 operational or proposed nuclear power stations (Table 7.10, Ref. 15).

If the EPA's Air Quality Standards are given the same weights that the AEC's radiation protection standards have been, then the significant contribution of nuclear fueled plants to limiting overall air pollution (or at least minimizing its increase) is obvious. This is especially so relative to coal and oil-fired power plants.

It is of interest to examine the consequences, were the air quality standards applied to a hypothetical individual at the "fence post" at the boundary of conventional fueled plants, as is the practice for nuclear power reactors. The results of such a calculation of average concentrations of conventional pollutants and of radioactivity at 500 meters distance from a 100 meter stack of the "reference" 1,000 MW(e) base load plants, are also indicated in Table V. Unless limited to low sulfur fuels, the reference coal and oil fueled plants would exceed the SO₂ concentration standard. All conventionally fueled plants would approach the NO₂ concentration standard. The reference BWR would approach the gaseous effluent and ¹³¹I radiation concentration standards. However, it should be noted in this connection that the current design provision of catalytic recombiners, which reduce the volume of the BWR off-gas, thereby permitting longer holdup and/or charcoal filtration prior to stack discharge, should materially reduce both of these components of their airborne effluent releases.

The foregoing comparisons admittedly omit the airborne effluent releases from fuel reprocessing plants. However, it appears that these will continue to be located at a relatively few sites more remote from surrounding populations than most future nuclear power reactor sites. If so, the "local" effects of their effluent releases appear less important on an integrated population exposure basis than those from nuclear power plants.

Data for similar comparisons of conventional radioactivity and pollutants in liquid effluents is not available. However, studies(19,20,21) to date in the vicinity of several nuclear power reactors suggest that their radioactive liquid effluents have been dosimetrically insignificant. Comparable studies of the degree of pollution of streams and ground water by releases peculiar to fossil-fueled plants have not appeared in the literature, so it also appears to be minimal.

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ENVIRONMENTAL TRITIUM AND THE DOSE TO MAN

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Abstract

The dose to man from acute or chronic intake of tritiated water³ has been determined using a three compartment model to describe the retention of tritium in loose water and in bound organic form in the body. The retention times determined from occupational exposure cases are reviewed. The total tissue dose, using representative retention half-times for the three components of 9, 30, and 450 days, is 84 mrad per 1 mCi intake by standard man, 84% of which is due to tritium in body water and 16% to bound tritium in tissue. The record of environmental tritium concentrations in surface waters has been compiled. Maximum levels caused by weapons testing occurred in 1963-64. Subsequent decrease is exponential with a half-time of 3.2 years. By relating environmental tritium levels to daily intake by man and applying the dose model, the dose commitment of 1.5 mrad from fallout tritium was determined.

Introduction

Tritium is produced naturally by cosmic ray interactions in the atmosphere. Superimposed on the natural tritium background are varying amounts of man-made tritium. Nuclear weapons testing activity introduced substantial amounts of tritium into the environment, although the levels of fallout tritium are currently declining. The expanding nuclear power industry is expected to cause increasing amounts of tritium to be released. These continual additions of man-made tritium to the environment are cause for repeated monitoring of environmental levels of tritium and for re-evaluation of the dose consequences.

In this study a detailed tritium dose model is presented which allows one to determine accurately the contribution to tissue dose from tritium in loose water in tissue and from tritium combined in tissue following acute and chronic intakes. The experience from occupational exposure cases is reviewed to indicate the average half-times of the retention components. A record of tritium concentrations in environmental surface water is compiled. By relating these levels to the daily tritium intake by man and applying the dose model, the dose commitment from weapons-produced tritium has been computed.

Tritium Retention in Man

The experience from observations of human cases of accidental tritium exposure is summarized in Table 1. The initial rate of elimination of tritium from the body is exponential with a half-time ranging from 4 to 18 days.¹⁻⁸ Identification of additional retention components is limited to the few cases reported with intakes large enough to allow relatively long-term monitoring of tritium excretion. In the 415 day observation of a case, Sanders and Reinig⁹ identified three retention components of half-times 6.1, 23, and 344 days. The first component can be associated with retention of free water in the body and the other two components with bound tritium in tissue. It should be noted, that a diuretic was administered in this case from the 3rd to 35th day following the exposure, which undoubtedly increased the turnover rate of body water.

TABLE 1
TRITIUM RETENTION HALF-TIMES IN MAN

Year	Investigator	Cases Studied	Notes	T ₁	T ₂	T ₃
1951	Pinson, Anderson ^{1,2}	9	range 9.3-13	11.3		
1957	Fallot <i>et al.</i> ³	20	range 5-11	8.5		
1960	Foy, Schnieden ⁴	10	high ambient temp.	7.5		
1962	Richmond <i>et al.</i> ⁵	5		9.5		
1963	Wylie <i>et al.</i> ⁶	7	range 6-12	8.5		
1965	Butler, Leroy ⁷	310	range 4-18	9.5		
1966	Osborne ⁸	30	range 6.4-14.4	10.5		
1968	Snyder <i>et al.</i> ¹⁰	1		8.7	34	
1968	Sanders, Reinig ⁹	1	diuretic used	6.1	23	344
1969	Minder ¹³	1			10-30	139-230
1971	Lambert <i>et al.</i> ¹²	1		9.1	36	
1972	Moghissi ¹⁴	3			21-26	280-550
Reasonable Range:				8.5-11	20-36	200-550
Assumed Average:				9	30	450

Snyder *et al.*¹⁰ studied another case for 255 days and identified two retention components of half-times 8.7 and 34 days. No particular treatment procedure was applied to this individual following exposure. Further analysis of this data has shown that the data are not inconsistent with a fit by three components of half-times 8.7, 30, and 550 days.¹¹

Another case was recently reported by Lambert, Sharpe, and Dawson.¹² Retention half-times of 9.1 and 36 days are very similar to the results reported by Snyder *et al.* The observation period, 161 days, is not quite long enough to allow a definitive fit with three retention components.

Approximate long-term retention components have been inferred from a few additional studies of occupational exposures.^{13,14} The differences in retention times in the cases reported allude to the variability which can be anticipated due to differences in metabolism, age, water intake, ambient temperature and treatment procedures. Reasonable ranges of the retention components have been indicated in Table 1. Approximate average retention half-times of 9, 30, and 450 days have been assumed for the model calculations.

Tritium Dose Model

Acute Intake

The dose to tissue following an acute intake of tritium arises from tritium in body water and from bound tritium in tissue. In computing the dose to tissue, the combined tritium in tissue is often neglected, it being regarded as an insignificant contributor to the total dose. Alternatively, the dose to body water (or a tissue with 100% water) is computed, this dose being a conservative estimate of the total tissue dose.¹⁵ Since the tissue dose is more a measure of the hazard than the water dose, it is useful to determine accurately the dose to tissue.

A three compartment model is necessary to reflect the three retention components that have been identified. Sanders and Reinig⁹ suggested the model represented by the diagram in Figure 1. A is the body water compartment, and B and C are bound hydrogen (tritium) compartments. The transfer coefficients represents constant fractional exchange rates of the compartment hydrogen (tritium) contents.

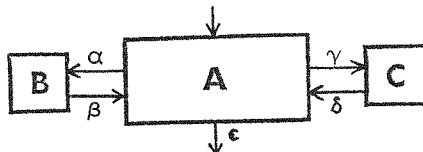


Figure 1. Three compartment model of tritium in the body.

A Markov chain calculation is used to determine the tritium concentrations in the three separate compartments at various times after intake. The fraction of assimilated

tritium in each compartment, f_A , f_B , f_C , are computed for as many iterations as may be required (each iteration representing one day). In analyzing the occupational exposure cases, combinations of exchange and elimination rates were selected so that close agreement was obtained with the measured fractional assimilated tritium in the body water.^{9,11} For other calculations the transfer coefficients correspond to assumed retention half-times and compartment sizes.

The cumulative dose to tissue from an acute intake of HTO is computed from this model as follows:

$$\text{Dose (wet tissue)} = .75 \int_0^{\infty} q_A f_A dt + .25 \int_0^{\infty} (q_B f_B + q_C f_C) dt$$

The first term is the dose to tissue due to tritium in loose water within tissue. The second term is the contribution to tissue dose from tritium combined in tissue. Wet tissue is assumed to consist of 75% water and 25% tissue solids. A 70 kg man contains an average 42 kg water and an estimated 10 kg active tissue solids (70 kg - 42 kg water - 13 kg fat - 5 kg mineral bone).¹⁶ The specific activities of tritium in the free water and bound compartments following an intake, I (mCi), are $q_A = I/42$ mCi/kg and $q_{B,C} = I/10$ mCi/kg times the fractional amount of the intake in these compartments, f_A , f_B and f_C , respectively. The relationship .29 mrad per mCi day/kg is used to convert the units.

A calculation based on average retention half-times of 9, 30, and 450 days results in an estimated total tissue dose of 84 mrad per mCi intake.¹¹ This dose is due 84% to tritium in water within the tissue and 16% to combined tritium in tissue. Fifty percent of the total tissue dose is delivered within 11 days, and 90% of the dose is delivered within 200 days. The bound hydrogen compartment sizes were inferred to be 120 g for B and 600 g for C, consistent with the total amount of tissue solids (10 kg) consisting of ~7% hydrogen. A maximum of 0.7% of the tritium intake becomes combined, this maximum being reached in about 20 days. The rapidly declining concentration of tritium in the body water allows only relatively small amounts of the intake to become combined. The bound tritium in tissue is, thus, not the major contributor to the total dose to tissue following a single intake of HTO, but its contribution (16%) is not insignificant.

In additional model calculations with the first component half-time T_1 varying from 6 to 12 days with 30 and 450 day combined components, an empirical relationship is obtained.

$$\text{Tissue Dose} = 9 T_1 + 3 \text{ mrad per mCi intake}$$

The relationship reflects a direct dependence of the amounts of bound tritium on the amount of tritium in the source reservoir, the body water compartment. For the assumed compartment sizes and within reasonable variation of transfer rates from the bound compartments, the second and third component half-times are not required in this relationship. The formula should be generally useful in providing an approximate estimate of the tissue dose following observation of the initial removal rate.

Intake of tritium other than in the form HTO may require special consideration. Bound tritium compounds in food may be more directly assimilated into bound compounds in tissue. Further study will be required to give the significance of the form of the tritium intake.

Chronic Intake

The model can be used for chronic intake situations by computing the compartment contents on a daily basis. For chronic intake of $1 \mu\text{Ci}/\ell$, the equilibrium dose rate to active wet tissue is 95 mrad/yr. The dose to body water is

$$.001 \frac{\text{mCi}}{\text{kg}} \times .29 \frac{\text{rad/d}}{\text{mCi/kg}} \times 365 \frac{\text{d}}{\text{yr}} = 106 \frac{\text{mrad}}{\text{yr}}$$

The dose to tissue containing 75% water is $106 \times .75 = 80$ mrad/yr, neglecting the dose due to tritium combined in tissue solids. The dose to active tissue is thus 1.2 times the dose due to HTO in tissue.

The dose model assumes that the tritium becomes uniformly combined in actively

metabolizing tissue and that all of the hydrogen of active tissue solids is exchangeable. These assumptions lead to conservative estimates of the dose.

In the model calculations, a water balance of 3.08 l/d has been assumed, based on the 42 kg body water compartment size and transfer coefficients corresponding to the 9, 30, and 450 day retention half-times. For an HTO intake of 1 mCi/d, the equilibrium specific activity is

$$1 \text{ mCi/d} \div \left[3.08 \frac{\text{kg H}_2\text{O}}{\text{d}} \times \frac{1 \text{ kg H}}{9 \text{ kg H}_2\text{O}} \right] = 2.92 \text{ mCi/kg H}$$

The tritium content of each compartment at equilibrium (the hydrogen content times the specific activity) is 13.6, .35, and 1.75 mCi for A, B, and C, respectively. Radioactive decay allows the long half-time compartment, C, to reach only about 90% of the intake specific activity. The equilibrium dose rate to wet tissue is

$$\begin{aligned} \text{A} \quad & \frac{13.6 \text{ mCi}}{42 \text{ kg H}_2\text{O}} \times \frac{.75 \text{ kg H}_2\text{O}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 70 \frac{\text{mrad}}{\text{d}} \\ \text{B} \quad & \frac{.35 \text{ mCi}}{10 \text{ kg solids}} \times \frac{.25 \text{ kg solids}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 2.5 \frac{\text{mrad}}{\text{d}} \\ \text{C} \quad & \frac{1.75 \text{ mCi} \times .90}{10 \text{ kg solids}} \times \frac{.25 \text{ kg solids}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 11.4 \frac{\text{mrad}}{\text{d}} \\ & \text{Total:} \quad 84 \frac{\text{mrad}}{\text{d}} \end{aligned}$$

As expected, the dose commitment following a single intake (84 mrad per mCi intake) becomes the equilibrium dose rate for chronic intake (84 mrad/d per 1 mCi/d intake).

Fallout Tritium

Levels in Surface Waters

Tritium is produced by fusion bombs and also by neutrons released during fission bomb explosions and in small amounts by the fission process itself. The amount of bomb-produced tritium is uncertain. Eriksson estimated that 1900 MCi were produced from 1952 through 1962.¹⁷ This compares with natural tritium production of around 2 to 6 MCi/yr. Additional amounts of tritium have been released into the atmosphere by French and Chinese weapons testing conducted since 1964, but the total is a small fraction of that released during the earlier testing period.

The effect of weapons-produced tritium on concentrations of tritium in the environment, in drinking water, food and in man has not been monitored in great detail. Data from the U. S. Geological Survey's program of analyzing river water for tritium content provides the most useful data for ascertaining the tritium levels in environmental waters in the U. S. The data for 1961-68 for 20 streams throughout the U. S. have been published.¹⁸ Preliminary data for 15 rivers for 1969-70 have also been obtained.¹⁹ Tritium data from the earlier testing period, 1952-61, are not nearly as complete. Measurements of Mississippi River water for 1954-57^{20,21} and of Rio Grande River water for 1957-58²² have been included, and comparisons were made with Ottawa River data²³ in arriving at representative average tritium levels.²⁴

Figure 2 shows the average U. S. data. The concentration of tritium in environmental waters reflects the weapons testing activity. Sharp increases are indicated following the testing series in 1954, 1956 and 1958. The very active test period in late 1961 and 1962 caused the peak concentrations in U. S. rivers in 1963-64. The declines in concentration during the 1959-60 moratorium and after the 1963 Test Ban Treaty are evident. Recent high yield tests in the Northern Hemisphere by the Chinese may be responsible for the relatively higher tritium levels measured in 1969-70 as compared to the previously declining values.

The average U. S. river tritium concentrations declined with a half-time of 3.2 years from 1963 until 1969 and 5.0 years during 1969 and 1970. Tap water analyzed at the Health and Safety Laboratory in New York City reflecting the lower Hudson Valley watershed area, showed somewhat lower tritium concentrations than the U. S. average in 1970. More recent tap water samples would indicate that the U. S. average beyond 1970 resumes the 3.2 year half-time decline. The absence of high yield atmospheric tests in the Northern Hemisphere in the past two years would also support this assumption.

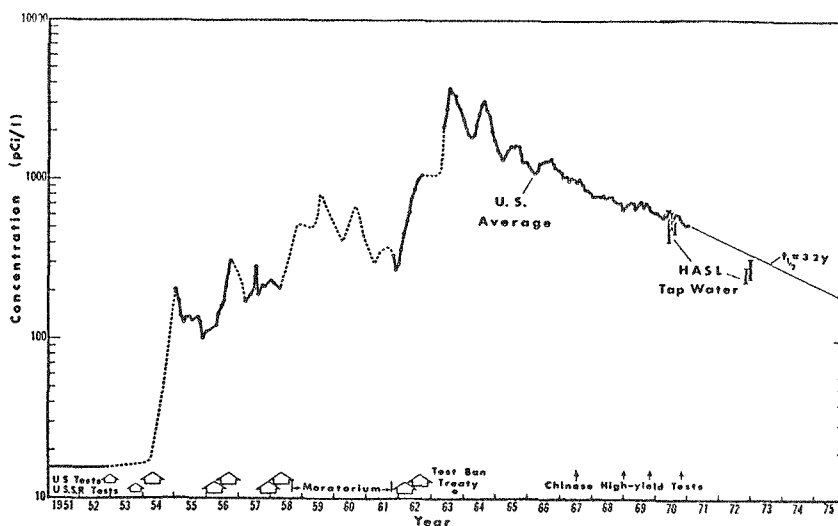


Figure 2. Environmental tritium in surface waters.

Tritium Intake

If a relationship can be found between the variations in tritium levels in environmental waters and the variations in the amounts of tritium intake by man or the levels of tritium in the body, the dose to man can be determined. The data of Bogen²⁵ show the relationship for New York in 1970 between the tritium levels of drinking water (530 pCi/l), loose water in food (860 pCi/l), oxidation water of food (2050 pCi/l), and water vapor in air (930 pCi/l). The drinking water intake by man can be assumed to be 1.4 l/d. Intake by inhalation and transpiration of water vapor is computed to be .13 l/d, based on average air temperature and humidity. The food water intake, based on analysis of food items of a standard consumption diet, was found to be 1.26 l/d loose water and .29 l/d oxidation water.²⁵ The effective concentration of the total tritium intake is thus:

$$(1.4 \times 530 + .13 \times 930 + 1.26 \times 860 + .29 \times 2050) \div 3.08 = 825 \text{ pCi/l}$$

These data indicate that, currently, one should assume somewhat higher concentrations of tritium intake than indicated by the tritium concentration in drinking water. During the earlier fallout period, when environmental levels of tritium were increasing, the tritium concentrations in water were probably higher than in food. Since specific data are lacking, it is probably most reasonable to assume that the tritium levels in surface waters reflect directly the tritium intake by man.

The Dose Commitment

The average natural concentration of tritium in environmental waters (16 pCi/l) results in a dose to man of 1.5 μ rad/yr, based on the three compartment dose model. The dose commitment to man from weapons-produced tritium is the dose due to the increases in tritium intake concentrations above the natural level. Extrapolation of the increased levels beyond 1972 is necessary to account for the exposure which is yet to be experienced. Assuming a 3.2 year half-time for weapons-produced tritium in the environment beyond 1970, a return to the natural background tritium level occurs in 1987.

The tritium intake is assumed to be 3.08 l/d times the effective tritium intake concentration (the average tritium concentration in surface waters). The dose model is used to determine the bound and loose tritium compartment contents and the dose for the entire period that the tritium levels are above the natural background. The dose commitment thus determined is 1.5 mrad. Details of the calculation are shown in Table 2. The highest annual dose due to weapons-produced tritium was .21 mrad in 1963 and 1964. The calculation assumes no further atmospheric weapons testing and takes no account of future tritium releases from nuclear facilities.

The ratios of specific activities (μ Ci ^3H per kg hydrogen) of the organic bound and

TABLE 2
TRITIUM TISSUE DOSE AND BOUND-LOOSE RATIO IN THE BODY

Year	Tissue Dose (mrad)		Bound-Loose Ratio	Year	Tissue Dose (mrad)		Bound-Loose Ratio	Year	Tissue Dose (mrad)		Bound-Loose Ratio
	Fallout	Total			Fallout	Total			Fallout	Total	
1952	0.0	1.5	.92	1965	144.8	146.3	1.21	1978	9.5	11.0	1.32
1953	0.1	1.6	.90	1966	122.0	123.5	1.17	1979	7.4	8.9	1.32
1954	3.8	5.3	.29	1967	97.0	98.5	1.35	1980	5.7	7.2	1.32
1955	10.5	12.0	.73	1968	76.4	77.9	1.28	1981	4.3	5.8	1.32
1956	16.4	17.9	.63	1969	68.4	69.9	1.21	1982	3.2	4.7	1.32
1957	18.4	19.9	.75	1970	58.1	59.6	1.21	1983	2.3	3.8	1.32
1958	30.3	31.8	.57	1971	47.4	48.9	1.26	1984	1.6	3.1	1.32
1959	53.1	54.6	.86	1972	38.0	39.5	1.28	1985	1.0	2.5	1.32
1960	47.2	48.7	1.12	1973	30.4	31.9	1.30	1986	0.5	2.0	1.32
1961	32.3	33.8	.95	1974	24.3	25.8	1.31	1987	0.2	1.7	1.21
1962	76.6	78.1	.61	1975	19.3	20.8	1.32	1988	0.1	1.6	1.08
1963	210.3	212.0	.73	1976	15.3	16.8	1.32	1989	0.1	1.6	1.01
1964	210.9	212.4	1.10	1977	12.1	13.6	1.32	1990	0.0	1.5	.97
									Dose Commitment		1.5 mrad

loose water tritium in the body, determined from this computation, are included in Table 2. The ratio would be 1.0 under natural conditions, except that radioactive decay does not allow complete equilibrium in the bound compartment with the slowest turnover time. As the concentrations of tritium intake increase, labeling of the bound components lag with respect to the specific activity of the loose water compartment. The bound-loose ratio is then less than one. During times of decreasing tritium intake, the bound tritium label is retained longer than the activity in body water, and the bound-loose tritium ratio becomes greater than one. The ratio is currently about 1.3 in man, according to this computation, which is not unlike the ratios actually being measured in human tissue.²⁶

Other Estimates of the Dose Commitment

Indirect estimates of the dose commitment can be made by comparing the total amounts of weapons-produced tritium with the natural production rate, as is done by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR).²⁷ The uncertainties involved, however, affect the accuracy of these estimates.

Most of the weapons-produced tritium, an estimated 1900 MCi,¹⁷ was introduced into the Northern Hemisphere. The stratospheric residence time of tritium (~1 yr) does not allow inter-hemispheric mixing to be an important factor. Ten percent may be a reasonable approximation for stratospheric tritium carried to the Southern Hemisphere.²⁸ The dose commitment for the Northern Hemisphere is consistent with the above direct computation if the natural production rate is taken as 3.4 MCi/yr (1.7 MCi/yr in each hemisphere). The natural background tritium dose to man is .0015 mrad/yr.

$$.0015 \text{ mrad/yr} \times \frac{1900 \text{ MCi} \times .90}{1.7 \text{ MCi/yr}} = 1.5 \text{ mrad}$$

UNSCEAR computed a range of possible dose commitments for the Northern Hemisphere, based on various assumptions of the natural tritium background and inter-hemispheric mixing of stratospheric weapons tritium. A conservative estimate for the Northern Hemisphere fallout tritium dose commitment of 4 mrad was obtained.²⁷

Tritium released from nuclear power facilities are as yet insignificant, compared to the amounts of weapons-produced tritium. It will be desirable, however, to continue to measure the environmental tritium levels and to ascertain the contribution from nuclear power activities. With use of the dose model presented here, the dose commitment from future activities involving release of tritium to the environment can be determined.

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RISQUE INDIVIDUEL, DOMMAGE COLLECTIF, ET CENTRALES NUCLEAIRES

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Abstract - The author specifies the way that leads from an individual risk to the theoretical collective damage in the matter of irradiation by nuclear power plants; in so doing he endeavours to show people's reactions towards nuclear power plants can be altered and on the other hand set free from any emotional loads. Finally he states the necessity of defining our concepts precisely and of giving them denominations that cause no anxiety.

1 - Introduction

Les termes "risque" et "dommage" foisonnent dans la littérature qui traite de radioprotection. De nombreux auteurs les emploient indifféremment; certains vont même jusqu'à désigner par risque, ou par dommage, les effets différés de l'irradiation... ou l'irradiation elle-même.

La personne (physique ou morale) qui crée un risque doit - sans contexte - réparation du dommage, s'il s'en produit. En ce qui concerne les centrales nucléaires le risque proviendrait, si l'on n'y prenait garde, des rayonnements ionisants. Le dommage s'exprimerait, par exemple, en cancers, en "morts génétiques". En matière de réparation, on rechercherait un lien de causalité entre le risque créé et le dommage incriminé, cas par cas, et a posteriori.

En 1955, à Genève, au cours de la conférence organisée par l'ONU sur les utilisations pacifiques de l'énergie nucléaire, le monde industriel et scientifique a cru - sans doute un peu prématurément - que cette forme d'énergie allait vite prendre le pas sur les autres formes, grâce aux centrales nucléaires.

L'hygiéniste, jusque là, observait l'évolution de l'état sanitaire de la population soumise à une pollution industrielle croissante; il suivait les résultats des recherches accomplies en vue de limiter cette pollution; au besoin, il engageait le législateur à prescrire des mesures de précaution. L'un et l'autre se résignaient: l'amélioration du niveau de vie et, partant, du bien-être et de l'état sanitaire, due au développement industriel, permettait de tolérer des inconvénients jugés raisonnablement supportables. Connaissant les dangers des rayonnements ionisants et, partant, d'un développement anarchique de l'industrie nucléaire, sous le couvert et sous l'autorité de la commission internationale de radioprotection (CIRP), il a émis des recommandations¹ suivant lesquelles le législateur a, préventivement, limité l'irradiation des personnes à un niveau jugé très prudent. Par la suite, n'ignorant pas l'importance des décisions prises, il a voulu prévoir quelles pourraient être les conséquences lointaines de cette irradiation. Ses prévisions, qu'il juge admissibles, alarment le profane.

Dans ce texte, de longueur limitée, destiné à souligner la nécessité de définitions, je me bornerai à reprendre des notions récemment exposées ailleurs²⁻⁴. Je vais seulement rappeler comment, pour asseoir ses prévisions, l'hygiéniste est passé de la notion de risque individuel à celle de dommage collectif; ce faisant, je mettrai en relief la nature toute théorique du risque qui sert de base au calcul de ce dommage, ainsi que la nécessité d'éviter les dénominations qui, sans raison, donnent prise à l'angoisse.

2 - Risque individuel

Préalablement, je proposerai une définition du risque individuel. La recherche de son expression quantitative se heurte à d'innombrables difficultés : l'hygiéniste a adopté une relation simplifiée, d'où découle un risque bien particulier. J'ai proposé de l'appeler : "risque individuel théorique", pour bien éviter de la confondre avec le précédent. Diverses dénominations utilisées, dont je donnerai quelques exemples, ne sont pas sans ambiguïté.

2.1 - Définition

D'une façon générale, on peut définir un risque individuel par la probabilité, pour une personne placée dans des circonstances particulières, de subir un préjudice du fait de ces circonstances.

Ici, les "circonstances particulières" sont dues à l'irradiation ajoutée par l'industrie nucléaire. Le préjudice se traduirait par des dommages corporels observables sur certaines des personnes irradiées (effets somatiques différés) ou seulement sur leur descendance (effets génétiques).

2.2 - Recherche d'une relation quantitative

Les affections susceptibles de résulter de l'irradiation ne lui étant pas spécifiques, un rapport de causalité entre risque individuel et irradiation ne peut être établi que statistiquement : on compare des groupes d'individus identiques, sauf pour ce qui concerne celle-ci. Dans chacun de ces groupes on détermine la proportion des personnes atteintes d'une affection déterminée; soit p sa valeur dans un groupe qui a été irradié, et p_0 sa valeur dans le groupe témoin, non irradié. La différence $(p - p_0)$ est fonction : du débit d'équivalent de dose (d), de l'équivalent de dose (D), du champ d'irradiation (f). La forme de la fonction dépend en outre de l'affection considérée (cancer, cause de mort prématurée, par exemple) de la population (âge, sexe, mode de vie, etc...); elle dépend aussi des conditions d'observation, laquelle débute au temps τ après le début de l'irradiation (suivant la valeur de τ , elle peut porter sur la population elle-même, sur sa progéniture, ou sur sa descendance lointaine) et se prolonge un temps t .

A condition d'être connus avant l'irradiation, ayant été déterminés sur des groupes semblables d'effectif suffisant, p et p_0 — et aussi leur différence — peuvent être assimilés à des probabilités. Pour en simplifier la détermination, le chercheur fixe les caractéristiques de l'irradiation (d , D et f), il arrête les conditions d'observation (τ et t), il précise les caractéristiques de la population observée, enfin, il choisit l'affection dont il recherchera le diagnostic. En irradiant plusieurs groupes, donnant à D autant de valeurs qu'il le juge utile, il peut, du moins en principe, pour un individu semblable à ceux qui constituent les groupes et pour l'affection considérée, traduire la variation du rapport de causalité graphiquement, et même mathématiquement, par une approche statistique, avec une relation de la forme :

$$(p - p_0) = \varphi(D). \quad (1)$$

Généralement trouvée positive pour des valeurs de D très élevées, la probabilité $(p - p_0)$ peut, lorsque D diminue : au mieux, s'annuler en changeant de signe, au pire, ne plus différer statistiquement de zéro. Cette dernière éventualité (due, soit à ce que l'irradiation n'a produit aucun effet traduit par l'affection recherchée, soit à ce que les effectifs des groupes étaient insuffisants) laisse le chercheur, et, partant, l'hygiéniste, dans l'incertitude.

Comme de fortes irradiations peuvent provoquer des affections nombreuses et comme, pour chaque affection, $(p - p_0)$, fonction de D, dépend de nombreux paramètres, pragmatique, la CIRP¹ a adopté la forme de relation quantitative la plus simple.

2.3 - La relation simplifiée adoptée

La CIRP a fait l'hypothèse que, pour toute affection que peut provoquer l'irradiation, la probabilité d'induction, indépendante de d, est proportionnelle à D. Si k représente la constante de proportionnalité, on peut écrire, pour l'affection considérée :

$$(p - p_0) = k D . \quad (2)$$

La valeur numérique de $(p - p_0)$ étant obtenue à partir de valeurs élevées de d et de D, il me paraît important, dans le domaine des faibles doses, d'écrire :

$$(\pi - \pi_0) = \chi D , \quad (3)$$

pour bien marquer, par des lettres différentes, l'hypothèse de la validité de la substitution de χ à k. Comme $(p - p_0)$, la valeur calculée $(\pi - \pi_0)$ est aussi dénommée "probabilité". Voyons de plus près.

2.4 - Dénomination proposée : risque individuel théorique

Dans l'expression (2) ci-dessus, la différence $(p - p_0)$ ne peut qu'être positive : cette variation de probabilité est donc un risque, un risque supplémentaire. On assimile $(\pi - \pi_0)$ au risque individuel ajouté par toute irradiation de D rems. Pour bien marquer que, dans le domaine des valeurs faibles de D, $(\pi - \pi_0)$ ne correspond à aucune observation ou expérimentation, je l'ai appelé : risque individuel théorique (RIT).

En pratique, l'observation porte sur des groupes d'individus qui diffèrent, mais que l'on recrute au hasard dans la population : la valeur $(p - p_0)$ qui en découle est une probabilité individuelle moyenne, donc $(\pi - \pi_0)$ est un RIT moyen.

Les prévisions ne valent que pour une population semblable à la population d'origine des groupes, que pour l'affection considérée, que pour les mêmes conditions d'observation. Le RIT est évalué, le plus souvent, soit pour la vie entière, soit en moyenne annuelle.

La constante χ est donc le RIT moyen supplémentaire de contracter l'affection considérée, par rem, soit pour la vie entière, soit ramené à l'année. Sa valeur est déterminée pour chacune des affections susceptibles d'être mises en cause (par effet somatique, par effet génétique).

Jusqu'ici, chacun pour ses besoins forge des dénominations particulières, cela ne va pas sans inconvénients pour interpréter les valeurs obtenues.

2.5 - Quelques dénominations usitées

On trouve des définitions éparses dans le rapport de 1964 du comité scientifique de l'ONU⁵ (UNSCEAR) et dans la publication 8 de la CIRP⁶. Le numérateur de la constante χ , non explicitée, est appelée le "risque estimé", par rem (ou par rad), par million de personnes, soit pour la vie entière, soit par an. En 1972 l'UNSCEAR⁷ l'appelle "coefficient de régression", ce qui a l'avantage d'en montrer la nature.

Dolphin et Marley⁸ donnent une expression que je peux traduire, avec mes notations :

$$\nu = \chi N D , \quad (4)$$

où $(\pi - \pi_0) = \nu/N$. Ils appellent χN le "coefficient de risque" qui, disent-ils, est exprimé habituellement en nombre de cas (pour l'affection considérée) par million d'hommerads ($ND = 10^6$), pour la vie entière.

Enfin, alors que la CIRP⁶ définit le "risque relatif" par le quotient : $(\pi - \pi_0)/p_0$, l'UNSCEAR⁷ le définit par : $(p - p_0)/p_0$.

A titre d'exemple, pour souligner, au besoin, la nécessité d'une codification de notre langue dans notre spécialité, je vais passer en revue les expressions usitées par une même école, celle d'Alice Stewart. Elle définit d'abord un "risque relatif" (1958)⁹, différent des précédents, puis un "risque absolu" (1970)¹⁰; enfin Kneale (1971)¹¹ a introduit un "risque relatif ajouté". Entre temps, la CIRP¹² avait rapporté des valeurs de la "proportion de cas attribuables à l'irradiation" (selon Stewart, 1968).

3 - Domage collectif

On doit, logiquement, appeler le dommage calculé à partir du RIT² : dommage collectif théorique (DCT). Il se déduit simplement de l'expression (3) :

$$(DCT) = \chi ND, \quad (5)$$

où N représente l'effectif de la population irradiée par l'industrie nucléaire; ou encore, si les membres de cette population sont différemment irradiés :

$$(DCT) = \chi (\Sigma D_i), \quad (6)$$

où ΣD_i est la somme des équivalents de dose individuels.

Cette dernière formule, tout à fait générale, s'applique aussi bien à la prévision d'effets somatiques différés qu'à celle d'effets génétiques à la première génération; des valeurs de χ ont été proposées⁶. Elles dépendent de l'effet somatique considéré. Une seule suffit pour l'ensemble des effets génétiques traduits, suivant H. J. Muller¹³, en "morts génétiques". Une formule semblable peut aussi s'appliquer aux générations lointaines : sous irradiation constante (D conserve la même valeur à chaque génération) la théorie montre qu'un nouvel équilibre finit par être atteint; les hypothèses de calcul supposent, soit des effets antagonistes de "pressions" de mutation (χ a même valeur à chaque génération) et de sélection, soit l'existence d'une "dose doublante". Quelques publications donnent des résultats de calcul de DCT à l'équilibre¹⁴, ou $(DCT)_\infty$; elles inquiètent.

4 - Discussion

Quel qu'il soit, un risque doit d'abord être bien défini, puis évalué par l'hygiéniste qui étudie, s'il y a lieu, à combien le réduire pour le rendre acceptable. C'est au législateur de prescrire les mesures nécessaires, compte tenu des considérations objectives de l'hygiéniste, et aussi, certainement, des réactions subjectives des intéressés.

En ce qui concerne l'énergie nucléaire, l'hygiéniste a innové. On sait que les recommandations de la CIRP¹ indiquent, pour les différents groupes considérés dans la population, des limites d'irradiation d'autant plus basses que leurs effectifs sont plus élevés (c'est logique: voir formule 5 ci-dessus). Vou-
lant prévoir ce que donneraient de telles limites, l'hygiéniste a calculé les principaux DCT, y compris le $(DCT)_\infty$. Les résultats ont permis à la CIRP de juger

que les valeurs trouvées étaient bien "admissibles", ou encore "l'enveloppe du risque, acceptable". Logique, conformément aux conséquences prévisibles suivant son hypothèse de proportionnalité, elle a aussi jugé prudent de prescrire que l'irradiation des personnes soit aussi réduite que possible. Cependant elle n'ignore pas que, pour obtenir un effet déterminé par l'irradiation il faut donner, d'abord à d, puis à D, une valeur supérieure au seuil correspondant à l'effet considéré. Les valeurs de ces seuils sont inconnues pour la plupart des effets différés, à moins qu'ils ne soient très élevés, comme, par exemple, pour la cancérogénèse de la peau par radiothérapie^{15,16}, ou du squelette contaminé par le radium 226¹⁷; ou encore pour l'obtention de mutants, dès la première génération, par irradiation de souris femelles^{18,19}. Sur des générations successives de rongeurs, malgré de fortes irradiations, le (D C T)_∞ demeure nul²⁰.

Il est même possible d'obtenir l'inversion de certains effets par diminution progressive de d (à D constant) ou de D (à d constant); par exemple, de façon spectaculaire pour la longévité des rats mâles²¹ (augmentée d'un tiers par 1000 rems en un an); et même, sans doute, pour une affection maligne : le lymphome de la souris^{22,23}

On comprend ainsi que des membres autorisés de la CIRP déclarent nettement qu'elle n'a aucune raison de publier des recommandations plus restrictives que les dernières¹.

Cependant, dans l'esprit du public, l'irradiation, traduite en leucémies et en malformations congénitales, frappe essentiellement les enfants. Et personne n'admet d'ajouter un tel risque à son compte, aussi faible soit-il. Et des personnes souvent bien intentionnées, des personnalités même, croyant savoir et censées savoir, mais n'ayant pas su interpréter les textes de la CIRP, y puisent des arguments pour alimenter leur propre inquiétude et pour la propager.

Dans les pays intéressés le législateur a calqué sa réglementation sur les recommandations de la CIRP. Voyant que la perspective de la multiplication rapide de centrales nucléaires provoque des réactions de défense, lorsque ce n'est déjà fait²⁴, il se sent disposé à prescrire des mesures restrictives. L'industriel est prêt à le suivre, voire à le précéder. L'inquiétude du public les rend perplexes, c'est le moins que l'on puisse dire : ils ne sauraient négliger les facteurs subjectifs.

Demain, appelé à rechercher a posteriori une relation de cause à effet, le juriste aura bien des difficultés pour disculper la centrale nucléaire tant que subsistera l'hypothèse d'une relation de proportionnalité. Il faudrait lui en donner les moyens; je l'avais proposé il y a une dizaine d'années²⁵.

5 - Conclusion

La CIRP est sereine, mais l'interprétation de ses textes, difficile. C'est à nous, spécialistes de radioprotection, de rassurer en informant. Commençons par réviser notre nomenclature spécialisée dans un sens qui facilite cette tâche. J'ai proposé de présenter cette communication dans cette intention.

Dans les textes de cette commission on trouve les D C T sous la dénomination "enveloppe de risque". Une telle dénomination, a fortiori, si on lui associe des valeurs numériques, provoque des réactions subjectives qui rendent bien malaisée toute information objective. Aussi, en terminant, me permettrai-je de souhaiter vivement que la CIRP désavoue son hypothèse de proportionnalité.

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AN EVALUATION OF NUCLEAR GAS STIMULATION IN TERMS
OF POTENTIAL RADIATION EXPOSURE TO THE PUBLIC^{a,b}

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Abstract

Experience gained from Projects Gasbuggy and Rulison and their follow-up studies indicates that natural gas produced from a nuclearly stimulated well field will contain small amounts of man-made radioactivity as it leaves the gas processing plant and enters commercial distribution channels. Individual and population doses have been estimated for hypothetical uses of such gas. For example, it is estimated that residential use of nuclearly stimulated gas in unvented cook stoves would result in an average total-body dose to the house occupants of approximately 0.2 millirem/year. Radon concentrations measured in natural gas at various locations in the United States average approximately 20 pCi per liter. Assuming this concentration of radon in the unvented cooking case mentioned, the lung dose is estimated to be 1.5 millirems per year. All of the dose estimates discussed are used to give perspective to the additional radiation exposure of the public which could occur due to use of gas from nuclearly stimulated wells. Both somatic risk and genetic risk are considered in the assessment of relative hazard. Comparisons are made with other risks encountered in the normal activities of life in the United States. The studies summarized show that the radiological impact of either domestic or industrial use of the gas can be small.

Introduction

The U.S. Atomic Energy Commission's Plowshare Program is almost exclusively devoted at present to the development of the nuclear gas stimulation concept. Two experiments involving detonation of single nuclear explosives in low permeability rock formations, Gasbuggy and Rulison, have been conducted to date. Results of these experiments are considered very encouraging by the AEC and the industrial sponsors. Rio Blanco, the third experiment, involved the use of three nuclear devices in one well hole to stimulate gas production in thicker rock formations than would be possible with only one explosive. The explosives were detonated simultaneously on May 17, 1973.

The Health Physics and Environmental Sciences Divisions of the Oak Ridge National Laboratory have been investigating the radiological impact of potential uses of natural gas from wells stimulated with nuclear explosives. This paper summarizes the more important results of these studies.

Radionuclide Inventory

Radionuclides found in gas produced from the completed experiments, Gasbuggy and Rulison, were ^3H , ^{14}C , ^{37}Ar , ^{39}Ar , ^{85}Kr , and ^{203}Hg . of the radionuclides

^aFor presentation at the Third International Radiological Protection Association Meeting, Washington, D.C., September 1973.

^bResearch sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

found, ^3H , ^{14}C , and ^{85}Kr have been studied in greatest detail because they contribute over 99% of the potential dose equivalent* at the radionuclide concentrations observed, and at those projected for future wells (see Table 1).

Table 1. Relative Percentage of Total Estimated Dose from Man-made Radionuclides in Nuclearly Stimulated Natural Gas

Radionuclide	Projected Average First Year Concentration (pCi/cm ³)	Projected Average Lifetime Concentration (pCi/cm ³)	Percentage of Estimated Total Somatic Dose ^a	Percentage of Estimated Total Genetic Dose ^b
^3H	<20	1.0	60	93
^{14}C	<1	0.02	0.3	0.3
^{85}Kr	65	3.3	39	6
All others			<<1	<<1

^aDose to total body.

^bDose to gonads.

The tritium concentration used in this paper, 1 pCi/cm³, is the value projected for the average concentration during the lifetime production of future wells.¹ The concentrations for ^{14}C and ^{85}Kr are scaled to the tritium value on the basis of literature values or fission and activation yield.²⁻⁵ The tritium inventory is distributed in the H₂O-H₂-hydrocarbon system with less than 25% of the total production in the form of hydrocarbons or hydrogen. The remaining 25% appears as tritiated water (HTO) which would normally be removed at the wellhead. Carbon dioxide is also normally removed from natural gas before it enters commercial distribution channels. The tritium and ^{14}C present as hydrocarbons will be released as HTO and $^{14}\text{CO}_2$, respectively, when the gas is burned.

Estimation of Dose to Man

There are numerous pathways through which radionuclides present in gas from nuclearly stimulated wells may cause radiation exposure to man. Our studies have indicated that ^3H and ^{85}Kr are the critical radionuclides and that exposure to combustion products from unvented home usage of natural gas containing radionuclides is the critical exposure pathway.

We can calculate the dose to an individual resulting from combustion of gas in unvented home appliances and heaters based on the projected radionuclide concentrations given in Table 1. For a residence (93 m² floor space, 227 m³ volume) of normal construction with one air change per hour ventilation rate, we estimate the following potential total-body doses (mrem/year) for various unvented domestic uses: cooking, 0.16; water heater, 0.38; refrigerator, 0.20; and heating (5000-degree days), 2.8. The maximum doses estimated for an individual at the projected radionuclide concentrations and assumed exposure conditions with no venting of appliances or heaters is less than 20 mrem/year to the total body. Most cities and states in the United States now require venting of all heaters and appliances except those used for cooking. If this requirement is implemented, the estimated average dose to an individual in the exposed population would be slightly less than 0.2 mrem/year for gas containing the average

*Dose equivalent (rem) = Absorbed Dose (rads) x modifying factors. For the sake of convenience, "dose" will be used hereafter instead of "dose equivalent."

lifetime radionuclide concentrations listed in Table 1. This estimated dose includes a contribution from unvented cooking (0.76 m³ of gas per day, United States average) plus an average atmospheric contribution from all gas used in the area. This latter contribution was shown in a study of the Los Angeles Basin to be less than 10% of the calculated individual dose from unvented cooking.⁶ Even this small calculated average individual dose (0.2 mrem/year) would deliver a potential 200 man-rems per year to each million people exposed. The man-rem dose estimate is obtained by summation of all individual doses within the exposed population.

Use of nuclearly stimulated natural gas in power stations has been suggested as an alternative to residential use. Power station use of nuclearly stimulated gas was given consideration in the Rulison study^{7,8} by assuming that the Cherokee electricity generating plant located in the Denver, Colorado, metropolitan area burned 2.66 x 10⁶ m³ of gas per day (9.72 x 10⁸ m³ per year) contaminated with the projected average lifetime radionuclide concentrations listed in Table 1. The estimated dose to the population (1,500,000) in the Denver area due to power station use of that quantity of gas is 0.32 man-rem. The maximum individual dose estimate for the entire area is 0.006 millirem/year.

The same 9.72 x 10⁸ m³ of gas would supply 174,000 households for 1 year under the following assumptions: unvented cooking (0.76 m³/day), vented water heater (1.8 m³/day), and vented heating for 5000-degree days per year (13.4 m³/day). If each household is assumed to have 3.5 residents, the total number of persons exposed is 610,000, approximately one-third of the total population in the Denver metropolitan area. Then the comparable estimated population dose due to residential gas use is 110 man-rems for 9.72 x 10⁸ m³ of gas having the projected lifetime radionuclide concentrations. Thus, under the conditions specified, the population dose estimate for household use of the nuclearly stimulated gas is nearly 350 times that for power station use.

Assessment of the Estimated Dose to Man

Assessment of the dose estimate projected for use of gas from nuclearly stimulated fields can vary in form and complexity. We believe that the assessment should begin with the recognition that natural gas contains natural radioactivity and that one result of nuclear stimulation is an incremental change in the total radioactivity concentration to which gas users are exposed.

It has been known for nearly 70 years that natural gas contains a radioactive species, radon. Samples supplied by gas transmission companies were analyzed in 1972 and 1973 by scientists in four institutions to provide data on radon concentration in gas being supplied to several metropolitan areas in the United States.⁹ The average value (20 pCi/liter) for all sample locations is used in our dose estimations.

One exposure situation that we consider for radon daughters produced by decay of radon in natural gas is the same as that assumed in the previously described studies.⁷ An unvented kitchen range using 0.76 m³ of gas per day was assumed to be located in a house having a volume of 227 m³. We calculated the concentration of radon daughters in the home for air change rates varying from 0.25 to 2.0 changes per hour. We then estimated doses to the bronchial epithelium from radon daughters resulting from decay of radon introduced with the natural gas and compared these doses with those from an assumed concentration of 0.13 pCi/liter of radon (the average concentration from a number of radon measurements in the United States) and each of its daughters in ventilation air. The estimated dose rate to the bronchial epithelium due to radon and its daughters in the ventilation air was 1300 to 1400 millirem per year. Additional estimated

dose to the bronchial epithelium due to the radon (20 pCi/liter) present in natural gas ranges from 90 mrem/year, for 0.25 air change per hour, to 5 mrem/year, for two air changes per hour. At most, the estimated dose increase due to radon present in natural gas is less than 7%, considering only the two sources of radon. The relative importance of this natural activity in the gas is reduced still further if one considers the daughter activity due to the decay of radon and thoron emanating from home building materials which in some situations exceeds our assumed concentration in ventilation air by a factor of 10. It appears likely, therefore, that the dose which can be attributed to the radon in natural gas is small (<1%) compared to the total dose received in the home from all sources of airborne radioactivity. These localized radon-radon daughter dose estimates are for a limited tissue volume: the basal cells of the bronchial epithelium, which are assumed to be the critical tissue. It has been estimated that the corresponding dose to the total lung mass (1000g) is an order of magnitude lower.¹⁰ The dose to the total lung due to radon in the gas (1.5 millirem/year, assuming one air change per hour) is more suitable for comparison with the total-body estimates obtained for the man-made radioactivity in nuclearly stimulated gas. The comparison indicates that the projected concentrations of man-made radioactivity will contribute a dose which is approximately 12% of the dose due to radon in the gas. Local conditions may, however, alter this percentage significantly.

Another possible assessment, and one which will be required, is comparison of the dose estimate with applicable radiation safety standards. At this time, however, there are no standards which are specific to the use of nuclearly stimulated gas. The Federal Radiation Council (FRC) has established 170 millirem/year as the upper limit for the average total-body dose to a suitable sample of an exposed population group for radiation from all sources exclusive of natural background and medical exposures.¹¹ However, this single source of exposure must not be permitted to take up a disproportionate share of the 170 millirem/year total. The estimated average total-body dose for the population group expected to be exposed via home use of nuclearly stimulated gas is a small fraction (0.1%) of the dose limit of 170 millirem/year.

Our estimate of dose to the total body from nuclearly stimulated gas (0.2 millirem/year) may also be put in perspective by comparing it with dose estimates (millirem/year to the total body) for other sources of radiation received by members of the public: natural background radiation, 130; medical diagnostic X-rays, 110; nuclear weapons fallout, 2; consumer devices, 2; industrial uses of radiation, <1.¹²

A hypothetical assessment of the projected dose may be obtained by estimating the risks which the exposure represents in terms of additional deaths, additional death equivalents due to radiation-induced life span shortening, and additional genetic deaths. A total risk estimate was obtained by summing all three types in spite of the recognized inherent difficulties in combining somatic and genetic insults whose manifestations may differ so greatly. The factors used to convert estimates of radiation dose into estimates of risk are those suggested by the ICRP.^{13,14} Those factors are based on the conservative assumption that there is a linear relationship between dose and effect. The estimates of additional deaths calculated here are believed to be upper limits of risk for the low dose levels considered. The actual risk in fact may be zero, for at such low doses, there is no practical method to reliably determine the actual risk involved. The risk estimated for the projected gas usage is compared in Table 2 with similar estimates of risk for other sources of radiation exposure of the public.

The population dose (man-rem) estimates for the hypothesized gas uses may also be assessed, but to a lesser extent, as there have been no official numerical limits established with which the population dose estimates can be compared. We

Table 2. Comparison of Estimated Deaths Due to Man-made Radioactivity in Nuclearly Stimulated Natural Gas with Similar Estimates for Other Sources of Radiation Exposure of the Public in the United States

Sources of Exposure	Estimated Deaths per Million Individuals Exposed ^a
Natural background radiation ^b	17
Radioactivity in natural gas ^c	
Natural (radon + daughters)	0.3
Man-made	0.03
Other man-made sources of radiation	
Medical diagnostic x-rays	20
Fallout from nuclear weapons	0.4
Consumer devices	0.4
Industrial uses of radiation	<0.2
Power reactors	<0.2

^aObtained by summing estimated somatic and genetic effects; therefore, some of these estimated deaths will occur among the exposed individuals or the first generation of their offspring, but a large majority (over 80%) will occur in succeeding generations.

^b0.1 rem per year.

^cBased on projected radionuclide concentrations used in this study.

have shown that the man-rem dose to the local population is sensitive to the manner of gas usage. The population dose in the Denver area due to background radiation (~200 mrem per person) is nearly 3.0×10^5 man-rems per year, while that estimated for residential gas use (110 man-rems) is 0.037% of the background dose. Dose to the global population is another point to be considered for comparison. Based on dose conversion factors presented in a recent report of the United Nations Committee on the Effects of Atomic Radiation (UNSCEAR),¹⁵ the estimated infinite dose (integrated over infinite time) to the population of the northern hemisphere due to the release of the man-made radioactivity in that volume of gas is approximately 840 man-rems. Nearly all (99%) of that infinite dose is contributed by ¹⁴C due to its long radioactive half-life (5730 years). This estimated dose must be added to the estimated local population dose in assessing the total population dose incurred as a result of the release.

Summary and Conclusions

The radiological impacts of hypothetical uses of nuclearly stimulated gas for domestic and industrial purposes were studied. The critical exposure pathway was determined to be the release of combustion products from unvented appliances in the home. The estimated average total body dose from man-made radioactivity for that pathway is 0.2 millirem per year of gas use. The estimated lung dose due to natural radioactivity (radon) in the gas is 1.5 millirem/year. The critical man-made radionuclides are ³H, ¹⁴C, and ⁸⁵Kr. The largest fraction of the local population dose received via the critical pathway is due to ³H, followed by ⁸⁵Kr. Carbon-14 is of importance as the major contributor to the infinite population dose estimated for the northern hemisphere, with the total dose to the local population being very dependent on the manner of gas use. The radiological impact of the hypothesized gas use was assessed in terms of dose and in terms of risk, in the interest of incorporating radiological impact of gas use as an integral part of the cost-benefit analysis for the development of nuclear gas stimulation technology. The results of the assessment indicate that the radiological impact would be very small. Although the dose and risk estimates obtained in this study are small, the possible exposures still must be scrutinized to achieve the lowest practicable local and global doses.

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GENETIC DOSE LIMIT FOR GENERAL POPULATION, DOSE LIMIT AND DERIVED CONCENTRATION GUIDES FOR MEMBERS OF THE PUBLIC COMPULSIVE ACTION GUIDES FOR EMERGENCY SITUATIONS. A proposal for México, based on data available on Mexican people.

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Using methods recommended by ICRP & UNSCEAR, the mean age of childbearing and genetic dose limit was calculated for mexican population. Data for mexican population when available, complemented with internationally recommended data are used to derive concentration guides for several radionuclides in various environmental media. Derived concentration guides in sea water are calculated applying the specific activity concept. Emergency projected doses demanding action to be taken are proposed. Data lacking is emphasized in order to encourage further studies on habits, critical pathways and transfer factors through food chain for radionuclides in Mexico.

Introduction

Mexico is a developing country with great energetic needs and is now on its way to start using nuclear energy for power production. On this behalf, Uranium ore mining and milling and fuel fabrication is being considered on an industrial scale.

For radiation protection purposes the recommendations of the International Commission on Radiological Protection are now being applied, and regulations are to be issued in a near future, as needed, taking into account habits and characteristics of mexican population.

Genetic Dose Limit

ICRP¹ recommends a genetic dose limit of 5 rems. In the UNSCEAR Report to the General Assembly² there is a detailed discussion of the genetically significant dose, and applying the method outlined with data for Mexican population^{3, 4, 5}, a mean age of childbearing of 35 years was obtained, this gives an annual genetically significant dose limit of 0.144 man rem per 10⁶ inhabitants. Apportionment of this dose, on applying the recommendations of K.Z. Morgan⁶ and rounding off numbers is stated in Table I.

T A B L E I

GENETIC DOSE LIMIT FOR GENERAL POPULATION: 5 rems in 35 years		
ANNUAL GENETICALLY SIGNIFICANT DOSE: 0.14 x 10 ⁶ man rem per 10 ⁶ inhabitants		
APPORTIONMENT IN MAN REMS PER YEAR 10 ⁶ INHABITANTS		
Diagnosis	5 x 10 ³	Other environmental sources
Therapy	10 ⁴	Future Applications and
Nuclear energy		emergency situation
production	10 ⁴	Occupational exposure
		10 ⁵
		10 ⁴

It may be noticed that a substantial portion of the genetic dose available is allowed for medical exposure. The genetic dose due to this source tends to increase the benefits being for the present generation whilst the risk will

burden future generations, and so it has to be recorded and included in what would be an acceptable genetic risk from all uses of radiation.

Dose Limits and Concentration Guides

Radionuclides, sources and environmental pathways considered as most important for the transfer to man of radionuclides introduced in the environment, were obtained from the general guidelines for the growth of nuclear industry, as set forward by the Nuclear Energy Institute in its development program⁷ and on the nuclear power reactor of Laguna Verde, Veracruz to start operating in 5 or 6 years. By now, there is no experience on pathways for radionuclides from source to man in Mexico, and this approach is a purely theoretical one.

Some studies have been made in Mexico on characteristics and habits of Mexican population^{8,9,10,11,12}, this studies were performed for medical or nutritional reasons. They show that a wide difference exists specially on nutritional habits, in different zones of Mexico and even between habits of people living in the same area, but engaged in different economical activities, this does not permit to find the critical pathway for Mexico as a whole, although it is feasible with a well designed survey for a small area, as the one affected by effluents from a nuclear reactor.

Federal Radiation Council¹³ has been issuing concentration guides for average population applying a 1/3 safety factor to dose limits set forth by ICRP for individuals in the critical population. Since in Mexico there is also an incomplete knowledge of some data needed, an arbitrary safety factor of 1/4 is used instead. Morphological and physiological parameters obtained from studies on Mexican population^{8,11} are summarized in Table II.

T A B L E II	
DATA ON PHYSIOLOGICAL AND MORPHOLOGICAL CHARACTERISTICS FOR AVERAGE MEXICAN POPULATION	
PARAMETER	MEN
Body Weight	65 Kg
Daily water intake	1300 ml
Body fluids	39 Kg
Mineral bone	4.6 Kg
Calcium content	975 g
Thyroid	35 g

For other data needed ICRP and other sources were consulted^{14,15,16,17,18},
¹⁹ Data on food consumption were obtained from Zubiran et. al²⁰, Table III.

T A B L E III		
AVERAGE FOOD CONSUMPTION IN URBAN AND RURAL AREAS IN MEXICO		
FOOD	GROSS WEIGHT IN GRAMS CONSUMED BY	
	RURAL POPULATION	URBAN POPULATION
Corn	399	202
Bread and Pasta	25	129
Rice	5	10
Beans	45	45
Meat	47	76
Milk	62	241
Cheese	3	3
Eggs	5	13
Vegetables	81	114
Edible roots	11	20
Fruit	36	72
Sugar	39	77
Fats	13	26
Cacao	3	0
Other	0	12

Table IV shows concentration guides or working limits for average population.

T A B L E IV		
WORKING LIMITS OR CONCENTRATION GUIDES FOR AVERAGE POPULATION IN MEXICO		
RADIONUCLIDE	WORKING LIMIT OR CONCENTRATION GUIDE	IN
^{226}Ra	3 pCi/l	Drinking water
U^{nat}	3 $\mu\text{g}/\text{m}^3$	air
^3H (HTO)	$30 \times 10^{-3} \mu\text{Ci}/\text{m}^3$	air
^4A	$3 \times 10^{-2} \mu\text{Ci}/\text{m}^3$	air*
$^{85\text{m}}\text{Kr}$	0.1 $\mu\text{Ci}/\text{m}^3$	air*
^{85}Kr	0.2 $\mu\text{Ci}/\text{m}^3$	air*
^{87}Kr	$2 \times 10^{-2} \mu\text{Ci}/\text{m}^3$	air*
$^{88}\text{Kr} - ^{88}\text{Rb}$	$10^{-2} \mu\text{Ci}/\text{m}^3$	air*
$^{131\text{m}}\text{Xe}$	0.3 $\mu\text{Ci}/\text{m}^3$	air*
^{133}Xe	0.3 $\mu\text{Ci}/\text{m}^3$	air*
$^{135\text{m}}\text{Xe} - ^{135\text{m}}\text{Cs}$	$8 \times 10^{-2} \mu\text{Ci}/\text{m}^3$	air*
^{131}I	30 pCi/ m^3	air
^{131}I	110 pCi/day	Total Diet (6 months child)
^{131}I	1000 pCi/l	cow's milk (Through mother to breast fed babies).
^{137}Cs	300 pCi/ m^3	air
	5×10^3 pCi/day	Total Diet
	RURAL TYPE DIET	URBAN TYPE DIET
	1.3×10^4 pCi/Kg	2.6×10^4 pCi/Kg Corn
	12×10^4 pCi/Kg	12×10^4 pCi/Kg Beans
	9×10^4 pCi/l	2×10^4 pCi/l Milk
	11×10^4 pCi/Kg	7×10^4 pCi/Kg Meat

*Safety factor of 1/4 was not used for calculus.

The main food for children's diet from birth to about 2 years of age is milk, but in Mexico most babies are breast fed, and weaning starts between 1 and 2 years of age substituting the mother's milk with corn or/and beans, not with cow's milk, the pathway for radioiodine being from milk and food eaten by the mother to breast fed babies. A survey made at the Instituto de Nutrición by Perez H. et al²¹ gives an average of 400 ml milk in the mother's diet and so the derived working level or concentration guide for average individual in general population was calculated, on the basis of iodine transferred by mother to child, using data from Weaver et al²² for a mother with a milk production similar to the average mexican mother.

Strontium has a metabolism similar to calcium, but is discriminated against through its pathway from environment to man, the ratio of Sr 90 to calcium in bone needed to obtain a dose of 0.75 rems/year to bone, for individuals in the average population applying the method outlined by UNSCEAR²: is 4.5 m rad y^{-1} per pCi (gCa)⁻¹, and dose in rems is obtained multiplying by the "relative damage factor" 5, for Sr 90 in bone, giving 23 mrem y^{-1} per pCi (gCa)⁻¹ and 33 pCi (gCa)⁻¹ for 0.75 rem y^{-1} UNSCEAR² gives a transfer factor of 0.12 diet to bone and 275 pCi ⁹⁰Sr/gCa in diet produces 33 pCi ⁹⁰Sr/gCa in bone.

A high proportion of calcium is obtained in mexican diet through mineral calcium added to corn (150 mg of Ca/100g corn²³), in making much of the food based on corn and specially "tortillas" which are used instead of bread by most of mexican population. This calcium has a negligible contribution to ⁹⁰Sr contamination and in considering a uniform contamination of the biosphere, after UNSCEAR and C. L. Comar^{2,24} and calcium content in food items commonly consumed in Mexico, transfer factors are shown in Table V. Doses produced by Sr89 for a long period of time are 25 times lower per pCi/gCa than dose produced by ⁹⁰Sr, and the average concentration guides or working limits for ⁸⁹Sr and ⁹⁰Sr in food regardless of the actual quantity consumed are shown in Table VI.

T A B L E V

RELATIONS BETWEEN *Sr-Ca OF VEGETATION (100 *Sr-100Ca) and *Sr-Ca OF MAN

	% Ca IN DIET		PLANT PRODUCT	DIET BODY	*Sr PER 100Ca IN BODY	
	RURAL	URBAN			RURAL	URBAN
Cereals	5	7	1	0.12	0.6	0.84
Other plants	15	19	1	0.12	1.8	2.28
Dairy products	10	35	0.12	0.12	0.14	0.5
Mineral Ca	67	36	0	0	0	0
Total					2.6	3.7

*Sr RADIOSTRONTIUM

T A B L E VI

RADIOSTRONTIUM DERIVED WORKING LIMITS OR CONCENTRATION GUIDES FOR AVERAGE POPULATION IN MEXICO

	URBAN DIET		RURAL DIET	
	⁹⁰ Sr	⁸⁹ Sr	⁹⁰ Sr	⁸⁹ Sr
Corn	100 pCi/Kg	2x10 ³ pCi/Kg	130 pCi/Kg	3.5x10 ³ pCi/Kg
Milk	130 pCi/l	3.3x10 ³ pCi/l	180 pCi/l	4.5x10 ³ pCi/l
Beans	2x10 ³ pCi/Kg	5x10 ⁴ pCi/Kg	3x10 ³ pCi/Kg	7.5x10 ⁴ pCi/Kg
Vegetables	10 ³ pCi/g Ca	2.5x10 ⁴ pCi/gCa	1.3x10 ³ pCi/gCa	

Concentration Guides in Sea Water. The marine food chain to man is not well known in Mexico's coastal waters and the specific activity approach is likely to be the best under this circumstances. Following Kaye S.V. and Nelson D.J.^{2,5} concentration guides in sea water for some radionuclides of interest are obtained using the following:

$$\text{Concentration Guide in sea water } \mu\text{Ci/l} = \frac{2.8 \times 10^{-3} Y_1 W}{\Sigma EF (RBE) n Y_2} \left| 1 + \frac{T_b}{T_r} \right| \left| \frac{1}{1 - e^{-\frac{(0.693t)}{T_e}}} \right|$$

Where: Y_1 = concentration of stable element in sea water ($\mu\text{g/l}$); Y_2 = concentration of stable element in organ of reference ($\mu\text{g/g}$); W = weekly dose limit (annual dose limit/52); $\Sigma EF (RBE) n$ = effective energy in MeV per desintegration; T_b = biological half-life in days; T_e = effective half-life in years; $t = 70$ years.

The dose limit used was not affected by the 1/4 safety factor since there are already safety factors included, in omitting the effective half-life and growth factors for every link in the pathway from sea water to man. Data used for calculation were obtained from literature^{1, 15, 26}.

For radioisotopes with GI tract as critical organ, since the exposure is due to the absolute concentration of radionuclide in the tract, instead of using the specific activity approach, the method outlined by Aten²⁷ is used:

$$\text{Concentration Guide } (\mu\text{Ci/l}) = \frac{M PC_w \times 2200}{|(0.13 \times F_C \times K_C) + (0.13 \times P_f \times K_p)| \times 4 \times 10^4}$$

Where: MPC_w = maximum permissible concentration in drinking water for occupationally exposed personnel (168 h) $\mu\text{Ci/ml}$ ¹⁵; F_C = Concentration factor for shrimps²⁶; K_C = Shrimp fraction in marine food intake (1, 0.5, 0); P_f = Concentration factor for fish²⁶; K_p = Fish fraction in marine food intake (1, 0.5, 0); 0.13 = Marine food daily consumption in Kg¹².

A survey made by the Instituto de Nutrición, on food intake, of a fishing community, Alvarado, in the same state where the power reactor site is, although not in the same area, is used for calculus, food consumed were fish and shrimps, but no mention is made on the proportion of each, since both were grouped together for survey purposes. Since concentration factor from sea water to edible product are quite different for shrimps and fish, derived working levels or concentration guides were calculated considering fish 100%, shrimps 100% and 50% consumption of each. A factor of 1/10 for individuals in the critical population and 1/4 safety factor are included.

The concentration guides are presented in Table VII.

T A B L E V I I						
DERIVED WORKING LIMITS OR CONCENTRATION GUIDES FOR SOME RADIONUCLIDES IN SEA WATER						
RADIO-NUCLIDE	CRITICAL ORGAN	DERIVED WORKING LIMIT OR CONCENTRATION GUIDE IN SEA WATER				
		SPECIFIC ACTIVITY METHOD	SHRIMP 100%	FISH 100%	SHRIMP 50%	FISH 50%
⁵⁴ Mn	GI Tract		2.2x10 ²	5.3x10 ³	4.3x10 ²	
	Liver	6x10 ²				
⁵⁵ Fe	Spleen	1.7x10 ²				
⁵⁹ Fe	GI Tract		10 ²	1.6x10 ²	1.3x10 ²	
	Spleen	31				
⁵⁸ Co	GI Tract		8.5x10 ²	4.2x10 ⁴	1.7x10 ³	
	Whole body	4.6x10 ²				
⁶⁰ Co	GI Tract		4.2x10 ²	2x10 ⁴	8.3x10 ²	
	Whole body	1.7x10 ²				
⁶⁵ Zn	Whole body	1.2x10 ²				
⁸⁹ Sr	Bone	11x10 ⁶				
⁹⁰ Sr	Bone	4.7x10 ⁴				
⁹¹ Y	GI Tract			5x10 ²		
¹³¹ I	Thyroid	1.1x10 ³				
¹³⁴ Cs	Whole body	8.7x10 ³				
¹³⁷ Cs	Whole body	10 ⁵				
¹⁴⁰ Ba	GI Tract			1.6x10 ⁴		
	Bone	3x10 ³				
¹⁴⁴ Ce	GI Tract		2x10 ⁴	1.4x10 ⁵	3.7x10 ⁴	

Action Levels

In order to set action levels, the social cost together with the expected effectiveness in enforcing the corrective measures has to be balanced against the risk reduced, in this behalf a due study has to be undertaken and each place has to be analyzed in itself, and reviewed as changes happen.

In order for the nuclear industry to include the needed safety measures in design it is considered that people are prepared to move from one state into another and in so doing their risk of accidental death will change, varying from 2.1x10⁻⁴ in Quintana Roo to 11.1x10⁻⁴ in Colima³, so for individuals in the population, an increase in 10% the previous risk due to accidental death is acceptable, and action levels for whole body irradiation, should not produce significant early effects in the individuals exposed, a limit of 25 rems to whole body for men and 10 rems for women in reproductive age, delivered in a short period of time, and for organ irradiation, the enhanced stochastic cancer risks, should not be higher than 10% the actual risk from accidental death in Mexico, about 6x10⁻⁴ in 1969. Action levels for whole body and different critical organs are displayed in Table VIII, together with data on risk estimates considered.

T A B L E V I I I			PROJECTED ACTION GUIDES	
	ACTION GUIDE	ENHANCED STOCHASTIC CANCER RISK OF DEATH PER MILLION PEOPLE EXPOSED.		
For whole body				
Women in reproductive age	10 rems			
Men	25 rems			
For Thyroid	35 rems	3x10 ⁻⁶ per rem for children ^{2,8}		
		10 ⁻⁶ per rem for adults ^{2,8}		
		1.7x10 ⁻⁶ per rem for average mexican population*		
For bone	1.5 rem/year	10 ⁻⁵ for 0.3-3 rems/year ^{2,9}		
For lungs	6 rems	10 ⁻⁵ per rem ^{2,8}		
*Mexican Population includes 33% of children under 9 years ³ .				

Applying action levels in order to get concentration guides in some of the links of the pathway, should be done after studying the population at risk.

Conclusions

On assessing the dose to average population, the common procedure is to survey the environment, by measuring activity in suitable samples and data obtained must be translated into dose for people exposed, or compared with data set as based on dose limits, in any case, parameters are needed for dose assessment and the values calculated are as good as the actual numbers used.

On the other hand values for parameters are different among different people and it is important, to dedicate some effort in obtaining these parameters and governmental agencies, beside the Nuclear Energy Institute should be encouraged to do research in this field, in order to find for average Mexican population physiological and morphological data, on food consumption habits and transfer factors for common food in Mexico. This studies are of importance, specially in areas where nuclear industry will be developed.

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VADOSCA: A SIMPLE CODE FOR THE EVALUATION OF POPULATION EXPOSURE DUE TO RADIOACTIVE DISCHARGES

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Abstract

The code consists of two parts, one for liquid discharges (VADOSCA-LI) and one for gaseous discharges (VADOSCA-GAS), and it incorporates the transfer parameters of twenty-four radioisotopes in the case of liquid discharges, and of twenty radioisotopes in the case of gaseous discharges. It allows the evaluation of the concentrations of the various isotopes in all the compartments of the critical paths outlined in the ICRP Publication No. 7, and the evaluation of the annual doses for five critical organs (whole body, G.I. tract, thyroid, bones, lung) for the various critical groups of population, on the basis of environmental parameters, such as the time of residence in a certain area, diet, type of activity, hydrological regimen, irrigation methods, and meteorological conditions.

Although extremely simple, the code allows rapid performance of all the evaluations required to define the amount of radioactivity that can be released and the associated exposures.

Introduction

As the peaceful uses of nuclear energy expanded, a great number of criteria had been voiced on environmental protection against radioactive discharges from nuclear plant; in 1965 they were collected in an orderly form in the ICRP Publication No. 7. In the light of the acquired knowledge of reconcentration of radionuclides in the environment as a result of chemical-physical (absorption, sedimentation) and biological processes, ICRP suggested assessing the doses due to the discharges following all the possible paths from the plant to man. This approach calls for a wide knowledge of the environment and transfer parameters of all nuclides present in the discharges, and sophisticated computer programs¹.

In the meanwhile the public opinion had risen against environmental degradation to the point of objecting even to the construction and operation of nuclear power stations, and especially to their effluents. The Regulatory Agencies were thus pressed to lower the limits of discharged radioactivity and to request of each station an analysis of its impact on the environment, more or less in harmony with the ICRP recommendations. This attitude is very well illustrated in a recent AEC document², which analyzes the merits of the various types of waste treatment plants in the light of the reduction of the population exposure to "as low as practicable" values.

Likewise, in Italy CNEN has long ago adopted the criterion of issuing operating licenses containing limitations on the radioactive discharges based on the actual station requirements and on an analysis of the receptivity of the environment. ENEL, the national producer and distributor of electricity in Italy, responsible for providing to CNEN such an impact analysis for its nuclear stations, developed a computer code in two parts that simplify the evaluation of

of population exposure due to liquid and gaseous radioactive wastes (called VADOSCA-Li and VADOSCA-Gas).

VADOSCA-Li

In its present form, the code covers twenty-four radionuclides, of which some are fission products (^{89}Sr , ^{90}Sr , ^{91}Y , ^{95}Zr , ^{106}Ru , ^{131}I , ^{134}Cs , ^{137}Cs , ^{140}Ba , ^{144}Ce), others are activation products (^3H , ^{14}C , ^{32}P , ^{35}S , ^{45}Ca , ^{51}Cr , ^{54}Mn , ^{59}Fe , ^{58}Co , ^{60}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$, ^{124}Sb), plus an alpha emitter, ^{239}Pu . The choice of these nuclides was dictated by the frequency of their presence in the station discharges and by their radiotoxicity. Nuclides having a half-life of less than a few days are not considered. However, the code can handle thirty isotopes to accommodate particular situations with isotopes that are not listed above.

When the wastes are discharged to a closed body of water (sea or lake) the main dilution considered is that due to the condenser coolant; moreover, dilution coefficients are fed to the computer for each critical path and for each single case on the basis of hydrological and thermal considerations. Introduction of the commonly used diffusion models^{3,4} was avoided because they lack the required flexibility and they required a semi-empirical approach.

When the wastes are discharged into a river, they are considered diluted first by the condenser coolant and then by the average flow of the river itself. If the water table is involved, decontamination coefficients are fed to the computer to take into account the absorption due to the soils crossed by the liquid. Finally, additional dilution coefficients can be introduced for each pathway to take into account any other mechanisms, such as suitably large tributaries or sedimentation effects.

Fig. 1 provides a schematic illustration of the critical pathways considered for the liquid wastes. The code calculates the doses to four special groups, namely, fishermen, other workmen, local population, farmers. A separate calculation is made for the doses originated by drinking water. The doses are evaluated for five critical organs (whole body, gastro-intestinal tract, thyroid, bones, lungs) and they are expressed in mrem/year if the discharges are ex-

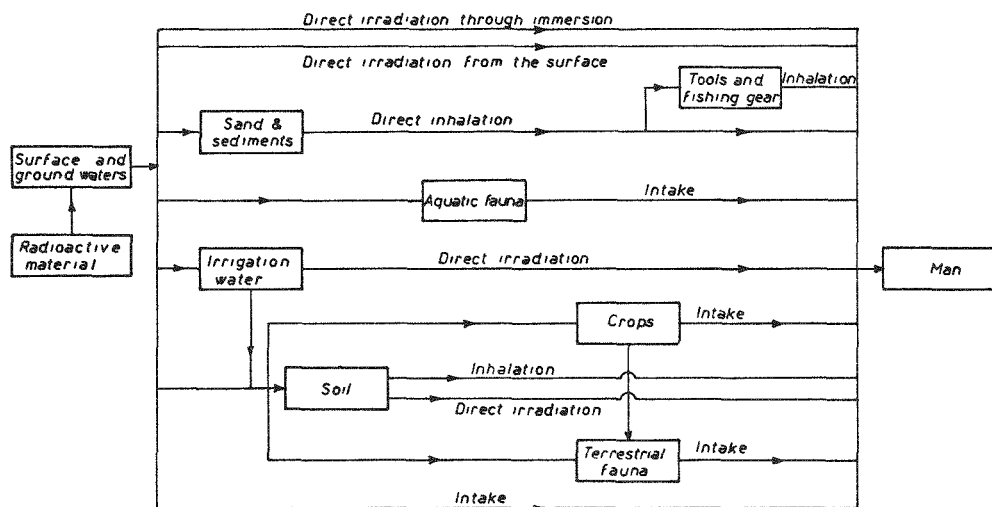


Fig 1 - Critical pathways for liquid discharge

pressed in Ci/year.

The concentrations in each compartment in Fig. 1 are obtained by means of a linear relation from the preceding compartment and with the transfer coefficients taken from the literature or from the concentrations of stable isotopes in the respective compartments or evaluated on the basis of other considerations, as illustrated below.

Marine biota and fresh water fish. Frecke's⁵ concentration factors were used for the former, whereas reference was made to the concentration factors found in the literature^{6,7,8} for the latter.

Irrigated crops. Six types of crops were considered, namely, rice, other cereals, leafy vegetables (including forage), radishes, other vegetables and fruit. The literature provides very scanty information on the relative concentration factors, so reference was often made to the method of specific activity; a standard concentration was assumed for fresh water and the concentrations for the vegetables were taken from the literature. When this method was not applicable, the concentration factors of chemically similar elements were used.

Terrestrial animals and products. The following critical pathways were considered: meat of herbivores (beef, rabbit, lamb, pork) and milk, poultry and eggs. The radionuclide transfer is considered from the forage for herbivores and from cereals for poultry. The transfer factors for C, P, S, Ca, Fe, Mn, I, Zn, Co, Sr were obtained by the specific activity method, the factor for Cs from the literature⁹, Cr, Co, Y, Zr, Ru, Ag, Sb, Ba, Ce and Pu were considered little assimilated by the GI tract, (i. e. 1% in respect of Cs).

Sediments. Lacking specific data that would any way strongly depend on local factors (meteorology, hydrology, type of river banks and soils), we assumed standard transfer factors that varied by a factor of 10 depending on the half life and type of sediment (sandy or silty). The tailrace was taken as the preceding compartment before complete mixing with the receiving body.

Irrigated tillage. The irrigating water was assumed to deposit its radioactivity in the first ten centimeters of soil and equilibrium concentration was assumed to be reached with a 15% leaching yearly.

Once the concentration in each compartment is known, the doses to the critical groups are calculated taking into account critical parameters, such as time of residence in water, on water, on sediments, on irrigated land, dietary habits, and working time. The doses due to irradiation from water and land were calculated with the formula¹⁰:

$$D_i = 1.06 \times 10^3 \times C_i \times S_i \times T$$

where D is expressed in mrem/yr, C_i is the concentration of isotope i (Ci/m³), S_i is the energy of beta and gamma rays emitted by the isotope i (Mev), T is the irradiation time (hours). The formula is based on the assumption of a plane infinite source and it overestimates the actual dose. When applied to sediments, the formula gave an excessive dose because of their high concentration factors; therefore, a more sophisticated mathematical treatment¹¹ was used, whereby allowance is made for the actual thickness of the sediments, for the overlying water layer and for the geometry effect of the source (semi-plane in the case of irradiation of a critical group residing on river banks or sea coast).

For the intake doses the CMAs given in the ICRP Publication No. 2 for the various organs, following the method proposed by Essig¹², and the yearly food consumption of the various population groups were used.

The doses due to inhalation of resuspended material (sediments and agricultural soil) are factored in by assuming a standard breathing rate of 0.83 m³/hr and a content of airborne particulates of 10 mg/m³. The latter value is a maximum obtained from samples of air taken in different conditions, such as open country, construction sites, residential areas.

The code is written in FORTRAN; it needs 20 K memories and it takes approximately 15 seconds in a GE 635 computer.

VADOSCA-Gas

Twenty nuclides are considered, of which eight noble gases (fission and activation products: ⁴¹A, ^{85m}Kr, ⁸⁵Kr, ⁸⁷Kr, ⁸⁸Kr, ¹³³Xe, ¹³⁵Xe, ¹³⁸Xe), various isotopes as particulates (⁶⁰Co, ⁸⁸Rb, ⁸⁹Sr, ⁹⁰Sr, ¹³¹I, ¹³⁴Cs, ¹³⁷Cs, ¹³⁸Cs, ²³⁹Pu), and ³H, ¹³N, ¹⁴C in the form of vapor. Short-lived isotopes were taken into account only when experience indicated that they might be present in the discharges. At any rate, the code can accommodate other five nuclides. For each nuclide it is necessary to supply the average yearly discharge.

Atmospheric diffusion is evaluated with Pasquill's theory¹³ adapted to the meteorological data available. In fact, two sets of information are handled by the code, namely,

Set A: Only a wind rose and an average distribution of the six stability categories are available. This is the general case described by P. Bryant¹³.

Set B: In this case, in addition to the wind rose, one must have the frequency distribution of the stability categories for each wind direction, subdivided by wind speed ranges. This is the sort of information used by May and Stuart for their diffusion analysis at Brookhaven¹⁴.

For simplicity, in its present form the code does not take into account particular effects such as the cloud depletion due to deposition, down draft and building effect; the particulate deposition rate is assumed constant at 3x10⁻² m/s.

Fig. 2 shows the critical pathways for gaseous discharges. Two sets of doses are considered: (1) doses due to irradiation and inhalation as a function of the distance from the stack and direction of plume travel; (2) doses due to intake of agricultural (vegetal and animal) produce. The former are calculated

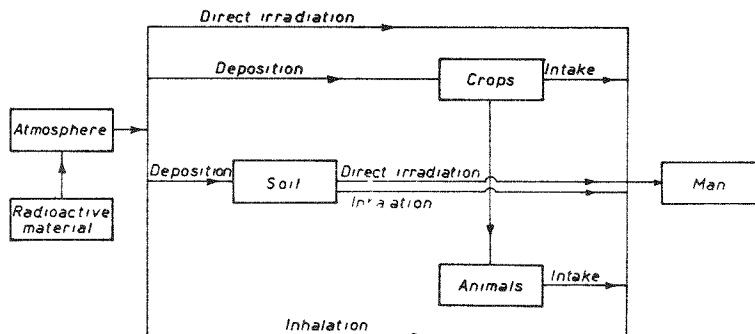


Fig 2 Critical pathways for discharges to the atmosphere

over a discrete number of distances between 0.3 and 50 km and for a maximum of sixteen sectors; the latter are calculated only at the point of maximum fallout and for two population groups (farmers and local population) and for the usual five organs.

The exposure due to the cloud is calculated with a method developed by Gammertsfelder¹⁵. This method takes into account the contribution made by a cloud of finite dimensions with a gaussian vertical concentration distribution and an average horizontal concentration.

The exposure due to inhalation is calculated from the ground concentrations, evaluated with Pasquill's formula¹³. The same formula is used to calculate the deposition on the crops, excluding the short-lived ⁸⁸Rb and ¹³⁸Cs.

The transfer factors for crops were calculated taking into account the growth time of the crops, the concentrations of the stable isotopes, the decontamination due to decay and processing. The related doses were calculated like those resulting from the liquid discharges.

The code is written in Fortran; it needs 60 K memories and it takes about 60 seconds in a GE 635 computer.

General Comments

Applied to the analysis of environmental impact, VADOSCA has proved to be a useful tool for the health physicist. While relieving him of painstaking calculations, it has compelled him to single out from the host of ecological data those that were most significant for radioprotection. In practical application, it was necessary to modify the standard transfer factors to adapt the code to the results of the surveys at the nuclear station sites. For instance, for the nuclear station on the Garigliano river at 11 km from the coast, the Cs transfer factor for fish was found to be much lower (200) than the standard value (1000). Instead, at Trino Vercellese on the upper course of the Po river the same factor was 1400 and for certain species of fish that feed on periphyton it was as high as 2500. This example is typical to illustrate the limitations of this type of code, which starts from standard values selected conservatively and then must be adapted through successive approximations.

In the negotiations with the safety authorities for the discharge permits, the availability of an agreed standard code facilitates mutual understanding and saves considerable time.

It is recognized that further improvements of the code are necessary in order to factor in all the information on radioactivity concentration and dispersion mechanisms in the environment that is building up. Moreover, the next step should be the use of dynamic models of the type already widely used in other branches of ecology^{1,16}, but this will be warranted only when a better understanding of the aforesaid mechanisms has been acquired and more complex problems are to be dealt with.

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MEDICAL EXPOSURE

NATIONWIDE EVALUATION OF X-RAY TRENDS (NEXT)

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Abstract

The impact of efforts by government agencies to minimize and control unnecessary patient exposure in medical x ray has been difficult to assess. Population exposure studies, such as those conducted in 1964 and 1970 of the United States Genetically Significant Dose, are expensive. Reports of the number of x-ray machines in compliance with equipment standards, such as for filtration and collimation, are traditional but are difficult to relate to patient or population exposures. Furthermore, such reports are limited to machine parameters and do not reflect other factors influencing patient dose such as operator training, film and screen selection and film processing.

A Task Force of State and Bureau of Radiological Health representatives, created in 1971, has applied the "standard man" concept as a method of evaluating the impact of government control efforts on medical x-ray exposure.

A limited number of randomly selected medical x-ray facilities are visited. The operator of an x-ray unit is requested to set the technique factors for a selected examination of a patient whose anthropometric characteristics have been standardized. Exposure data is collected using standardized procedures and equipment. Organ Dose Index values are generated for selected critical organs.

By eliminating patient size variation, the wide range of exposure technique factors currently employed in medical radiography has become readily apparent, together with the wide range of gonad doses encountered by a standard patient. It is expected that these can be correlated with governmental agency activities in radiological health and can be modified by their efforts.

The methodology of the NEXT Organ Dose Index System is simple. Fifty local, State and Federal radiological health programs in the United States are participating or planning to participate in the system which began operation in October, 1972. Some results of the first nine months of its operation are presented. The system appears to be far more efficient in assessing the impact of governmental program efforts in medical x ray than previously used methods. It avoids the complexity and cost of population exposure studies but can relate program impact in terms of patient exposure and dose.

Radiological Health Efforts in the United States

Historically, regulatory radiological health efforts in the United States trace their origins to the state and local levels, particularly as they affect medical x-ray use. Currently 50 States and the District of Columbia conduct radiation control programs under either general public health laws or specific enabling legislation.¹ Technical details of regulatory control are usually not included in authorizing legislation but are delegated to a radiological health agency or advisory group. More recently, under authority of Public Law 90-602, Federal performance standards for medical x-ray equipment have been prepared.²

In 1962 the Council of State Governments, in cooperation with the United States Public Health Service and the United States Atomic Energy Commission, developed Model Regulations for Radiation Control which included a section on the "Use of X rays in the Healing Arts."³ The original regulations have been updated twice and are undergoing further revision. Most States which have adopted regulations in the area of medical and dental x ray have followed this guidance thus providing some measure of uniformity. The Council of State Governments' regulations have been based upon the recommendations of various standard-setting bodies, particularly those of the National Council on Radiation Protection and Measurements.⁴

The major thrust of radiological health agencies' activities in the United States, to date, has been directed at upgrading the x-ray equipment used in the diagnostic healing arts to meet minimum regulatory standards. Consequently, the primary purpose of most medical x-ray inspection programs has been to determine that the equipment requirements of the regulations are being followed. However, advantage is frequently taken of the personal contact during an inspection of a medical or dental facility to discuss with personnel other items related to the use of the equipment.

For example, exposure technique and film processing may be reviewed with the operator. In such cases, the inspection visit becomes a mini-training program. As a result, in addition to a report of items which do not meet regulatory requirements, recommendations may also be provided to the user which, if followed, can improve the overall radiological health aspects of an x-ray facility.

Regular training courses are offered by many radiological health agencies, usually in conjunction with, or using the resources of, various Federal agencies. The Bureau of Radiological Health of the Food and Drug Administration has developed training packages for use by State and local agencies. Many of these agencies routinely offer the services of their staff to provide lectures for ongoing formal training programs in hospitals or other schools.

Many State and local radiological health agencies review and approve x-ray facility plans and specifications. Items such as adequacy of radiation barriers, film handling and processing facilities, type and location of radiation machines, and adequacy of ancillary equipment are subjected to critical review.

Radiological health agencies realize that ensuring that proper equipment is provided is only one parameter in the equation of optimizing the benefit of an x-ray exposure of a patient. With this in mind, three States and the Commonwealth of Puerto Rico have approved specific laws which establish minimum standards for education, training and experience for certain user groups that apply x rays to humans for diagnostic or therapeutic purposes.¹ The United States Public Health Service has developed guidelines for the establishment of

licensing programs for x-ray users in the healing arts.⁵

All of these programs are directed toward minimizing exposure in the environs of an x-ray installation and optimizing the benefits of patient radiation. But how do these agencies determine their program effectiveness? National X-ray Exposure Studies such as conducted by the United States Public Health Service in 1964 and in 1970 can and do provide information to answer this question.^{6,7}

However, such x-ray exposure studies require considerable investments of time, money and personnel at local, State and Federal levels. This time interval - 6 years - is too long for use by agencies requiring the kind of information which is necessary to justify budget requests, to plan program priorities and to evaluate past program effectiveness. These activities are performed on an annual basis and information must be available on an annual basis.

The usual information resources available to agencies are limited to reports of inspection programs such as the number of x-ray machines in compliance, the number of x-ray machines with deficient filtration, etc. Increasingly, such data has been found unsuitable for identifying specific problem areas, for justifying existing radiological health programs and budget requests, and for program planning. Such terminology is not meaningful to many public officials responsible for planning fiscal and personnel resource allocations to the various technical programs under their jurisdiction. Instead, program effectiveness or needs must be reported in people-related terms.

Nationwide Evaluation of X-ray Trends (NEXT)

In May 1971 the Conference of Radiation Control Program Directors called for the formation of a Task Force to design a uniform program for surveys of x-ray facilities.⁸ The Task Force, co-sponsored by the Bureau of Radiological Health of the Food and Drug Administration, Department of Health, Education and Welfare, was appointed in July 1971 and consisted of equal representation from State and Federal radiological health agencies.

The Task Force adopted the project name of "NEXT", an acronym for Nationwide Evaluation of X-ray Trends. In reviewing its charge, the Task Force identified four specific objectives⁹ it wished to meet:

1. Design a system to measure the effectiveness of radiological health programs,
2. Design a system which would enable program priorities to be assigned on a rational, documentary basis,
3. Identify the optimum components of a radiation survey, and
4. Provide for the uniform collection of data related to radiological health.

The development of an Optimum Survey Procedure Manual would partially satisfy these objectives. This is currently under revision and will not be discussed here.

While the production of a manual of optimum survey procedures will fulfill a long sought need by local and State radiological health programs, the NEXT Task Force recognized that additional parameters were required to meet the objectives of providing a system to measure program effectiveness and to assign program priorities. The Organ Dose Index System (ODIS) was devised to meet these objectives.

The Organ Dose Index System (ODIS)

The Organ Dose Index System is based on an annual survey of a statistically representative sample of the x-ray tubes within an agency's jurisdiction. The results are intended to provide the agency with a measure of its effectiveness in reducing unnecessary radiation exposure during diagnostic radiography. The system was not designed to replace compliance survey procedures now in use but is intended to be an adjunct to these procedures. The system provides specific organ doses called Organ Dose Indexes for selected x-ray procedures applied to a standard sized patient.

The term "Index" has been appended to "Organ Dose" because the calculated dose values are not an "average", nor representative of the population dose. They are the organ doses only for an individual who fits the physical characteristics of the "standard patient". (This patient was subject number 16 of the group who participated in the Johns Hopkins study to determine scatter to primary x-ray beam exposure ratios.⁶) Organ Dose Index, is therefore, a people-related quantity. Since it reflects a "standard patient" it removes the variable of patient size. It has the potential for evaluating variations of organ dose by type of facility, technique, operator training, beam size and shape, etc. The methodology of calculating radiation doses to the gonads used in the Organ Dose Index System is that used in the X-ray Exposure Studies of the United States in 1964 and 1970, with modifications.⁶ Other organs have been identified by the Task Force for which calculated radiation doses are desired. These are the thyroid, lens of eyes and bone marrow. Organ Dose Indexes for these organs are awaiting development of suitable dose models.

Twelve common diagnostic radiographic examinations are included in the Organ Dose Index System:

<u>Projection</u>	<u>Body Part of Interest</u>	<u>Body Part Thickness (centimeters)</u>
Chest (P/A)	Thorax	23
Skull (Lateral)	Head	15
Abdomen (KUB)		
Scout Film (A/P)	Abdomen	23
Retrograde Pyelogram		
Scout Film (Cysto Units) (A/P)	Abdomen	23
Thoracic Spine (A/P)	Thorax	23
Cervical (A/P)	Neck	13
Lumbo-Sacral Spine (A/P)	Abdomen	23
Full Spine (A/P) (14"x36" film size only)	Chest and Abdomen	23
Feet (Weight Bearing) (D/P) (Podiatrists Only)	Foot	8
Dental Bitewing (Posterior)	Left Bicuspid and Molars	-
Dental Periapical	Central Incisor (Maxillary)	-
Dental Cephalometric (Lateral)	Head	15

These projections were selected to provide a useful cross-section of x-ray examinations encountered in private and institutional medical care facilities.

To obtain Organ Dose Indexes, a statistically representative sample of the healing arts x-ray facilities within a participating agency's jurisdiction is drawn by the participating agency on an annual basis.

During the inspection of a selected x-ray facility, the inspector determines which of the twelve selected examinations are performed most frequently on the machine being inspected. The inspector asks the operator to set the technique (milliamperage, kilovoltage, exposure time, target-to-film distance, collimation, etc.) that would be used for this standard patient. For example, if a Chest P/A is the most frequent examination performed with the machine in question, the operator is asked to set the technique that would be used for a patient having a 23 centimeter chest. Appropriate measurements of x-ray beam exposure, quality and beam size are made utilizing standardized procedures and equipment.⁹ A Mean Ovarian Dose and a Testicular Dose are then calculated from the measurement data using computer programs.

Preliminary Results

The NEXT Organ Dose Index System began October 1, 1972. At the end of the first nine months operation, 32 States and 3 Federal agencies were participating. Additionally, NEXT data has been processed for one foreign government and for an international health agency. At the end of nine months of operation (June 30, 1973), data for 3,431 projections had been collected in the United States and submitted for processing. Pre-edit and quality control checks designed to eliminate erroneous data are applied to all submitted data. 2,316 projections passed these checks and were entered upon the NEXT ODIS master file.

The present data pool is not yet complete. Many participating agencies have not completed surveying their annual representative sample. Not only is it too early to attempt an identification of trends, but the baseline has not yet been established.

Nonetheless, preliminary analysis of existing data does seem to validate some of the concepts, and expectations of the system.

For example, data is available for 291 cases of the Lumbo-Sacral (A/P) projections. Registered radiologic technologists performed 129 of these projections, the others being performed by practitioners or other persons. For these 291 applications of this projection to our standard patient:

1. The reported kVp ranges from 50 to 110,
2. The reported mAs ranges from 10 to 400,
3. The measured tube target-to-film distance ranges from 30 to 72 inches,
4. In view of the ranges in the above 3 categories it was not surprising to find the calculated exposure at skin entrance varied by 2 orders of magnitude.
5. X-ray Field Size at the film varied from well collimated beams limited to the spinal column, e.g., 5"x16" to large circular beams, e.g., 49", and even large rectangular beams, e.g., 22"x47",
6. In consequence of these variations, the Mean Ovarian Dose Index ranged from 4 mrad to 951 mrad and the Testicular Dose Index ranged from <0.5 mrad to about 2,300 mrad.

This kind of variation is not unique to the Lumbo-Sacral Spine examination. As another example, 42 cases of the Retrograde Pyelogram Scout (Cysto only) (A/P) were collected. This is a specialized projection rarely performed outside a hospital or major private practice facility. The x-ray machine operator in 34 of the 42 cases was a registered radiologic technologist.

1. The reported kVp varied from 68 to 90,
2. The reported mAs varied from 20 to 250,

3. The measured tube target-to-film distance varied from 31 to 59 inches,
4. The calculated exposure at skin entrance varied from 205 mR to 1,200 mR,
5. Beam size at the film varied from a 25" diameter circular beam to a 10"x13" rectangular beam,
6. The mean Ovarian Dose Index varied from 45 mrad to 507 mrad and the Testicular Dose Index varied from 1 mrad to about 1,000 mrad.

It is evident that the range of variation for this projection is smaller than for the Lumbo-Sacral Spine, but it is still quite large. As noted earlier, this projection is a specialized procedure restricted to a limited number of facilities. In comparison to the Lumbo-Sacral projection, a larger proportion of the operators were registered radiologic technologists and one may speculate on the influence of this factor.

Even so it is perplexing to find such wide variation in technique for the same examination for the same standard patient.

NEXT, Now and Future

These preliminary results suggest that opportunity does lie ahead for reducing unnecessary diagnostic x-ray exposure by identifying facilities using high exposure techniques. The NEXT Organ Dose Index System, by eliminating patient size variation and utilizing standard survey techniques, provides an objective method of accomplishing this.

Presumably, a radiological health agency effort, directed at identifying the high exposure facilities, followed by efforts to change their techniques, if successful, should be reflected by a trend, over time, to lower average organ dose indexes. The NEXT Organ Dose Index System will monitor these trends.

The NEXT Organ Dose Index System will not provide estimates of population dose, nor will it provide per capita dose information. It will provide information that can be used to effect changes in population dose. It will monitor changes in medical diagnostic x-ray application and trends in medical x-ray exposure.

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THE GENETICALLY SIGNIFICANT DOSE DUE TO MEDICAL X-RAY EXAMINATIONS IN THE NETHERLANDS

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Summary

Recent in situ measurements of the gonadal doses of 2500 male patients in seven Dutch hospitals make a reappraisal of the genetically significant dose (GSD) due to X-ray examinations in the Netherlands possible. It was found that the gonadal doses of nearly all examination types in all hospitals form a log normal distribution with a standard deviation of the same order of magnitude as the mean doses. The mean doses differ largely between the hospitals. Measurements in many hospitals will be necessary to obtain a reliable national mean dose for each examination type. The number of examinations for each type varies appreciably per district and needs careful consideration. The child expectancy, which did change drastically during the last 4 years, did not influence the GSD.

Introduction

The Genetically Significant Dose (GSD) due to X-ray examinations in the Netherlands was estimated by Beekman¹⁾ over 1959 and by Beentjes²⁾ over 1967. The former estimation resulted in a GSD of 7 mrem, the latter in different values, ranging from 19 - 40 mrem. Beekman used a set of relatively low gonadal doses obtained from phantom measurements with minimum beam size. In this respect her result is a minimum value. The number of examinations was obtained from the registers of all medical services in the city of Leiden and surroundings. For this confined population the total number of examinations was 0.55 per year in 1959, dental radiography and mass survey excluded. Beentjes used different sets of doses obtained from the literature and the phantom measurements of Beekman. The frequency of examinations was obtained from health insurance companies operating in different districts and covering 9 million people out of a total population of 13 million. The number of examinations per caput per year varies per district from 0.25 to 0.45 with a mean of 0.37. Dental radiography and mass radiography are excluded. The latter would add 0.21. Since no in situ dose measurements were available in our country we decided to perform a survey in the hospitals in order to obtain dose values as they really occur. The preliminary results from measurements on male patients now available permit some conclusions about the procedure to be followed for gonadal dose measurements and their influence on the GSD estimate. They will be discussed in this paper.

Methods

We used thermoluminescent dosimeters, consisting of three extruded lithium fluoride ribbons (1/8 x 1/8 x 0.035 inch) from Harshaw, wrapped up in plastic film. Up to this moment we performed measurements on male patients only. The dosimeter is attached to the scrotum during the X-ray examination. The measured exposure permits to calculate the testis dose if the following factors are properly taken into account.

- a) The difference between the skin exposure and the testis exposure is estimated to be about +5% in the direct beam and -5% in scattered radiation.
- b) A conversion from testis exposure to absorbed dose, chosen as 0.91 rad per röntgen.
- c) The sensitivity of LiF for diagnostic X-rays compared with 0.662 MeV ¹³⁷Cs radiation is estimated to be between 1,25 and 1.38.

The absorbed dose can be obtained from the exposure (calibrated with ¹³⁷Cs) through division by an overall correction factor, deduced from a, b and c between 1.55 and 1.40 for direct radiation and between 1.40 and 1.30 for the scattered radiation. An arbitrary value of 1.4 was chosen.

Results of the measurements

The results of the measurements on about 2500 male patients in seven Dutch hospitals are given in table 1.

The mean gonadal dose d_{ij} of each examination type (i) is indicated separately for each hospital (j) with the number of patients n_{ij} and the standard deviation s_{ij} of the doses. The X-ray departments of the hospitals have slightly different characters. They comprise one academic hospital, one military hospital and five peripheral hospitals in different cities. The hospitals numbered 3 and 4 did not use lead shielding at all, while hospital 2 always used lead shielding for the examination types IVP, lumbo-sacral region, pelvis and hip. In hospital 5 an image intensifier was consistently used for examinations (fluoroscopy and radiography) of the lower gastro-intestinal tract.

For each examination type and hospital the distribution of the measured doses proved to be log normal. An example is given in fig. 1 and 2.

The standard deviation (s_{ij}) is often larger than the mean dose (d_{ij}). Since the accuracy of each measured dose is better than 10 % (except of measurements at the edge of the direct beam where the localisation is uncertain) this standard deviation reflects the real difference between the individual doses. The relative standard deviation of the mean dose (d_{ij}) is

$$\frac{s_{ij} \cdot 100}{d_{ij} \sqrt{n_{ij}}} \%$$

and it is used to calculate an upper and lower confidence limit of the doses (95 % confidence) obtained in each hospital.

The mean gonadal doses for one examination type differ significantly between the hospitals. Local circumstances influence the doses to a large extent. For each examination type (i) the mean \bar{d}_i of the doses d_{ij} obtained in the 7 hospitals is calculated, together with its standard deviation s_i . From table 1 we see that s_i has the same order of magnitude as \bar{d}_i .

Calculation of the GSD

The measured gonadal doses make a reassessment of GSD possible.

Our first measurements happened to be in the hospitals 3 and 4 where relatively high doses were obtained. These results gave us the impression that the doses of Penfil and Brown ³⁾ are appropriate for the Netherlands. However, further measurements, performed in the other hospitals, make this conclusion doubtful. It appears that the data out of a restricted number of hospitals are subject to such a large variation that this causes one of the major sources of error in the estimation of the GSD, as follows also from the calculated standard deviation s_i of \bar{d}_i .

For the calculation of the GSD due to the examination of male patients we used the mean of the doses d_{ij} weighted according to the number of measurements performed in each hospital (see table 2, column 3). The frequency of examinations of each type is taken from Beentjes and the child expectancy over 1971 is used. At the bottom of the table a correction of 10% per year for the increase of the number of examinations during the years 1967-1971 is given ⁴⁾. The resultant value of the GSD, 28 mrad, is subject to a large error for the following reasons.

- 1e. The influence of errors in the doses d_{ij} on the GSD is estimated with the 95% confidence limits of the mean doses. The deviations of the contribution to the GSD of the eight examination types proved to be plus or minus 36%.
- 2e. A much larger error is to be expected due to the restricted number of hospitals used in the calculation, as is already mentioned. The standard deviation of the mean \bar{d}_i indicated that measurements in about hundred hospitals should be necessary to obtain this figure accurately, unless the hospitals can be classed into groups, for instance academic and peripheral hospitals. In order to study this point a calculation of the GSD was made with, for each examination type, the maxima and the minima of the mean doses d_{ij} found in all hospitals. The resultant GSD for the eight examination types was 19.8. and 1.2 mrad respectively for male patients over 1967. It seems unlikely that for male patients further dose measurements will lead to a higher GSD than 32 mrad over 1971, which is the maximum calculated in this way (table 2, column 4-7).
- 3e. The frequency of examinations of each type is not precisely known and influences the GSD to a large extent. Beentjes calculated the GSD on the basis of frequencies found in six different districts. For each examination type the contribution to the GSD in the six districts varies with a standard deviation between 30 and 40%.

We conclude that an estimate of the GSD for the whole country, obtained by multiplication of the mean doses with the mean number of examinations can lead to large errors.

Table 1. Gonadal doses of male patients per examination type (i) and hospital (j). Doses in mrad.

d_{ij} = mean gonadal dose \bar{d}_i = mean of the mean doses for examination type i
 s_{ij} = standard deviation s_i = standard deviation of d_i
 n_{ij} = number of patients

j \ i	Intravenous pyelography			Lower gastro-intestinal tract			Lumbosacral region			Abdomen (general)			Pelvis			Hip			Femur			Stomach		
	Hospital no.	d_{ij}	s_{ij}	n_{ij}	d_{ij}	s_{ij}	n_{ij}	d_{ij}	s_{ij}	n_{ij}	d_{ij}	s_{ij}	n_{ij}	d_{ij}	s_{ij}	n_{ij}	d_{ij}	s_{ij}	n_{ij}	d_{ij}	s_{ij}	n_{ij}	d_{ij}	s_{ij}
1	228	346	71	761	1154	15	204	324	290	65	164	45	893	901	53	—	—	—	560	630	12	6	5	100
2	86	216	238	107	104	90	30	111	127	—	—	—	99	168	8	36	24	17	23	36	6	24	30	9
3	1262	2560	102	367	728	134	—	—	—	136	264	46	—	—	—	—	—	—	—	—	—	24	28	134
4	1580	1871	64	106	71	56	1048	855	93	184	350	17	807	797	20	350	453	5	—	—	—	—	—	—
5	322	670	81	48	122	35	460	950	33	58	32	5	—	—	—	347	636	20	113	97	3	2	6	70
6	167	269	83	133	44	13	156	279	18	482	421	12	—	—	—	—	—	—	—	—	—	4	7	92
7	205	416	211	59	46	5	36	31	63	164	373	6	—	—	—	—	—	—	56	61	3	—	—	—
mean \bar{d}_i	550			226			322			182			536			244			188			12		
standard deviation s_i	606			259			388			156			377			180			251			11		
number of measurements n_i			850			348			624			131			101			42			24			405

Table 2. Different estimations of the GSD over 1971. GSD in mrad per year, gonadal doses in mrad.

Examination type.	Measured gonadal doses						Gonadal doses of Penfil and Brown 3)		Gonadal doses of Beekman 1)		
	mean doses		set of highest doses		set of lowest doses		Gonadal dose	GSD	Gonadal dose	GSD child exp. '67	GSD child exp. '71
	Gonadal dose	GSD	Gonadal dose	GSD	Gonadal dose	GSD					
<i>Hip</i>	223	0.27	350	0.43	36	0.044	1064	1.29	3323	4.06	4.04
<i>Femur</i>	307	0.67	560	1.22	23	0.050	96	0.21	91	0.20	0.20
<i>Pelvic region</i>	793	2.91	893	3.27	99	0.363	717	2.63	157	0.58	0.58
<i>Lumbosacral region</i> (lumbar spine and abdominal aortography included)	290	1.84	1048	6.69	30	0.191	2268	14.46	60	0.32	0.32
<i>Intravenous urography</i> (retrograde urography and urethro cystography included)	411	1.43	1580	5.51	86	0.299	2091	7.29	640	2.11	2.10
<i>Abdomen (general)</i>	148	0.29	482	0.94	58	0.112	254	0.49	92	0.18	0.18
<i>Lower gastrointestinal tract</i>	229	0.49	761	1.63	48	0.102	1585	3.39	45	0.10	0.10
<i>Stomach and duodenum</i> (oesophagus included)	11.2	0.09	24	0.19	2	0.016	137	1.10	4.8	0.03	0.03
Total		7.99		19.88		1.177		30.86		7.58	7.55
Other examinations		2.01		2.01		2.01		2.01		0.03	0.03
Total GSD for males		10.00		21.89		3.19		32.87		7.61	7.58
Total GSD for females		9.13						8.88			9.13
Total		19.13						41.75			16.71
Total corrected for increase of number of examinations '67 - '71		28						61			24

- 4e. In our country the child expectancy is decreasing rapidly during the last years. In order to get an idea of the influence of this factor we made two calculations of the GSD (male patients), one with the child expectancy over 1967 and one over 1971. The estimations were calculated following the method of Beentjes; for both years the set of doses reported by Beekman and the frequency of examinations of Beentjes was used. Although there is a strong shift in the number of live births and child expectancy in all age classes (see table 3), the resultant differences in the calculated GSD are insignificant (7.58 and 7.55 mrad for male patients over 1967 and 1971 respectively).

Table 3.
Male child expectancy in the Netherlands

age	1967	1971
-0.75 - 0	2.644	2.232
0	2.676	2.255
0 - 4	2.720	2.307
5 - 9	2.742	2.314
10 - 14	2.748	2.319
15 - 19	2.745	2.309
20 - 24	2.564	2.071
25 - 29	1.921	1.405
30 - 34	1.080	0.809
35 - 39	.494	0.294
40 - 44	.186	.100
45 - 49	.056	.028
50 - 54	.014	.005
55 - 59	.002	—
60 ⁺	—	—

Deduced from tables of the Netherlands Central Bureau of Statistics.

Conclusion

In spite of 2500 gonadal dose measurements and intensive efforts to obtain the frequency of each type of examination per year the estimation of the GSD is still unreliable. Improvement is only possible if an appreciable number of measurements in many hospitals is available. Relatively large statistical errors in the dose measurements may be tolerated due to the large standard deviation in the dose distributions. In our country the rather important changes in child expectancy did not influence the GSD of the male patients significantly.

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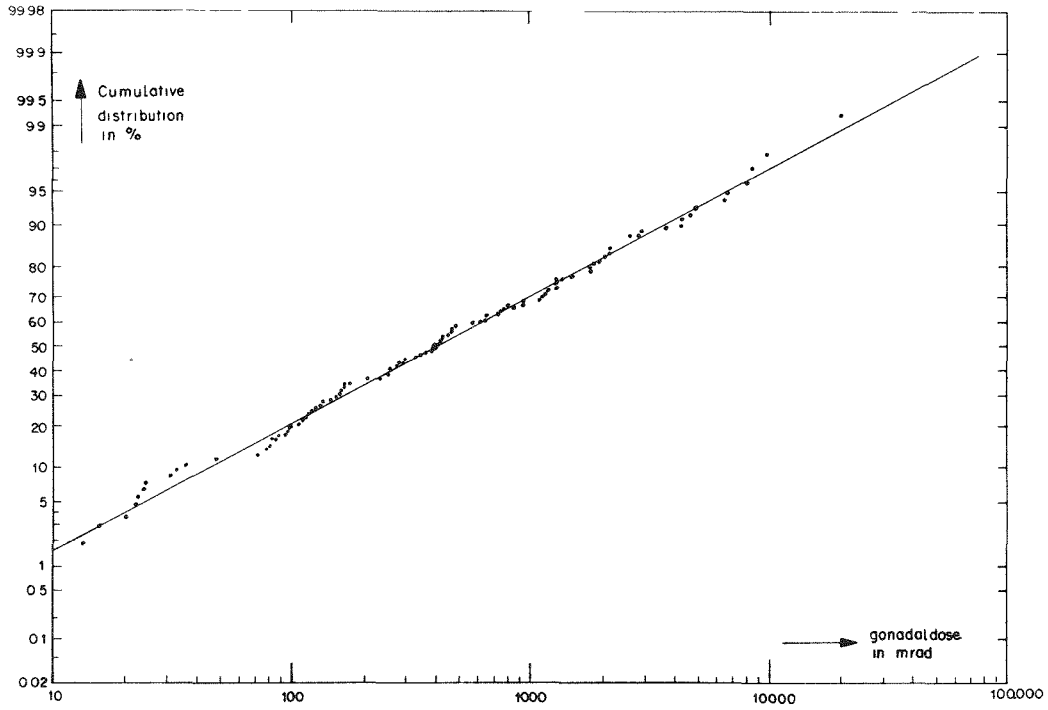


Fig. 1. Cumulative distribution of male gonadal doses from intravenous pyelography; hospital no. 3; $d_{ij} = 1300$ mrad; $s_{ij} = 2600$ mrad; median = 370 mrad; 102 patients.

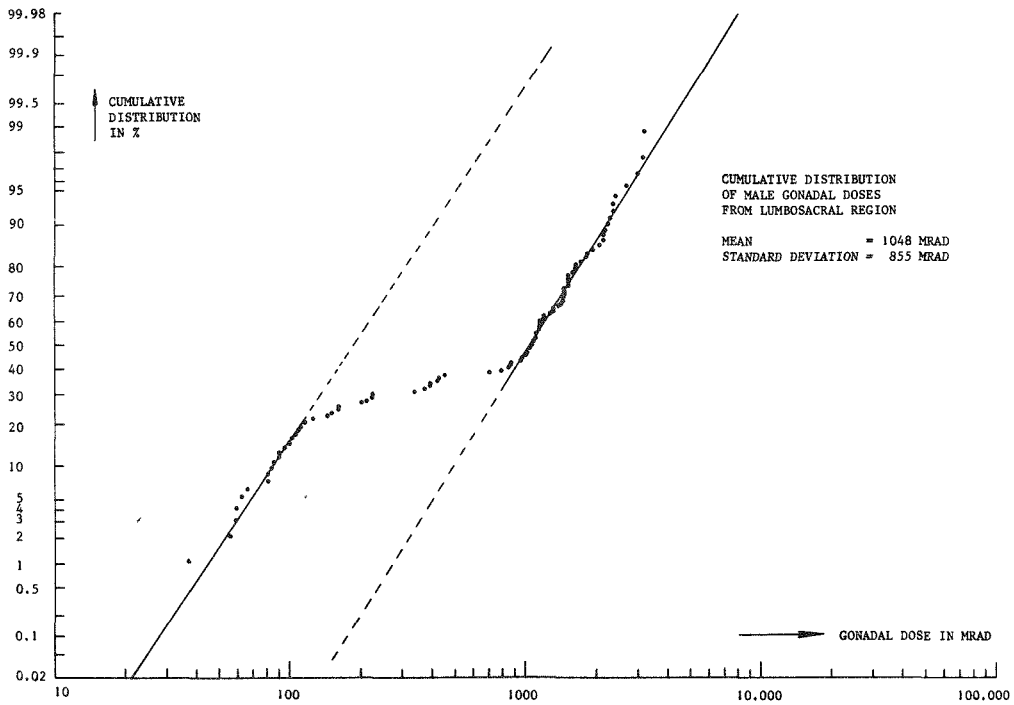


Fig. 2. Cumulative distribution of male gonadal doses from intravenous pyelography; hospital no. 4; $d_{ij} = 1048$ mrad; $s_{ij} = 855$ mrad; median = 1079 mrad; 93 patients. This examination type forms the unique exception on the log normal distribution found in all other types.

EVALUATION OF DIAGNOSTIC X-RAY CONTRIBUTION TO
THE ANNUAL GENETICALLY SIGNIFICANT DOSE
EQUIVALENT OF TAIWAN URBAN POPULATION

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Abstract

Thermoluminescent dosimeters (LiF-*teflon* discs) were distributed to clinics and hospitals using X-rays for diagnostic purposes in Hsinchu which is a medium size city of Taiwan with a total population of 213,735. The dosimeters were placed inside a small pocket stitched on a piece of cloth. Before irradiation the patient was covered with this cloth of which the location of the pocket was adjusted to be against the gonadal region of patient's body.

The detailed record of the irradiation conditions and the status of patients were provided by the clinics and hospitals concerned. Dosimeters were replaced and read in a one-month interval. Based on the formula given by the United Nations Scientific Committee of the Effects of Atomic Radiation the annual genetically significant dose equivalent was calculated with proper corrections for gonad dose of both sexes.

Since one half of Taiwan population is living in the cities nowadays, and surveys of medical radiation sources in Taiwan have been carried out twice since 1970, the annual genetically significant dose equivalent calculated for Hsinchu is extended to Taiwan urban population. It shows that the annual genetically significant dose equivalent of Taiwan urban population was in the range of 3 to 4 mrem in 1972.

Introduction

The hazard of radiation exposure of most concern has shifted from relatively high doses received by a few people to low doses received by a large segment of the population. The diagnostic X-rays belong as a contributor to the latter category. This paper is concerned with doses from diagnostic X-ray received by gonads of the Hsinchu population during an eight-month investigation period as recorded by thermoluminescent dosimeters.

Hsinchu is a medium size city of Taiwan with a total population of 213,753 (1972 census). Of all inhabitants 113,921 are male and 99,814 are female. The medical radiation sources surveyed in 1970 and again in 1972 show that there are 15 diagnostic X-ray units in operation, i.e., 70 X-ray units per 10^6 population.¹ Of the 15 hospitals and clinics equipped with diagnostic X-ray units, only 5 major hospitals or clinics have frequent use of the equipment. The other 10 hospitals and clinics have a few exposures taken every year. Based on this information, the

major hospitals and clinics were chosen for this investigation and the characteristics of their X-ray units are shown in Table 1.

Table 1. The Manufacturers, Applied Voltages, and Beam Currents of X-ray Units in Hsinchu Major Hospitals and Clinics.

Hospital No.	Manufacturer	Filter (mm Al)	Radiography			Fluorography		
			kVp	mA	sec	kVp	mA	sec
1	Aroma	2.0	95	300	0.1	95	4	60
2	Shimadzu	2.0	60	300	0.5	60	2.5	180
3	Toshiba	2.5	100	500	0.05	70	3	60
4	Toshiba	2.5	70	200	0.25	70	2.5	120
5	Picker	2.5	75	100	0.05	70	3	60

Procedures

The 13 mm diam. x 0.4 mm thick discs of ^7LiF -Teflon of Teledyne Isotopes were used because of the consideration of the maximum sensitivity. The minimum dose defined as three times the standard deviation of the background is 15 mrad. The tissue-equivalence of LiF -Teflon dosimeters enables meaningful estimates of dose in X-ray radiation from a single measurement of a dosimeter at the monitoring site. The response is independent of photon energy to within $\pm 30\%$ down to 20 keV. In addition the LiF -Teflon discs are unaffected by extremes of humidity and environmental temperatures which do exist in Taiwan. Readout LiF -Teflon discs was performed on Teledyne Isotopes Model 7100 TLD instrument. Nitrogen was supplied to suppress the spurious thermoluminescence during readout procedure. The standard deviation was about 3.6%.

Two LiF -Teflon discs each were placed inside a small pocket stitched on a piece of cloth for radiography and fluorography, respectively. Before irradiation the patient was covered with this cloth of which the location of the pocket was adjusted to be against his-her gonadal region. The detailed record of the irradiation conditions and the status of patients were replaced and read in a one-month interval. Table 2 present the results of an eight-month investigation period in 1972.

Table 2. The Average Exposure per Capita due to Diagnostic X-rays Monitored with LiF-Teflon Discs at the Gonads.

Month	Hospital No.	mR/man-exposure		Number of Patients							
				Radiography				Fluorography			
		Radio- graphy	Fluoro- graphy	Age			Total	Age			Total
				<18	18-45	>45		<18	18-45	>45	
1	1	28	243	11	141	31	183	1	2	3	6
	2	15	303	15	28	13	56	1	4	4	9
	3	16		10	17	42	69				
	4	62	273	3	28	13	44		7	5	12
	5	25	242	5	46	5	56	5	18	14	37
2	1	32	218	32	261	42	335	2	2	2	6
	2	78	233	12	36	27	75		7	9	16
	3	11		2	35	63	100				
	4	7	453	1	103	24	128		13	6	19
	5	42	363	4	18	6	28	5	63	5	73
3-4	1	23	145	65	476	67	608		3	4	7
	2	48	414	17	90	50	157	1	16	15	32
	3	38			24	81	105				
	4	17	487	4	98	49	151		34	10	44
	5	37	212	10	77	22	109	44	229	56	329
5-6	1	30	342	83	1101	52	1236	1	2	6	9
	2	46	320	25	99	75	199	1	19	7	27
	3	18			5	22	27				
	4	22	256	1	207	82	290	1	54	18	73
	5	38	285	9	68	23	100	45	210	61	316
7-8	1	25	246	90	1320	48	1458	1	3	9	13
	2	40	381	30	110	86	226	1	20	8	29
	3	32		2	30	88	110				
	4	36	298	4	102	67	173		51	11	62
	5	40	240	13	86	31	130	46	253	61	360
Average or Total		32	298	448	4606	1109	6153	155	1010	314	1479

The annual genetically significant dose equivalent to the population is a measure of the genetic significance of the yearly dose equivalent received by the population's reproductive organs (gonads). To calculate this dose equivalent, one should consider the gonad dose and the future number of children expected by each member of the population as listed in Table 3.

Table 3. Age-Group and Average Expected Children Census

Age	Population (%)	Average Expected Children
0-18 years		
(M)	19.9	4.00
(F)	18.4	4.00
18-45 years		
(M)	23.6	2.30
(F)	21.7	2.30
>45 years		
(M)	9.8	0.015
(F)	6.6	0.015

Table 3 was taken from population and birth rates census data released by the National Health Administration of Republic of China.

The genetically significant dose equivalent described here has the same meaning as that given in the report of the UNSCEAR.² The genetically significant dose equivalent can be calculated with the following formula:

$$D = \frac{\sum_j \sum_k (N_{jk}^{(F)} W_{jk}^{(F)} d_{jk}^{(F)} + N_{jk}^{(M)} W_{jk}^{(M)} d_{jk}^{(M)})}{\sum_k (N_k^{(F)} W_k^{(F)} + N_k^{(M)} W_k^{(M)})} \quad (1)$$

where

D = annual genetically significant dose.

N_{jk} = number of individuals of age-class k, subjected to class j exposure, i.e., either radiographic or fluorographic X exposure.

N_k = total number of individuals of age-class k.

W_{jk} = future number of children expected by an exposed individual of age-class k subsequent to a class j exposure.

W_k = future number of children expected by an average individual of age-class k.

d_{jk} = gonad dose per class j exposure of an individual of age-class k.

(F) = female.

(M) = male.

Since the radiation levels and exposure frequency from diagnostic X-rays are quite low, the number of expected children will be the same for individuals after irradiation as it was before. Therefore, for the purposes of these calculations W_{jk} will be assumed to be the same as W_k .

For calculation of male and female gonadal dose, the correction factors K_m and K_f can be used, where

K_m = Depth dose at male gonads/skin exposure,

K_f = Depth dose at female gonads/skin exposure.

The central axis depth dose factors used to calculate depth dose to gonads from air dose were $K_m = 72\%$ and $K_f = 11\%$.³

During the investigation period, it was assumed that the number of exposures taken was equal to the number of patients being examined. According to the data provided by the hospitals and clinics, patients under age 18 were less than 10% of all patients concerned. Hence, it was assumed that the gonad dose was independent of age-class.

Results and Discussion

The gonad doses thus obtained were as follows:

1. Radiography
 $d^{(F)} = 3.52$ mR
 $d^{(M)} = 23.04$ mR
2. Fluorography
 $d^{(F)} = 37.78$ mR
 $d^{(M)} = 214.56$ mR

For practical purposes, in X-ray diagnosis, an exposure of 1 R can be regarded as delivering to soft tissue a dose of 1 rad or a dose equivalent of 1 rem.⁴ The annual genetically significant dose equivalent in Hsinchu City due to diagnostic X-ray only was 3.83 mrem in 1972.

The major cities in Taiwan are Keelung, Taipei, Taichung, Tainan, and Kaohsiung with a total population of 5,034,267 of which 2,683,264 are male and 2,351,003 are female. The number of diagnostic X-ray units is known.¹ Based on the data surveyed at Hsinchu City, it can be estimated that the annual genetically significant dose equivalent due to diagnostic X-rays in major cities of Taiwan was 3.64 mrem in 1972 while that of U.S.A. was 5 mrem. Since one half of Taiwan population is living in the cities nowadays, it is concluded that the annual genetically significant dose equivalent due to diagnostic X-rays of the Taiwan urban population was in the range of 3 to 4 mrem in 1972.

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THE ROLE OF THE HEALTH PHYSICIST IN REDUCING MEDICAL RADIATION

by

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Abstract

Medical use of x ray continues to be the greatest manmade source of exposure to the United States population, in spite of substantial improvements in equipment and technique. Reduction of this exposure is possible through training and motivation of allied health professionals.

In addition to proposed x-ray equipment standards and current upgrading of technologist proficiency criteria, increased involvement of health physicists is needed. This paper examines the role of health physicists in the health care environment, stressing their potential for effecting exposure reduction through close working relationships with allied health professionals.

Health Physics emerged as a child of the nuclear age even before the first atomic bomb was exploded. As the use of atomic energy rapidly increased, so did the concern for the hazardous consequences of misuse and carelessness. Although the term Health Physicist may not have been used as it is today, scientists and engineers became increasingly involved in the problems of radiation protection and control. This was a logical development in view of the potentially high levels of radiation exposure to man, not only from the obvious threat of nuclear warfare but also from the rapid increase in radiation use by industry and research.

It is interesting to note, though, at the beginning of this nuclear age, medical radiation had already been in use for half a century, ever since Wilhelm Roentgen in 1895 accidentally discovered the mysterious rays that could penetrate matter including human tissue, and despite the extensive use of medical x rays for many years, only in the past few decades has there been an emerging medical specialization from the ranks of the radiation physicists.

In the scientific community, specifically that portion concerned with the uses of radiation, it is generally accepted that the medical and dental use of x rays constitutes the greatest single source of manmade radiation exposure to the general population. Yet, a relatively small percentage of the overall effort for safe, effective use of ionizing radiation is directed toward medical use. Since the Health Physicist has selected a profession which is dedicated to the prevention of unnecessary exposure to people, it follows that the Health Physicist would want to do something about the source of 90%¹ of the manmade radiation exposure. In 1971, there were approximately 3,000 members of the Health Physics Society. A statistical analysis of 2,862 members was done from application form information. An item asking for area of professional interest by first, second, and third choice resulted in the following figures: As a first choice, only 4.5% indicated the medical field, and ranked behind Health Physics (52.3%), Physics (8.7%), Biology (6.5%), Chemistry (5.7%), Engineering (4.8%), and other areas not specifically listed (6.0%). Even as the second and third choice, medicine was only 5.4% and 5.9% respectively.²

Dr. Dade Moeller, who conducted this analysis, offers this general comment:

"It would appear that a Society with such a high percentage of members with undergraduate backgrounds in physics would have many of the qualifications necessary for making a major impact in the field of medical radiation physics and the control of associated x-ray exposures. This is particularly true in light of the fact that the Society has such a high percentage of people with Doctoral degrees (18%) of whom well over a fifth are medical doctors and dentists. Undoubtedly, the reason that the Society has not been more active in this field is that most graduate programs in radiation protection, as currently organized, simply do not offer the opportunity for Health Physicists to receive the specialized training required for professional work in Medical Physics."³

We would add that perhaps an equally important reason is a general lack of communication between the health physics profession in general and the majority of those scientists (be they Physicists, Physicians, Biologists or whatever) presently working primarily in the area of medical radiation. There is a need for greater awareness of and active involvement in medical radiation exposure problems by Health Physicists.

A recent manpower study⁴ estimates that in 1971 there were approximately 500 physicists working in the medical field and that double this amount would be required by the end of this decade.

Considerable public awareness and concern has been generated regarding radiation pollution of the environment by nuclear power plants. Much effort and money has been spent in the analysis of real and potential hazards of radiation exposure from nuclear power. But how does this compare to medical radiation exposure? The following statements from a study by the National Academy of Sciences (BEIR Report) can give one an idea of the relative magnitude of the two problem areas:

"Based on experience to date and present engineering judgement, the contribution to radiation exposure averaged over the U. S. population from the developing nuclear power industry can remain less than about 1 mrem per year (about 1% of natural background) and the exposure of any individual kept to a small fraction of

background." [Provided certain controls are maintained.]
[Whereas] "In the foreseeable future, the major contributors to radiation exposure of the population will continue to be natural background with an average whole body dose of about 100 mrem/year and medical applications which now contribute comparable exposures to various tissues of the body."¹

We derive great benefit from medical radiation in terms of public health and therefore we accept a certain degree of risk. This is a reasonable argument which will usually meet with little resistance and which is applicable to many areas of public concern such as the use of automobiles or the taking of drugs. The criteria, though, in each case, is to maintain an imbalance in the benefit/risk scale--that is, the benefits must outweigh the risks. There must be a constant effort to minimize the risks without adversely affecting the benefits.

The increase in numbers, types and complexities of medical radiation procedures challenges the allied health professionals just to keep pace with the medical and logistic considerations before one even considers the radiation protection problems which may be involved. We believe that increased efforts by Health Physicists who can develop good working relationships and communication lines with the medical professionals can further tip the scale to the benefit side.

The benefits of medical radiation are well known and need no elaboration. But just what are the risks? The answer, of course, is unknown for a particular individual involved in a particular type of medical radiation procedure. One can only talk about probabilities when large numbers of people are subjected to low levels of radiation, such as those used in the range of medical procedures. Even then, lack of sufficient human data precludes accurate predictions.

The BEIR Report contains this consensus regarding risks from radiation:

"Until recently, it has been taken for granted that genetic risks from exposure of populations to ionizing radiation near background levels were of much greater import than were somatic risks. However, this assumption can no longer be made if linear non-threshold relationships are accepted as a basis for estimating cancer risks. Based on a knowledge of mechanisms (admittedly incomplete) it must be stated that tumor induction as a result of radiation injury to one or a few cells of the body cannot be excluded. Risk estimates have been made based on this premise and using linear extrapolation from the data from the A-bomb survivors of Hiroshima and Nagasaki, from certain groups of patients irradiated therapeutically, and from groups occupationally exposed. Such calculations based on these data from irradiated humans lead to the prediction that additional exposure of the U. S. population of 5 rem per 30 years could cause from roughly 3,000 to 15,000 cancer deaths annually, depending on the assumptions used in the calculations. The Committee considers the most likely estimate to be approximately 6,000 cancer deaths annually, an increase of about 2% in the spontaneous cancer death rate which is an increase of about 0.3% in the overall death rate from all causes."¹

Adopting the assumption of linear relationship between dose and biological damage then, the prudent course is to minimize all unnecessary or unproductive exposure. Two predominant types of unproductive radiation exposure in

the medical realm are radiation from procedures considered medically unnecessary, as in the case of patient self-referral, and radiation from improper performance of equipment and/or operator.

Both types of unproductive radiation exposure are sensitive and highly controversial issues that can be dealt with and eliminated without a decrease in medical benefit to the patient.

The use of radiation in medicine is usually divided into three general categories: Diagnostic Radiology, Radiotherapy, and Nuclear Medicine. Reference 3 indicates that Physicists working in medicine devote the greatest effort to Radiotherapy, with the remaining effort about equally divided between Diagnostic Radiology and Nuclear Medicine. The remainder of this paper is concerned specifically with the area of Diagnostic Radiology. Diagnostic x rays involve the greatest segment of the population.

Approximately one hundred and thirty million persons had one or more diagnostic x-ray examinations done in 1970* with an estimated 660 million radiographic films being taken. Data taken from the 1970 X-ray Exposure Study is now being compared with the earlier 1964 Study. Preliminary figures seem to indicate that average exposure to the population may have been reduced. It is comforting to know that progress has been made. However, we believe that much greater reduction is still possible.

In the scope of a diagnostic x-ray examination, there are three main functions:

- 1) selection of the patient
- 2) performance of the examination
- 3) interpretation of the results

Health Physicists can, by active involvement, be the catalyst in improving the use of medical radiation on the public. By investigating and analyzing uses and abuses of medical radiation in these three functions of an examination, physicists can and must take decisive action to influence changes as required to minimize exposure to the population. One of the immediate areas that requires change is the chest x-ray screening procedures for cardiopulmonary disease. As you well know, this method of screening has been used for many years. It has recently been clearly identified as a procedure that should not be done. In view of the undesirability of using radiation without clear evidence of significant benefit, the liaison committees of the American College of Radiology, the American College of Chest Physicians and the United States Public Health Service issued on February 18, 1972, a policy stating:

"Community chest x-ray surveys among the general population as a screening procedure for the detection of tuberculosis, other pulmonary disease and heart disease are not productive and should not be done."

With reference to the detection of tuberculosis, the policy states that chest x-ray examinations should be restricted to individuals evidencing a positive reaction to the tuberculin skin test. Mass chest screening procedures should be eliminated also, because they are examples of the practice of patient

*Preliminary estimates from the U. S. Public Health Service 1970 X-ray Exposure Study.

self-referral, that is the patient himself, not a qualified physician, decides he should have an x-ray examination. This is undesirable because the physician is much more capable of weighing the benefits to be derived against potential risk.

This policy statement has been effective in discontinuing mass screening surveys in some areas, but has not completely eliminated the situation. Health Physicists should investigate the practices in their own areas and individually and through their societies make sure that the mobile chest vans are located in those areas only where there is the possibility of high incidence of chest disease.

Another area which should be given strong consideration by the physicists is the extensive use of pre-employment chest x rays. This again is the use of x rays primarily for the detection of tuberculosis and is used extensively as a prerequisite in hiring of food handlers. This is another situation where the x radiation is applied to people indiscriminately, many of them very young, without prescription by a physician. Although there is admittedly a higher risk of the spread of tuberculin infection through food handlers, the criterion for a chest x ray should still be a positive skin test. By coordinating their efforts the physicists can affect this situation by investigating the reasons for pre-employment chest x rays and suggesting methods that would obtain the same results but without the use of radiation.

The second function of a diagnostic examination "performance of the examination" is where the scientific and technical knowledge of the Health Physicist can be put to very great use. The taking of a radiograph involves operation of complex equipment usually with many options and variables under the control of the operator. In many cases, although the operator may be knowledgeable and skilled in particular areas of health care, he or she may have little or no knowledge of the physical principles involved in the production of x radiation. If there is no real understanding of what happens when the button is pushed, this operator cannot effectively exercise the options available in order to obtain maximum benefit with the least radiation exposure to the patient.

Health Physicists can improve this situation by exercising their role as teachers and educators. They should become an integral part of the education of all those who apply radiation to other humans. If greater awareness of and concern for radiation safety can be instilled at the grass roots level through teaching in the medical, dental, and x-ray technology schools, the more difficult task of correcting poor practices can be considerably lessened.

A similar issue that Health Physicists could become involved in is that of credentialing of operators of radiation emitting equipment. In the United States presently there are only 3 States that have mandates requiring the licensure of x-ray machine operators. Although the licensing procedures vary in these States, all of them require by law that operators meet certain minimum educational requirements and possess knowledge and skills required to deliver health care x-ray services with minimum amounts of radiation. Health Physicists can play an important role in this issue by becoming aware of the provisions in the licensure bills on the State and Federal levels and assume the responsibility to influence these bills with regard to reducing unnecessary exposure.

The third function "interpretation of the results" - that is making a diagnosis from a radiograph - is the responsibility of the physician. However, the physician's interpretation is limited by the quality of the radiograph which he interprets. Proper performance of the operator and the equipment are essential to quality radiographs. It is true that one can use poor radiation

safety practices and still obtain diagnostically acceptable radiographs. However, most methods used to minimize exposure will also improve the film quality. This is an important fact which much be emphasized to the users of diagnostic x rays. A false assumption by many is that a reduction of exposure to the patient is automatically accompanied by a reduction in quality or diagnostic information on a radiograph. Here again, the Health Physicist through educational methods, can help to modify such beliefs.

We have talked primarily about exposure to the general population meaning the patients undergoing radiographic examinations. With the development of higher energy equipment and complex procedures which require both a large number of successive radiographs and the presence of greater numbers of health care specialists, occupational exposure becomes an increasing problem. In many of these procedures, serious medical conditions exist and far outweigh radiation exposure hazards to the patient. However, the health professionals who perform these procedures on a daily or weekly basis, can be subjected to extremely high exposures unless constant safeguards provided through innovative techniques, equipment modifications and procedural analyses are established and maintained. For the professional, the cost of the exposure would of course not be compensated for by the benefit of the treatment.

A fourth area where Health Physicists may be very effective is in the area of assistance to the State and local radiation control programs. An effective dialogue between the two groups can result in an interchange of experience which can lead to the effective solution of common problems and assist the local radiation control programs in broadening their efforts beyond facility compliance with State codes. A mutual effort at working with users of medical x ray can lead to a significant reduction in the unnecessary exposure received by the population.

The challenge is there. We believe that the Health Physicist with a desire to enter the medical field can meet it. He need only take heed of the following principles:

1. Be sure that the problem being tackled is recognized as such by others involved.
2. Try to find solutions to exposure problems which also improve the quality of the radiological service.
3. Be sensitive to changes in attitudes of the medical profession toward recognizing the health benefits of improved radiological practices.
4. Be persistent but not with the "hard sell" approach.

In general, the medical profession is open to many changes in radiologic practice, even more so when the changes result in meaningful improvements in the quality of the service they perform. Sell yourself as a service and making the changes which they recognize as valuable to their department will make the elimination of unnecessary exposure easier to achieve.

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DENTAL TECHNIQUE NORMALIZATION:
DEVELOPMENT AND PILOT TEST

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Abstract

A two phase program designed to help control excessive radiation exposure in dental radiography is described and the results of a two state pilot test presented. The first phase is a data collection system for measurement of radiation exposure from dental x-ray machines and the identification of machines producing excessive exposures. It was found that 54% of the machines surveyed have outputs within "acceptable" exposure limits. Personal in office visits to the other 46% constitutes the second phase of the program. The cause of the excessive exposure was identified and proper radiographic techniques were presented. The result was a reduction of 80.4% in the average exposure at end of cone (mR/film),

Introduction

The Bureau of Radiological Health, FDA, in conjunction with the radiation control agencies of several States has developed and tested a program using thermoluminescent dosimeters (TLD's) to help control excessive radiation exposure in dental radiography. The magnitude of this problem has been discussed and documented in literature. The skin entrance exposure for the average adult dental bite-wing examination has been shown to vary tremendously from facility to facility.¹ The PHS X-ray Exposure Study of 1970² indicated that dental exposures range from 20 to 13,000 mR even in facilities which comply with appropriate local State and federal radiation standards. These programs have traditionally been oriented to equipment and facility regulation and reduction of occupational exposure. As a result the user and his impact on the patient dose has not been directly affected. This program has been developed to deal directly with the user to improve his performance, and lower patient x-ray exposure.

The program described in this paper has been designed for use by radiation control agencies and provides methodology for reducing population exposure from dental radiography to levels in accord with good radiographic technique.

This program consists of two phases: identification and action. The major emphasis of this discussion concerns the data collection system for measurement of radiation exposure from dental x-ray machines and the identification of machines producing excessive exposures. An action program designed to reduce these excessive exposures will also be presented. The development of the system and the results of a pilot test in two states are presented.

Methodology

The thermoluminescent dosimeter in the form of lithium fluoride (LiF) chips has, in recent years, found many applications in the field of radiation measurement. This project was undertaken to determine the effectiveness of mailing TLD's in a data collection and screening system for dental x-ray exposures. Specifically we wished to determine whether the information collected by such a method could be used to determine and set priorities in such a way as to accomplish the greatest exposure reduction with the most efficient use of time and resources of state radiation control personnel.

Acceptable Dental Exposure Range

As a first step in the development of this program, it was necessary to determine acceptable exposure levels for diagnostic dental radiographs. A search of the literature was conducted and a laboratory investigation performed to establish these levels.

Travis and Hickey³ have empirically determined values for the exposure at the tip of the cone which are useful as a guide to the production of diagnostic quality dental radiographs. They reported a single tip of cone exposure value, capable of producing a diagnostic quality dental radiograph, for selected operating kVp values.

It is known, however, that for a given kVp and properly filtered x-ray beam, there exists a range of tip of cone exposure values which produce radiographs of acceptable diagnostic quality. In order to determine this range, laboratory studies were conducted on a dental phantom consisting of the mandibular and maxillary sections of a human skull imbedded in a transparent, non-granular plastic which has the same absorption and secondary radiation-emitting characteristics of living tissue. The part of the phantom that would correspond to the inside of the mouth was notched to accept a standard (1-1/4" x 1-5/8") dental x-ray film positioned behind the teeth to represent the bite-wing examination.

A series of radiographs were then produced at selected kVp settings from 45 to 90 kVp to represent the operating kVp found on most dental units in dental offices. The exposure range at each kVp setting was broad enough to produce films too light to be of diagnostic quality at the lower exposure and too dark to be of diagnostic quality at the upper exposure levels. The tip of cone exposure was measured using Harshaw LiF (TLD-100) chips and read on an Eberline Model TLR-5 Reader. Exposure values were recorded for each radiograph. A panel of 24 dentists read these radiographs and selected those they felt were diagnostically acceptable. All radiographs were produced using speed group "D" dental film and were processed using fresh solutions and the time and temperature developing technique recommended by

the film manufacturer. Speed group "D" film was used because of its wide acceptance by the Dental Profession, and its ability to produce high quality diagnostic radiographs at reduced patient exposure. Figure I illustrates the range of cone tip exposure that yields diagnostic radiographs as a function of operating kVp. The upper curve is the maximum exposure. With proper development, overexposure results in a black film and underexposure produces a film which is too light. Figure I clearly indicates that there is a finite exposure range which will produce dental radiographs of acceptable diagnostic quality.

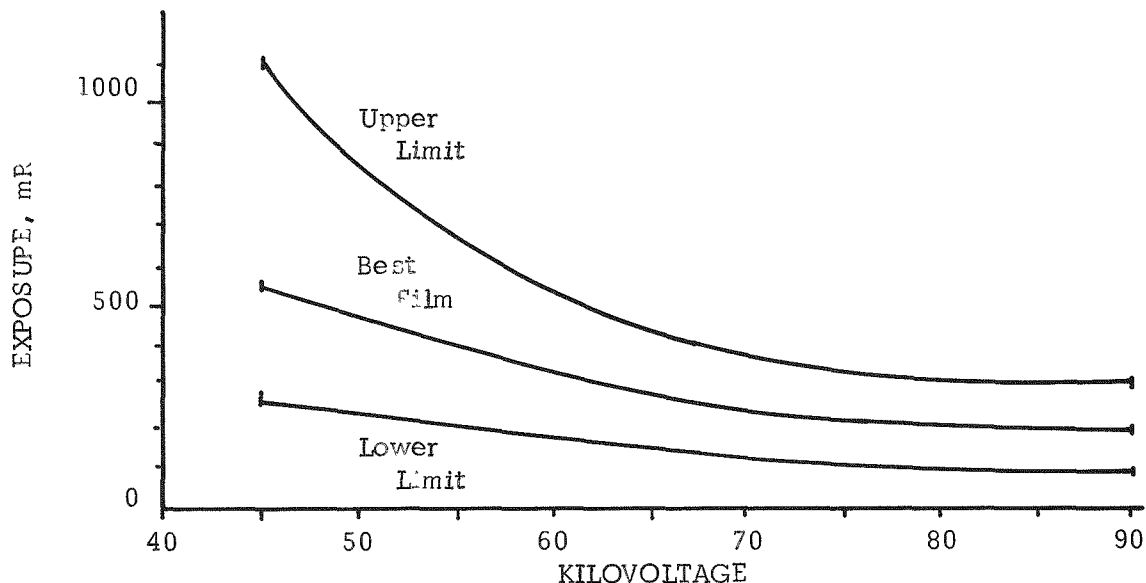


FIGURE 1 DENTAL EXPOSURE RANGE TO PRODUCE RADIOGRAPHS OF DIAGNOSTIC QUALITY

Identification Phase

The program described in this paper was developed to answer two questions: 1) how many dental x-ray machines used in the routine practice of dentistry fall within the acceptable exposure range; and 2) how can these specific machines producing excessive radiation output be efficiently identified so corrective action can be taken? To find the answers, Radiation Control Program Directors in two states, Rhode Island and New Hampshire, were invited to participate with the Northeastern Radiological Health Laboratory in the development and testing of a dental inspection program which made use of a mailable card system to obtain cone tip exposures with thermoluminescent dosimeters.

A 5" x 7" card (folded to 3-1/2" x 5") was designed to carry three TLD's. An identification number, such as the machine registration number, was written on the card before mailing to relate the exposure to a specific machine. A card was sent for each x-ray machine registered by the dentist in the participating states. Instructions and a sketch indicating how to properly make the end of cone exposure were printed on the card and spaces for the exposure data were provided. The data collected includes the radiographic

technique and film type normally used. A cover letter of instructions was included briefly describing the program and inviting the dentist's participation. Upon receipt of the card, the dentist was requested to expose the TLD's attached to the card and return the card or cards, with TLD's still attached, in a self-addressed stamped envelope to the Health Department for reading and evaluation. For this pilot test all TLD's were read at the Northeastern Radiological Health Laboratory.

Action Phase

Once the dental exposures had been determined, an action program to correct deficiencies was begun. In this study, the individual State Radiological Health Program Directors identified the exposure level at which corrective action was to be taken. This action point could be a constant value for all operating kVp or a sliding scale depending on the exposure range required for a given kVp. Any machine producing an exposure greater than required for diagnostic radiographs based on the operating kVp could be visited. Priority in this system normally was given to those facilities having the potential for the greatest exposure reduction to the population.

Field visits to offending x-ray units showed that the chief causes of excessive radiation are: 1) use of slow speed film; 2) overexposure and underdevelopment (sight development) of film; or 3) use of depleted developers. During a field visit, the inspector identified the cause of the excessive exposure, informed the dentist of the problem and took the necessary corrective action. The success of this action phase of the program lay in the approach the inspector used during the visit. The dentist was instructed in how to produce quality radiographs at lower patient exposure and was left with an improved radiographic technique.

During the inspection visit the x-ray machine output was adjusted to produce an end of cone exposure which fell within the acceptable exposure range presented in Figure 1. The inspector made the necessary adjustments of the exposure time, x-ray tube current (mA), voltage (kVp), or installed additional aluminum filtration to obtain the required exposure. A recommended technique was left for each machine after it had been adjusted. The new technique would produce an acceptable radiograph for all common dental exams when processed in fresh developing solution at 68° for 5 minutes.

The value of the new technique was reinforced by asking the dentist to witness a demonstration. Two bite-wing radiographs were taken of the dental phantom; first using the equipment, exposure technique, film and processing technique routinely employed by the dentist; the second radiograph was made using speed group "D" film and the newly recommended exposure and processing techniques. Before the films were processed the dentist's darkroom was checked to insure that adequate conditions for film development were present. If not, the inspector would develop the film produced by the new technique, using fresh solutions in small portable developing tanks which he carried. The film which had been exposed using the pre-adjusted equipment and timer settings specified by the dentist, was developed according to the dentist's customary manner of development. The group "D" film taken with the correct exposure factors was developed by the time and temperature technique specified by the manufacturer of the film.

Films exposed to the proper amount of radiation, as determined in Figure I, and developed according to the recommended time and temperature technique proved to be consistently superior to those improperly exposed and developed. The fact that the dentist could observe a film developed in his own facility which was of both low patient exposure and good diagnostic quality, served to insure that the recommended techniques left by the inspector would be followed.

Results

The dental profession in the two pilot test States have responded enthusiastically to this new program as indicated by both the number of cards returned and the active endorsement by the Dental Associations in both States. The return of the TLD cards ranged from 75% in New Hampshire with a modest follow-up effort, to 99% in Rhode Island with a comprehensive follow-up effort. The results from both States were compiled within a 3-month period.

State Radiation Control Program Directors' acceptance of this program is primarily due to the ease and convenience with which it can be performed without significant increased cost of program personnel and time.

Identification Phase

The exposure data obtained by the cards was compared to the exposure ranges shown in Figure 1. Table I lists by operating kVp, the number of machines that fell within the exposure range prescribed. Fifty-four percent of the dental x-ray machines surveyed in this two State pilot study had outputs which fell inside the acceptable range. Forty-six percent of the machines surveyed were found to produce excessive radiation and contribute to the unnecessary exposure of the population even though with few exceptions, these dental facilities comply with existing local, State, and Federal regulations.

TABLE I

Results of the TLD Card Survey of Two States

Operating kVp	Acceptable Range mR	Total No. of of Machines Surveyed	Machines Inside Range		Machines Outside Range	
			No.	%	No.	%
90+	100-315	179	124	69.3	55	30.1
70-89	115-330	152	92	60.5	60	39.5
60-69	190-570	410	200	48.8	210	51.2
50-59	250-800	93	37	39.8	56	60.2
	Total	835	453	54.3	381	45.7

Action Phase

The action phase of this program has been completed in New Hampshire, and the results are encouraging. The point at which action was initiated in New Hampshire was 800 mR. A total of 105 dental x-ray units were found to lie above this limit and were visited by inspection personnel. The average

exposure at end of cone, mR/film, was reduced by 80.4% for the x-ray machines visited during the action phase. A total of 269 dental x-ray machines were surveyed in the identification phase of the project in New Hampshire, the net reduction in the average exposure at end of cone, mR/film, for all of the machines involved in the study is a very respectable 56%.

Conclusions

The dental profession in the two pilot test States responded enthusiastically to this new program as indicated by both the number of cards returned and the endorsement by State Dental Association in both States. Radiation Control Program Directors' acceptance of a card system has been accomplished through the ease and convenience with which the program is conducted without significant increased cost of program personnel and time. Based upon the results of this pilot study it is estimated that one Radiological Health Specialist working one-quarter time and one Secretary working one-half time can complete a 1,000 unit program in 3 months. The estimated cost of such a survey of 1,000 units is \$4.50 per machine. This cost includes the purchase of all needed equipment, printing, postage and personnel. This cost would be greatly reduced in those States already having TLD capabilities.

It has been demonstrated that TLD's sent through the mail can be used as effective screening devices to identify dental x-ray equipment producing excessive patient exposures. With this information, Radiation Control Agencies are able to plan their program efforts to lower exposure to the population from dental radiography according to a defined priority system.

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NASHVILLE DENTAL X-RAY STUDY

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I. INTRODUCTION

In January of 1972, personnel from the Bureau of Radiological Health, U.S. Public Health Service and members of the Department of Radiology, Vanderbilt University, met with representatives of the Nashville, Tennessee Dental Society to discuss dental radiological health practices. When comparing data from the 1964 and 1970 National X-ray Exposure Studies (XES)^{1,2} it was pointed out that the use of slow and intermediate speed dental film decreased substantially, yet the average incident skin exposure remained disproportionately high. In 1964 about half of the dentists were using the slow and intermediate film (speed group A, B and C) and 50% were using fast film (speed group D). In 1970 only 20% of the dentists used slow or intermediate film while 80% were using the fast film. However, the mean incident skin exposure in 1964 was 1,138 mR, whereas in 1970 it was 910 mR, a decrease of only 20%.

The group raised the following questions: "What are dentists doing which contribute to overexposure of their films and their patients? Are they using good equipment, following recommended procedures, and exercising good judgment? Furthermore, can the dentist be motivated to correct any deficient findings or improper radiographic practices through an educational approach?"

In an effort to answer these questions, a pilot project was initiated by Vanderbilt University, under contract with the Bureau of Radiological Health, and in cooperation with the Nashville Dental Society. An effort would be made to test the effectiveness of a non-regulatory educational approach to dental x-ray exposure reduction. The primary objective of the project would be to motivate the participating dentist toward improved dental radiological practices with emphasis on efficient and effective use of X-radiation.

The project was divided into three phases. Phase I was a detailed assessment of radiology practices and equipment in a random sample of private dental offices in the Nashville area. Phase II consisted of a continuing education program individualized to each office. Specific recommendations based on Phase I information were given to each participating dentist for modification of his x-ray equipment and radiology practices to achieve optimum diagnostic quality with minimum exposure. Phase III was a follow-up survey, one year after Phase II, to determine if this educational effort to improve radiographic practices was successful.

Phases I and II of the project have been completed and are the subject of this report. Data are being further evaluated to develop a profile of dental radiographic practices in Nashville. Such information will be useful in setting priorities for corrective action programs locally and possibly in indicating nationwide problems. A more complete report will be available from the Bureau of Radiological Health in the near future.

II. METHODOLOGY - PHASE I

Phase I consisted of a physical survey of 72 dental facilities selected at random from a total of 209 offices operated by 250 members of the Nashville Dental Society conducted during the summer of 1972. Offices having intraoral x-ray equipment were contacted by telephone and appointments were made. The surveyors were trained by members of the Radiology staff at Vanderbilt University and members of the Bureau of Radiological Health. The two man survey teams collected such information as: number of x-ray machines; cone types; speed of intraoral dental films; processing procedures; and, film mounting and viewing. Physical measurements were made of the beam size, filtration, exposure/film, and stability of the tube head.

III. RESULTS AND DISCUSSION - PHASE I

A total of 72 dental facilities were surveyed, involving 80 dentists and 110 x-ray machines. The following tables show some of the more significant findings.

Table 1.

<u>BEAM SIZE</u>	
<u>INCHES</u>	<u>MACHINES</u>
2.0-2.4	15
2.5-2.8	85
2.9-3.1	8
3.2+	2
TOTAL	<u>110</u>

The beam diameter was measured by making an exposure on direct print paper. Results from Table 1. show 100 machines have a beam diameter of less than 2.8 inches. Only 2 of the 110 machines had a beam diameter larger than the Tennessee State regulation of 3.0 inches. The largest beam diameter recorded was 3.4 inches.

Table 2.

OPERATING kVp BY CALCULATED HVL (mm Al)

kVp	Machines	HVL (mm Al)		
		< 1.1	1.2-2.0	> 2.1
< 50	1	--	--	1
50-70	75	3	47	25
> 71	34	--	9	25
TOTAL	<u>110</u>	<u>3</u>	<u>56</u>	<u>51</u>

The operating kVp was determined by the setting on the dial of the x-ray machine used by the dentist for a periapical exposure. The half value layer (mm Al) was determined using the Organ Dose Index System³. Table 2. shows that 76 machines operate up to 70 kVp and 34 machines operate over 70 kVp. Twelve of the 110 machines were deficient in filtration according to the proposed Federal Standards⁴. Three 50-70 kVp machines had a HVL of < 1.1 mm Al and nine machines operating over 70 kVp only had a HVL of less than 2.0 mm Al.

Table 3.

FILM SPEED - FACILITIES

	Speed Group				Total
	B	C	D	Unk.	
Periapical	1	2	71	--	74*
Interproximal	1	2	70	1	74*

*Two facilities used two different film speeds.

Table 3. shows that over 95% of the dental offices in Nashville are using speed group D dental film. This is considerably better than the national average of 80% as reported in the 1970 National X-ray Exposure Study. The two offices using speed group C film also used speed group D.

Table 4.

AVERAGE mR/FILM

All	542
Sight Develop	730
Occ. Sight Develop	536
Never Sight Develop	404

The exposure per film at the end of the position indicating device (cone) was determined by making three exposures on low energy dosimeters using the Organ Dose Index System³. Table 4. shows the average incident skin exposure for all 72 facilities was found to be only 542 mR. This is low compared to the national average and is to be expected when one considers the high percentage of dentists using the fast film. However, it was also noted that 43 of the 72 offices sight-developed, 24 routinely. These "sight developing" dental offices are overexposing and underdeveloping dental films. Consequently, the average exposure per film was calculated for the sight developing offices, the occasional sight developing offices and the never sight developing offices. The results are seen in Table 4; sight developing offices 730 mR, occasional sight developing offices 536 mR, and the never sight developing offices only 404 mR.

This is felt to be significant because apparently by just properly exposing the film and changing the processing technique to one of time-temperature, the sight developing dental office can reduce the exposure to its patients by at least 45%.

Table 5.

mR/FILM (INTERPROXIMAL) BY kVp

mR/film	Machines	kVp			
		< 50	50-60	61-70	> 70
55-200	23	--	3	10	10
201-400	37	--	--	23	14
401-600	17	--	--	10	7
601-999	15	--	1	13	1
1,000+	18	1	4	11	2
TOTAL	110	1	8	67	34

Table 5. is a cross tabulation of mR/film by kVp listed by x-ray machine. Seventy-seven of the 110 machines were producing an exposure of 600 mR or less. The exposures ranged from less than 100 mR to slightly over 2,500 mR. Although the average exposure was 542 mR, the median exposure was 404 mR. The kVp was determined by the setting on the dial of the x-ray machine used by the dentist for an interproximal exposure. Table 5. indicates that in general, the dentists operating higher kVp equipment are giving their patients less skin exposure than dentists using lower kVp techniques. It is well established that incident skin exposure increases as kilovolt peak is reduced, but it is not known exactly what the effect is on the integral absorbed dose to the patient.

Table 6.

POSITION INDICATING DEVICES (CONES)

Type	Machines
Pointed	46
Open lined	50
Open unlined	13
Unknown	1
TOTAL	110

A breakdown of position indicating devices (cones) on equipment showed 46 pointed cones; 50 open lead-lined cones; 13 open unlined cones; and 1 unknown. There is more scattered radiation associated with pointed cones than with open end cones. Tennessee State regulations require that after July 1, 1973, all x-ray apparatus designed for intraoral radiographic use shall be equipped with open end cones (cylinders).

Table 7.

<u>TUBE DRIFT AND VIBRATION</u>		
	<u>Drift</u>	<u>Vibration</u>
	<u>Machines</u>	<u>Machines</u>
Yes	34	18
No	76	92
TOTALS	110	110

Tube head drift was determined by fully extending the tube head and noting any drifting or pulling back of the head from its set position. Thirty-one percent demonstrated drifting. Drifting can cause inferior radiographs by cone cutting and/or improper angulation of the primary beam.

Vibration was determined by extending the tube head to near maximum and noting any vibrations. Sixteen percent of the tube heads vibrated. Excessive vibration can cause increased penumbra and blurring of the radiograph.

IV. CONCLUSIONS

After analyzing the preceding data, one can conclude that in general the dental x-ray equipment in Nashville, Tennessee, meets recommended standards. Most dental x-ray machines have properly collimated and filtered beams. Within the near future all dental x-ray equipment in Tennessee will have open end cones (cylinders). Almost all of the dentists are using the fastest speed film available. Yet the incident skin exposure is higher than necessary because the dentists fail to use proper processing techniques. Instead of overexposing films and using a sight developing technique, dentists should properly expose the films and use the recommended time-temperature processing procedure.

V. METHODOLOGY - PHASE II

At the completion of Phase I, the results from each survey form were read and reviewed. Dental radiology experts from Vanderbilt University and the Bureau of Radiological Health took one facility at a time and in detail listed the deficiencies in the office on a Check List Form. Then recommendations were made for improving radiographic practice on a Reporting Form. The report also included the approximate cost involved to bring the facility up to the recommendations.

Phase II consisted of a second visit to the dental office by a consulting team composed of one of the dental students and a dentist knowledgeable in dental radiology from Vanderbilt University or the Bureau of Radiological Health. As in Phase I, the telephone was used for making definite appointments. It was requested that the dentist and all his staff attend this meeting.

While in the dental office, the consulting teams performed the following: presented and reviewed the findings of the initial survey; discussed the written recommendations for any changes to improve the dentist's radiographic practice and left a copy of the "Reporting Form" and "Survey Check List"; answered all questions from the facility personnel; gathered from the dentist certain judgmental factors, such as criteria and frequency for making examinations; collected missing or questionable information from the initial survey; and left with the dentist selected reprints and pamphlets for future reference.

Phase III was started in July of this year with a follow-up visit to the original 72 dental offices where the equipment was resurveyed. Analysis of the data will determine the effectiveness of the project in motivating the Nashville dentists to correct any machine deficiencies and to improve their radiographic practices. If the project proves successful, the Bureau may assist other interested dental societies, universities, and health agencies in implementing similar educational programs.

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ABSORBED DOSE TO SELECTED INTERNAL ORGANS

FROM TYPICAL DIAGNOSTIC EXPOSURES *

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Estimates of dose to internal organs from exposure to diagnostic x-ray beams are usually based on measurements of the entrance and exit dose and/or the use of depth dose curves for homogeneous media. This paper presents the results of a series of Monte Carlo calculations which mock-up typical diagnostic x-ray procedures. Results are presented for 22 internal organs as well as red and yellow bone marrow for two typical procedures. The calculations employ measured x-ray spectra from 45 kVp, 1-mm Al to 105 kVp, 2-mm Al and are for a field size of 14" x 17". In addition, depth dose profiles in various sections of the heterogeneous phantom are presented for each x-ray beam.

Introduction

It is well established that x-rays, particularly medical and dental x-rays, contribute the largest exposure to the population of any man-made source of ionizing radiation. The fundamental objective of the medical use of radiation is to obtain optimum diagnostic information with minimum exposure to the patient, and the radiological personnel concerned, and the general public. However, the problems posed when one attempts to estimate the doses received by various organs of the body from a medical exposure are among the most difficult problems the radiological physicist must face. The geometrical complexities and inhomogeneities of the body and the various organs make experimental simulation of the human body extremely difficult and usually unsatisfactory.

Monte Carlo techniques currently in use on high-speed digital computers have greatly facilitated the solution of these complex problems. These techniques have gained wide use in the field of radiation protection because the method allows one to perform an experiment by use of the computer. Many experimental arrangements and physical parameters, which can be described mathematically, can be operated on by the computer to produce the desired results.

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A Monte-Carlo-type computer program has been developed at the Oak Ridge National Laboratory, which can be used to estimate dose due to external photon beams typical of those employed in x-ray diagnosis.

Description of the Study

Two diagnostic x-ray procedures were simulated on a computer for these studies. Eight computer runs consisted of a set of exposures each of which simulated a chest x-ray and eight runs consisted of a set of exposures each of which simulated a G. I. x-ray.

The target for these studies was an anthropomorphic phantom, which may be considered to be two coexistent phantoms. One is that of an adult human body and some of its internality. It has been variously described ^{1,2,3,4,5} and in its present form represents a worthwhile target for these studies. It contains 23 internal organs including gonads, lungs, and four parts of the G. I. tract; it has ten skeletal parts with provisions for red and yellow marrows; and there is skin and there is tissue which includes muscle. The bone marrow and the bone are mixed homogeneously in the skeleton of this phantom.

The other phantom is called a geometric phantom. Whereas, in toto, it has the same outer dimensions and the same mass and composition of the human phantom, it is divided into dose regions by cutting planes and curves. For example, the trunk of the phantom has five layers, is divided into five concentric cylinders, and is cut by four vertical cross planes. This results in 85 subregions in which depth dose may be determined.

Both phantoms are heterogeneous by virtue of their composition which consists of 3 distinct media: tissue, lung, and bone with their concomitant densities and attenuation and absorption properties.

Each of the 16 exposures consisted of a collimated 36 cm x 44 cm (14 x 17 in) beam of 120,000 parallel photons incident on the posterior (P-A) of each phantom. In these calculations the source input was a set of eight measured x-ray energy spectra due to Epp and Weiss⁶ at the Sloan-Kettering Institute for Cancer Research in New York City. The spectra range from 45 kVp, 1-mm Al filtration to 105 kVp, 2-mm Al filtration. The energy of each photon was determined from a normalized distribution of relative photon fluences per unit energy interval between 10 keV and 102 keV. Monte Carlo methods were used to follow the transport of each photon through the phantoms, determining the scattering angles, absorption sites, etc., and permitting the estimates of absorbed dose in units of absorbed dose per unit incident exposure (rad/R). The absorbed dose was calculated in the internal organs of the

adult human phantom as well as in the volume elements of the geometric phantom.

Depth dose distributions in the trunk for the simulated chest x-ray exposures are presented in Figure 1. These data are for 36 x 44 cm beams incident on the posterior of the phantom. Illustrated are the effects on dose of the reduction in average energy of the beams and the attenuation of the beams as they pass through the phantom. For the high energy beam, the dose from the back to front drops off by a factor of 10. For the low energy beam, it is reduced by a factor of about 130. The average dose in the first 2 cm of tissue for the 105 kVp beam is 1.7 times higher than for the 45 kVp beam. Near the exit surface the dose for the 105 kVp is 23 times higher than that for the 45 kVp beam. Data for the simulated G. I. exposure are similar in magnitude and ratio negating the necessity to discuss these results in detail.

Often the radiologist uses the dose at 5 cm depth as an indicator of the average dose to the red bone marrow. Table I presents such a comparison of the data derived from the simulated chest exposures. The last column of the table is the ratio of the red bone marrow dose to the 5 cm depth dose and shows that the indicator mentioned above might lead to a 40% error.

Figure 2 shows dose to selected organs for a simulated chest x-ray as a function of average beam energy. There appears to be three pairs of curves. The pair with the highest dose represents organs definitely within the beam. The next highest pair, the upper large intestine and the thyroid gland, represents organs outside but near the edge of the beam. The last pair represents organs definitely outside the beam.

Lowering the beam location to a position which simulates a G. I. exposure caused a 50% increase in the dose from the low energy beam and a 20% increase in dose from the high energy beam to the red bone marrow. This result is due to the exposure of the pelvis which contains about 32% of the red bone marrow and was outside the beam during the simulated chest exposure. The dose to other organs, such as the uterus and the upper and lower large intestine, was increased by at least a factor of three at this lower exposure.

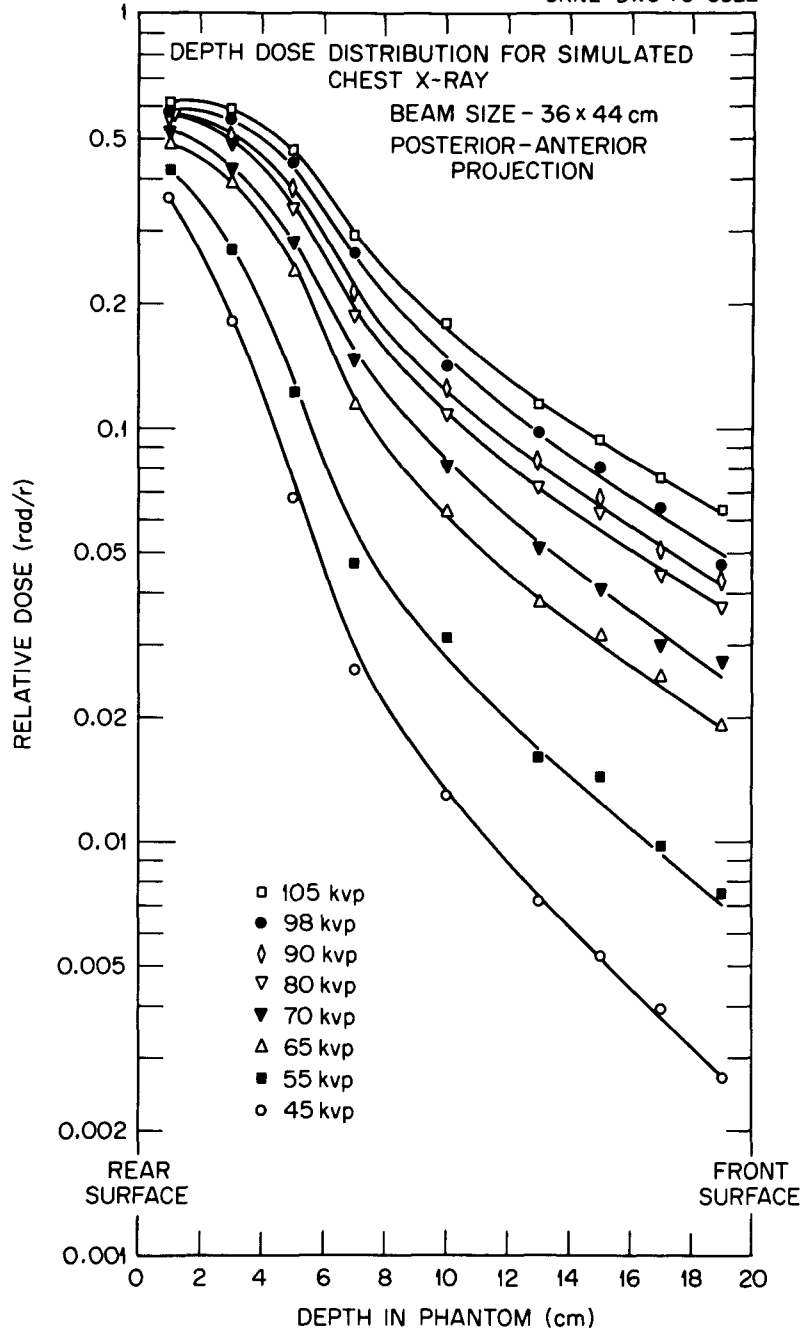
This study represents only a beginning in that it demonstrates the versatility of Monte Carlo techniques in the simulation of diagnostic procedures. The computer programs used allow various source descriptions, such as point sources located at various source to skin distances, divergent beams, etc. In addition, the beam size, shape, and angle of incidence on the phantom may be specified.

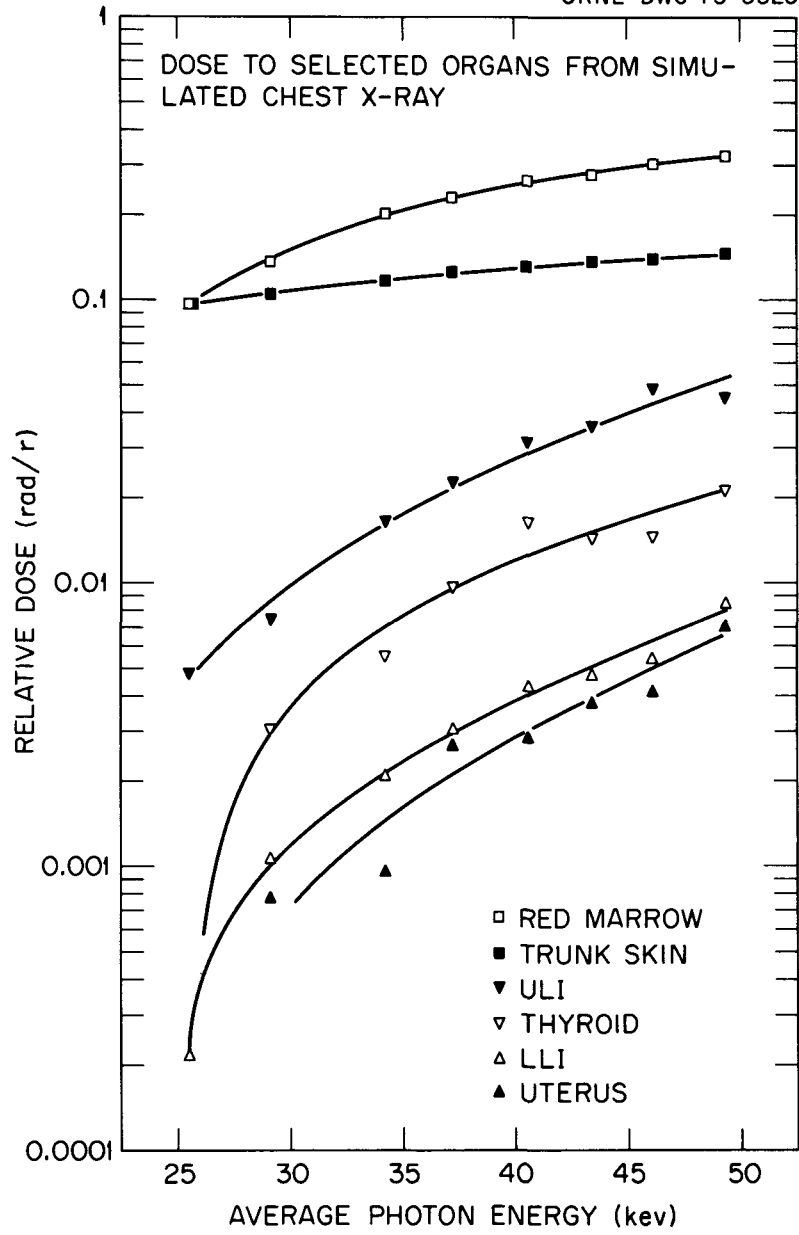
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TABLE I. Comparison of Average Absorbed Dose at 5 cm Depth to
Average Absorbed Dose to Red Bone Marrow

Avg. Beam Energy (keV)	Avg. Dose at 5 cm Depth in the Beam (rad/R)	Dose to Red Bone Marrow (rad/R)	Ratio
25.5	0.164	0.242	1.48
29.1	0.303	0.343	1.13
34.2	0.564	0.503	0.892
37.1	0.707	0.573	0.810
40.6	0.836	0.657	0.786
43.4	0.944	0.696	0.737
46.1	1.08	0.754	0.698
49.3	1.18	0.805	0.682





HANDY DEVICE FOR PROTECTION OF
THE TESTICLES IN X-RAY EXAMINATIONS

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Abstract

We have modified the Picker lead plastic gonad capsule by mounting leaf springs in the two ducts along its opening slit, thus making it "self-supporting" and able to adjust itself to the scrotum so that the testicles are covered as completely as possible.

Results are given of an investigation of the practical applicability and the shielding effect of the new device.

It is demonstrated that the shielding effect of the modified Picker lead plastic gonad capsule is as good as that of the common two-piece lead capsule device. From a practical point of view it is far better, because it is easily applied and causes the patient no discomfort.

Introduction

It is generally admitted by now that the gonads in men in the reproductive age should be protected by shields of capsule type whenever X-ray examinations exposing areas close to the gonads to the primary beam are carried out, i.e. by shields which fit tightly around the scrotum in order that the testicles may be efficiently protected against primary radiation as well as against scattered radiation coming from the body volume exposed to the primary beam. Even so, it is our impression that this type of shielding of the gonads is used only rarely, at least in Denmark. The main reason is probably that application of the common two-piece lead capsule device in general is considered inconvenient in the routine.

Accordingly, we tried in 1970 to find other means of protection and chanced to find a lead plastic gonad capsule produced by the firm Picker; it is provided with a slit-formed opening the edges of which are in the form of two ducts. It was suggested that it might be desirable if the ducts were provided with leaf springs which tentatively were fitted in. The result was a capsule which for one thing remains in place automatically, no matter the patient's movements, secondly it fits around the scrotum, its opening being the smallest possible. The original Picker capsule and the modified type are illustrated in Fig. 1. The problem of hygiene is solved by a disposable plastic bag which prior to each application is to be inserted into the capsule and turned over its edges.

A few tests on patients showed beyond doubt that the new device was by far more convenient than the solid two-piece capsule and besides, the patients found

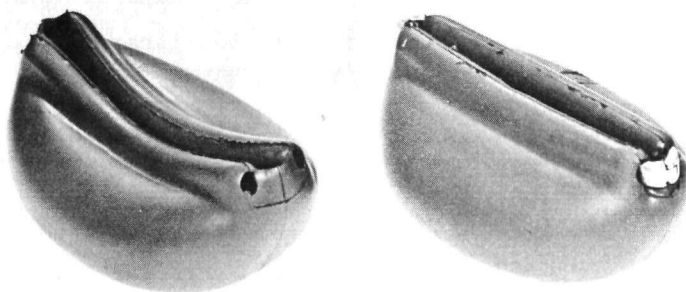


Fig. 1. Picker's original lead plastic gonad capsule and the modified capsule.

that they might easily apply it themselves. Thus, it seemed as if we actually had found a gonad shield which was more handy than the two-piece capsule and, in fact, everybody who at present have reported on the experience gained in the use of the new capsule have shared our opinion. Fig. 2 illustrates how patients in standing position manage to apply the capsule.

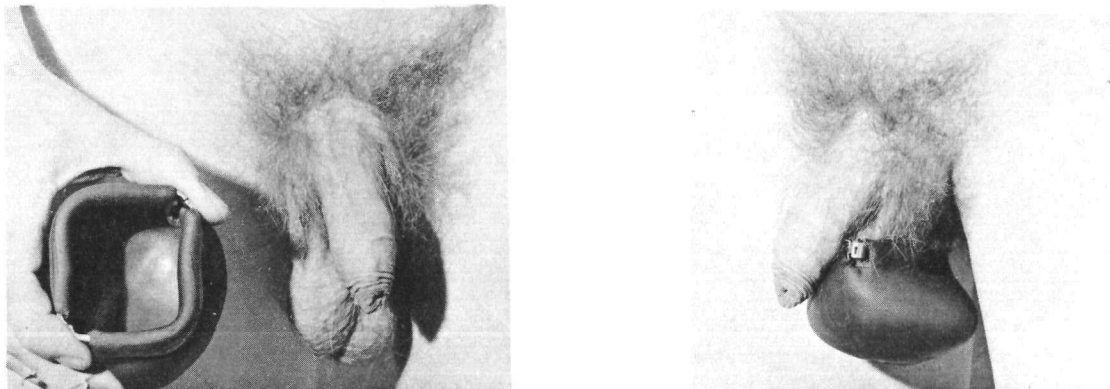


Fig. 2. Patient demonstrating how to apply the modified gonad capsule.

The question remained, however, whether the shielding effect of the new capsule was sufficiently satisfactory. Its lead equivalent is stated to be only 0.5 mm in contrast to an equivalent of 1 mm in the case of the two-piece capsule made by Mavig and consequently, it provides less protection against the primary beam; on the other hand, the two-piece capsule must be assumed to provide a less satisfactory protection against scattered radiation because its opening around the scrotal basis is about 15 cm² (large model) at optimal fitting while the new capsule leaves an opening of only 10-15 cm², dependent on the anatomy of the patient. The shielding effect of the new capsule was therefore tested, partly by measurements during urography of hospitalized patients and outpatients, partly by phantom-measurements. The new capsule was finally tested in practice, mainly in order to obtain an impression of the patients' capacity to apply the capsule correctly.

Dose Measurements on Patients

In one series of patients the shielding effect was examined by measurement of doses accumulated during 10 routine urography examinations using no shielding and during another 10 examinations using each of the following three types of shielding: common lead rubber sheet, Mavig's two-piece capsule, and the new capsule. LiF thermoluminescence dosimeters were used for the measurements; the results are recorded in Table 1.

As regards the three groups in which shielding was used, a dosimeter was placed on top of the shielding device in addition to the gonad dosimeter on the scrotum; the former dosimeter recorded approximately the dose to be given to

	No shield	Lead rubber	Mavig capsule	Modified Picker capsule
No. of urographies	10	10	10	10
No. of films used	56	61	60	56
Mean dose externally on shielding device	(130)	380	130	85
Mean dose to gonads	130	40	24	8

Table 1. Mean gonad doses in mrad/urography with different types of shielding.

the gonads under the said conditions in the absence of shielding.

It appears from the table that the dose to the gonads was remarkably low if the new capsule was used. Even though the measurements involve a high degree of uncertainty owing to the non-standardized experimental conditions, it gave us reason to believe that the shielding effect of the new capsule was sufficient.

In order to obtain a further insight into the individual variations in doses, we continued our experiments in a minor series in which doses were measured separately during each urography, the latter including five exposures. The dosimeters were arranged as described above. The results appear from Table 2 and are to be interpreted to the effect that the two capsules are of equal value.

The conclusion to be drawn on the basis of the two tests on patients is that our measurements during urography failed to disclose any significant difference in shielding effects of the two-piece capsule and the new lead plastic capsule. The tests are described in further detail in¹.

Patient	Mavig capsule							Modified Picker capsule				
	1	2	3	4	5	6	7	8	9	10	11	12
Dose externally on shield	138	73	108	125	71	85	40	110	35	75	170	30
Gonad dose	34	24	47	29	14	35	14	23	15	8	90	12
Mean dose to gonads	28							30				

Table 2. Individual gonad doses in mrad during 7 urographies using the Mavig two-piece capsule and during 5 urographies using the new capsule.

Dose Measurements on Phantom

Phantom-measurements were subsequently performed in order to determine the shielding effect under reproducible conditions. A therapy equipment with Greinacher coupling was used for the exposures. The Alderson-Rando phantom which for the occasion was provided with a gonad phantom is depicted in Fig. 3 in midline sectional view.

Two LiF dosimeters were placed centrally in the gonad phantom. The thickness of the stalk of the latter, on which the size of the opening of the new capsule depends, was chosen at 20 mm, providing about the maximum size of openings of capsules applied to patients, namely 15 cm², and thus an opening similar to that of the two-piece capsule. Owing to the construction of the Alderson-Rando phantom, the opening of the capsules is unfortunately turned in the posterior-cranial direction and hence their orientation is not quite in agreement

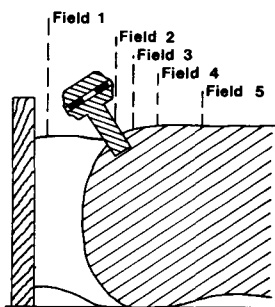


Fig. 3. Phantom in midline sectional view. Caudal field edges are sketched in.

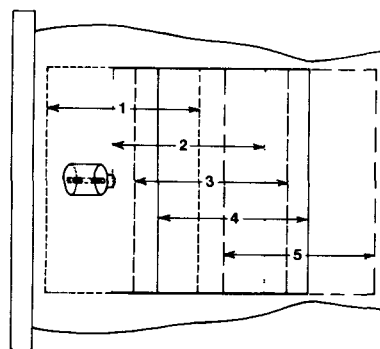


Fig. 4. Location of fields on the anterior surface of the phantom.

with that of capsules used on patients. A treatment applicator providing a field size of 17 x 25 cm on the anterior surface of the phantom was used at a distance of 70 cm from focus. The position of the fields used appears from Fig. 4.

At field 1, the gonad phantom is in the primary beam; as regards fields 2-5, their caudal field edges are at distances of 2.5, 5, 7.5, and 12.5 cm, respectively, from a point centrally between the dosimeters. Doses were measured at 60, 90, and 120 kV (total filtration equivalent to 4 mm Al). The results are recorded in Table 3, except the results obtained by common lead rubber sheet since these values, when outside the primary beam, were almost the same as those obtained without shielding. The doses are expressed in $\mu\text{rad}/\text{mAs}$ at the given focus-skin distance.

	No shield			Mavig capsule			Modified Picker capsule		
	60 kV	90 kV	120 kV	60 kV	90 kV	120 kV	60 kV	90 kV	120 kV
Field 1	8200	21000	38000	75	350	920	42	270	700
Field 2	550	1900	3700	72	270	570	39	165	400
Field 3	270	1050	2200	48	230	470	43	150	350
Field 4	135	540	1150	19	73	200	16	71	170
Field 5	32	155	350	3	20	61	3	20	50

Table 3. Gonad doses in $\mu\text{rad}/\text{mAs}$ measured on phantom.

It will be noted that the shielding effect of the new capsule was not in any case found inferior to that of the two-piece capsule. It should be mentioned that the results are impaired by some uncertainty because due regard has not been paid to the exact reproducibility of the orientation of the capsules. This may explain why the dose at 60 kV with the new capsule was found to be higher in field 3 (43 $\mu\text{rad}/\text{mAs}$) than in fields 1 and 2.

The diagram in Fig. 5 illustrates the results obtained at 90 kV, including the results obtained by lead rubber sheet shielding. As mentioned, the shielding effect of a lead rubber sheet against scattered radiation is seen to be almost negligible.

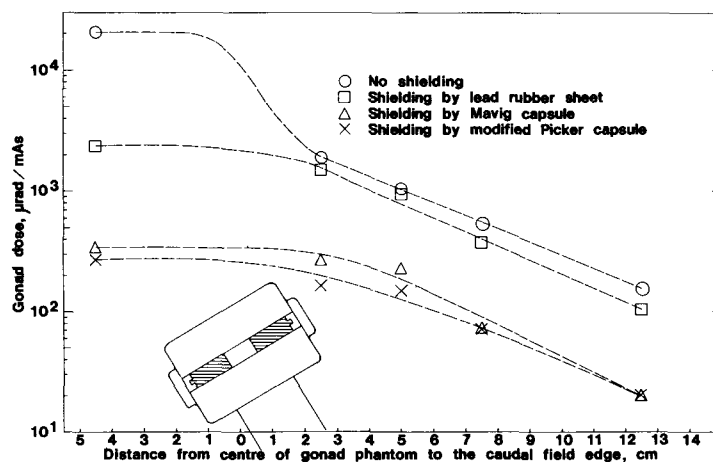


Fig. 5. Graphical representation of the results obtained by measurements on phantom at 90 kV.

The results obtained by the Mavig two-piece capsule and recorded in Table 3 have been compared with those of similar measurements performed by STIEVE² in an autopsy series; The reduction factors in our series (ratio of dose measured externally on the capsule to gonad dose) were generally found to be 2-3 times higher than those reported by STIEVE. According to our opinion, part of the explanation may be that, as already mentioned, the opening of the capsules in our phantom set-up is turned more backwards than is normally the case if they are used on patients.

It is not easily comprehensible why the shielding effect of the new capsule appears to be superior to that of the two-piece capsule, taking into consideration that the lead equivalent of the latter is highest (1 mm versus 0.5 mm) and that the sizes of openings were almost identical during measurements on phantom. The explanation may be that the form and orientation of the openings of the two capsules have a marked influence on the amount of scattered radiation to pass through the opening and that doses contributed by radiation through the capsular walls at 0.5 and 1 mm lead equivalent are negligible as compared with the doses contributed by scattered radiation through the opening.

Experience gained in Practice

The results obtained by testings in practice of the new lead plastic capsule are discussed below. The main object was to learn how many patients would manage to apply the new capsule correctly on themselves. With this end in view, a brief, illustrated instruction was prepared. The tests were performed on out-patients who met for examination in a diagnostic X-ray hospital department throughout two months; it must be admitted that it proved impossible to include all patients who appeared during the said period, partly because one physician, always the same, had to be present and supervise that application was correct in all cases, partly because we were interested primarily in the applicability of the device among young patients. It must also be admitted that the patients were not selected at random in the statistical sense of the word since selection was dependent on various practical circumstances. As the object of the investigation merely was to obtain an impression of the applicability of the device, without aiming at a direct collation with other gonad shields, the bias thus introduced is hardly of any significance.

A total of 46 patients received a lead plastic capsule - provided with a disposable plastic bag - and the written instruction immediately after they arrived in the changing room. Two or three minutes later, the examiner would appear and supervise that the capsule had been correctly applied. Thirty-one out of the 46 patients managed to apply the capsule completely correctly within the allowed interval of time. Seven patients found application rather difficult or they applied it slightly incorrectly which, however, had no essential influence on its shielding effect. Application was unsuccessful in three cases, either because of some genito-anatomical deviations or because surgery recently had been performed on the scrotum; in two of these cases, the examiner managed to apply the capsules to the patients in supine position. Two elderly patients failed to apply the capsule because they had not brought their glasses and could not read the instruction. Three patients had applied the capsule in such a way that one testicle was above the opening although there was no anatomical explanation of the phenomenon. They were all able to apply the capsule correctly after they had been told of their mistake. Not a single patient refused to participate and all tried to apply the capsule (except the two patients who had not brought their glasses). Nobody found it inconvenient to wear the capsule, and nobody complained of having found it too difficult to apply. It is our impression, however, that application might be facilitated in some cases if the capsule were a little larger, but if so, its capacity to remain in place might be reduced in other cases and thus, we cannot recommend any changes in size until further experiments have been carried out. Application may be facilitated if the scrotum and the disposable plastic bag are sprinkled with talc powder.

der prior to application; it is not necessary, however, and was not done in any case in the present investigation.

Thus, the result of the testings in practice was that about 80% of the patients (38 out of 46) managed personally to apply the capsule sufficiently correctly. If the instruction could be revised on the basis of the experience gained, the results would probably be better.

Conclusion

The new lead plastic capsule seems to fulfil all reasonable requirements to a gonad shield to be used by men and it fulfils also the first five out of the six requirements set up by STIEVE²: (1) It must be suitable for all types of examination and hence, it must fit tightly around the scrotum; (2) The opening admitting the root of the scrotum must be as small as possible; (3) It must be easily applicable, preferably by the patient himself; (4) It must be as small as possible; (5) It must be hygienic in use. The sixth requirement set up by STIEVE, namely that the gonad shield must attenuate primary radiation to 2%, is not fulfilled, however, since this would require a lead equivalent of 1 mm at 150 kV in stead of the 0.5 mm in the lead rubber capsule. In consideration of the applicability of the capsule, it might be reasonable to be content with the 0.5 mm which is apparent also from the results of measurements performed in the present investigation from which it may be inferred that the gonad dose contributed by primary radiation through the lead rubber wall, even at high voltages somewhat beyond 120 kV, is not of great consequence as compared with the dose inevitably contributed by scattered radiation through the capsular opening.

As already mentioned, it is our impression that gonad shields of capsule type are used only on too rare occasions during X-ray examinations of areas close to the gonads. We are of the opinion that one reason is that the hitherto used capsules are highly inconvenient in use and another that a certain sense of modesty may be in evidence. Such obstacles are apparently eliminated by the new capsule which patients may apply to themselves. Accordingly, there is no longer any excuse why an effective gonad protection should not be used in all cases in which areas close to the gonads in men in the reproductive age are exposed to the primary beam. In this context, X-ray examination of areas close to the gonads refers to all types of X-ray examination in which the gonads either are in the primary beam or are less than about 10 cm from the edge of the beam.

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DOSE REDUCTION BY ELECTRONIC RADIOGRAPHY

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ABSTRACT

High resolution electronic radiography has been developed for use in diagnostic radiographic and fluoroscopic procedures. These methods are being developed for IVP pelvimetry, intrauterine transfusions, gastrointestinal examinations, stereotaxic procedures, and selective catheterization. Preliminary clinical data show that dose reduction can range from 10 to 10,000 times depending on the resolution requirements of the particular examination.

I. INTRODUCTION

The use of new electronic image amplifiers (1) television systems, and electronic recording devices represents a major advance in radiological techniques by allowing a significant reduction in radiation dose and an improvement in the diagnostic quality of images. The medical radiation dose in diagnostic procedures (2) has become of increasing concern as the result of mounting evidence for serious somatic effects of diagnostic x-rays such as an increased incidence of leukemia in young children whose mothers received diagnostic x-ray examinations during pregnancy, (3) as well as increasing evidence for a growing genetically significant dose from diagnostic procedures. Recent calculations have estimated that from 3,000 to 6,000 cancer deaths annually are caused by exposure of the American public to present levels of diagnostic x-rays. In addition, ill health results from genetic damage caused by the exposure. (4)

At resolutions adequate for most diagnostic purposes the amount of radiation required for radiography, even when utilizing the best intensifier screens and fastest films, is far in excess of what would be required if x-ray quanta could be utilized as efficiently as in present electronic image intensifiers

for fluoroscopy. This arises from the fact that although one or more grains are typically sensitized for each x-ray photon absorbed in the intensifying phosphor screen, the presence of unavoidable fog at low densities causes these grains to be lost in the statistical fluctuations of the background grains.

With a high gain x-ray phosphor and television camera system, a detectable signal above noise can be produced for single x-ray quanta absorbed in the phosphor screen. This high efficiency of visible photon conversion in the photo-electric effect, amounting to as much as 65%, is constant for all fluxes of x-ray photons, independent of both of accumulated dose and dose-rate, unlike the case of film where the quantum efficiency for visible photons is generally less than 0.1 percent due to the inherent non-linearity of the photographic process since 5 to 10 visible-light photons must be absorbed by a given grain before it becomes developable.

Theoretical considerations and experimental evidence indicate that the ultimate limitations to the reduction in x-ray dose set by quantum fluctuations of the x-ray flux permit substantial reductions far below the doses presently realized with films. Even at resolutions of about 4 lp/mm and a contrast of 5 to 20% actually utilized in most clinical radiographs, significant dose reductions should be possible by the use of electronic radiography which maximizes the utilization of x-rays while minimizing the radiation exposure to the patient in diagnostic procedures. This method permits the radiologist to reduce the patient exposure depending on the resolution requirement of a particular examination. With film relatively large dose are required to attain adequate densities which

reduces the image quality because of focal spot and motion blurring. In film radiography the inability to achieve short exposures with very small focal spots limits magnification techniques, as well as the "air-gap" technique. Contrast enhancement by the use of heavy selective filtration to produce monochromatic radiation from the continuous spectrum of x-rays is severely limited by the high radiation exposure required by film. All these limitations are reduced by the substitution of the more efficient electronic technique for recording radiographic images, improving the diagnostic value of radiograms beyond that possible with chemical photography.

Basically, electronic radiography is an x-ray recording technique which involves the substitution of the highly effective photo-electric effect for the relatively inefficient photo-chemical effect taking place in film.

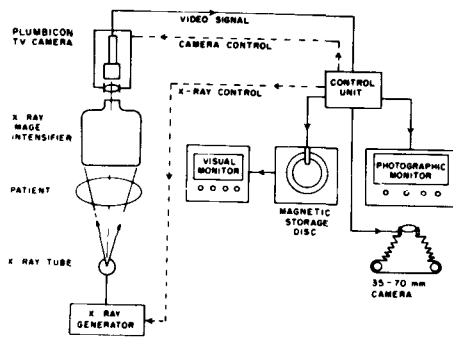


Figure 1: Block Diagram of the Apparatus

Electronic radiographs are recorded as follows (Figure 1). X-rays produced in the usual manner are incident on the object, and those that penetrate impinge upon an x-ray luminescent phosphor screen of an x-ray image intensifier. Light emitted by the phosphor screen releases photo-electrons from a thin photo-cathode in optical contact with the x-ray phosphor. The photo-electrons released are accelerated by some 30,000

volts and focused onto a small output phosphor where they give rise to a greatly brightened image which is viewed by a television camera. The optical image on the output phosphor of the intensifier is converted to an electronic charge pattern on the target of the camera tube. The charge pattern produced on the target is converted to a video-signal by means of a scanning electron beam and recorded in three alternative methods. (1) The electrical signal of one television frame or a series of frames may be recorded on a magnetic disc and then immediately replayed over a flicker-free frozen radiographic image for continuous viewing without appreciable deterioration. (2) The signal representing an individual frame may be recorded on a silicon image storage tube and immediately replayed over a TV monitor. However, this image gradually deteriorates and is completely erased after 5 to 10 minutes of continuous viewing. (3) A single video frame or many frames may be photographed by camera focused on a remote television monitor.

Systems incorporating these components have increased diagnostic information, reduced procedure time, decreased patient trauma, reduced radiation dose and permitted new types of procedures to be undertaken. Electronic radiography is used in gastrointestinal examinations (5) selective catheterization (6) pelvimetry (7) and repair of intracranial aneurysms and arteriovenous malformations (8).

In addition to improving the image quality, electronic radiography eliminates some other disadvantages of film radiography such as chemical processing time and expenses and permits a reduction of radiation exposure to the absolute minimum dictated by the quantum noise limit for the particular degree of detail and contrast needed for a given purpose.

II. ELECTRONIC SPOT IMAGING FOR GASTROINTESTINAL FLUOROSCOPY

We have developed a high resolution technique of electronic spot imaging, to replace standard spot-filming in gastrointestinal fluoroscopy. The system consists of a standard

fluoroscopic unit electronically interfaced to a magnetic disc recorder. Key components of the system are: (1) a small focal spot x-ray tube, (2) a CsI intensifier tube, (3) a Plumbicon television camera, and (4) a 400 track magnetic disc recorder. The principle of operation is the storage of single television fluoroscopic frames on individual tracks of a magnetic disc. Once recorded, the signals can be reviewed over a television monitor at the end of the examination and later photographed for a permanent record.

At the start of an examination the unit is placed in the fluoro-record mode which allows monitoring of dynamic motion. A standard television fluoroscopic image is first generated by depressing a foot pedal. Further depression of the pedal closes a second switch which automatically boosts x-ray output, and records a single television fluoroscopic frame on the magnetic disc. In contrast to normal spot-filming this recording technique does not interrupt the fluoroscopic sequence and permits the fluoroscopist to monitor the patient continuously. At the termination of an examination, the fluoroscopist can review the images by turning the hand switch to reverse. Then, every time the foot pedal is depressed, the recording head of the disc recorder is moved one track in the reverse direction and the monitor displays a frozen radiograph previously recorded as a flicker-free electronic spot image. After arriving at the initial image of a given examination the fluoroscopist by switching to the forward mode, may then leisurely study each image in the order that it was recorded. Image brightness and contrast can be adjusted on the television monitor. After checking the recorded images for completeness, the fluoroscopist is ready to proceed with the next examination. At the end of a day's fluoroscopy, the disc recorder is moved to the reporting area, where corresponding radiographs are displayed. The electronic spot images and overhead films are then

reviewed before rendering a final diagnostic report. Key electronic spot images are photographed to provide a permanent record while allowing reuse of the magnetic disc.

The radiation exposure for a single electronic spot image is about 1/50 of that required for conventional film-screen cassettes and 1/5 of the requirement for 105mm. spot-filming respectively. The exact dose depends on the resolution and contrast requirements of the particular examination.

We have compared the electronic technique to 105mm. spot-filming with regard to diagnostic accuracy in a clinical study (Fig. 2,3). Small ulcers (1-5mm. in diameter), varices, colonic polyps, various diverticula and other pathology were readily diagnosed by both techniques. In a double blind study six radiologists independently arrived at diagnoses in 21 patients. Their interpretations were then scored against those of the fluoroscopist and evaluated using the analysis of variance. The average correct scores were 76.5% and 75.5% for the electronic spot imaging and 105mm spot-filming methods respectively. Although both methods yield clinically acceptable results, we have found electronic spot imaging to be a more convenient technique of recording and reviewing fluoroscopic information.

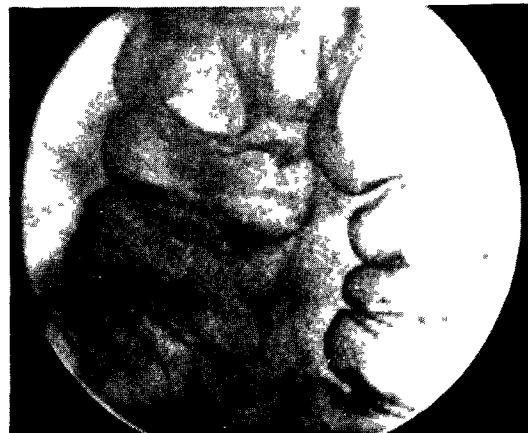
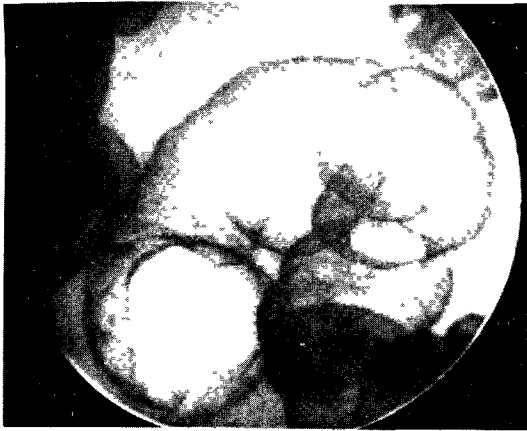


Figure 2: Electronic Spot Image of Splenic Flexure Taken During Double Contrast Barium Enema



**Figure 3: Electronic Spot Image
Taken of Sigmoid Colon**
**III. PELVIMETRY WITH SMALL RADIATION
EXPOSURE**

The apparatus for polaroid television technique consists of a polaroid camera focused at a remote television monitor of a standard television fluoroscopic unit. The principle of the method is to photograph an entire fluoroscopic exposure on the polaroid film which is rapidly developed. Since television fluoroscopic units have high gain image intensifiers and television cameras, the image produced on the television screen can easily be adjusted to be noise limited. Then, the radiation exposure to the patient would be reduced to a minimum which is about 1/100 of that for standard film radiography.

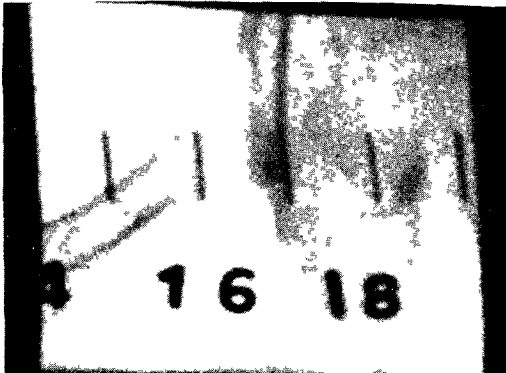
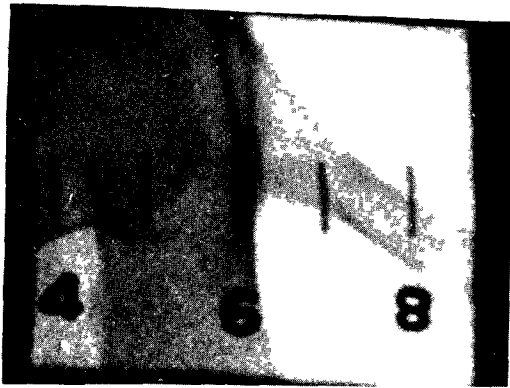
The polaroid television technique is achievable with low-cost modification of most standard television fluoroscopic units. The polaroid image is obtained in the following sequence of operations: (1) the foot-pedal is compressed, initiating the x-ray exposure, (2) the camera shutter is manually opened for a 1/2 second exposure, (3) the camera shutter automatically closes, (4) the foot-pedal is released terminating the exposure, (5) the polaroid film is developed in 15 seconds.

Since in most television chains the automatic gain control built into the television camera keep the average video output constant, the image quantum noise can be varied by changing the kVp and ma setting in the fluoroscopic exposure.

In our technique which is adapted from the orthometric pelvimetry used with standard film, a radiopaque ruler is placed between the patient and the intensifier tube. The apparatus is positioned to be centered first on one and then the other side of the anatomical structures to be measured and a polaroid fluoroscopic television image is taken. Because the field size is small, the geometric distortion is limited for each picture, and the radiation exposure is reduced from scattered radiation. The measurement of the pelvic inlet and mid-pelvis in lateral and AP views are obtained by imaging the ruler and the anatomical structure at their intersection and subtracting the two readings to obtain the distance of interest.

The fluoroscopic apparatus was slightly modified to facilitate accurate and quick measurement of the pelvis. We developed a set of clamp on lights to act as a collimator during the orthometric pelvimetry. These lights which could easily be removed at the termination of the examination, were held in place by springs and by a permanent magnet. To support the radiopaque ruler for the pelvic measurements, a plastic table was positioned on the fluoroscopic table top.

In the clinical setting, the polaroid television images of the mid-pelvis and pelvic inlet had adequate detail for the required calculations, (Fig. 4, 5). The average mid-pelvic radiation exposure to the mother in the direct beam is 1/100 of that for film cassetts radiography while the fetal exposure is reduced another factor of 100. The fetal exposure reduction is a result of removing the fetus from the direct beam and from using a small field size of 4 cm x 4 cm which reduces the scattered radiation. By further reducing the field size to 2cm x 2cm for each polaroid image. We can further lower the exposure. Since the polaroid fluoroscopic television technique for pelvimetry is an inexpensive and simple method that gives adequate clinical data with a radiation exposure much reduced compared to standard radiography.



Figures 4 & 5: Polaroid Television Images of Mid-Pelvis
IV. STEREOTAXIC POSITIONING FOR NEUROSURGERY

Electronic radiography has been developed for neurosurgical procedures by coupling a two track magnetic disc recorder to a portable television fluoroscopic unit. The apparatus, enables rapid stereotaxic needle positioning for the treatment of intracranial aneurysms.

In the clinical setting for the repair of the intra-cranial aneurysm, the C-arm was placed in the AP position centered over the approximate location of the aneurysm. First, the apparatus was switched into the "MAP" mode and an electronic angiogram showing the location of the aneurysm was made. Then with the apparatus switched into the mode "STORED + MAP" successive electronic images were taken showing the needle guide superimposed with the frozen image of the aneurysm as the guide was moved into position, (FIG. 6).

After positioning had been completed, the stereotaxic needle was inserted through the skull into the aneurysm. To monitor this insertion, the apparatus was placed in a lateral position and a new angiogram, taken

in the MAP mode, was superimposed with the needle as it was advanced through the brain into the aneurysm using the FLUORO - MAP mode. Tissue adhesive was injected into the aneurysm via the needle. Using the FLUORO mode the extent of occlusion of the aneurysm was evaluated by injecting contrast material into the carotid artery to outline the vessel.

The electronic technique permits successful localization and treatment of an intracranial aneurysm with procedure time and patient trauma markedly reduced compared to the traditional craniotomy.



Figure 6: Electronic Radiograph of Stereotaxic Needle Guide Superimposed with the Electronic Angiogram of the Aneurysm

The radiation dose to the patient for each electronic radiograph was 1/100 of that for one minute of continuous fluoroscopy. By superimposing single electronic radiographs on a stored map, the surgeon required only a few images, each of the order of a milliroentgen significantly reducing the radiation exposure to personnel in the operating room.

V. SELECTIVE CATHETERIZATION

By coupling a 150 track disc recorder to a plumbicon television fluoroscopic unit, we have developed electronic radiography for selective catheterization to reduce procedure time, radiation exposure, and the volume of injected radiopaque contrast material. There are three levels of refinement of electronic catheterization which can be applied depending on the degree of difficulty encountered in the procedure. These

methods have been clinically applied to selective catheterization of both neuro and visceral vessels.

A. Individual Electronic Radiograph

In this technique the angiographer takes a single electronic radiograph which he studies with the x-rays off to determine the amount and direction the catheter should be manipulated to achieve successful selective catheterization. These images showing the catheter relative to bony landmarks such as vertebrae and ribs substitute for live fluoroscopy. The progress of the manipulated catheter can be recorded on successive electronic radiographs.

B. Electronic Radiograph Guided by Map

An electronic angiogram is recorded during an aortic injection of radiopaque contrast. The arterial map showing the aorta with the branch vessel is electronically combined with successive individual electronic images of the catheter and the combined image is displayed over the TV monitor. By studying the position of the catheter tip relative to the branch blood vessel, the angiographer can evaluate the amount and direction of repositioning required for successful selective catheterization.

C. Arterial Map with Live Image Superimposed

This method, as in the previous one, uses the electronic angiogram (arterial map) as a guide for selective catheterization. In this method however the live fluoroscopic image of the catheter is electronically superimposed on the map. In a preliminary clinical trial in 20 patients, the three variations described were found to simplify the procedure and reduce the radiation dose. By obviating the need for repeated test injections of radiopaque contrast, the electronic arterial maps greatly decreased the total amount of contrast injected during catheterization. By utilizing a series of images stored on a magnetic disc, electronic radiography replaces continuous live fluoroscopy by the production of electronic images requiring much less radiation than that for

conventional fluoroscopy. This technique reduces procedure time, radiopaque contrast, and the radiation exposure.

VI. CONCLUSION

In the proper clinical setting electronic radiography will reduce the radiation exposure from 10 to 10,000 times of that with standard techniques in diagnostic radiology. The principle advantages of this method are that the clinical information determines the level of radiation exposure required and as a consequence of the efficient use of radiation various image enhancement techniques will yield a final electronic image with higher resolution and contrast than standard spot films in some cases. In addition to these advantages electronic radiography is convenient and opens the way for new treatment methods.

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MEDICAL RADIATION PROTECTION IN THE EASTERN MEDITERRANEAN REGION

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Abstract

Six hundred and fifty diagnostic X-ray installations, representing over 50 percent of the existing ones, were surveyed in sixteen countries of the Middle East in 1969/70. A number of technical and human deficiencies were observed. Sixty-five percent of the X-ray units surveyed lacked one or more of the following radiological safety features: Adequate filtration; beam no larger than needed to cover the X-ray film; adequate operators' protection; and adequate protection of neighbours and all other personnel. The measures taken to tackle these problems will be described.

Introduction

The Eastern Mediterranean Regional Office of WHO undertook early in 1969 to assist the X-ray departments in hospitals, and medical and dental departments in the countries of this Region in the evaluation and eventual improvement of the radiation safety of patients and of medical and auxiliary personnel exposed to ionizing radiation in the course of diagnostic or therapeutic procedures.

From 14 February to 7 November 1969, the WHO Technical Offices in X-rays visited ten countries of the Eastern Mediterranean Region.

During the stay in these countries he has visited 154 institutions which included medical schools, hospitals, cancer centres, tuberculosis clinics, dental schools, dentists, and private physicians (Table I); surveyed and measured field radiation levels on a total of 334 X-ray installations (Table II); instructed and demonstrated practical means of reducing dose levels to 744 X-ray operators which included radiologists, physicists, and technicians (Table III); repaired, adjusted, and recalibrated approximately 50 X-ray units; instructed dark-room personnel on processing methods leading to improvement of film quality; discussed with public health and hospital administrators the need for introducing radiation protection legislation and for establishing film-badge services; and at construction sites advised responsible authorities on the design and construction of adequate premises to accommodate new X-ray installations.

Observations

The main shortcomings observed were:

- 2.1 human deficiencies
- 2.2 technical deficiencies

2.1 Human Deficiencies

The medical and para-medical personnel the Technical Officer normally met were (Table III) radiologists, radiation health physicists, X-ray engineers, and X-ray technicians.

Of the 104 radiologists met, approximately forty were expatriate doctors employed on government contracts. It is difficult to estimate precisely the shortage of radiologists. It is likely however, that the number of radiologists required is twice the number presently available.

In the ten countries visited, only five qualified radiation health physicists are available. No information could be obtained on the number of health physicists undergoing training abroad.

Radiological health inspectors do not exist in these countries.

Only four x-ray engineers are employed by their respective governments in the ten countries visited by the WHO Technical Officer. A few commercial firms keep qualified engineers on their staff in a few countries.

Training schools for X-ray technicians are operating in four of the countries visited and an attempt at training assistant X-ray technicians is now starting in a fifth one.

Approximately one third of the 634 X-ray technicians met have attended training courses varying in duration from six months to two years. As the diagnostic radiology departments are heavily dependent on the activities of this category of personnel, at least twice the number presently available is required to adequately cope with the current workload.

Low standards of exposure control and collimation were prevalent in the radiograms performed by technicians or non-radiologists. Considerable effort in improving this situation should be made and educational programs directed to this end are of great importance. Fluoroscopic examinations were often carried out by non-radiologists and even by X-ray technicians. Referring physicians often request radiological examinations without sufficient reasons, thus the yield in terms of diagnostic information is very little and patients are unnecessarily exposed to radiation.

Where radiologists are not available and other medical officers must perform radiological examinations, an adequate radiological training should be required from them.

2.2 Technical Deficiencies

2.2.1 Radiological

Sixty-five percent of the X-ray units surveyed lacked one or more of the following safety features: (Table IV)

- adequate filtration
- beam no larger than needed to cover the X-ray film
- adequate operator's protection
- adequate protection of neighbours and all other personnel (Table IV)

A number of these defects were actually rectified in the course of the visit. In most cases this could be done at small cost. Only in a few cases did the Technical Officer advise to stop further operation of the X-ray units as the

defects were highly dangerous from the point of view of radiation and/or electrical safety.

2.2.2 Electrical

Approximately 40% of the X-ray units seen were connected to electrical mains supply which could not provide the required power. It was also noticed that the X-ray units were connected to the same lines as other high consumers of electricity (elevators, sterilizers, etc.) thus being subject to gross power fluctuations. Most of the electric outlets (plugs) in wards where portable units are connected (bedside radiography) lacked a proper earth wire, thus exposing operators and patients to electrical hazards.

2.2.3 Dark Rooms

About 40 to 50% of the dark rooms have serious defects. Among the most frequently seen: lack of ventilation, light leaks, unprotected electrical fixtures, no safe-lights or incorrect filter used. Some do not have running water.

More than 80% of the dark rooms lacked one or more of required accessories, i.e., thermometers, timers, driers. Damaged cassettes, intensifying screens, hangers were often seen.

Discussion

The analysis of the data contained in this report leads to some considerations on the adequacy of:

1. Radiological Services (Table V)
2. Radiation Protection (Tables IV, VI, VII)

1. Radiological Services

Table V shows in a very striking way the insufficiency of radiological services in the ten countries so far surveyed.

There is an average of 72,000 people (range 11,250 to 317,000) for each diagnostic X-ray unit, as compared with 1,000 people/unit in the United States.

The estimated average film consumption in the countries surveyed, 0.063 films/person-year, represents only one-fortieth of the average film consumption in the U.S.A. (2.46 films/person-year).

This should be kept in mind in order to place the radiation hazards to the population at large into a proper perspective.

2. Radiation Protection

We have seen in Table IV that only 48% of the operators and 58% of all other personnel occupationally exposed to ionizing radiation could be adequately protected by suitable structural or movable shielding, lead-glass screens, distance, etc. And yet only one-fourth of the operators (physicians, radiographers) in the ten countries surveyed (Table VI) are equipped with personnel monitoring devices. Since the number of people occupationally exposed (operators and all other personnel) is much larger, the personnel being monitored represents only a small proportion (perhaps less than 10%) of those exposed to ionizing radiation. In the U.S.A. about one-third of the personnel occupationally exposed are equipped with personnel monitoring devices.

Table VII pools the results of the survey of 334 installations in ten countries showing the percentage of units complying with some of the most essential radiological safety features.

Conclusions and Recommendations

The insufficiency of radiological services both in personnel and equipment in some of the countries surveyed is obvious.

The careful study of this report leads to evident conclusions regarding some of the remedial measures that should be taken without delay. They are as follows:

A. Stepping Up Training:

1. Of radiologists and radiological physicists.
2. Of X-ray technicians, through national courses.
3. Of X-ray technician-tutors and of technicians specialized in the maintenance and repair of X-ray equipment.
4. Of radiological health inspectors.

B. Promulgating Radiation Health Legislation:

Empowering the Ministries of Health:

1. To establish a system of registration, inspection, and licensing of X-ray, radioisotope teletherapy, and unsealed radioisotope sources and their users.
2. To promulgate rules, codes of practice, and regulations for the safe use of radiation sources.

C. Setting Up or Expanding National Services

1. For monitoring of personnel occupationally exposed to ionizing radiation.
2. For radiological health inspections.

TABLE I
TYPE AND NUMBER OF INSTITUTIONS VISITED

Country	Private Physician	Dentist	Hospital	Tuber- culosis Clinic	Total
1)	-	-	4	-	4
2)	-	-	7	1	8
3)	-	-	18	3	21
4)	-	-	6	2	8
5)	10	-	7	2	19
6)	3	1	23	1	27
7)	-	-	21	3	24
8)	-	-	15	4	19
9)	2	2	5	1	10
10)	-	-	12	2	14
Total					154

TABLE II
TYPE OF INSTALLATION SURVEYED

Country	Dental	Fixed Rad.	Fluoro.	Therapy	Portable	Photo-fluoro.	Combined Rad. and Other Fluoro.		Total
1)	-	2	-	-	-	2	5	-	9
2)	1	-	1	-	-	2	7	-	11
3)	1	6	4	-	1	3	21	-	36
4)	-	-	1	2	2	2	9	1	17
5)	-	6	4	4	1	2	22	-	39
6)	1	7	-	4	-	2	38	-	52
7)	-	7	17	2	7	2	29	-	64
8)	-	2	5	6	16	4	23	1	57
9)	4	2	1	2	4	1	6	-	20
10)	2	3	3	3	3	-	14	1	29
Total									334

TABLE III

Country	Radiologists	Physicists	X-ray Operators
1)	1	0	22
2)	0	0	27
3)	12 (f)	0	79
4)	10	2	42
5)	15	0	92
6)	27	1+1 WHO	107
7)	14	1	120
8)	10 (f)	0	92
9)	6	1	30
10)	9 (f)	0	23
Total	104	6	634

TABLE IV

Country	A			B			C			D			Total units surveyed	% of units in which one or more of A,B,C,D, features were missing
	Filtration adequate			Beam no larger than needed to cover X-ray film			Operator can be adequately protected			All other personnel within permissible limits				
	Yes	No	% Compl.	Yes	No	% Compl.	Yes	No	% Compl.	Yes	No	% Compl.		
1)	6	3	66.5	4	4	50	6	3	67	4	4	50	9	50
2)	3	8	27	6	5	54.5	5	6	45	3	8	27	11	73
3)	28	8	78	26	3	89	24	12	67	26	12	68.5	36	33
4)	9	8	53	9	2	82	11	6	65	16	1	94	17	47
5)	12	27	31	17	10	63	21	18	53	28	9	76	39	69
6)	20	31	39	39	6	87	29	23	56	47	4	92	52	61
7)	7	52	12	28	13	68	29	35	45	29	35	45	64	88
8)	13	44	23	27	13	68	23	34	40	12	38	33	57	77
9)	9	11	45	11	5	69	6	14	30	13	6	68.5	20	70
10)	9	20	31	15	3	83	6	23	21	5	24	17	29	83
Total	116	212	35 (Av.)	182	64	74 (Av.)	160	174	48 (Av.)	183	141	56 (Av.)	334	65 (Av.)

TABLE V
RADIOLOGICAL SERVICES

Country	Population (1)	Estimated no. of diagnostic X-ray units	Population per diagnostic X-ray unit	No. of physicians (2)	No. of physicians per X-ray unit etc	No. of radiologists	No. of radiographers	No. of operators/unit etc	Estimated annual X-ray film consumption	Estimated no. films/person-year
1)	260,000	15	17,333	117	7.8	1	22	1.5	98,500	0.378
2)	2,755,000	20	137,750	86	4.3	0	27	1.3	171,500	0.062
3)	23,782,000	75	317,093	320	4.3	12	79	1.2	464,500	0.019
4)	2,251,000	20	112,550	505	25.0	10	42	2.6	182,750	0.081
5)	5,724,000	50	114,480	978	19.5	15	92	2.1	266,250	0.046
6)	2,588,000	230	11,252	2,025	8.8	27	107	0.58	574,250	0.221
7)	4,463,000	100	44,630	666	6.7	14	120	1.3	590,250	0.132
8)	1,675,000	100	16,750	530	5.3	10	92	1.0	519,500	0.310
9)	620,000	40	15,500	460	11.5	6	30	0.9	165,250	0.266
10)	5,100,000	35	145,714	84	2.4	9	23	0.9	30,500	0.015
Total	49,218,000	685	71,851 (Av.)	5,771	8.4 (Av.)	104	634	1.08 (Av.)	3,113,250	0.063
USA	200,000,000	206,560 (3)	970 (Approx.)	387,422 (3)	1.83			1.5 (3)	506,000,000 (4)	2.446

- (1) Demographic Yearbook, 1966, UN.
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 (3) Lawrence R. Fess, Summary of Diagnostic X-ray Statistics Relating Facilities, Equipment, and Personnel by Healing Arts Professions, Radiological Health Data and Reports, Vol. 10, No. 9, Sept. 1969, pp. 379-380.
 (4) John H. Knowles, Radiology - A Case Study in Technology and Manpower, New England Journal of Medicine, 280, 1271-1278, (19).

TABLE VI
PERSONNEL MONITORING
(Film-Badges)

Country	Total No. of Operators	Personnel Monitored	%
1)	23	0	0
2)	27	0	0
3)	91	21	23
4)	52	52	100
5)	107	0	0
6)	134	68	51
7)	134	16	12
8)	102	6	6
9)	36	23	63
10)	32	2	6
Total	738	188	26

TABLE VII
MAIN RADIOLOGICAL SAFETY FEATURES
Pooled Results of Ten Countries
(% Units Complying)

EQUIPMENT	Filtration adequate	35.4
	Beam no larger than needed to cover film	74
	Tube housing leakage within normal limits	100
	Table top dose < 10 R/min	96
	Fluorescent screen interlocked with tube	91
	Fluoroscopic shutters adequate	92
	Lead glass on fluorescent screen adequate	97
OPERATORS	Operator can adequately be protected	48
	Exposure of all other personnel within permissible limits	56

SOURCES OF UNNECESSARY IRRADIATION DURING FLUOROSCOPY.

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Abstract

644 gastrointestinal and 731 chest fluoroscopies performed by 6 radiologists were investigated concerning : the duration in time, the area exposed, magnitude of exposure in X-ray beam, the gonadal and bone marrow dose.

The patient exposure was found to depend on : 1.mean time duration of the examination, which varied for the six different radiologists, from simple to double (from 169 to 259 sec. for gastroduodenal fluoroscopy, 48 to 80 sec. for intestinal, and 11 to 62 sec. for chest fluoroscopies) ; 2. quality and exposure rate of primary beam (the exposure) rate ranged from 3 to 12 R/min.) ; 3. area exposed during fluoroscopy (cross-section of primary beam varied from 26 to 230 cm² for gastroduodenal, from 38 to 383 cm² for intestinal and from 47 to 490 cm² for chest fluoroscopy ; 4. the value dose area product proved to be the most interesting parameter, variations between 420 and 3160 R.cm² for gastroduodenal, 124 to 2091 R.cm² for intestinal and 100 to 473 R.cm² for chest fluoroscopies were recorded, the corresponding integrated dose values were from 3.5 to 26.8 kg. rad for gastroduodenal, from 4.0 to 16.6 kg. rad for intestinal and from 0.8 to 4.0 kg. rad for chest fluoroscopies.

The diagnostic informations being practically the same, the variations of the operational parameters encountered to the six investigated radiologists, evinces the existence of at least three sources of unnecessary irradiation : 1. the use of a too large beam (large surface and integrated dose) ; 2. the dose rate of primary beam too high ; 3. too long time of irradiation ; this situation maybe related to the knowledge level of the radiologist.

1. Introduction

Previous research has shown that fluorescopic examinations represent the major contribution to medical irradiation of the population in Romania. Due to the great number of fluoroscopies performed in this country, over 300 per 1000 inhabitants, instead of 80-110 in other countries, we were interested in the assessment of the sources of unnecessary irradiation during fluoroscopies.

Fluoroscopy is a nonstandardizable type of X-ray examination, its performance being entirely dependent on accuracy of the radiologist's knowledge and on the interest he pay, to the benefit vs.risk ratio.

The higher benefit/risk ratio maybe obtained by performing the fluoroscopy in that way to find maximum of diagnostic information with minimum of irradiation dose to the patient.

Objectivation of sources which contribute to lowering the above mentioned ratio in view of their control is the main aim of this paper.

2. Material and method

Six radiologists working in three outpatient clinics in Bucharest were subjects of the present investigation.

Measurements were done during 644 gastrointestinal and 731 chest fluoroscopies concerning : 1. time duration ; 2. cross-section of useful beam ; 3. dose area product ; 4. dose rate of X-ray machine ; 5. doses received by some tissues and organs (skin, bone marrow, gonads, thyroid, eyes, a.s.o.). Technical parameters (KV, mA) used by the radiologist were also recorded.

For measurements we have used : 1. the Diamentor (PTW Py-chlau) for dose area-product ; 2. the VA-J-15A Dosimeter (Vakutronick) for dose rate ; 3. thermoluminescent dosimeters TLD loo (readed at a 2000 Harshaw apparatus) for tissue and organ dose. All dosemetric facilities were calibrated at the WHO-IAEA Regional Reference Centre for Secondary Standard Radiation Dosimetry in Bucharest.

3. Results and discussions

Table 1 is showing the mean values data obtained for g.i. fluoroscopies.

Table 1.

Time, dose. area product, exposed area, integrated dose and dose rate used by six radiologists in Bucharest during gastrointestinal fluoroscopies (mean values).

	Dr.P.	Radiologist				
		Dr.G.	Dr.I.	Dr.L.	Dr.T.	Dr.S.
<u>I. Upper g.i. fluoroscopy:</u>						
Time (seconds)	82.5	212.3	161.8	258.8	206.2	194.7
R.cm ²	1880.0	3160.0	1300.0	2430.0	900.0	420.0
cm ²	210.0	95.0	220.0	230.0	53.0	26.0
kg.rad	15.0	26.8	10.4	19.3	7.4	3.5
dose rate (R/min.)	7.3-12	10.6-11.3	3.0	3.0		5.0-8.2
<u>II. Lower g.i.fluoroscopy:</u>						
Time (seconds)	51.9	-	48.1	64.1	79.8	36.9
R.cm ²	2091.2	-	556.9	735.7	480.6	124.1
cm ²	383.2	-	337.5	222.8	67.7	37.6
kg.rad	3.9	-	4.5	6.0	3.1	0.8

Data obtained for chest fluoroscopies ~~are~~ shown in table 2.

Table 2.

Time, dose, area product, exposed area, integrated dose and dose rate used by six radiologists in Bucharest during chest fluoroscopies (mean values).

	Radiologist					
	Dr.P.	Dr.G.	Dr.I.	Dr.L.	Dr.T.	Dr.S.
Time (seconds)	11.2	27.3	42.8	62.4	54.3	25.4
R.cm ²	473.2	432.0	356.0	494.0	371.0	99.7
cm ²	490.0	122.8	302.4	363.9	83.6	47.1
kg.rad	3.9	3.6	2.9	4.0	3.1	0.8
dose rate(R/MIN)	5.2	7.7	1.6	1.6	4.7	5.0

Both tables 1 and 2 are displaying the same phenomenon - the irradiation of the patient during g.i. and chest fluoroscopy is due mainly to cross-section of useful beam and not to the intensity and quality of the beam used.

If we compare, for the six radiologists we have studied, the dose area product, exposed area and integrated dose with time and dose rate the above mentioned phenomenon becomes more evident.

For g.i. and chest fluoroscopies the differences between maximum and minimum values for dose area product, exposed area, integrated dose, time and dose rate are the following :

	upper g.i.	lower g.i.	chest
dose.area product (R.cm ²)	420 to 3160 7.9	124 to 2091 16.9	99.7 to 494 4.9
exposed area (cm ²)	26 to 230 8.8	37.6 to 383.2 10.2	47.1 to 490 10.4
integrated dose (kg.rad)	3.5 to 26.8 7.7	0.8 to 6.0 7.5	0.8 to 4.0 5.0
time (seconds)	82.5 to 258.8 3.1	36.9 to 64.1 1.7	11.2 to 621 5.6
dose rate (R/min.)	3.0 to 12.0 4.0		1.6 to 7.7 4.8

From the above figures results that the greatest difference between minimum and maximum mean values obtained for the six radiologists was that of exposed area. An order of magnitude is separating the minimum and maximum values for the above mentioned parameter both in g.i. and chest fluoroscopy.

The dose.area product which reflect not only the cross-section but also the intensity of the primary beam is also very relevant about the unnecessary exposure of patient. For lower g.i. fluoroscopy especially dr.S. has obtained the diagnostic information with 1/17th part of the roentgen area product used by dr. P. Concerning upper g.i. and chest fluoroscopies the differences are smaller but still important (1/8 and 1/5).

Integrated dose, measuring the true tissue dose, is much

more important from the radiological point of view. This parameter also reveals differences between absorbed dose in patient of 5 fold in chest fluoroscopy and 7.7 or 7.5 in upper and lower g. i. fluoroscopy.

By comparison the differences concerning the duration of procedures were only 1.7 to 5.6 fold and the dose rate of primary beam 4.0 - 4.8 fold.

Data above discussed are consistent in showing that the main sources of unnecessary irradiation of the patient during fluoroscopy are the following, in order of their importance :

1.- the cross-section of the primary beam-expressed in our study as the exposed area of the patient ;

2.- the intensity of the primary beam ; use of higher m. values produces, together with larger beam, greater dose.area products and integrated dose values ;

3.- the duration of exposure, which results from the fact that the radiologist try to obtain as many information is possible from fluoroscopy and not from combining the use of fluoroscopy and radiography.

A realistic programme for control of unnecessary irradiation during fluoroscopy requires the improvement of the knowledge of radiologists concerning :

a) - use of smallest possible cross-section of primary beam during fluoroscopy except for a very brief initial general view ;

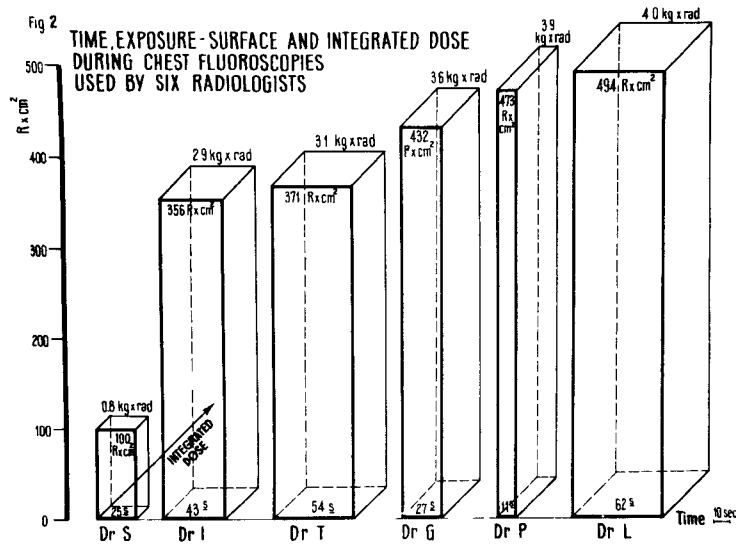
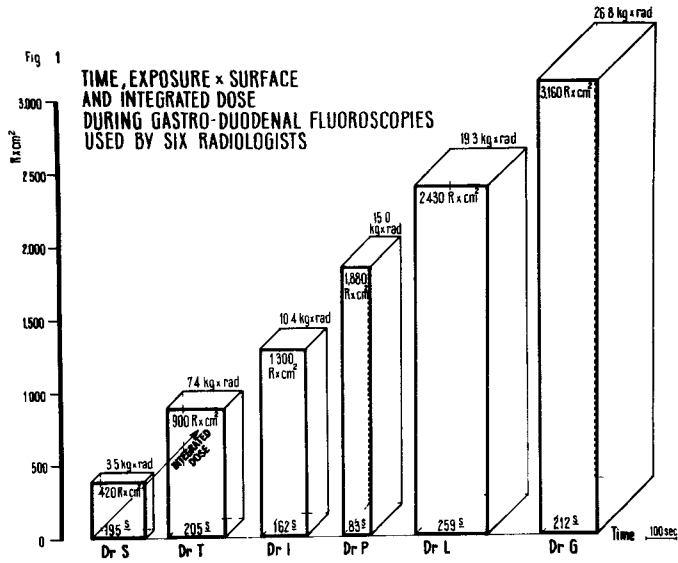
b) - decrease of intensity of tube current and increase of KV_p in view of obtaining the lowest dose rate for the useful beam;

c) - take of radiographs during fluoroscopy in cases which are not clear enough on fluoroscopic screen avoiding the prolongation of the patient exposure.

As dose.area product and integrated dose are also a measure of the irradiation received by the radiosensitive tissues and organs (bone marrow, gonads, a.s.o.) the significance of these parameters from the viewpoint of radiation protection is highly relevant. The decrease in roentgen.area product and kg.rad values have a real value in control of unnecessary irradiation during radiological diagnostic procedures.

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MEDICAL IRRADIATION OF THE POPULATION IN ROMANIA
DURING 1970

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Abstract

X-ray examinations have increased in Romania between 1953-1970 from 429 to 1,012 per 10^5 inhabitants, photofluorographies (54 to 452) and radiographies (37 to 238) being developed while radiosopies remained at the same level (338 and 322).

Age group 16 to 30 and over 31 years are the most X-rayed averaging 1.22 examinations/inhab./year. Males are predominantly investigated at all ages and the most frequent examinations concern the chest, G.I.tract, upper and lower members, vertebral column, pelvis, teeth, a.s.o.

Were calculated for 1970 the genetically significant dose and the mean bone marrow dose, the value obtained been 28.5 and 382 mrad respective. Radiographies of the addominal and lumbar regions in female had the main contribution to genetically significant dose and the G.I.series, chest fluoroscopies and photofluorographies to mean bone marrow dose.

1. Introduction

Medical use of ionizing radiations is still the major source of artificial irradiation of the population throughout the world.

The present work has attempted to estimate population's genetically significant dose and mean bone marrow dose due to medical irradiation in Romania.

2. Increase of the X-ray diagnostic procedures in Romania

Statistical records concerning all medical activities were developed in Romania and since 1953 we have yearly data referring X-ray procedures in all medical units throughout the country.

The data show the fact that radiographies and photofluorographies have increased 6.4 and 8.4 times during the last 18 years; at the same time radiosopies have increased very slowly until 1964 and are decreasing after that year.

3. Distribution of X-ray diagnostic procedures.

Using data recorded during 1970 by 98 different X-ray diagnosis departments throught the country we have established on 145,525 radiosopies, 115,271 radiographies and 231,800 photofluoroscopies the distribution of X-ray procedures on: sex, age

group (0-15,16-30 and over 31 years) and body area exposed. The result of the examination coded as positive and negative was also mentioned.

Due to the shortage of space we are obliged to present only in a very condensed form these results.

The number of examinations per person in total population were as follows :

- up to 15 years	- boys	0.50 examinations
	- girls	0.48 examinations
- between 16-30 years	- male	1.36 examinations
	- female	1.09 examinations
- more than 31 years	- male	1.27 examinations
	- female	1.17 examinations

Data show that the age group 16-30 years, which is very interesting from the point of view of genetically significant radiation dose, performs an increased number of examinations, especially the men, fact which must be taken into account in any programme of reduction of medical irradiation.

4. Exposure of patient during the radiological examinations

The exposure received by the patients was determined by direct measurements on the patient during different kind of examinations, using pocket ionization chambers and TLD-100 Harshaw dosimeters.

The measurements were performed in 62 medical X-ray diagnosis units, during 5,370 radiographies, 8,750 radioscopies and 9,370 photofluorographies.

The dosimeters were calibrated in the energy range from 60 KVp (0.056 mm Cu HVL) to 120 KVp (0.400 mm Cu HVL) at the WHO IAEA Regional Reference Centre for Secondary Standard Radiation Dosimetry in Bucharest.

The measurements of the exposure received by patients during X-ray examinations were done by positioning the dosimeters at several points on the surface of the body. The points for gonadal dose determinations were the anterior and posterior projections of the ovary on the skin in female and testes in male. Bone marrow determinations were done at the most important skeletal part with red bone marrow, in the vicinity or directly in the primary beam. The values obtained in such a way are skin doses and we used during the calculation a correction factor of 0.3 (experimentally determined) for the true bone marrow dose (table 2).

For a realistic estimation of patient dose during radioscopies, the average time duration of radioscopies measured during our field investigations was used : 68% of chest fluoroscopies were until 30 seconds and 11% over 60 sec., as for G.I. fluoroscopies 41% were until 80 sec., 54% between 80-240 sec. and 5% more than 240 sec.

5. Genetically significant dose

The following simplified equation for the genetically significant dose (GSD) was used (see also reference 1) :

$$GSD = \frac{\sum D_i N_i P_i}{\sum N_i P_i}$$

where :

- D_i = mean gonadal dose from certain type of examination received by a patient of age group "i" ;
 N_i = number of persons of age group "i" who were exposed to the specified type of examination during 1970 ;
 P_i = expected number of children per person of age group "i" and mentioned sex ;
 N_i = number of persons of age group "i" and mentioned sex in the total population.

Using data from chapters 3 and 4 we found the gonadal dose of the population in Romania during 1970 (table 1, part I).

The calculus of genetically significant dose using the equation mentioned above and data from table 1 gives a value of $GSD=26.5$ mrad/year/inhabitant, which is comparable with the values obtained in others countries as USA (1954)-55 mrad, Sweden (1955)-28 mrad, Japan (1960)-39 mrad, Denmark (1956)-22 mrad, United Kingdom (1957-1958)-14 mrad and New-Zeeland (1963)-12 mrad.

6. Mean bone marrow dose

The mean bone marrow dose was calculated in a simple way by integrating of all values for bone marrow dose produced during a certain type of investigation in persons of a specified age group and dividing by the total number of persons of that age group.

Using data from chapters 3 and 4 and values of distribution of population from table 1, we found the bone marrow doses of the population in Romania during 1970 given in table 2 and finally as result, the mean annual bone marrow dose per inhabitant per type of examination (table 2, last column).

As it can be seen, the bone marrow dose averaged over entire population resulting from various X-ray examination was 302 mrad/year, in comparison with 32,4 mrad/year estimated in United Kingdom by Auriar Committee (1957-1958, reference 5).

The mean bone marrow dose arising from chest radioscopies was 92 mrad/year/inhabitant in comparison with 50 mrad (Austria), 10 mrad (Belgium), 600 mrad (France), 8 mrad (Spain) and 14 mrad (Switzerland).

Bone marrow dose seems to be the most important health consequence of the medical irradiation. We have not a good estimation of leukaemia expectancy in Romania so that we couldn't calculate the leukaemogenic significance of bone marrow irradiation.

The fact that age group 10-30 years, which has enough large life expectancy (in 1970 in Romania the mean life expectancy was 66 years for men and 70 for women) receive an important part of medical irradiation, gives us the real idea referring the programme for reduction the unnecessary irradiation.

7. Conclusions

The results of our work concerning the medical irradiation of the population in Romania allow to point out the following conclusions :

a.- medical irradiation in Romania constantly increases; an increase of 2.4 times in 1970 can be presented for total X-ray diagnosis procedures in comparison with 1953 (6.4 times for radiographies and 0.4 for photofluorographies) and a slowly decrease for radioscopies (322 radioscopies in 1970 per 10⁵ inhabitants instead of 337 in 1953) ;

b.- the number of examinations carried out during 1970 by one inhabitant varied with age group and sex. It was 0.50 for boys between 0-15 years, 0.48 for girls, 1.26 for men between 16-30 years, 1.27 for men over 31 years, 1.09 for women between 16-30 years and 1.17 examinations for women over 31 years ;

c.- the genetically significant dose in Romania during 1970 was 28.7 mrad per inhabitant, a medium value in comparison with those established in other countries in the world;

d.- the mean bone marrow dose per inhabitant in Romania during 1970 arising from x-rays diagnosis was 382 mrad, a value which put in evidence the necessity of a programme for reduction of the unnecessary medical irradiation.

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Table 1. Gonadal dose of the population (rad), distribution of population and children expectancy, per age group and sex in Romania during 1970.

	Sex	Age group			Total
		0-15 years	16-30 years	over 31 years	
		Gonadal dose (rad)			
I. Type of examination:					
A. Radiographies	M	4,100	19,475	101,680	125,255
	F	26,035	164,000	776,130	966,165
B. kadioscopies:					
Chest	M	410	1,743	3,075	5,228
	F	717	1,743	4,205	6,755
G.I.	M	615	4,100	12,505	17,220
	F	1,845	5,535	18,450	25,830
C. Photofluorographies	M	3,075	1,743	2,562	7,380
	F	7,175	7,790	14,555	29,520
Total		43,972	206,129	933,262	1.183,363
II. Distribution of population (x10 ³)					
	M	2,972	2,578	4,920	10,270
	F	2,829	2,338	5,063	10,230
Total		5,801	4,716	9,983	20,500
III. Children expectancy					
	M	2.953	2.798	0,453	
	F	2.885	2.402	0,310	

Table 2. Bone marrow dose of the population (rad) per age group and mean marrow dose per inhabitant (mrad) in Romania during 1970.

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Type of examination	Age group			Total (rad)	Mean annual bone marrow dose per inhabitant (mrad)
	0-15 years	16-50 years	over 51 years		
A. Radiographies	22,072	62,359	299,974	384,405	19
B. Radioscopies :					
Chest	241,427	559,373	1,112,915	1,913,715	92
G.I.	100,390	495,274	3,160,082	3,756,346	103
C. Photofluorographies	80,770	038,000	1,083,133	1,201,903	88
T o t a l	425,067	1.756,286	5,656,104	7,837,457	382

Use of Medical X-Ray Diagnostic Units in Iraq

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Abstract

The medical use of X-rays represents a major source of population radiation exposure. A special consideration is given to assess the genetically significant dose due to diagnostic radiology.

All medical institutions, hospitals, private physicians and dentists in Iraq, were visited to record: the number and type of X-ray units, the frequency and kind of X-ray diagnostic examinations and number of workers in such units. In addition the protection of the workers from the radiation and the services of personnel monitoring were also observed.

It was found that, up to the end of 1972, there are 407 X-ray units, all over the Republic used for whole body medical roentgenodiagnosis and 146 X-ray units for dental radiography, carrying out more than 4×10^6 and 15×10^4 examinations per year, respectively.

The study gives a clear map of the distribution of the X-ray units in the different parts of the country and can be used as a guide for future radiological health programs. It might also serve as a model for radiation survey and inspection. Also, the figures could be useful in the estimation of the annual genetically significant dose received by the 10 million Iraqis from exposure to diagnostic X-rays.

Introduction

X-ray diagnosis in the application must be used only when highly specifically indicated. The radiation dose received on an individual ground varies with the examination used, the number of exposures during their reproductive period of life and the techniques used.

Diagnostic X-rays started to be used for medical purposes in Iraq since 1917 / 1 /. No scientific measures, concerning protection of personnel and patients were taken until 1972 when a highly specialized committee was established to study this problem.

A legalized regulations were issued by this committee applying what the ICRP suggested for radiological protection and permissible doses / 2-3 /.

Therefore in response to the need of radiological health program for an effective method of surveying of all radiation sources, the Radiation Control Board requested all medical institutions and hospitals, to register radiation sources and give full details of their uses.

The survey covered approximately 99% of the radiation machines, which constitute the subject of this paper for the year 1972.

Materials and Method

Regulations were issued restricting the sale and import of diagnostic X-ray films or any spare parts without permission. Such permission is now given only after a visit to the X-ray machine in order to record the number and type of the unit, the frequency and type of X-ray examinations performed, output measurements at various tube voltages and the different age groups of the patients of both sexes. In addition, the protection of the workers from the radiation and the services of personnel monitoring were also observed.

With regard to the dose measurements, skin dose estimation either of the critical organs was made directly on patients during different types of X-ray examinations. Film badges type L.R.P. 30 Black Spot, such as personnel monitoring film badge with Kodak radiation-monitoring films were used for these measurements for practical reasons and availability. The contribution of radiography to gonadal dose is a complex function of applied peak kilovoltage, tube target to skin distance, volume of the tissue in the primary beam, the sex and age of the patient. / 4 /.

The dose received by gonads in 70 patients exposed to radiation for diagnostic purposes was calculated. The genetically significant dose was obtained from the following formula / 5 / :

$$D = \frac{\sum_{j,k} \Pi_{jk}^{(F)} W_{jk}^{(F)} d_{jk}^{(F)} + \sum_{j,k} \Pi_{jk}^{(M)} W_{jk}^{(M)} d_{jk}^{(M)}}{\sum_k \Pi_k^{(F)} W_k^{(F)} + \sum_k \Pi_k^{(M)} W_k^{(M)}}$$

where

D = (annual) genetically significant dose.

Π_{jk} = (annual) number of individuals of age-class k, subjected to class j exposure.

Π_k = total number of individuals of age-class k.

W_{jk} = future number of children expected by an exposed individual of age-class k subsequent to a class j exposure.

W_k = future number of children expected by an average individual of age-class k.

d_{jk} = gonad dose per class j exposure of an individual of age-class k.

(F) and (M) denote "female" and "male" respectively.

Results

A. Number of machines and area distribution :

A total number of 553 X-ray machines were used in Iraq for diagnostic purposes (Table I), 407 conventional X-ray units are used for general diagnostic medical purposes, the remaining 146 units are used only for dental purposes. Some of these machines are quite old (since 1934).

30.5% of the conventional X-ray machines are found in private clinics, while in the case of dental X-ray machines the number of the machines in the private clinics were higher than the governmental machines (53.3% of the total dental machines).

With regard to the area distribution, Baghdad (the capital) has the higher number of machines; Basrah came the next and Nineva came the third. Other provinces are more or less similar in the number of the X-ray machines used.

Table 2. shows the different firms of both conventional and dental machines used in Iraq. Since there are approximately 10 millions inhabitants in Iraq at the end of 1972 /6/, it means that the average is 1 medical diagnostic X-ray machine for each 25×10^3

inhabitants and 15 dental X-ray machines for each million inhabitants.

Table I. Distribution of X-ray machines by Governorates (Muhafadha).

Muhafadha	Inhabitants %	Diagnostic			Dental		
		Public	Private	Total	Public	Private	Total
Duhok	11.1	4	-	4	1	-	1
Nineva		18	9	27	6	5	11
Arbil	4.5	12	2	14	1	-	1
Kirkuk	5.9	13	6	19	2	3	5
Sulaimaniye	4.7	11	3	14	3	-	3
Diyala	4.5	9	1	10	1	-	1
Baghdad	25.5	107	79	186	29	53	82
Al-Anbar	3.9	11	1	12	2	-	2
Babylon	5.7	11	2	13	1	2	3
Karbala	4.2	11	4	15	4	3	7
Al-Nadisiye	6.8	9	2	11	1	1	2
Al-Muthana		5	1	6	1	-	1
Wasut	4.2	7	2	9	2	-	2
Thi-car	6.4	9	1	10	4	3	7
Measan	4.3	12	1	13	3	1	4
Basrah	8.4	34	10	44	7	7	14
Total	100	283	124	407	68	78	146
		69.5%	30.5%	100.6%	46.6%	53.4%	100%

Table 2. Distribution of X-ray machines in Iraq by manufactures.

Manufacturer	Diagnostic			Dental		
	Public	Private	Total	Public	Private	Total
Siemens	53	46	99	6	7	13
Phillips	48	30	78	1	7	8
General Electric	29	10	39	11	-	11
Generay	85	-	85	-	-	-
Watson	30	7	37	-	-	-
Tur	29	6	35	-	-	-
Explor	-	-	-	-	29	29
Lavo	-	-	-	24	-	24
Honda	-	-	-	11	-	11
Ritter	-	-	-	1	5	6
Other Types	8	15	23	6	25	31
Unknown	3	10	13	8	5	13
Total No.	283	124	407	68	78	146
%	69.5	30.5	100	46.6	53.4	100

B. Number of X-ray Examinations :

In the estimation the number of radiographs taken during fluoroscopic examinations were included in the list under radiography and one examination means one exposure. All types of examination concerning radiography and fluoroscopy were classified to 10 forms as shown in Table 3, which also illustrates details regarding the age, sex and area examined in a total number of one thousand patients examined in different diagnostic X-ray machines except

5

mass miniature radiographies and dental, because it was difficult to know the sex of patients in these last two types of examinations.

Table 3. Frequency in thousand of diagnostic examinations by age, sex and type examination.

Type of Examination	years												Total				
	M	15		16-20		21-30		31-45		46-50		50+		M	F	%	
Hands	12	5	5	1	15	3	3	3	3	1	1	3	39	16	5.5		
Head&Neck	12	25	10	12	37	27	13	22	3	3	8	17	83	1.6	18.9		
Feet	7	3	2	7	5	7	7	3	5	1	1	1	27	22	4.9		
Chest	25	25	5	32	27	35	23	37	15	10	32	8	127	147	27.4		
VertebralCol.	3	3	3	10	22	7	17	7	5	7	12	7	62	41	10.3		
Gall Bladder	1	1	1	1	3	3	1	5	1	5	1	1	8	16	2.4		
Stomach	1	1	3	15	32	25	25	22	7	8	12	5	80	76	15.6		
Urinary Tr.	15	2	8	10	17	18	11	13	2	11	3	6	56	60	11.6		
Belvis	3	3	1	3	1	5	1	1	1	1	2	1	9	14	2.3		
Bre,nancy	-	-	-	5	-	5	-	1	-	-	-	-	-	11	1.1		
Total	M	79		38		159		1.1		42		72		491		1000	
	F	68		96		135		114		47		49		509			
%		14.7		13.4		29.4		21.5		8.9		11.1		49.1		50.9	100%

27.4% of the total examination in Table 3 were performed for chest X-ray examination, while the head and neck X-ray examination came to be second in frequency (18.9%), stomach and the surrounding organs (gall bladder and liver) constituted about 18% of the total X-ray examination done. Pelvis X-ray diagnosis constituted about 3.4% of the total X-ray tests. The urinary tract X-ray diagnostic tests constituted 11% of the total examinations.

With regard to age grouping, Table 3 shows that 14.7% of the test were done on patients under the age of 15 years of both sexes. While 13.4% of the tests were done on patients between 16-20 years old.

The age group 21-45 years which constitute the active reproduction age specially in women, reached 51.9% of the total number examined in this table. With respect to the other older age groups, it appears that a relatively small percentage of patients were examined. Regarding the total male to female ratio of the one thousand patients examined in this work, the ratio was nearly one. The official census of the population indicates that there are 5,073,600 males and 5,000,600 females in Iraq at the end of 1972/6/.

The total number of radiographic examinations was 4.2×10^6 diagnostic examinations. Nearly 21% them were mass miniature radiographies. While the annual total number of dental X-ray examination was about 150×10^2 examinations. The estimation shows that the patients performed in average 2.2 examinations each visit. Therefore it appeared that the frequency is 2 persons from 10 inhabitants were undergone diagnostic examinations yearly. Moreover, about 15 persons out of 1000 had dental X-ray examinations annually.

Table 4a shows the total number of X-ray examinations done on different parts of the body of both sexes and the gonad dose measured from each particular examination.

The man-rad/year received, was the highest in performing the abdomen, which is in the range of 7.8×10^5 man-rad/year. The mass chest X-ray miniature constituted the second man-rad/year received. Other clinical examinations gave a significant decrease in man-rad/year dose in comparison with the above mentioned diagnostic tests. The total man-rad/year for all types of

examinations 8×10^5 man-rad/year.

Table 4a. Total gonad dose in man-rad due to examinations of both sexes (Figures are taken 1000 exm.per year).

Type of Examination	Male		Female		Total	
	No. exm.	gonad dose man-Rad	No. exm.	gonad dose man-Rad	No. exm.	gonad dose man - rad
Mass miniature extremities	225	0.220	129	0.130	354	0.350
Head & Neck	282	0.280	360	0.360	642	0.640
Chest	423	0.860	500	1.000	922	1.860
Abdomen	731	329	741	445	1472	774 99%
Dental					150	0.300
Total	Gonadal dose 779.85×10^3 man-rad/year.					

Table 4-b represents the genetically significant dose received by both sexes of the Iraqi population. It is apparent that the dose received from the use of the dental machines is significantly smaller than the dose received from the conventional X-ray diagnostic procedure. The calculation of the annual genetically significant dose resulted in a value of 52 m rad for 1972.

The accuracy of this result is probably of the order of 60%. The number and technical data for measurements are reported by the author in a separate paper/7/.

Discussion Table 4b. Genetically significant dose by sex (mrad/person per year).

There is no general standard system to distribute the X-ray machines over the different parts of Iraq. But one can make his own conclusion from table I, which reflects the relation

Sex	Type of exam.		Total G.S.D. mrad/year	%
	Diagnos. mrad /year	Dental mrad /year		
Male	25.62	0.15	25.77	49.65
Female	26.03	0.15	26.18	50.35
Total	51.65	0.30	51.95	100

between the density distribution of the population and the X-ray units. The relatively high number units of (45.7% and 56.1%) of the total conventional diagnostic and dental units respectively in Baghdad area is due to the number of its inhabitants (25.5% of the total population of the country) and the same situation is true in Basrah and Nineva. Another reason for this distribution may be attributed to the preference of most doctors to live and work in large cities. On the other hand, wherever the official numbers of units are high, the number of private units are high too. This may be due to the permission is given only to specialist doctors in radiological fields to possess X-ray units in private clinics. This permission is given to any dentist. It is found that the private specialists in medical radiology themselves are mainly the official ones. Sometimes specialists with high qualification in other fields might be granted a permission too.

A total number of 407 conventional X-ray machines performing an annual frequency of 420 X-ray examinations per 1000 persons. This figure of examinations is similar to the figures obtained in many other countries which had carried out comprehensive survey /5/ while our number of instalations per 1000 of total populations are less. From Table 2 it is shown that there are only 6 firms

which supplied about 90% of the conventional units and 7 firms supplied about 70% of the dental X-ray machines. This situation makes it easy for detestation of the dose received from all machines. The average operating peak kilovoltages has been found to be 70 KVP in conventional machines and (50-60).KVP in dental.

Data of Table 3 included sex categories by sex and age. Each category encompassed the types of examinations. It seems that 20% of the total patients are less than 30 years old. The last percentage of patient is to be considered when discussing genetic effects.

The abdominal (stomch, vertebral column, gall bladder and urinary tract) examinations which comprised 33.7% of all the examinations, give exposure values representing about 99% of the average gonadal exposure as shown in Table 4a. In estimation of genetically significant dose, the individual gonad dose is weighed with a factor taking in to account the future number of children expected. However the total gonadal dose is about 780×10^5 man-rad/year, the genetially significant dose resulted is 52 mrad/year which is a high figure in comparison to that of other countries having frequencies between 8 and 44 mrad/year/4, 5, 8, 9/. However, this dose is still below the dose recommended by ICRP, which is 5 rem over a period of 30 years.

Conclusions

1. The study gives a clear picture of the X-ray unit distribution in different parts of Iraq, which is somewhat satisfactory.
2. The annual number of radiographic examinations is expected to be soon more than 5×10^6 . It is thought advisable to start a local film production. As long as films are, for the time being, imported, this study will help very much in the estimation of the annual need of these films.
3. Since the genetically significant dose (52 mrad/year) is higher than that in other countries, beside that we have no data for the past years about the number of diagnostic X-ray examinations per year, it seems that there is an urgent need for the reduction of the annual genetically significant dose from X-ray examinations, although very few people are being exposed.

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RADIOBIOLOGICAL SIGNIFICANCE OF ^{125}I MICRODOSIMETRY

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Abstract

Significant difference in the microscopic dose distribution due to ^{125}I and ^{131}I in the thyroid gland is reported. Its radiobiological significance is studied using varying doses of these two radionuclides in one month old rats. The total body weight increase, thyroidal retentivity of radioiodine, 24 hour uptake and the thyroid weight at the end of the treatment are measured in both the treated and control groups. Thyroid weight is taken as an index of thyroid cell population and any reduction in its increase with age compared to that in controls (which is taken as 100% cell survival) is considered as due to the impairment of proliferative capacity of the thyroid cells. Average gland doses due to ^{125}I and ^{131}I for 50% cell survival are 40500 and 18500 rad respectively. The 24 hour uptake is significantly reduced in 71 uCi ^{131}I treated rats whereas in all other treated ones there is no change compared to controls. Body weight increase is impaired in both the treatments, more so with ^{125}I . These differences are explained in terms of differences in dose distributions across a thyroid follicle due to ^{125}I and ^{131}I .

Introduction

For the same activity administered the mean dose to the thyroid gland is 2-3 times less with ^{125}I compared to ^{131}I . Microscopic dose distribution across a thyroid follicle, which is a basic unit of the thyroid gland, shows that with ^{125}I the dose at the colloid-cell interphase is 2-6 times and 4-10 times that over the nucleus and that at the basal membrane respectively depending upon the colloid content in the gland whereas with ^{131}I it is practically uniform¹⁻³.

The radiobiological significance of this difference in microscopic dose distribution was studied in the adult rat thyroid by different workers⁴⁻⁶. However, the normal adult rat thyroid has a closed, well differentiated cell system with little proliferation and upon irradiation (with about 500 rads of X-rays) shows little change as judged by weight, cell counts or DNA and RNA content⁵. So to see the effects of irradiation on thyroid cell population, particularly on its proliferative capacity, it is necessary to promote the cell multiplication artificially, say by administration of methyl thiouracil.

Instead, young rats of one month age were chosen for the present investigation. At this age their thyroid cells are rapidly proliferating⁷ and the thyroid weight increases by a factor of 2-3 within a span of 2 months. So a study of the radiation effects is possible without the administration of a drug.

Materials and Methods

One month old Holtzman strain male rats were used for the study. They were fed with standard dry pellet Hindustan Liver rat food and tap water ad libitum. 5 rats were taken for each dosage. One of the following doses in a volume of 0.2 ml was injected intraperitoneally to each group: 100, 50, 25, 12.5 and 2.5 uCi of Na^{125}I ; 71, 24.2 and 4.8 uCi of Na^{131}I . Two groups of 5 rats each served as respective controls for the two radionuclides. The total body weight and thyroid retention of radioiodine (by in vivo counting with an end window

G.M. Counter over the thyroid) were measured periodically.

When the rats attain 3 months age they were injected intraperitoneally a tracer dose of ^{125}I for measuring the thyroid uptake at the end of the treatment. 24 hours after administration of the tracer the rats were sacrificed. The two lobes of the thyroid from each rat were taken out and weighed correct to 0.2 mg. The thyroidal activity was obtained using a Nuclear Chicago Autogamma well counter and was expressed as the percentage of the injected activity per unit thyroid weight.

Results and Discussion

Variation of Thyroid Weight and Radiiodine Uptake with Age:

Fig.1 shows the variation of thyroid weight and 24 hour radioiodine uptake as a function of age. The mean thyroid weight of 30 d old rats is 6.8 ± 2.3 mg for a mean body weight of 45.2 ± 7.3 g while that of 100 d old ones is 24.2 ± 2.6 mg for a mean body weight of 210 ± 18.1 g corresponding to an increase in the thyroid weight by a factor of 3.5. However, the variation in thyroid weight expressed as a fraction of body weight is only from 0.154 ± 0.006 at 30 d to 0.115 ± 0.006 at 100 d. The 24 hour thyroid radiiodine uptake expressed as a percentage of the injected dose per unit thyroid weight in mg varies from 0.059 ± 0.0049 to 0.016 ± 0.0049 with the age of the rat. Higher uptake per unit thyroid weight in the one month old rats indicates the hyperactivity of their thyroids.

Biological Half-life of Radiiodine in Thyroid:

The *in vivo* thyroid activity is monitored periodically for all the animals and Fig.2 gives the retention pattern in arbitrary units as a function of time. In case of ^{125}I , the retention could be expressed as a single exponential with a half-life of 10-13 d, irrespective of the dose administered. So only the data corresponding to 100 uCi group is presented in Fig.2. The pattern of retention of ^{131}I was more complex, consisting of at least two components. The size and half-life of the two components depended on the dose administered. The half-life of the fast component increased and that of the slow component decreased with the decrease of dose.

Estimation of Average Gland Dose:

The average gland dose, D, is estimated for each treated rat from the knowledge of its 24 hour uptake, U, the thyroid weight, m, in g and the effective half-life, T, in h using the well known equation

$$D = 1.44 \text{AUT} \sum \Delta_i \phi_i / m \quad \text{rad}$$

where A is the activity administered in uCi, \sum the summation sign, Δ_i , the equilibrium absorbed dose constant in g-rad/uCi-h, and ϕ_i , the absorbed fraction in the thyroid gland for i th component. The value of $\sum \Delta_i \phi_i$ is calculated to be 0.0506 for ^{125}I using the values of Δ_i and ϕ_i listed in MIRD pamphlets⁹⁻¹¹ and includes both penetrating and non-penetrating components. For ^{131}I the contribution due to photons is neglected. Considering the thyroid as a sphere the absorbed fraction for ^{131}I beta radiation is obtained using Berger's tabulation of ϕ for different sphere sizes. The value of ϕ is about 0.8 for the rat thyroid¹⁰.

The mean gland dose corresponding to 100 uCi ^{125}I treated group is 34322 ± 9560 rad. The value for 71 uCi ^{131}I treated group is 21820 ± 4034 rad.

Body Weight vs. Days after Administration of Activity

Figs. 3 and 4 give the ratio, Y, of the body weight on the day of observation to that on the day of the administration of activity, plotted against the time, X, elapsed from the day of administration of activity for the ^{125}I and ^{131}I treated groups respectively. The data in each case is statistically analysed and the respective regression equations, correlation coefficients, r, and the standard errors, S_{XY} , are given in the same figures.

The data shows that there is a reduction in the body weight ratio of the treated groups in comparison to the control groups and the reduction is more pronounced in the case of ^{125}I . The reduction in the body weight indicates that there is probably a disturbance in the release of the right amount and right type of iodinated compounds which affect body growth and that the disturbance is pronounced with ^{125}I .

24 hour Radioiodine Uptake at the end of Treatment:

The 24 hour uptake was obtained for all the treated and control groups by administration of a tracer dose of ^{125}I , just one day prior to sacrifice. The results are given in Table 1. The uptake value for all the treated groups (except that of 71 uCi ^{131}I) is the same as that of the corresponding control group(C). The lower uptake of 71 uCi ^{131}I group may be due to a damage of the iodine trapping mechanism and/or due to a decrease in the number of follicular cells per unit thyroid weight.

Table 1: Radioiodine Uptake at the end of ^{125}I and ^{131}I treatments

^{125}I treatment		^{131}I treatment	
Mean Gland Dose(rad)	24 hour uptake	Mean Gland Dose(rad)	24 hour uptake
34322(100uCi)	0.028±0.0059	21800(71uCi)	0.020±0.0094
24867(50uCi)	0.033±0.0092	15793(24.2uCi)	0.044±0.0099
12604(25uCi)	0.030±0.0078	2500(4.8uCi)	0.044±0.0047
6182(12.5uCi)	0.025±0.0073	C	0.037±0.0047
1299(2.5uCi)	0.032±0.011		
C	0.029±0.0065		

Percentage Cell Survival vs. Average Radiation Dose to the Thyroid:

In Fig.5 the percentage cell survival is plotted against the average gland dose for both ^{125}I and ^{131}I treated groups. The thyroid weight at the end of the treatment normalised to the then body weight is taken as an index of cell survival. The percentage cell survival is calculated by the following equation:

$$\% \text{ cell survival} = (m/M)_j \times 100 / (m/M)_0$$

$$\text{where } (m/M)_j = \frac{\sum_{i=1}^{n_j} (m_{ij}/M_{ij})}{n_j} \text{ and } (m/M)_0 = \frac{\sum_{i=1}^{n_0} (m_{i0}/M_{i0})}{n_0}$$

Here m_{ij} and M_{ij} are the thyroid and body weights of the ith rat in jth treated group consisting of n_j number of rats. m_{i0} and M_{i0} are the thyroid and body weights of ith rat in the control group consisting of n_0 rats.

The values of D_{50} (the gland dose for 50% cell survival) for ^{131}I and ^{125}I respectively are 18500 and 40500 rad. Their ratio, 0.46, suggests that RBE for ^{125}I compared to ^{131}I is less than one which is contrary to expectation. This may be explained in terms of the differences in microscopic dose distribution across a thyroid follicle due to ^{125}I and ^{131}I .

Using Berger's scaled absorbed dose distributions¹², the dose distribution across a thyroid follicle was computed assuming colloid mass as 50% of the gland mass. In each follicle it is considered to be in the form of a sphere of 25 μm radius surrounded by follicular cells of length 9 μm with their nuclei 3 μm away from the apical membrane. In Fig.6, the ratio of dose rate at any point R_s , to that at the center of the colloid, R_0 , is plotted against the distance s . The values of R_s/R_0 at the colloid cell interphase, over the nucleus and at the basal membrane are 0.49, around 0.25 and 0.14 respectively. The average gland dose corresponds to that at colloid-cell interphase as the colloid content is assumed as 50% of the total gland mass with all the thyroid iodine concentrated in it².

So in case of ^{125}I treatment, the nuclei of the follicular cells at 50% cell survival level received only about 20000 rad as against the gland dose of 40500 rad. For ^{131}I , however, dose distribution across the follicle is more or less uniform and so the gland dose (18500 rad for 50% cell survival) itself is the dose to the nucleus. Thus, if the dose to the nucleus is considered the 50% survival dose is same with both the nuclides.

The same microdosimetric considerations explain the differences found in 24 hour uptake at the end of ^{131}I and ^{125}I treatments and their respective controls. The percentage cell survival at which the significant reduction in 24 hour uptake was observed with ^{131}I is 44.3%. The corresponding gland doses with ^{131}I and ^{125}I are 21800 and 46500 rad respectively. The dose at the basal membrane (which is considered as the site for trapping of iodine) of the follicular cell of 9 μm length is about one fourth that at the colloid-cell interphase, that is, about 12000 rad in case of ^{125}I . So the absence of reduction in 24 hour uptake in ^{125}I treated groups is due to the inhomogeneity in dose distribution resulting in only about 12000 rad at the site of trapping compared to the gland dose of 46500 rad. Since in case of ^{131}I treated group the gland dose (21800 rad) itself is the dose to the basal membrane also the reduction in 24 hour uptake observed can be expected.

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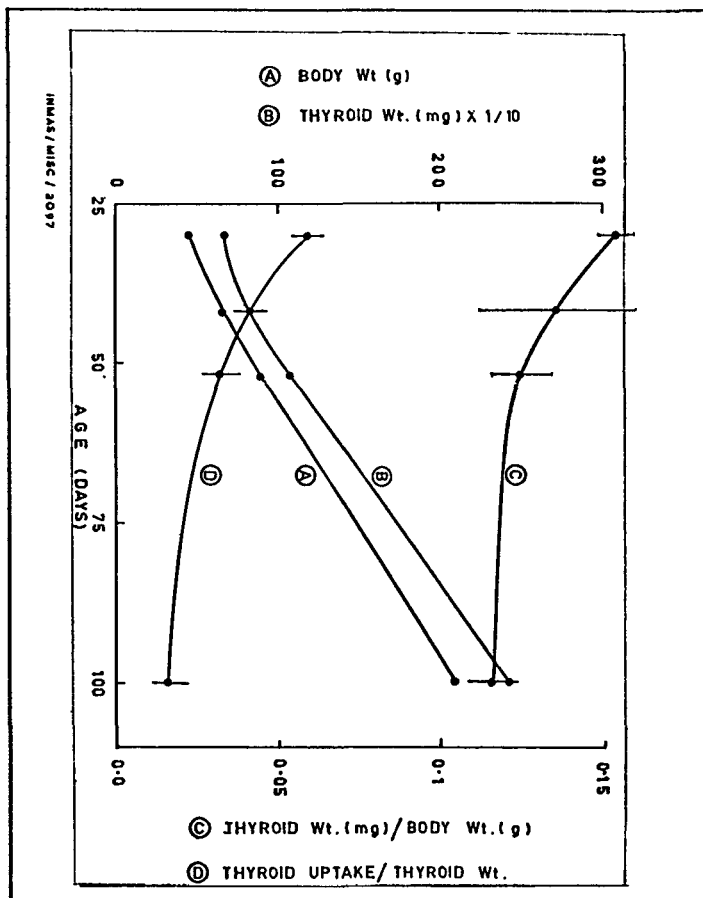


FIG. 1. Variation of body weight (A), thyroid weight (B), thyroid weight expressed as a fraction of the body (C), and 24 hour thyroid radioiodine uptake per unit thyroid weight (D) with age of rats.

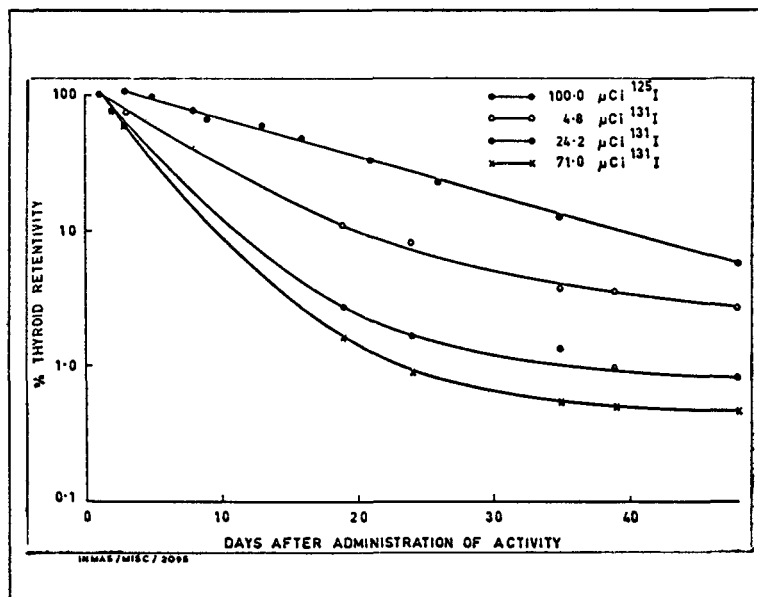


FIG. 2. Thyroid retentivity of radioiodine as a function of time in days after the administration of the activity.

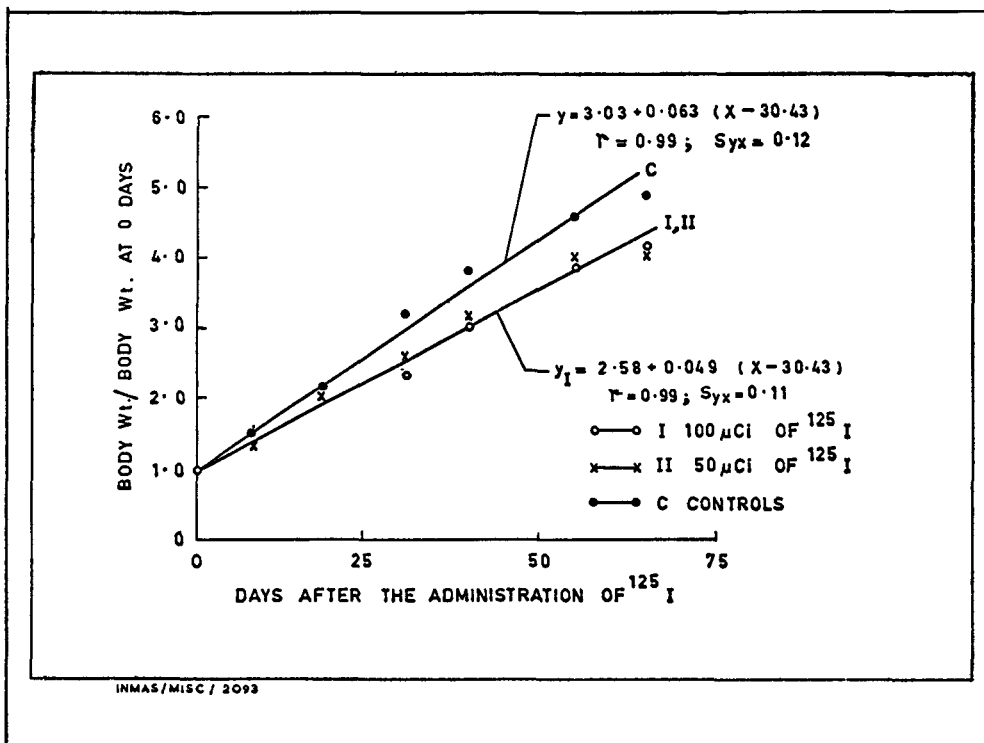


FIG. 3. Variation of the body weight ratio of 125 I treated groups and the respective controls as a function of time after the administration of the activity.

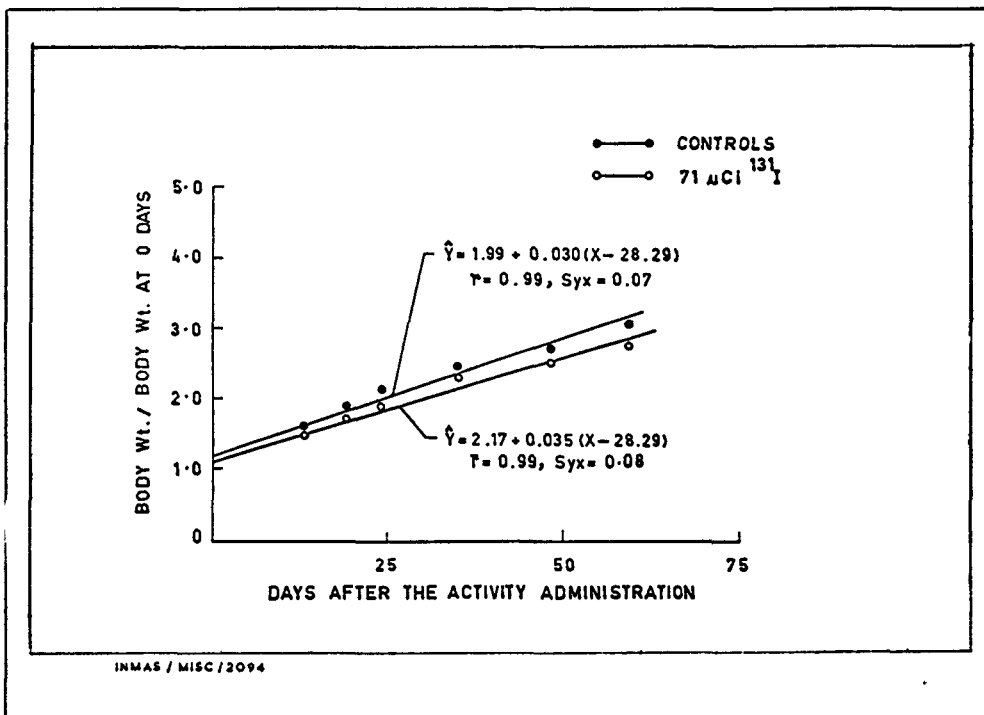


FIG. 4. Variation of the body weight ratio of 131 I treated groups and the respective controls as a function of time after the administration of the activity.

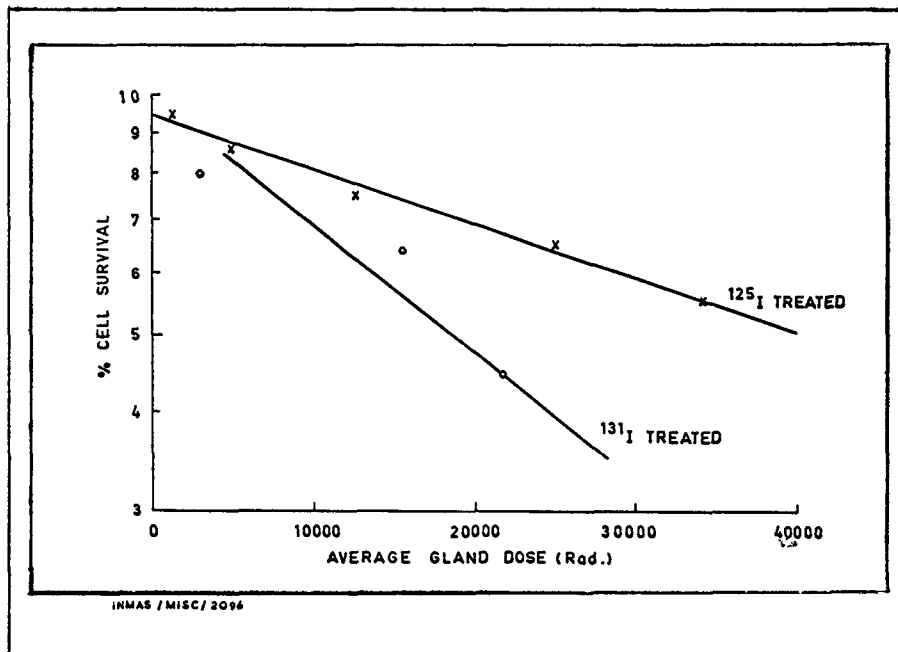


FIG. 5. Cell survival curves for ^{125}I and ^{131}I treated groups.

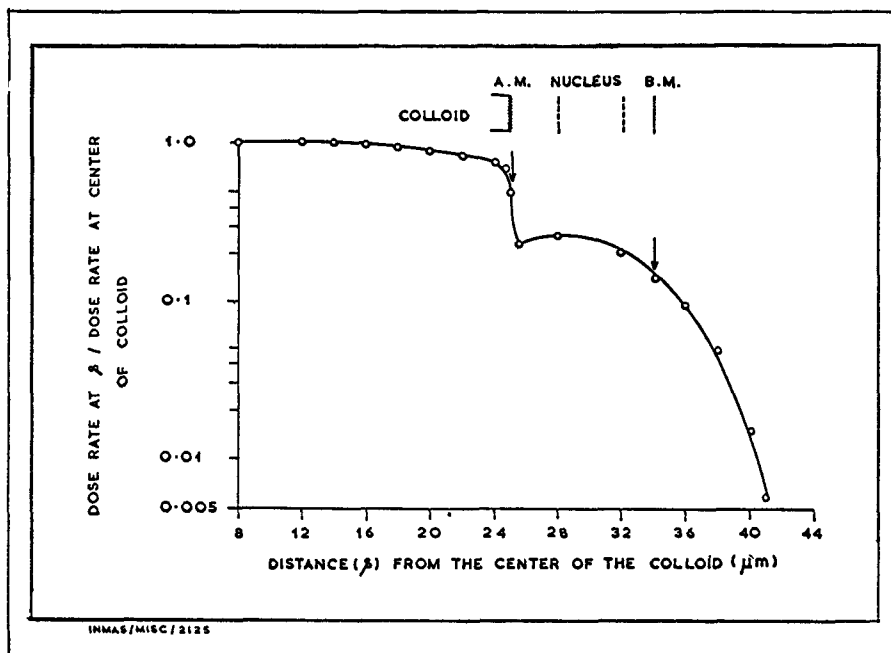


FIG. 6. Dose rate ratio, R/R_0 , for ^{125}I as a function of distance from the center of the colloid. At distances below $8\ \mu\text{m}$ the ratio is 1.0. The two arrows point to the positions of Apical Membrane (A.M.) and Basal Membrane (B.M.) of the follicular cell. Position of the nucleus is also shown.

DOSIMETRY OF INTERNAL EMITTERS IN NUCLEAR MEDICINE
AND RADIATION PROTECTION : AT WHAT LEVEL

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Abstract

The inadequacy of conventional dosimetry at the organ level assuming a uniform activity distribution has been brought out in specific examples. In each case dosimetry at different anatomical levels is presented bringing out the probable understanding and lacuna in the radiobiological consequences of such dosimetry. Bone dosimetry of alpha and beta emitters is presented. Dose to whole kidney and differential doses to medulla and cortex from ^{203}Hg -neohydrin are described. The mean dose to lung from ^{131}I -MAA, the mean local dose to capillary bed and to capillary epithelium adjacent to an MAA particle are reviewed. The importance of Auger electron emission and the consequent transmutational effects is discussed with the examples of ^{125}I in thyroid, ^{125}I -UDR in proliferating cells, ^{59}Fe in erythrocytes. High doses to limited portions of fingers while handling $^{113\text{m}}\text{In}$ and $^{99\text{m}}\text{Tc}$ labelled pharmaceuticals is presented.

Introduction

In nuclear medicine the clinician wants to know what are the potential hazards to a patient if he undergoes a particular investigative procedure which is expected to yield diagnostic information of clinical value. In the case of radiation protection the similar question is: what are the potential hazards to the worker under given exposure situation. The focus of interest is the biological effect which is the end-point in a complicated chain of events at the physical, chemical and physiological levels. Conventional dosimetry is mainly concerned with the primary event, viz. physical step, and the absorbed dose is an indicator only of physical events that happen at the macroscopic level. But we are far from understanding the biological significance of the absorbed dose. Partial consideration to the secondary events is attempted to be given by assigning a somewhat arbitrary value for the RBE or QF and quoting a rem dose.

For a proper evaluation of the biological effect, we should know the microscopic spatial and temporal distribution of the primary and secondary events. This leads to a detailed consideration of several factors, some associated with the radiation alone like radiation quantity and dose rate, others associated with the target as well, like location of radionuclide in the cell, the biochemistry of the labelled compound, bond rupture resulting from nuclear recoil after beta emission, chemical effect of nuclear transmutation on functional integrity of molecule, effect of sudden changes of charge on daughter nuclide (particularly important for isotopes decaying by electron capture), oxygen tension, cells at risk, radiosensitivity of cells in question, etc.

Further, concepts like LET and absorbed dose are macroscopic quantities or 'expectation values'. As the volume over which the absorbed energy is computed is reduced, the fluctuations associated with the stochastic nature of the interaction process assume increasing importance; concepts like 'event size', 'local energy density' and 'event size spectrum' have then to be introduced. This approach has not yet been made in practical situations of concern in internal dosimetry.

Very often we are not quite clear as to what is the biological end-point that is of relevance, although it is generally accepted that for comparatively

low doses as are encountered in routine occupational exposure or diagnostic nuclear medicine procedures, the effects may be classified into two categories, viz (i) those leading to the impairment of the functional integrity of the organ (this may be due to reduction in number of functioning cells or fibrosis with scarring) and (ii) induction of malignancies¹.

With all these complications, the question arises: At what level should dosimetry be done? Can we be satisfied with the conventional calculation of absorbed dose at the organ level assuming a uniform concentration of radionuclide? Or should we go down to the tissue, cell and even subcellular level? Should we consider the stochastic nature of the interaction process and enter into details of the microdosimetric concepts? What degree of sophistication is necessary and what degree sufficient?

The problem is discussed in terms of some well-known examples of practical interest.

Bone Dosimetry

Bone dosimetry is a classical example of the inadequacy of the conventional organ dose computation for an assessment of the potential risks. The inhomogeneous structure of bone and bone cavities (where the linear dimensions of the inhomogeneities are frequently of the same order as the range of the ionizing particles), the varieties of cells at risk and the non-uniformity of distribution of the radioisotope make the situation complex. A good deal of effort and ingenuity have gone into the solution of the problem from both the theoretical and experimental sides. From a consideration of the critical tissues for radiation damage, it is usual to calculate the following separately²:

- i) Dose to a very small tissue-filled cavity in the bone matrix, D_0 (to evaluate risk to osteocytes, cells lining Haversian canals and blood vessels in Haversian systems which are concerned with maintaining the functional integrity of bone as a living tissue).
- ii) Mean dose to endosteal cells near the surface of bone trabeculae in the marrow cavities, D_S (osteogenic sarcoma risk).
- iii) Mean dose to active marrow in trabecular cavity D_M (leukaemogenesis risk).

Typical results for radium and strontium-90 are shown in Table 1².

Table 1

Dose rates in rad/year from skeletal burden of 1 uCi

	Radiation	D_0	\bar{D}_M	\bar{D}_S/\bar{D}_M
^{226}Ra series	α	36	10.5	7
$^{90}\text{Sr} + ^{90}\text{Y}$	β	2.7	1.1	0.5

It is only by a detailed consideration of the cells at risk that we are able to perceive a major difference between the alpha and beta emitters. In view of the limited range of alpha rays, the bone marrow dose is only a small fraction of the endosteal dose in the case of alpha emitters. This is borne out by experience where we find that the incidence of leukaemia in radium

poisoning cases has been negligibly low and osteogenic sarcomas (and cancers of paranasal sinuses) are more common. With ^{90}Sr both leukaemia and osteogenic sarcomas have been induced in animals.

We shall next consider the question of non-uniformity of distribution of radium (and strontium) in bone. In addition to a diffuse distribution in the bone matrix, hot spots also occur where the local concentration may be 30-40 times the average concentration³. For ^{226}Ra , the range of variation of concentration is faithfully reflected in a corresponding range of variation of dose rates between the different concentration sites (factor of 16). Due to the longer range of beta rays from ^{90}Sr , the local dose rate variations are not that marked (factor of 3). We still do not know whether the hot spots play a role in radiation damage to bone and what the biological significance of the non-uniformity of damage is.

Neohydrin Dosimetry

Controversy was intense a few years ago whether ^{203}Hg -neohydrin should not be banned as a radiopharmaceutical for kidney and brain scanning in view of the high kidney doses. Neohydrin concentrates primarily in the cortex from where it is eliminated only very slowly. Since the cortical mass is about half that of the kidney, the dose to cortex would be twice that to the kidney as a whole had the isotope been uniformly concentrated. The cortex dose can be taken, as 146 rads and the medulla dose as 77 rads per millicurie of ^{203}Hg -neohydrin⁴.

We may discuss the question a little further. Is there firm evidence to show that 100 rads to the cortex is necessarily more harmful than 50 rads to the kidney as a whole? What is the biological end-point we are looking for? The natural incidence of malignant tumours of the kidney is quite small; also there is as yet no established case of radiation-induced kidney tumour. If malignancy is not the critical end effect, we have next to consider impairment of functional integrity. At the levels used in diagnostic procedures, gross impairment like acute or chronic nephritis is ruled out and much milder damage, which cannot be unambiguously pinpointed, must be considered. It appears that the fine vasculature is the histological site of damage of primary importance in the pathogenesis of radiation induced nephrosclerosis; the renal epithelium is relatively resistant but it may degenerate as a result of damage to the fine vasculature⁵. Since the proximal and distal parts of the tubules lie mainly in the cortex, it is not inconceivable that secondary tubular damage may be somewhat more intense from 100 rads to cortex than 50 rads to total kidney. On the other hand, it has been pointed out⁶ that the effective surface area of the renal cortex is about 4 times larger than the surface area of the kidney. Hence the escape of the beta radiations from the cortex will reduce this dose variation factor of 2 by an amount which has not yet been computed. The uncertainty remains.

^{131}I -Human Serum Albumin Macroaggregates for Lung Scanning

Uncertainty in the effective tissue mass to be considered in the dose computation can be illustrated by ^{131}I HSA macroaggregates in lung scanning. If a homogenous distribution of MAA in lungs is assumed, average dose to lung is about 1.5 rads for 30 μCi ⁷. If a more realistic volume distribution is assumed, viz. the capillary bed of the lungs with a mean diameter of 10 μm , the average local absorbed dose is nearly 5.5 rad. At the cellular level, absorbed dose to capillary epithelium adjacent to an MAA particle is several orders of magnitude higher than the average local absorbed dose, although this extreme dose is received only over a distance of one or two cell thicknesses. The problem is the anatomical level at which the dose is to be evaluated. If the induction of malignancy is the end-point of interest, the integral dose is

probably a valid indicator of the potential hazard. We do not yet know the biological significance of the very high level of local absorbed dose.

A somewhat similar situation exists in the field of radiation protection for assessing hazards from plutonium inhalation.

Significance of Auger Effect

Radionuclides decaying by electron capture and isomeric transitions are attractive in *in vivo* applications since they do not emit particulate radiations which give a radiation dose to the organ but do not contribute any diagnostic information. Several such radionuclides are now in common use, e.g. ^{51}Cr , ^{55}Fe , ^{57}Co , ^{58}Co , ^{67}Ga , ^{75}Se , ^{85}Sr , ^{123}I , ^{197}Hg and $^{99\text{m}}\text{Tc}$, $^{113\text{m}}\text{In}$. Certain special features of the electron capture process and the process of internal conversion consequent on gamma photon emission are of great relevance. After these two processes, the K or L shell vacancy initiates orbital excitation and electrons fall down from outer orbits successively into the vacancies in inner orbits. Excess energy is lost by X-ray emission in part but a large part of de-excitation occurs through the emission by the Auger effect of several electrons of low energy with a range in tissue of less than a micron. They, therefore, give a very high local dose to the tissue over a micron length. In addition their LET is very high and hence a correspondingly high RBE/QF will have to be postulated, leading to an intense local rem dose. We are still far from understanding the precise biological significance of this peculiar feature but some indications are available in the case of ^{125}I dosimetry which we shall touch upon subsequently.

Another consequence of Auger electron emission needs attention. As a result of the release of several Auger electrons, the daughter atom is left with a strong positive charge. If this charged nuclide is bound within a molecule it attracts electrons from various molecular positions and the positive charges are distributed throughout the molecule. The various positively charged atoms within the molecule strongly repel each other, which may lead to a virtual 'Coulombic explosion' of the molecule.

^{125}I Dosimetry

Conventional macroscopic dosimetry of ^{125}I in thyroid has been shown to be entirely inadequate in view of the special characteristics of the radiations from the radionuclide, and one has to go down to subcellular microdosimetry for obtaining a better understanding of the possible biological effects of ^{125}I . Since the range of Auger electrons is small compared to the dimensions of the thyroid cell, the cell-colloid interface or the apical membrane, which is the seat of thyroid hormone biosynthesis, receives a high dose. On the other hand the nucleus which is farther away gets only about one-fourth the dose to apical membrane. The variation between the nuclear and apical membrane doses is accentuated in thyrotoxic conditions in view of the greater distance of the nucleus from the apical membrane of the columnar cell of the thyrotoxic gland. The clinical significance of these observations has led to intense interest in the use of ^{125}I for therapy of thyrotoxicosis in preference to ^{131}I . However the clinical experience as well as the follow-up periods are as yet too small for an unambiguous conclusion to be drawn⁸.

The biological significance of the high LET of the Auger electrons and the possibility of Coulombic explosion mechanisms have not yet been clarified. Perhaps since the ^{125}I decay takes place mainly in the colloidal gel outside the apical membrane the latter effect may not be critical⁹.

On the contrary if ^{125}I is attached to essential structures such as DNA the influence of the charge transfer processes and the associated Coulombic explosion effects may be comparable to if not outweigh the radiation effects in producing the biological effect. This has been demonstrated^{10,11} while studying the relative effect of varying doses of ^{125}I -UdR, ^{131}I -UdR and ^3H -TdR on proliferating mouse cells *in vivo*. The toxicity of ^{125}I -UdR was reported to be 10 times greater than that with ^3H -TdR when these specific precursors of DNA were utilised by either bone marrow cells, or proliferating cells in the whole body in general. A similar finding was reported with ascites tumor cells also. The greater radiotoxicity of ^{125}I -UdR has been explained as due to a variety of factors including (i) differences in energy deposition in the cell nucleus per disintegrating atom (ii) greater inhomogeneity in the distribution of energy around the site of decay in the case of ^{125}I (iii) transmutational effects of ^{125}I , specially the consequences of molecular explosion.

^{55}Fe Dosimetry¹²

^{55}Fe , an electron capture radionuclide, is an important neutron activation product found in fallout. Levels as high as 3 pCi/mg blood have been recorded in New York residents. It has been computed that the dose to erythrocytes is about 10 times that to whole blood since the Auger electrons deposit their energy entirely within the erythrocyte itself where the ^{55}Fe is tagged. Dose to aggregates of ferritin molecules in which highest concentration of iron is found has been calculated to be about 200 times that to erythrocyte. However, the critical tissue in this case is perhaps the erythrocyte precursor cells in the bone marrow wherein a concentration around one-third of that in erythrocytes has been found, leading to a dose around 3 times the blood dose.

Skin Dosimetry

Skin dosimetry has acquired some urgency in view of the increasing use of short-lived isotopes like $^{113\text{m}}\text{In}$ and $^{99\text{m}}\text{Tc}$ in nuclear medicine. In the milking of the generator, the preparation of the radiopharmaceuticals and injection to the patient, levels of several tens of millicuries have to be handled at a time. The tips of two or three fingers, in particular, get exposed to significantly high doses (around 10 mrem/mCi-min). If the ICRP dose limit of 75 rem/year to hands is not to be exceeded, we would be severely curtailed in the scope of work; not more than two or three brain scanning preparations can be handled per person per week.

In this connection the health physicist turns to ICRP for guidance. A report of an ICRP Task Group¹ has something to say on skin dosimetry. The report recognises that the end-point of relevance here is not carcinogenesis, since the skin is relatively highly radioresistant, but radiation dermatitis. According to the report, in the case of irradiation of part of a tissue, the significant parameter is the mean dose to the entire tissue. If only a fraction of tissue is exposed, the dose allowed can be 1/f of dose limit for the whole organ. On this basis, if the dose limit for entire skin with an area of about 2 square meters is 30 rem/year, the dose to 1 cm² (of the order of high exposure areas of finger tips) could go as high as 60 kilorems. So why need we worry?

But the situation is not that simple. There is a limit beyond which the above concept cannot be extended. A vital consideration is the range of dose rate over which effective linearity of dose response can be assumed to hold. The point at which departure from linearity occurs will depend on the precise cellular mechanisms involved and the extent to which abscopal effects come into play. It may happen that at high doses the response may be higher than

would be predicted from a linear hypothesis since a number of contiguous cells are affected and irreparable physiological damage occurs. The Report says that linear response can be assumed up to hundred rems per year and possibly several hundred rems per year, and recommends that 'the present limit of 30 rem/year averaged over 1 cm² of skin be increased to at least 100 rems in a year with a proviso that irradiation of the same area year after year should be avoided if possible'. The health physicist wishes that the ICRP Task Group had categorically set a specific limit, say 500 rems per year, instead of vaguely leaving it at, 'at least 100 rems per year', so that he could ask the technicians, with some authoritative sanction behind him, to accept a higher work load with the generators. Note again the words, 'if possible'. In the present case of working with generators the irradiation is going to be received by the same area year after year unless a right handed person could be persuaded to become left-handed. Of course there is no sanctity about the limit of 1 cm² which is taken as the 'significant area' for averaging and the Report says '1 cm² seems reasonable on grounds of operational convenience'.

Conclusion

In this present paper dosimetry at different anatomical levels has been presented with the help of specific examples. In each case probable biological significance of such dose estimation also has been brought out. However, still there are several uncertainties in the biological significance of such detailed dose estimates and the importance of the possible transmutational effects with electron capture radionuclides.

Acknowledgement

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CONTAMINATION OF AIR AND SURROUNDINGS BY PATIENTS TREATED WITH LARGE QUANTITIES OF IODINE ^{131}I FOR THYROID CARCINOMA

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Abstract

On six occasions patients were treated with radioiodine for thyroid carcinoma and were isolated. The administered doses were in the range of 100 - 200 mCi of ^{131}I . All items from the room were collected, to determine the typical contamination deposited during the therapy. Continuous air monitoring was also performed. The total contamination collected on such items as bedding, towels and eating utensils was typically of the order of 1 mCi. Contamination of these items varied erratically during the first three days of therapy. Abnormally high levels of contamination were associated with certain medical disorders. Air burdens were in the range of 10 - 200 pCi per litre during the first day and usually declined progressively during the course of therapy.

Introduction

In radioiodine therapy of thyroid carcinoma, 100 mCi or more of ^{131}I is administered to the patient orally. This presents a unique radiation protection problem in the hospital since the patient is both a radiation source and a source of environmental contamination. The problem of managing the patient as a source of radiation is one which is common to other forms of therapy where radioactive material is administered or implanted.¹ The magnitude of the contamination problem with ^{131}I is not generally recognized. Most of the material is excreted in the urine and feces, but a few percent is excreted in saliva and perspiration.² During the initial hours after administration the saliva is especially rich in ^{131}I . This leads to contamination of the air and items in contact with the patient and other items in the room. Previous studies have attempted to determine the resulting ^{131}I burdens in medical and para-medical personnel caring for such patients^{3,4} and the dose to relatives exposed to outpatients treated with relatively small quantities for hyperthyroidism^{5,6}. We have described preliminary results of a study in which patients were isolated for a few days following administration of material for carcinoma therapy, during which time, room air contamination was determined. This paper is an extension of this latter study.

Experimental Details

This experiment has been in progress since May 1970. Five patients were treated and followed in this study, and one received two treatments during this interval, and was thus studied twice. The administered doses were in the range of 100 - 200 mCi as shown in table 1. Each patient was located in a private room and isolation precautions were observed. The nurses were supplied with over-gowns and gloves which they wore each time they entered the patient's room. Routine room cleaning operations were suspended during the days in which the patients were isolated. Food was supplied on disposable dishes and with disposable utensils. Each day the bedding and towels, and the disposable dishes and utensils and remaining food items were sealed into plastic bags and labeled accordingly. Some areas of the floor and particularly the bathroom floor, were covered with heavy brown paper which remained in place until the end of the period of observation. Air was sampled by drawing room air through two tubes in series filled with silver nitrate solution, using a pump of approximately two litres per minute capacity. This trap was placed about 1 meter from the bedside. The patients were allowed to move about within the room and were not unduly distressed by these special precautions, which were continued until the quantity of material in the patient had fallen below approximately 30 mCi. The rooms were well ventilated and the air changed approximately once per hour.

After the patient had been discharged, the bags of potentially contaminated items were counted in the whole body facility at New York University Medical Center. This consists of a single 8 inch sodium iodide detector which was placed sufficiently far from each of the bags of contaminated items to give less than 20% variation in detection sensitivity over the volume of each bag.

The air sampler flow rate was determined with the full impedance of the silver nitrate traps present. The quantity of iodine trapped in the silver nitrate during each sampling interval was determined by counting in a standard well-type scintillation counter. Comparison of the quantity trapped in the first and second tube indicated that this trap could be considered essentially 100% efficient for this source of air-borne radioiodine.

Results

The contamination collected on bedding, towels, dishes, utensils and other items is summarized in table 1. These values were all corrected for decay of the radioactive material between the time of collection and the time of measurement. The quantity of contamination found on bedding and utensils varied erratically during the several days of the therapy as shown. The average total of ^{131}I contamination on bedding, dishes and utensils averaged over all patients and normalized to 150 mCi administered dose is shown in table 2 for successive days of treatment. The concentration of radioiodine in the air is summarized in table 3, for the same five patients. Air sampling was not performed during the first treatment of patient A. Average air concentrations (normalized to 150 mCi administered dose) are also shown in table 3.

Patient	Administered Activity (mCi)	Contamination on Bedding & Towels (uCi)				Contamination on Dishes & Utensils (uCi)				Contamination on Bathroom Floor (uCi)
		1	2	3	4	1	2	3	4	
A1	200	370				20	180	-	-	-
B	200	31	25	25	3	66	27	-	-	12
C	200	127	37	39	390	570	105	110	490	-
A2	150	69	320	250		8	6	0	5	14
D	150	2	40	17	6	2	9	72	3	9
E	100	340	76	98	150	328				200

TABLE 1 Contamination measured on Bedding, Towels, Dishes, Utensils, and Bathroom Floor

Patient	Concentration in Air (pCi/litre)		
	Day 1	Day 2	Day 3
B	59	-	-
C	26	124	77
A2	178	73	48
D	15	7	5
E	10	8	-
Normalized Mean	55	47	37

TABLE 3 Measured Concentration of ¹³¹I in Air and Mean Concentration Normalized to 150 mCi Administered Activity

Day	1	2	3	4	TOTAL 1-4
Bedding & Towels	140 (2-500)	100 (20-300)	90 (20-250)	130 (2-300)	400 (60-1000)
Dishes & Utensils	100 (2-400)	50 (6-150)	50 (5-100)	180 (3-400)	300 (15-950)
TOTAL					700 (150-1500)

TABLE 2 Mean and (Range) of Contamination Measurements Normalized to 150 mCi Administered Activity

Items	Activity (uCi)
Nurses Gown	1
Thermometer	60
Telephone Mouthpiece	0 3
Nightgown	71
Water Jug	3 7

TABLE 4a Contamination Measured on Representative Items

Items	Activity (uCi)
Paper Tissues	3400
Respirator	252

TABLE 4b Exceptional Contamination on Individual Items

Contamination found on some individual items of special interest is listed in table 4. The figure of approximately 1 microcurie total contamination on the nurses isolation gowns was typical for all patients except where contamination by vomitus, stool or urine occurred. Following accidental contamination by stool, about 100 uCi was detected on an isolation gown. Thermometers, toothbrushes and containers for dentures were always contaminated. The telephone mouthpiece was always slightly contaminated.

Discussion

The air burdens and the levels of contamination were related to the behavior of the patients and to their medical problems. For example, levels of contamination on bedding were clearly related to the radioactivity in perspiration, to the time each patient spent in bed, and to medical problems such as incontinence. In general, contamination was either due to contact with saliva, perspiration, urine, feces or vomitus, or to exposure to iodine in air. The latter was assumed to be due to saliva spray but could also involve oxidation and volatilization of iodine on contaminated items.

The pathways of iodine metabolism, the time interval after ingestion of the isotope and the thyroid functional status of the patient influence the excretion and dissemination of radioiodine. Early on, the material is largely in inorganic form and extra-thyroid in location. It is cleared from the blood efficiently by the salivary glands and kidneys leading to abundant contamination of saliva and urine and some radioactivity in sweat. After 24 hours radioiodine is mainly present in the form of thyroxine, a small proportion of which is excreted in stool. In hypothyroid individuals hormone synthesis is reduced and a relatively larger proportion of the ^{131}I is excreted in the saliva and urine than in euthyroid or hyperthyroid patients. Furthermore, in the former patients the isotope persists in these biologic fluids for prolonged periods.

In general, iodine levels in air declined progressively after the day the radioiodine was administered (table 3) with the exception of patient C. This patient had copious salivation during periods of nausea. Since this patient had large metastases to the liver and subsequently developed granulocytopenia, the nausea and salivation were attributed to symptoms of mild radiation sickness. As a result of the excess salivation approximately 3mCi of ^{131}I was deposited in disposable paper tissues during the first four days of therapy. The air burden also differed from the general case since the concentration increased during the first two days before declining. This is shown in detail in figure 1. Figure 1 also shows the pattern observed for patient A2 who was also associated with a high air burden.

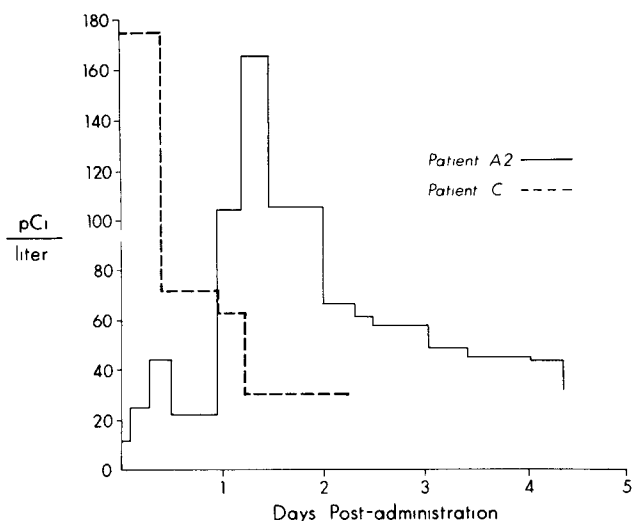


Figure 1. Concentration of ¹³¹I in Air After Administration of Radioiodine to Patients A2,C.

Total contamination on bedding, towels, dishes and utensils was greatest for patients C and E. The high value for patient C was due to exceptionally high contamination on dishes and utensils and this was consistent with the excess salivation problem. Patient E had an enlarged prostate and suffered from hesitancy in voiding and dribbling after completion of the urinary stream. Abnormally high levels of contamination on the bathroom floor and on towels and bedding were associated with these problems. One patient suffered a transient cerebral ischemic episode and required the use of a respirator, which was consequently contaminated as shown in table 4.

The measurements of air burdens have been used to estimate the potential burdens to other persons exposed to this source of radioactive material. Assuming that there is constant occupancy at the point of air sampling, that the concentration in air falls with a 1.5 day effective half life in the presence of the patient, the subject breathes 2×10^4 litres of air per day, 23 percent of inhaled iodine enters the thyroid and decays with a 7.6 day effective half life,⁸ and the initial concentration in air is in the range shown on day 1 in table 3, then the maximum thyroid burden is in the range 0.1 - 1 uCi and is reached in 4 days. This represents the situation which might be approached if two patients shared the same room or if relatives were exposed to an outpatient over long periods. The situation would presumably be worse if the room air were not changed by a central air conditioning system and were either static or recirculated by a single air conditioner. These potential burdens are reasonably consistent with burdens predicted for relatives of thyroid carcinoma patients by normalizing results obtained for hyperthyroid outpatients to doses used for carcinoma therapy.⁶ Medical and paramedical personnel are only exposed to air contamination for a relatively short time if the situation is well controlled and nursing duties are rotated. Such a person exposed to the levels observed on day 1 for a period of 10 minutes would potentially accumulate 0.5 - 8nCi. This is reasonably consistent with

thyroid burdens in the range of 0.1 - 1.8 nCi reported for medical and paramedical personnel from this institution following administration of 100 mCi doses of radioiodine to patients for thyroid carcinoma.³ Thyroid burdens in paramedical personnel of approximately 0.1 uCi reported for other institutions⁴ are consistent with our measurements of air burdens only if long periods of exposure are assumed, or if the rate of air change is less than in this institution.

Conclusions

The levels of contamination following radioiodine therapy for thyroid carcinoma are much larger than those normally tolerated in an open laboratory. Air burdens exceeded the MPCa values for controlled areas (9 pCi/litre, 40 hour week and 3 pCi/litre, 168 hour week⁸). However, thyroid measurements on medical and paramedical personnel and predictions based on our observations of air burdens indicate that such personnel will not accumulate excessive thyroid burdens if the patient management is well controlled. The problem of managing these patients is discussed in NCRP 37¹, where it is stated that patients may contaminate dishes, utensils and bedding, whereas we have found this to be invariably the case. The problem of air contamination is not discussed in the report. We consider that it is important to recognize that the contamination problem continues to be significant for 3 or 4 days following administration of radioiodine and that significant air burdens are present during this time. Radioiodine therapy is of proven value and importance but we consider that patients should generally be hospitalized for 3 or 4 days and should be managed with attention to the contamination and air burden problems. The considerations for radiation safety are closely related to the nature of the patient's medical problems, the pathways of iodine metabolism and the routes of iodine excretion.

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A COMPREHENSIVE APPROACH FOR THE EVALUATION OF COMPARATIVE
DOSIMETRY OF INTERNALLY ADMINISTERED RADIOPHARMACEUTICALS

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Abstract

Recent innovations both in instrumentation and radiopharmaceuticals are helping nuclear medicine to develop as a discipline in medical practice primarily for diagnosis with an emphasis on scanning. Radiation safety of a radiopharmaceutical is based on the assessment of the radiation exposure to the critical organ from a tracer dose. It is also important to consider relative tracer doses necessary for an optimal result for selecting one of the several radiopharmaceuticals available for a similar investigation. A comprehensive formulation of an index has been attempted for the relative dosimetry integrating mainly (i) conventional method of dose calculation, (ii) physical properties of radionuclide, (iii) available nuclear instrumentation, (iv) metabolic fate of radiopharmaceutical, and (v) gross considerations for radio-sensitivity of organs and dose rate. This approach has been illustrated considering the situation of brain scanning using Hg-197 chlormerodrin, Tc-99m pertechnetate, Tc-99m DTPA, short-lived In-113m DTPA and long-lived Yb-169 DTPA.

Introduction

Nuclear medicine is appearing as a discipline in medical practice which involves primarily the use of radiopharmaceuticals for diagnosis. Recently scintigraph has attained tremendous importance due to availability of advanced nuclear instruments and short-lived radiopharmaceuticals. Agents with high photon yield with low radiation dose are considered most desirable for clinical use.

Large number of radiopharmaceuticals are being developed in order to scan various organs and diseased conditions, and to improve the existing methods. Acceptance of any new product for clinical use depends on its cost, efficacy, specificity, toxicity and dosimetry. Although radiation exposure is an important consideration, it is difficult to control the medical applications by any formal method due to the fact that the primary interest lies in the immediate benefit to the patient.

The present exposition has been aimed to provide a guideline for the evaluation of relative dosimetry while choosing one of the several similar agents.

Comparative Dosimetry

Conventional method of dose calculation has been improved and standardized greatly¹. It is based on the utilization of physical properties of radionuclides, absorbed fraction of the radiation, and the metabolic fate of the radiopharmaceutical. It provides a practical value for the concept of safety with a recommended tracer dose for a clinical study. It could be expressed by

$$D \text{ (rads/mCi)} = 1000 \text{ c/m } 1.44 T_e \Sigma \Delta \phi = R.A \quad \dots\dots\dots(1)$$

where, R is the maximum dose rate (rads/mCi/hr) to an organ of mass m gm (=1000 $\Sigma \Delta \phi$ /m), and A is the area under time-concentration curve (approximately equal to 1.44 c T_e where c is the maximum fractional concentration and T_e is the effective half-life in hours).

Comparative dosimetry becomes important when several similar agents are available for a particular investigation. Usually the tracer dose is adjusted to keep the radiation dose within a safe limit. However, an appropriate approach should be to find out the relative amounts of tracer doses that would give similar measurable count rate at the time of study. Occasionally this method has been adopted². The relative tracer dose could be expressed by

$$T \text{ (mCi)} = 1/(f.e.d) \quad \dots\dots\dots(2)$$

where, f is the fractional yield of the photons used for detection, e is the detection efficiency of the instrument or at least the photo-peak interaction coefficient, and d is the activity remaining after decay or elimination by the mean time of the study. It is assumed that the target to non-target ratio for the different agents remain approximately the same.

The variations in radiosensitivity of critical and other organs have always raised questions in mind, but it is difficult to assign any quantitative value. However, it is well known that bone marrow is most radiosensitive. It would be reasonable to ascribe an empirical sensitivity factor (S₀) of 3, 2, and 1 respectively for bone marrow (and gonads during reproductive age), gastrointestinal, and the rest of the organs. Further, it is also desirable to incorporate a sensitivity factor for increased dose rate (S_r). Empirically, D/R (= 1.44 T_e) = 10⁻⁽ⁿ⁺¹⁾ may be used for a normalization of dose-rate sensitivity: assuming n=0 for normal condition (S_r = 1), one could arrive factors like S_r = 1/(n+1) or (n+1) for +n or -n values, the intermediate values could be obtained graphically. As an example, S_r = 2 if 10 rads are delivered at the rate of 10 rads/hr instead of 1 rad/hr.

$$\text{Then, } S = S_0 \cdot S_r \quad \dots\dots\dots(3)$$

In summary, the normalized radiation dose (NRD) for different radiopharmaceuticals used for similar investigations can be expressed by

$$\text{NRD} = R \cdot A \cdot T \cdot S \quad \dots\dots\dots(4)$$

It is assumed that the radiopharmaceuticals are pure, otherwise the contributions of any radionuclide impurity and any radiochemical impurity should be taken into consideration.

Example of Brain Scanning

Mercury-197 labeled chlormerodrin has been and is being used for brain scanning³, although technetium-99m pertechnetate has become the most preferred agent⁴. Chelates (DTPA) labeled with Tc-99m⁵, In-113m⁶ and Yb-169⁷ can be used for brain scanning. Kidneys, upper large intestine and bladder could be taken as the critical organs for chlormerodrin, pertechnetate and chelates, respectively.

The radiation doses for the different agents were calculated by taking values of nuclear parameters (except for ytterbium-169⁸) and absorbed fractions of energy for different organs from MIRD pamphlets¹. Biological factors (such as, concentrations in organs, biological half-lives, time of study after administration of tracer dose, and usual tracer doses) were assumed. However, these assumptions were based on various publications (such as the summary in a text book⁹). Estimation of relative tracer dose was based on the useful photon yield, photo-peak interaction coefficient of useful photon energy in 2-inch NaI crystal (thickness), and the fractional activity remaining after effective loss by the mean time of study. Relative tracer doses were then normalized to 10 mCi of Tc-99m pertechnetate. Further, it was assumed that a 100% of the dose was initially uniformly distributed in the total body and remained uniform although the fractional concentrations in different organs were different. The empirical sensitivity factor for dose rate was obtained using a semi-log plot of 10^{+n+1} (for 1.44 T_e values) against $+n$.

Table 1 and Table 2 represent the basic physical and biological data. Table 3 shows the aspects of dosimetry for the total body. Table 4 summarizes the results for the critical organs.

Table 1: Basic physical data for the radiopharmaceutical

Labeling nuclide	Chemical agent	Physical half-life (hr)	Useful photon energy (keV)	Useful photon yield (%)	Usual tracer dose (mCi)
Hg-197	Chlormerodrin	65.0	67-81	90.8	0.75
Tc-99m	Pertechnetate	6.0	140.5	88.3	10.0
Tc-99m	Chelate (DTPA)	6.0	140.5	88.3	10.0
In-113m	Chelate (DTPA)	1.67	393.0	65.4	15.0
Yb-169	Chelate (DTPA)	763.2	177 & 198.0	55.3	10.0

Table 2: Basic biological data for the radiopharmaceutical

Agent	Photo-peak interaction coefficient (2-in NaI)	Biological half-life (hr)	Time gap of study (hr)	Relative tracer dose (mCi)	$\Sigma\Delta\phi$ for total body
Hg-197 Chlor.	1.00	6	3.0	13.6	0.225
Tc-99m Pert.	0.98	48	1.0	10.0	0.130
Tc-99m DTPA	0.98	2	0.5	11.6	0.130
In-113m DTPA	0.77	2	0.5	24.7	0.474
Yb-169 DTPA	0.97	2	0.5	17.2	0.554

Table 3: Calculation of dosimetry for the total body

Agent	Dose rate rads/mCi/hr	Area under conc.-time curve (1.44 cT _e)	Radiation dose (rads/mCi)	Rads per usual tracer dose	Normalized rads/test
Hg-197 Chlor.	0.0032	7.91	0.0253	0.018	0.38
Tc-99m Pert.	0.0019	7.68	0.0146	0.146	0.16
Tc-99m DTPA	0.0019	2.16	0.0041	0.041	0.08
In-113m DTPA	0.0068	1.31	0.0089	0.089	0.41
Yb-169 DTPA	0.0079	2.87	0.0227	0.227	0.60

Table 4: Calculation of dosimetry for the critical organ

Agent	Concerned organ	Fractional conc. (c)	Biological half-life (hr)	Rads per usual tracer dose	Normalized rads/test
Hg-197 Chlor.	Kidneys	0.2	1704	8.2	75.9
Tc-99m Pert.	UL intest.	0.1	12	0.8	2.0
Tc-99m DTPA	Bladder	0.5	2	1.5	2.8
In-113m DTPA	Bladder	0.5	2	4.5	20.8
Yb-169 DTPA	Bladder	0.5	2	10.6	28.1

Discussion

Radiation dose rate (R) can be calculated with a high degree of accuracy except when the mass of the concerned organ is variable or uncertain (such as bladder with urine¹⁰). Considerable uncertainty could be inherent with 'A' due to difficulties in obtaining distribution patterns of the radiopharmaceuticals in human organs under normal and diseased conditions. Relative tracer doses could be calculated with sufficient accuracy if instrumental sensitivity for different agents (radionuclides) are determined experimentally. Quantitative assessment for radiosensitivity would remain a radiobiological problem particularly with reference to dose rate. However, it does not seem very unreasonable, at present, if a factor of 2 is used to increase the index of radiation dose when 10 rads are delivered at the rate of 10 rads/hr instead of 1 rad/hr.

Reduction of radiation doses in diagnostic uses of radioisotopes has remained an important consideration from the point of view of exposure to patient and population. In earlier days, usually low level counting techniques have been considered to reduce the tracer dose¹¹. In recent days, it appears that the diagnostic values are being enhanced by quantitation of scans with computers in studies with multi-millicuries of short-lived radiopharmaceuticals¹². In practice, one has to compromise to certain extent the radiation dose with the specificity, efficacy or the cost of the radiopharmaceutical. In the growing phase of the development of radiopharmaceuticals, it is hoped that the present consideration would help in the selection of the agent to reduce the radiation dose in an investigation under optimal condition.

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REDUCTION OF RADIATION EXPOSURE TO NUCLEAR MEDICINE PERSONNEL
BY THE USE OF NEW "INSTANT" TECHNIQUES FOR PREPARATION
OF TECHNETIUM RADIOPHARMACEUTICALS

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Abstract

Radiation exposure to the hands from the preparation of ^{99m}Tc -Sn-MAA from an instant kit and ^{99m}Tc -HAM from a non-instant kit has been measured by thermoluminescent dosimetry (TLD). The exposure to the fingers from the preparation of 100 mCi of ^{99m}Tc -HAM exceeded the exposure from preparation of the same amount of ^{99m}Tc -Sn-MAA. An exposure of 153 mR/100 mCi of prepared radiopharmaceutical to the left middle finger was the highest value recorded in the study.

Introduction

Use of short half-lived radiopharmaceuticals such as ^{99m}Tc and ^{113m}In compounds has greatly reduced patient exposure in nuclear medicine while requests for diagnostic procedures using these radionuclides have increased. Because of the short half-lives of ^{99m}Tc and ^{113m}In , these radiopharmaceuticals must be prepared on the day of use. Pre-tested vials of sterile, pyrogen-free reagents (kits) have streamlined the preparation of various radiopharmaceuticals but personnel contact with the radionuclide extends 10 to 90 min beyond the normal time for generator elution and transfer of patient dose to a syringe.

This study was designed to measure the radiation exposure to experienced nuclear medicine personnel preparing the lung scanning agents, technetium- 99m macroaggregated albumin (^{99m}Tc -Sn-MAA) using an instant kit and technetium- 99m human albumin microspheres (^{99m}Tc -HAM) using a conventional kit. An instant kit is one which requires a minimum number of manipulations and can be prepared quickly. Our kit for ^{99m}Tc -Sn-MAA meets these requirements. Technetium- 99m human albumin microspheres is prepared from a conventional kit requiring many manipulations and 25 min elapsed time. The methods of preparing ^{99m}Tc -Sn-MAA and ^{99m}Tc -HAM will be described to illustrate the various steps in which unnecessary exposure to personnel may occur.

Table 1 lists the various ^{99m}Tc radiopharmaceuticals prepared in nuclear medicine laboratories and the typical maximum quantities of ^{99m}Tc used. The exposure time is the period during which personnel handle the radionuclide directly or remotely on a daily basis.

If one person prepared all the radiopharmaceuticals without shielding, on a typical day he could be exposed to 200-300 mCi of

^{99m}Tc for 30-40 min. This, in fact, is not the case since an L-block shield¹ and lead generator eluate shields (cylindrical shields)² are used to minimize the exposure. Handling the vials restricts the exposure to the fingertips, wrists, and forearms.

Table 1 Quantity of ^{99m}Tc used and daily exposure times for typical radiopharmaceuticals

	Quantity (mCi)	Exposure Time (min)
^{99m}Tc -sulfur colloid	50	5
^{99m}Tc -human serum albumin	30	1
^{99m}Tc -macroaggregated albumin (^{99m}Tc -MAA)	120	10
^{99m}Tc -macroaggregated albumin (^{99m}Tc -Sn-MAA)	120	2
^{99m}Tc -DTPA	15	1
^{99m}Tc -diphosphonates	70	1
^{99m}Tc -human albumin microspheres (^{99m}Tc -HAM)	140	14

It can be seen from Table 1 that the exposure from ^{99m}Tc -MAA or ^{99m}Tc -HAM would exceed the exposure from other radiopharmaceuticals if only the quantity and the exposure time are considered. The technique for preparation of ^{99m}Tc -Sn-MAA, developed by this laboratory³ requires less than 5 min elapsed time, and involves an exposure time of only 2 min. It was thought that this shortened exposure time would be of advantage in terms of personnel effort and radiation exposure.

Methods and Materials

The modification of the method of Robbins et al.³ outlined in Table 2 was used for preparing ^{99m}Tc -Sn-MAA. Fifty to one hundred twenty mCi of ^{99m}Tc were handled in its preparation.

Table 2 Preparation of ^{99m}Tc -Sn-MAA

1. Add 1 ml tin stock solution to a vial containing albumin macroaggregates
2. Swirl
3. Add $^{99m}\text{TcO}_4$ (5 ml)
4. Swirl
5. Centrifuge and withdraw supernatant solution
6. Reconstitute with 6 ml saline
7. Assay in dose calibrator

Technetium- 99m human albumin microspheres were prepared according to the method described in the 3M Company package insert using the

3M brand ^{99m}Tc -HAM kit.⁴ An outline of this procedure is given in Table 3. Fifty to one hundred forty mCi of ^{99m}Tc were handled in this procedure.

Table 3 Preparation of ^{99m}Tc -HAM

To the labeling vial, containing microspheres and a thiosulfate tablet:

1. Add $^{99m}\text{TcO}_4^-$ -(10 ml)
 2. Sonnicate 2 min
 3. Agitate in boiling water 6 min
 4. Cool in water bath 1 min
 5. Withdraw all liquid from labeling vial
 6. Add 10 ml saline suspending solution
 7. Sonnicate 1 min
 8. Withdraw liquid from labeling vial
 9. Add 10 ml saline suspending solution
 10. Assay in dose calibrator
-

The 3M brand albumin microsphere labeling system, composed of the labeling and suspending unit, and rinsing unit and a shield with lead-glass window for the tagging vial were also employed throughout the ^{99m}Tc -HAM preparation.

Exposure data were obtained from duplicate sets of TLD's placed on the distal medial aspect of the thumb, the distal lateral aspect of the middle finger, the distal medial aspect of the ring finger, and the anterior wrist surface of each hand. The TLD's were worn during five or more preparations of each radio-pharmaceutical so that the exposure was accumulated while handling more than 500 mCi of ^{99m}Tc . An average exposure was calculated from the two measurements at each point. The exposure is expressed as mR per 100 mCi prepared. LiF chips consistent to within $\pm 5\%$ were used in these studies (TLD 100's, $1/8 \times 1/8 \times 0.035$ in). Six TLD's, arranged two each at 3, 5 and 8 cm from the central point source, were exposed to 17 mCi ^{99m}Tc for 20 min for use as standards. The TLD's were readout in a TLR-5 Eberline Reader using standard techniques.

Results

Table 4 contains the results of measuring hand exposure during preparation of ^{99m}Tc -Sn-MAA and ^{99m}Tc -HAM. While handling a total of 523 mCi of ^{99m}Tc for nine batches of ^{99m}Tc -Sn-MAA, the total quantity prepared was 497 mCi with an average tagging yield of 95%. For ^{99m}Tc -HAM, a total of 518 mCi of ^{99m}Tc handled in six runs provided 362 mCi with an average tagging yield of 70%. The results in Table 4 show that ^{99m}Tc -HAM contributes more radiation exposure to the fingers than does the ^{99m}Tc -Sn-MAA. The unusually high exposures to various fingers are indicative of the preparation techniques. In both procedures transferral of the $^{99m}\text{TcO}_4^-$ from its storage vial to the syringe for addition to the reaction vial is considered the cause for high exposures. Due to

the technique in performing this step, the exposure to the left ring finger (for $^{99m}\text{Tc-Sn-MAA}$) was higher than the other fingers. This technique is also considered one of the causes of high exposure to the left thumb (for $^{99m}\text{Tc-HAM}$). Use of a screw-cap cylindrical shield, which exposes only the rubber septum of the pharmaceutical vial, is thus recommended for each of these procedures.

Table 4 Average exposure to the hands during preparation of $^{99m}\text{Tc-Sn-MAA}$ and $^{99m}\text{Tc-HAM}$

	mR/100 mCi prepared	
	$^{99m}\text{Tc-Sn-MAA}$	$^{99m}\text{Tc-HAM}$
<u>Right Hand</u>		
Wrist	9	7
Thumb	47	86
Middle Finger	55	109
Ring Finger	45	46
<u>Left Hand</u>		
Wrist	10	19
Thumb	53	151
Middle Finger	60	153
Ring Finger	94	76

In preparing $^{99m}\text{Tc-Sn-MAA}$, the left ring finger received additional exposure when the reaction vial was held in the left hand to withdraw the supernatant solution (Step 5, Table 2). Holding the vial with tongs in the left hand is recommended for this step of the procedure. The left thumb and middle finger received added exposure from the microspheres in Step 4, Table 3 wherein the tagging vial was wiped with a paper towel held in the left hand. This step is necessary to prevent rusting of the interior of the special shield for the tagging vial. Use of remote wiping technique is also recommended in this step. Monitoring of the exposure to the face, chest and body trunk from both of these procedures was less than detectable by the TLD techniques employed (<10 mR total cumulated exposure).

Conclusions

The exposure from instant kits is less than with other methods because the simplicity of the instant procedures permits shorter exposure time and less contact with the pharmaceutical vial. The use of instant kits is to be encouraged.

Furthermore, while the radiation exposure to the fingers from preparation of $^{99m}\text{Tc-HAM}$ is greater than that received from $^{99m}\text{Tc-Sn-MAA}$, the exposure level from both procedures could be reduced by adherence to procedural recommendations in the discussion. Additionally, all procedures should periodically be evaluated in terms of exposure, especially to the hands. The techniques monitored in this study are just two from the various ^{99m}Tc radiopharmaceuticals prepared daily by most nuclear medicine laboratories. Based on the highest exposure recorded in this study (153 mR/100 mCi to the left middle finger), a person

preparing 200 batches of 80 mCi each of ^{99m}Tc -HAM during the year would receive about 24 R/year to the hand. While this is within the recommended limits of NCRP (75 rems/year to the hands)⁵, it represents the exposure from ^{99m}Tc -HAM preparation only. The use of instant kits, such as ^{99m}Tc -Sn-MAA and ^{99m}Tc -DTPA⁶ along with an adequate knowledge of the procedural steps will contribute to a lower total exposure from all phases of nuclear medicine.

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REDUCING PERSONNEL EXPOSURE IN NUCLEAR MEDICINE

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ABSTRACT

Nuclear medicine procedures are growing at a rate greater than 15%/yr and the radiopharmaceutical industry over 25%/yr. A nuclear medicine facility in a medium-large hospital routinely handles 300-700 mCi of ^{99m}Tc /day plus prepares ^{99m}Tc radiopharmaceuticals and handles millicurie amounts of other isotopes. As this specialty grows, so must exposure reduction techniques. Periodic assessment of personnel handling the molybdenum-technetium generators and ^{129}Cs were made using TLD's. These data are reviewed with comments about potentials for personnel exposures and exposure reducing methods. Additional shielding and new handling techniques have been incorporated and are discussed.

INTRODUCTION AND BACKGROUND

The Nuclear Medicine Laboratory is located and operated in collaboration with the Radioisotope Laboratory. In these laboratory facilities, the following contribute primarily to personnel radiation exposure:

1. daily handling of 500 mCi to 1.4 curies of ^{99m}Tc ,
2. weekly processing of mCi amounts of ^{129}Cs ,
3. preparing throughout the week a variety of ^{99m}Tc labeled radiopharmaceuticals from kits, and
4. weekly handling of mCi amounts of ^{131}I , ^{133}Xe and ^{113m}In .

Each of these can increase the daily exposure to laboratory personnel and result in exposure levels that exceed what we would have predicted just a few years ago.

Max Lombardi¹ reported on a 12 month survey of 69 hospitals that nuclear medicine procedures grew at a rate of 16% per year and that the radiopharmaceutical industry grew 25-26% in a similar 12 month period. Accompanying this growth in radiopharmaceutical usage and new nuclear medicine laboratories in many hospitals, the problem of personnel protection has been recognized by some but unrecognized by many.

The National Council on Radiation Protection and Measurements (NCRP)² recommends 75 rems/year, 25 rems/qtr., as the permissible dose equivalent to the hands. They have further characterized this as an "interim concession" in "comment" to this section and have also indicated that "all reasonable efforts should be made to keep exposure of the hands and forearms within the general limit for skin, 15 rems/year".

A study 4 years ago by Neil³ on the radiation exposure to the hands from handling ^{99m}Tc showed a maximum dose equivalent of 10 rems/curie/minute for the index finger and thumb with lesser dose equivalents for other parts of the hands. Using Neil's data, a physician that gives 400 injections/year of 10 mCi each and in only 30 seconds handling time would receive 30 rems for that year for that portion of his hand alone, if handling of any other radioisotopes is neglected. (^{131}I , ^{133}Xe , ^{18}F , ^{129}Cs , etc.)

The need for additional personnel protection for all radioisotopes in the laboratory resulted in a complete rearrangement of our "hot" lab area. Lead-lined housings with sliding lead glass doors were specially built* for two molybdenum-technetium generators, figure 1. The two end doors must be moved to the middle where there is a third fixed plate of lead glass. These form a body shield while the hands may be inserted through the openings on either side of the lead glass. We have also built ^{99m}Tc eluate organizer racks, figure 2, that provide $\frac{1}{4}$ " lead shields for each eluate bottle. Each shield has a lid with a hole for the head of the eluate bottle. The bottles are placed on a slant pointed away from the operator. Each tier of eluate shields is color coded to identify the eluate by the parent generator. Added positions are available for ^{99m}Tc radiopharmaceuticals, prepared from kits. An additional tier permits storage of other radiopharmaceuticals in use that day. All the ^{99m}Tc and most radioiodine doses are prepared behind the face and body shield⁴ seen just in front of the organizer rack.



Figure 1. Mo-Tc house built of plywood, Pb lined epoxy painted, fluorescent light and 3 Pb glass doors. Base made as a tray to contain spills.



Figure 2. Eluate organizer rack for ^{99m}Tc placed immediately behind a face and body shield. Most radiopharmaceutical doses are prepared at this work station.

Most of the diagnostic administrations of radioisotopes in nuclear medicine are given at some location other than the hot laboratory. For injections, a transporting tray, with a lead syringe holder anchored to the side of the tray is used and is shown in figure 3. In addition, the tray is equipped with other items the physician will need for injection. This special tray has been helpful in reducing exposure and contamination and has proven very easy and convenient to laboratory personnel.

We conservatively estimate that the generator housing reduces major body exposure 2 times during the elution procedure. The eluate shields probably reduce personnel exposure by a factor of 5, and the face and body shield reduces the technicians total body exposure an estimated factor of 5. The personnel exposure reduction realized from the dose tray is difficult to calculate, but it is our conservative guess that the hand exposure is reduced a factor of 5 to 15 depending on the distance the dose syringe must be carried, etc.

* These were designed for one specific company's generator and might require modification for a different brand generator.

In order to assess the personnel exposures more specifically, we have studied some hand exposures during routine handling procedures using thermoluminescent dosimeters (TLD's). For ^{99m}Tc and ^{129}Cs the procedure used was to place TLD* dosimeters in the anterior and posterior positions on one or two fingers, the thumb, and wrist of each hand. Additionally, TLD's were placed on the forehead or glasses frame, chest and gonadal area of the body. Unless otherwise stated the TLD's on the fingers were in the finger ring position. We assumed that all radiation absorbed by the lithium fluoride chip was from either ^{99m}Tc or ^{129}Cs . This actually is not the case; for instance, the aluminum holder for the target material in the cesium production was emitting high energy gamma rays from ^{22}Na and ^{24}Na and likewise for ^{99m}Tc the 740 and 780 keV ^{99}Mo radiation is emitted through the generator shielding. For each study two lithium fluoride chips were placed at each dosimetry point; each was measured separately and the count data averaged.

At the time of this study, two molybdenum technetium generators were received each week; each rated at 400 mCi 5 days after receipt. One technician had the responsibility for removing the old generator, installing the new one, and obtaining the daily elutions required (about 5 curies/week). For this study, the TLD's were worn only during these procedures. The resultant TLD data, Table I, reflects the expected higher exposure on the anterior surface of each hand and higher exposure to the right hand. Data from the forehead, chest and gonadal area of the body were all under 10 mR/week. The highest finger exposures averaged 131 mR/week which over 50 weeks would amount to 6.5 R maximum cumulative exposure. In some other laboratory with only one 400 mCi generator and using only the shielding provided with the generator, the annual exposure could be as much as 20 R for the same person performing this task.

Table I. TLD PERSONNEL DATA FOR ^{99m}Tc IN mR
- Handling Generators Only -

		<u>Weekly Average</u>	
Fingers - Anterior Surface		61	109
		36	94
Thumb - Anterior Surface		46	198
		54	124
Wrist - Anterior Surface		19	33
		10	19
Totals - Fingers		49	131
- Hands		37	109

50 Week Total - Fingers (131 x 50) = 6550 mR

A more recent study has compared the preparation of ^{99m}Tc macroaggregated albumin prepared according to the method of Robbins⁵ and ^{99m}Tc human albumin microspheres labeled according to the method described by the manufacturer (3M Company) using their equipment. For this latter study TLD's were placed as shown in figure 4. The TLD's were on the lateral and medial borders of the finger tips so as not to interfere with operator's finger tip sensitivities. The dosimeters were worn during five or more preparations of each radiopharmaceutical, each study involving the handling of more than 500 mCi of ^{99m}Tc . The exposure expressed is in mR/100 mCi of the prepared product. An analysis of this exposure data is given in Mr. Robbins' paper⁶. The higher exposure levels for ^{99m}Tc -HAM are due to the higher levels of ^{99m}Tc handled for the resulting

*The chips used in these studies, TLD-100 (1/8 x 1/8 x .0035 inch) were selected to have sensitivities within ± 5 percent of each other. In some ^{99m}Tc studies, two sets of TLD's were exposed, one at twice the activity level of the other. These became the calibration standards for the two isotopes.

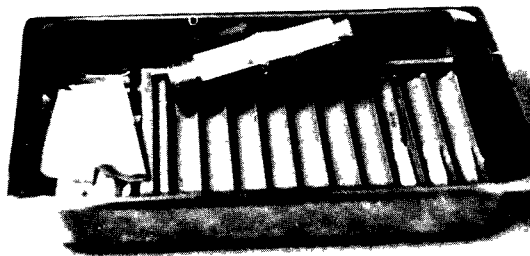


Figure 3. Transporting tray with lead holder for dose syringe plus items needed at injection site.

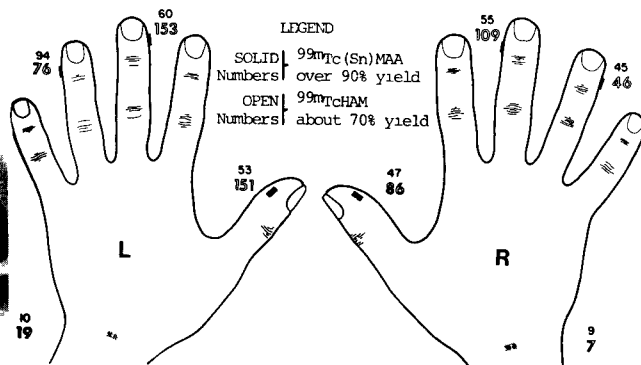


Figure 4. ^{99m}Tc Exposure in mR/100mCi ^{99m}Tc Prepared. Wrist TLDs anterior side only.

yield, and difference in handling procedures required. Unusual exposure data can be accounted for in specific handling operations. Yet, based on the left hand finger exposures recorded in this study, the preparation of 200 batches of 80 mCi each of ^{99m}Tc during a year would result in approximately 17 R/year to the hand. While this is within the 75 rem/year NCRP limit, (it exceeds the 15 rem/year skin exposure recommendations) it only accounts for exposure from the preparation of one radiopharmaceutical.

A new radioisotope for tumor and heart scanning, ^{129}Cs is produced by cyclotron* irradiation and is air shipped to Cincinnati where the target material is chemically processed to extract the cesium as cesium chloride. The chemical extraction takes about 60 minutes. In the first few production runs, the ^{129}Cs yield from each bombardment was 2 to 4 millicuries total. As the procedure was refined and the cesium yield increased, more shielding was incorporated throughout the extraction-purification process. Handling devices were likewise incorporated into the process. Figure 5 shows a pair of vice-grip pliers used to hold the aluminum target. A long bolt has been substituted for the adjustment screw on the pliers to add distance between the target and hands, yet maintain reasonable operation.

Figure 6 shows the special tools assembled for this procedure; tweezers permanently attached to tongs for handling the target cover, a long handle with a funnel stopcock in the slotted right end of the handle, a flexible pick-up tool and an allen wrench built into a long handle for removing the target cover. At the time of the cover removal the target was emitting more than 500 R/hour at 1 centimeter. One other device, figure 7, is a remote hydraulically operated syringe. By coupling two syringes tip to tip with small plastic tubing, one acts as a piston controlled by the other. The piston can push or pull the primary syringe plunger to deliver or take up liquid. The beauty of this system is that the personnel radiation exposure is essentially eliminated for this part of the procedure. Refinements on the procedure occurred over a period of 3 months and with each succeeding week we noted decreases in radiation exposure. Table II summarizes this exposure for the hands. Immediately after run #1 techniques to reduce the dose were incorporated. As will be noted, the effect was dramatic on run #2, one week later. With succeeding weeks, and with the employment of new shielding and tools, the average exposure dropped. For brevity of data presentation runs #3 and #6 are eliminated but fit as expected in the step-by-step exposure reduction - a 6-8 times reduction. If the exposures recorded in run #1 were received over a 50 week period, it would result in more than 8 rem exclusive of exposures in other duties with other radioisotopes. However, employing the techniques and shielding described, the ^{129}Cs exposure is about 1 R/year.

* In cooperation with the Naval Research Laboratory cyclotron, Washington, D.C.

Table II. TLD HAND DATA FOR ^{129}Cs EXTRACTION

		Run	1	2	4	5	7
FINGERS AND WRIST	Right						
	Average(mR)		149	46	35	28	20
	Reduction		← x3	✗	→ x2	→	→
	Left						
Average(mR)		147	35	22	21	16	
Reduction		← x4	✗	→ x2	→	→	
FINGERS ONLY	Right						
	Average(mR)		167	48.3	31.8	31.2	18.8
	For 50 Weeks(R)		8.4	2.4			1.0
	Left						
	Average(mR)		162	40	27	26	17
	For 50 Weeks(R)		8.1	2.0			0.9
Week Total(R)		8.2	2.2			1.0	

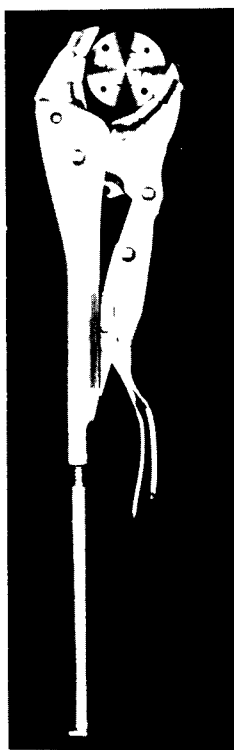


Figure 5. Modified vise grip pliers for holding irradiated cyclotron target secure and at a safe working distance.

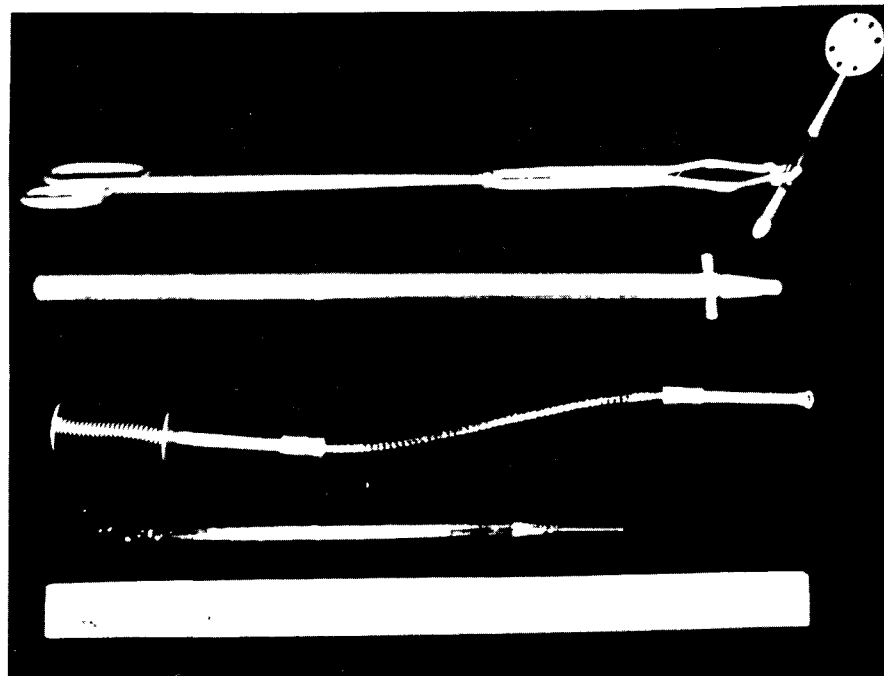


Figure 6. Special tools for ^{129}Cs extraction. Top to bottom: tongs and tweezers, long handle with funnel stop-cock (right end), pick-up tool, and allen wrench in a long handle.

In summary, we have indicated some of the ways we have assessed and reduced personnel exposure during routine radioisotope handling in our laboratory. The results are within or close to the recommended guideline of 15 rems/year for the specific function analyzed. From our experience, personnel exposures in nuclear medicine laboratories can and must be reduced by modifying current techniques and practices. In a growing clinical field radiation

exposure assessments should be made every few months to determine where improvements can be made for the protection and exposure reduction of the laboratory personnel.

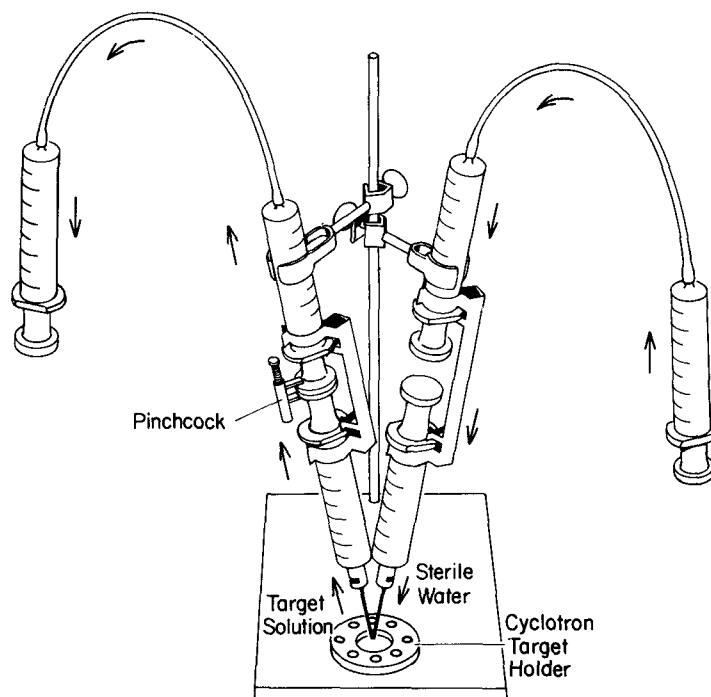


Figure 7. Remote control syringes used for target dissolution. The system on the right is used to wash the target, the one on the left is used to pick-up the solution.

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RADTOPROTECTION IN A NUCLEAR MEDICINE UNIT

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Abstract

The main aspects of the radioprotection program of a Nuclear Medicine unit organically included in the services of an Occupational Diseases Institute are described and discussed. Analysis of the records of the environmental and personal dosimetry points out to the opportunity for monitoring particularly of the internal contamination. The need for employing usual diagnostic equipment for regular monitoring of the working staff is stressed. Due to the increasing diffusion of WBC for diagnostic applications, scheduling of this tool for monitoring of the personnel is recommended.

A transportable WBC of the shadow type susceptible to be transferred from the laboratory to a mobile unit in less than half an hour is described for clinical and radioprotection applications including monitoring of operators handling gamma emitters in medical units and in industry when no facilities for internal dosimetry are available.

Monitoring of the excreta of patients submitted to radiometabolic therapy has been one of the aims of the health physics programs in this Nuclear Medicine unit. An original system for monitoring liquid radioactive waste is described as a first step for ecologic control of spreading of radioactive contamination.

Introduction

Problems related to safe handling of radioactive isotopes in medical practice are well known and have been widely discussed in a number of reports ¹⁻⁵. When applied to single medical institutions general regulations have to be tailored for the particular situation, the problem being often to meet requirements for high standards of safety as necessary in nuclear technology with the not unlimited resources of small units.

Starting a Nuclear Section in an Occupational Diseases Institute we were faced with a number of problems related to diagnostic, therapeutic and research applications of isotopes as well as to radioprotection of workers handling radionuclides.

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In this paper some of the main features of the radioprotection program of a Nuclear Medicine unit will be reported and discussed based on the observations made during first 3 years of practice. Present stage of development of the unit can be evaluated as comparable to a medium sized diagnostic laboratory , therapeutic applications being still limited to some experience with radioiodine treatment.

I.-Remarks after 3 years experience with a radioprotection program in a Nuclear Medicine unit .

General points of the radioprotection program are:

I) a stressing accent on autoradioprotection as far as collection, registration and first evaluation of data are concerned : weekly check for white coats, fortnightly surveys for detection of laboratory contamination , monthly registration of film dosimetry data for environmental and personal monitoring, quarterly whole body examinations. The bulk of this job is committed to the physician of the operating staff (1 physician and 2 technicians).

II) a decision to avoid heavy shielding structures in the laboratory. Location of the unit in a partly underground floor made preferable to keep windows which opened over a street of the town.

III) a rigorous organization of the unit with a clear definition of the controlled zone and of compulsory pathway with a monitoring unit at the end. Part of the controlled zone are rooms for metabolic therapy and for waste disposal.

Planimetric organization of the laboratory, consumption of radio-nuclides, dosimetry of the surveilled zone and summary of data of the external irradiation and external and internal contamination of the working personnel are reported in fig.I and tables I,II,III. The following concluding considerations are drawn:

I) environmental monitoring is worthwhile in a Nuclear Medicine unit if special shielding of walls and ceilings are not available because safe limits can be overcome if the amount of handled isotopes rises enough.

II) personal monitoring is important especially as far as internal contamination is concerned. Notwithstanding observation of safety rules , internal contamination can occur: ^{75}Se and ^{131}I were mostly found. Uptakes and scanning instruments available in each nuclear medicine laboratory are of course useful for detecting contaminations of the order of a fraction of 1 microcurie of the used gamma emitters: periodical checks with a WRC as frequently as possible for satisfactory internal dosimetry however is suggested.

II.- Presentation of a transportable WRC of the shadow type.

In the country efficient WRC are available but a need is felt for monitoring of workers in organizations where WRC facilities are not provided as in some Nuclear Medicine units and in industry . Many examples of transportable WRC have been described⁶⁻⁸. A main feature of the present model is that it can be used both in the laboratory set up as in a mobile unit and that in 30 minutes its

t 1.5, damped, can be transferred, by an elevator, from the laboratory to the car, ready for emergency. At least 1% of most restrictive WPCB for gamma emitters with an accuracy of $\pm 20\%$ can be appreciated in 20 minutes. This WPC is composed by 2 blocks of comparable weight: a 5 in. x 4 in. NaI(Tl) crystal shadowed by a 10 cm thick Pb shield elliptically collimated and a bed plane shielded by 5 cm Pb. Remote devices can move electrically the detector vertically from 66 cm to 114 cm from ground level and can adjust the segments of a chair geometry. The apparatus is equipped with a 800 channels analyzer with computation facilities, analogue display, digital data printing and magnetic tape recording. In the laboratory as well as in the mobile unit are provided a room for external decontamination, air filtering and conditioning; a whole body scanning bed is in progress.

III.- Control of radioactive waste disposal

For gaseous waste a ventilation circuit is operating in the laboratory from the over-pressure WPC room to the filter of the laboratory cupboard (fig. 1). At present the only gaseous waste is ^{133}Xe expired in perfusion studies in a closed circuit and partly adsorbed at room temperature on a shielded activated charcoal, which is then deposited in a plastic closed bag to decay in the waste room, whereas the not absorbed fraction is ventilated through a short stack on the roof of a 3 floor building. Absorption procedure is not practical but it is suggested by the very closed proximity of other houses.

Solid waste, collected in plastic bags, are stored for some months until activity is less than $1 \mu\text{Ci}/10 \text{ kg}$: ^{75}Se contaminated material is kept distinct and stored for longer time.

For liquid radioactive waste we assumed that effluents from the laboratory can be evaluated, on the consideration of MPC_W . around 10% of the effluents from a proportionate radiometabolic therapy unit using ^{131}I . So, if a treatment of radioactive waste is decided, the main point is to remove the activity released from treated patients. It has been calculated that, if 75 litres a day are released from the WC of the patient, 1 clarifier and 3 tanks in series with an individual dilution volume of 1500 litres, can provide a 37% reduction each of the concentration of the radioactivity so that only a fraction of 2% can reach the well. Two electrovalves allow discharge into the well from the laboratory and from the patient line at a concentration lower than $10^{-3} \mu\text{Ci}/\text{ml}$. Two G.M. detectors shielded by 9 cm Pb are operating to remove the radioactivity released from the laboratory and to measure the total discharged activity which is given by the product of the known volume voided by the pump (1240 litres) by its measured radioactive concentration. Due to further dilutions (700 m^3/day in the sewerage collector and 4200 m^3/min in river Ticino) present release is less than $1/10000$ of the ^{131}I MPC_W for the population. The counterpart is the care which is requested for looking after the system (as for repair of valves, pumps, cleaning of detectors).

Table I

Isotopes (mCi) handled in first 3 years of laboratory activity

Year	¹³¹ I	^{99m} Tc	¹⁹⁸ Au	¹⁹⁷ Hg	⁷⁵ Se	^{87m} Sr	⁶⁷ Ga	¹⁶⁹ Yb	¹³³ Xe	¹²⁵ I	³ H
1	43	80	16	5	11	-	-	-	-	0.04	0.02
2	64	1340	39	6	21	68	-	-	-	0.06	0.02
3	101	4546	67	7	55	80	40	3	475	0.22	0.03

Table II

Summary of dosimetry records of operators (subjects A,B,C)

Year	μCi/yr internal contamination			μCi/yr white coat contamination			mrem/month external irradiation (film)			
	A	B	C	A	B	C	A	B	C	
1	0.07	-	-	0.2	0.3	0.08	20	20	20	
	(⁷⁵ Se)									
2	-	0.01	-	0.4	0.5	0.09	20	20	20	
	(¹³¹ I)									
3	0.33	-	0.01	3.3	0.5	0.9	20	20	20	
	(¹³¹ I) (¹³¹ I)									

A= physician : examines the patient - administers diagnostic and therapeutic doses ; B= technician : cleans glassware, stocks radioactive waste ; C= technician ; performs radiochemical work. Every one does in vivo investigations (scanning, fast dynamic studies).

Table III

External dosimetry of surveilled zone in the laboratory (mrem/month)

Year	door of the room for waste disposal	<u>room of sources deposit</u>			all other spots
		wall	window	upstairs	
1	15	37	6	0	20
2	29	50	12	20	20
3	44	89	49	20	20

The following spots have been monitored: uptake room, scanning room, in vivo dynamic studies room, radiochemical laboratory, animals rooms.

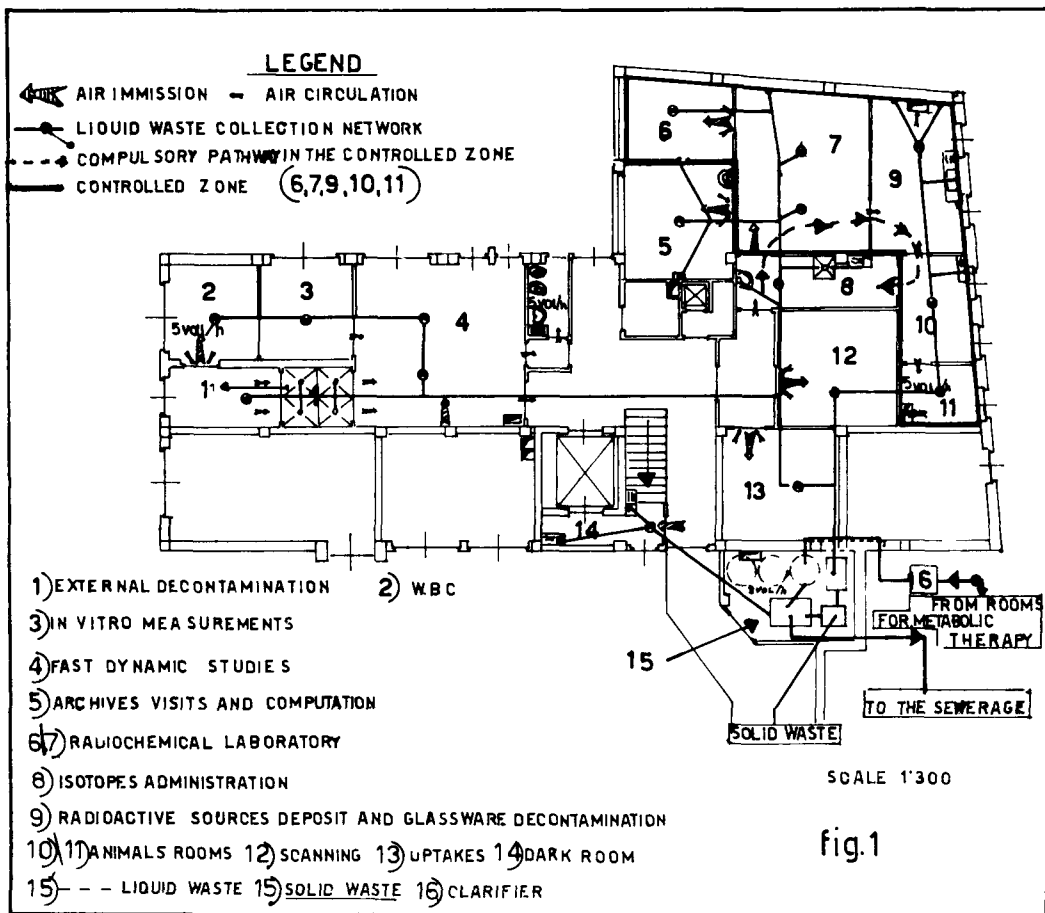
Table IV

Percentage of frequency of detection of contamination in absorbing coverings higher than 1×10^{-4} μCi/cm² (fortnightly surveys).

Year	percentage
1	21.2
2	14.1
3	19.4

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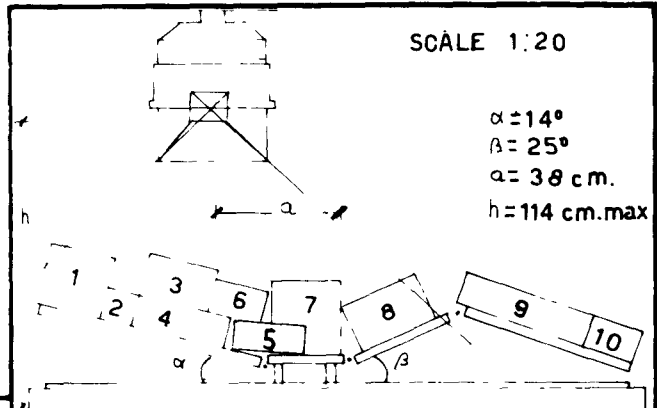
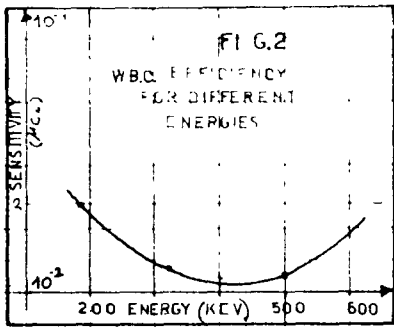
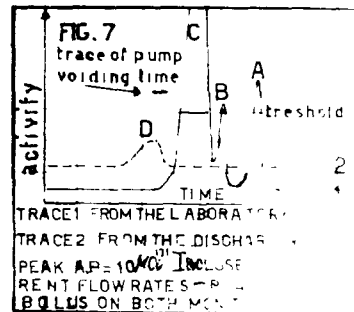
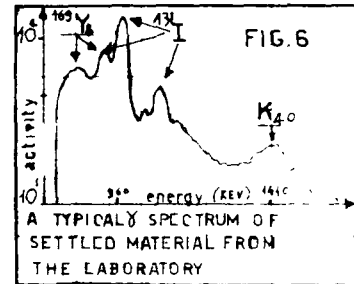
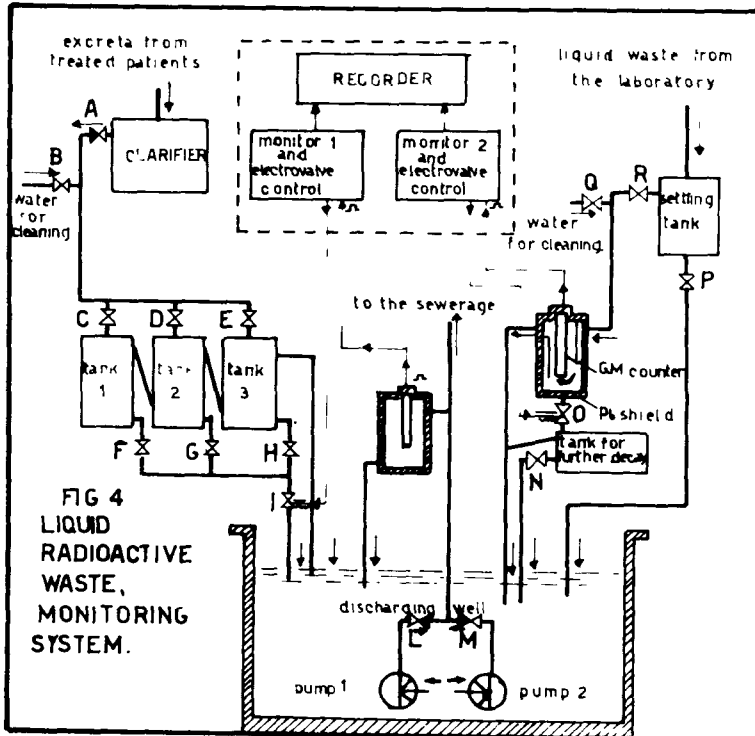
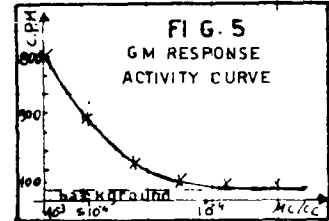


FIG. 3
WBC SHADOW IN TYPE A GEOMETRY— EFFICIENCY FOR DIFFERENT BODY SEGMENTS AND ENERGIES

	BODY SEGMENTS	I-131 364-kev	I-131 637-kev	FE-59 1095-kev	FE-59 1292-kev
1	HEAD	0.027 %	0.027 %	0.018 %	0.017 %
2	NECK	0.136 %	0.147 %	0.045 %	0.026 %
3	UPPER TRUNK	0.051 %	0.059 %	0.029 %	0.021 %
4	ARM	0.050 %	0.047 %	0.027 %	0.024 %
5	FORE ARM	0.023 %	0.058 %	0.007 %	0.007 %
6	MIDDLE TRUNK	0.056 %	0.060 %	0.032 %	0.032 %
7	LOWER TRUNK	0.032 %	0.036 %	0.021 %	0.014 %
8	LIMB	0.019 %	0.019 %	0.018 %	0.014 %
9	LEG	0.001 %	0.001 %	0.001 %	0.001 %



RADIATION PROTECTION IN A NUCLEAR PHARMACY

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Abstract

Regional nuclear pharmacies are emerging throughout the world to meet the increased demands for radiopharmaceuticals. A radiation protection program for a nuclear pharmacy encompasses facility design, quality control, dispensing and documentation, and provides for increased utilization of radiopharmaceuticals. The resultant radiation protection program is synergistic between health physics and pharmacy principles.

Introduction

Regional nuclear pharmacies provide radiopharmaceutical services for multiple hospitals located over wide geographic areas. Such pharmacies provide the necessary pharmaceutical expertise for preparing radioactive chemicals into pharmaceuticals and also provide an increased assemblage of radiopharmaceuticals at reduced costs to many hospitals that cannot individually afford nuclear pharmacy services.

A regional nuclear pharmacy may have inventories of 5-10 Curies of radioactive materials, with greater than Curie quantities of Iodine-131, Xenon-133 and Molybdenum-99/Technetium-99m.

The University of Michigan Hospital Regional Nuclear Pharmacy, over the past two years, has evaluated radiation protection as applied to nuclear pharmacy practice. Facility Design, administered Doses, product Dispensing and quality control Documentation are considered to be the principle axioms for the development of a radiation protection program in nuclear pharmacy.

Nuclear Pharmacy Design

Nuclear pharmacies must always incorporate health physics principles associated with "wet" radiochemistry laboratories. In addition, special consideration must be given to pharmaceutical techniques, i.e. aseptic preparation of parenteral products, synthesis of radiolabeled organic compounds, dispensing of radioactive gases, repeated handling of syringes containing radioactive materials, maintenance of product quality and potential contamination of non-radioactive pharmaceuticals.

Traffic flow patterns within a nuclear pharmacy must be defined for efficient utilization, as well as radiation protection planning. A general consideration of functional separation of activities within the pharmacy aids in radiation protection and pharmaceutical quality. The package receiving and shipping area should be a separate room to minimize potential radioactive contamination and to reduce airborne dust and particulate matter from entering the compounding area. A dispensing area, separate from the compounding area, will minimize traffic where bulk quantities of radioactive material are used and parenteral products formulated. In addition, a dispensing window will reduce the access of unauthorized personnel. The quality control laboratory should be housed in a separate room, as it is predominately an instrumentation facility, and considered only to contain tracer quantities of radioactive materials. Finally, because adjunctive (non-radio-

active) pharmaceuticals are often stocked and dispensed with radiopharmaceuticals, a separate room for pharmaceuticals again provides radiological and pharmaceutical quality assurance.

An adequate floor plan for efficient nuclear pharmacy design is shown in Figure 1. The compounding and dispensing rooms form an integral, limited access area for the storage, preparation and dispensing of radiopharmaceuticals. Low traffic flow in the compounding area reduces potential spread of contamination. Dispensing of unit-dose radiopharmaceuticals to medical personnel is conducted through the dispensing window; a procedure which guarantees limited access only to authorized individuals and further reduces the potential spread of radioactive contamination. The non-radioactive pharmaceutical dispensing area and quality control laboratory are located across the hall from the compounding and dispensing rooms. Adjacent to the nuclear pharmacy is a separate radiochemical laboratory where packages are received and shipped. Included in the radiochemical laboratory is an absolute filtered radiochemical hood for the storage and dispensing of iodine-131 and xenon-133.

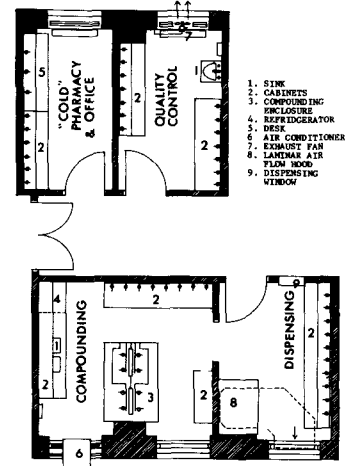


Figure 1
Floor Plan

Too often, one segment of a laboratory bench is chosen for all radiopharmaceutical preparation. Such a design can offer satisfactory health physics considerations, but poses a serious potential risk of product cross contamination and erroneous product selection and dispensing. Nuclear pharmacy design should provide separate work areas for the compounding and dispensing of radiopharmaceuticals. To facilitate both health physics and pharmacy requirements, the University of Michigan Nuclear Pharmacy has designed a lead laminated plywood ($\frac{1}{4}$ inch lead) compounding enclosure shown in Figure 2.

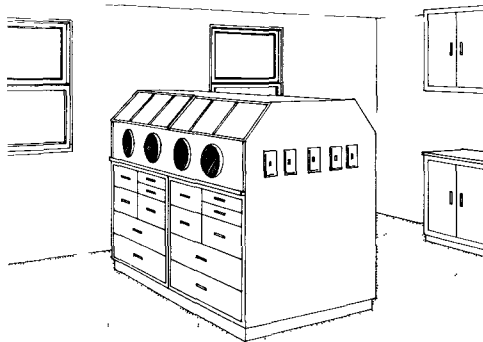


Figure 2
Compounding Enclosure

This installation is specific for technetium-99m compounding and contains sliding lead glass (1.7 mm lead equivalent) doors across the top. The primary design consideration of this enclosure was consolidated shielding with separation of compounding involved with technetium radiopharmaceuticals: a union of radiation protection with pharmaceutical quality assurance.

Within a nuclear pharmacy, laminar air flow hoods are used to provide a sterile work area for aseptic subdivision of parenteral products and not to vent volatile radioactivity. Laminar flow hoods are manufactured to provide either horizontal or vertical air flow. Since horizontal air flow is directed towards the operator and could lead to a severe personnel contamination hazard, it must be stressed that only vertical laminar air flow hoods should be chosen for use in nuclear pharmacy. Sterile air is provided in a laminar air flow hood by a series of HEPA filters which are designed to remove all

dust and bacteria from the air before entering the working space. To insure the sterility of the working compartment, articles should be autoclaved or suitably disinfected before entering the hood.

Several problems associated with shielding are evident with a nuclear pharmacy. Too often personnel without health physics training fail to consider the energy of associated radiation and assume that shielding designed for technetium-99m is suitable for fluorine-18, iodine-131 or phosphorous-32. The designation of separate work areas for different radionuclides allows for optimal shielding design associated with each product. Such considerations, however, should be flexible to allow for adaptation of new procedures and new radionuclides. Nuclear pharmacies should be cautioned against stacking heavy, free-standing, lead blocks, since they may present serious hazards, i.e. traumatic injury to legs and feet from accidental falling blocks, radiation exposure through non-interlocking joints, and excessive weight induced structural damage to cabinetry. Inexpensive and highly effective shielding for low energy radionuclides can be obtained with lead perchlorate shields suggested by Barnett and Harris.¹

Radiopharmaceutical Dispensing

The proper dispensing of radiopharmaceuticals will affect the absorbed radiation dose to the patient and the pharmacist. Maximal patient protection is achieved by the utilization of unit dose radiopharmaceuticals. All radiopharmaceuticals are dispensed from the nuclear pharmacy on prescription. The prescription indicates the requested study, the preliminary diagnosis, patient name, height, weight, age and the time that the patient is to receive the dose. With this information, the pharmacist can correlate the radiopharmaceuticals with the proposed study and patient information in such a manner that the patient will receive the optimum dose. Each product is dispensed precalibrated to the time of administration, and receives duplicate assays of the radioactive contents. The radiopharmaceutical is dispensed with a label indicating the patient's name, time and route of administration, date, prescription number, physician's name and pharmacist's initials. Unit dose dispensing has reduced the potential for dispensing and administration errors associated with major drug delivery systems.²

It is believed that the concept of unit dose dispensing of radiopharmaceuticals is an efficient and effective method of reducing errors associated with the administration of radioactive pharmaceuticals.

As an additional radiation protection device for the pharmacist, as well as maintaining the pharmaceutical quality of the products, our pharmacy utilizes a shielded syringe-valve dispensing system as modified from Hoar.³ A schematic diagram is given in Figure 3. The solution to be unit dose dispensed is drawn from the shielded vial (A) into the shielded large volume syringe (B) via a 3-way valve (C). A unit dose is subsequently dispensed by attaching a small volume syringe (D) to the dispensing part of the 3-way valve, drawing off the desired volume, attaching a needle, assaying the contents and affix-

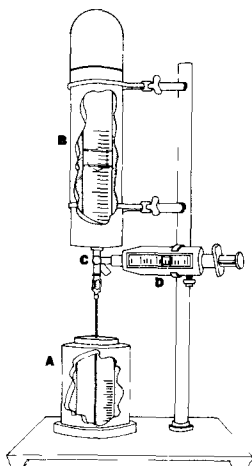


Figure 3
Dispensing System

ing the label. This dispensing process is conducted within the laminar air flow hood to insure sterility of the product.

The use of the 3-way valve dispensing unit not only insures pharmaceutical quality of the product, but decreases the hand and finger doses to the pharmacist with an average dispensing time of 25 seconds. Several reports ^{4,5,6} indicate the potential problem hand and finger doses to medical personnel. Implementation of a 3-way valve dispensing system significantly reduces exposure problems since the pharmacist need not handle the barrel of the syringe.

Administered Radiation Dose

The reduction of medical exposures and patient protection have always been the goals of nuclear medicine personnel. Tremendous gains in reducing medical exposures have been accomplished by using shorter half-life radiopharmaceuticals, by increasing biological turnover times, and by selection of the best radionuclide for labeling purposes. However, little consideration has been given to the administered dose. The relative assessment of benefit versus risk can only be accomplished if the administered dose is tailored to the individual patient. Too high an administered dose results in increased absorbed dose. Too low an administered dose may result in a missed diagnosis or a readministration of the drug. The Nuclear Pharmacy has evaluated the administered dose regarding the genetically significant population, the method of dose determination and the pharmacists role in product selection.

An evaluation of 200 randomly selected patients receiving a radiopharmaceutical indicated that 12.5% were less than 18 years old; 34.5% were between 18 and 45, and 53% were greater than 45 years old. The genetically significant population (less than 45) represents a 13% increase as compared to the national average of 1970. Leblanc and Johnson⁷ have also indicated that exposures from nuclear medicine procedures increased from 5% gonadal exposure/admission in 1964 to 11% in 1968.

Administered radiopharmaceutical doses vary from clinician to clinician. The selection of an administered dose may be from a table of doses determined from past experience,⁸ the application of body weight, e.g. mCi/Kg or from a series of rules, e.g. Young's rule, Clark's rule or surface area.^{9,10,11}

While adult administered doses are fairly well established, large variations in pediatric doses are apparent. Administered doses based upon age show great limitation when one considers the variability of a given age. For example, the 3rd percentile of a 10 year old girl is 53.2 lbs., while the 97th percentile is 101.9 lbs.¹² Administered doses determined by weight, while better than doses determined by age, usually underestimate the requisite clinical dose. The underestimated dose is due to 1) weight changes as a function of the cube of linear dimensions while the necessary photon fluence for adequate lesion localization varies with the square of linear dimensions, and 2) ratios of organ/body weights in infants are greater than those observed in adults.

To provide a uniform and reliable method of computing administered radiopharmaceutical doses, for the broad spectrum of patients seen in our clinic, Nuclear Pharmacy employs body surface area as modified by height and weight. Administered doses, in mCi/m² are usable for all patients regardless of variations in weight, height, age or sex.

As a further consideration of the absorbed radiation dose, our pharmacists participate in the selection of the radiopharmaceutical for the patient. Qualified nuclear pharmacists have the

necessary training in radiopharmaceuticals, biopharmaceutics and metabolism to guide the physician in selecting the optimum drug to obtain the maximum diagnostic information. In addition, the nuclear pharmacist has played a vital role in identifying patients that may have drug interactions which prevent the meaningless use of a radiopharmaceutical. For example, a request for a red cell survival test one week after a gallium-67 scan.

Radiopharmaceutical Documentation

Good radiation protection principles require adequate record keeping to evaluate personnel methods and product control. Pharmacy requires substantial record keeping to validate a product's suitability for human use. To meet the requirements of both health physics and pharmacy, a product quality control system has been developed. A schematic diagram of the quality control program is shown in Figure 4.

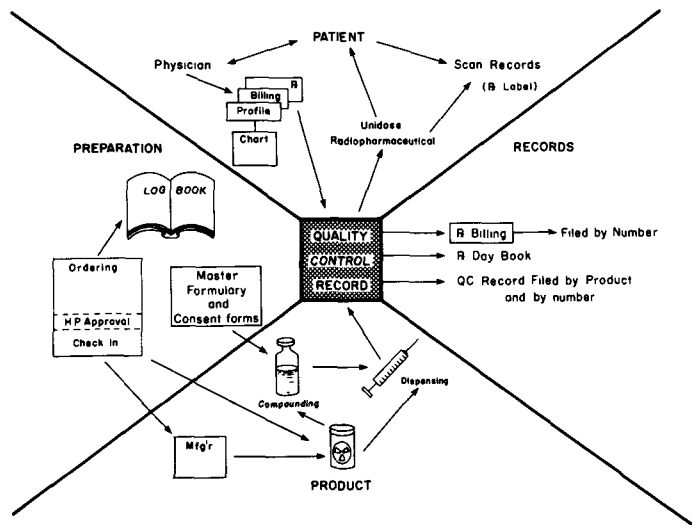


Figure 4
Quality Control Program

Each product received into or compounded by the nuclear pharmacy has an assigned quality control card (center). Each product is further designated by a number indicating the date received or compounded. All compounded radiopharmaceuticals are prepared in accordance with a master formulary. All records of tests, e.g. sterility, apyrogenicity, particle size, radiochemical purity, chemical purity, radionuclidic purity, pH, etc. are indicated on the quality control card. All unidoses dispensed from the product are also recorded with the

quality control record. In this manner, a complete history of any radiopharmaceutical can be ascertained with a brief glance at the quality control record card.

As part of our quality control program, we require that used syringes be returned to the nuclear pharmacy. This alleviates the hazard of loose contaminated syringes and also allows the syringe to be reassayed. In this manner, complete assessment of the administered dose can be determined. LeBlanc and Johnson¹³ and Abdel-Dayem¹⁴ have reported on retained activity of Xenon-133 within syringes. Freedman¹⁵ has described a similar deposition of technetium-99m sulfur colloid in the rubber plunger of disposable syringes. Our reassay procedure indicates that many radiopharmaceuticals have residual activity in syringes, especially radio-labeled proteins.

As part of the documentation concern for radiopharmaceuticals, our pharmacy has assumed the responsibility for the maintenance of all records necessary for clinical trials of new radiopharmaceuticals, as well as validating drug interactions or adverse reactions. Within our hospital, the patients chart is available to the pharmacist during the prescription preparation. A review of previous or existing drug therapy by the pharmacist can offer lead to the

explanation of modulation in drug distribution. When an adverse reaction occurs, our nuclear pharmacy coordinates the compilation of associated data and the reporting of the reaction to the professional societies and concerned authorities.

Conclusions

1. The objectives of pharmacy and health physics are synergistically compatible in reducing personnel and patient radiation exposure.

2. Methods for reducing radiation exposures within regional nuclear pharmacies while maintaining large inventories and increased utilization of radiopharmaceuticals requires adequate facility design, consideration of administered dose, unidose dispensing and quality control documentation.

3. Surface area measurements used in consideration of administered doses have aided in providing uniform and reliable scans between patients, especially in pediatric nuclear medicine.

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AEROSOLS AND LUNG MODELS

РАДИОАКТИВНЫЕ АЭРОЗОЛИ КАК ФАКТОР ВНУТРЕННЕГО ОБЛУЧЕНИЯ ПРИ ИСПОЛЬЗОВАНИИ РАДИОАКТИВНЫХ ВЕЩЕСТВ

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In this report we consider the problem of determining the probable accumulation of the isotopes in the organism. These questions are solved on the basis of values of mean air contamination.

The experimental results were applied to the regulation of the air contamination.

Authors have proposed to use the term "the standard work conditions" in radiation protection practice. These "conditions" are characterized by the hygienical safety factor equal to 5, which should be included in the values of regulated PMS.

Общеизвестные принципы организации производства позволяют обеспечить безопасные условия труда для персонала только на базе норм радиационной безопасности, в основе которых лежит лимитирование дозовых нагрузок (первичные дозиметрические характеристики), определяющих степень опасности непосредственно для индивидуума.

Одновременно действует статус среднегодовых допустимых концентраций (вторичных дозиметрических характеристик), относящихся к параметрам среды, в которой работает персонал, и характеризующих безопасные условия труда.

Именно здесь, по мнению авторов, существуют принципиальные трудности как в подходах к определению этих норм РБ, так и в оценках лучевых нагрузок по показателям загрязненности воздушной среды.

В первую очередь в проблеме "Радиоактивные аэрозоли как возможный фактор внутреннего облучения" возникает задача перехода от величины экспериментально определяемого среднего загрязнения воздуха к вероятному накоплению изотопа в организме на основе данных по статистическим закономерностям распределения концентраций и различным дозиметрическим характеристикам радиоактивных аэрозолей.

Во всех случаях при качественной характеристике вдыхаемого радиоактивного вещества фактически ведется речь о модели аэрозолей, в той или иной степени аргументированной. Например, некоторая модель аэрозолей может представляться как суперпози-

ция в том или ином соотношении газообразных соединений изотопа, мелкодисперсной и грубодисперсной аэрозольной компоненты различной растворимости.

В качестве примера остановимся на влиянии дисперсности пыли на величину отложения в дыхательном тракте. Радиоактивные аэрозоли дезинтеграции "горячих" лабораторий, как правило, относятся к классу грубодисперсных, а распределение размеров аэрозольных частиц с достаточной точностью описывается логарифмически нормальным законом, характеризующимся в случае плутония следующими параметрами: $\sigma_g = 1,6 \pm 3,4$ мкм, $\beta_g = 2,0 \pm 2,7$ аэродинамический $\sigma_g^{аэ} = 2,4 \pm 5$ мкм и активным среднегеометрическим аэродинамическим радиусом $r_{аэ} = 20 \pm 50$ мкм. Учитывая вышеприведенную дисперсность, можно считать, что мгновенное распределение активности по дыхательной системе характеризуется следующими величинами: для носоглотки мгновенный общий коэффициент задержки активности равен K_{a_1} общ = 90%, для области трахеи - бронхиального дерева K_{a_2} общ = 4%, для собственно легких (от дыхательных бронхиол и ниже) K_{a_3} общ = 2% /1/.

Таким образом, первоначальное отложение активности в собственно легких в этом примере будет значительно в 5-10 раз ниже, чем по принятой в настоящее время модели расчета СДК.

Весьма важным для ряда изотопов, так и в дисперсионной среде (например, тритий, углерод, ртуть, иод и др.), является вопрос об относительной роли перкутанного поступления в организм из воздуха.

На примере паров ртути сравним перкутанный и ингаляционный пути поступления. Авторами совместно с Ю.М. КОВАЛЕНКО было показано, что равновесное содержание ртути в организме в мккюри для первого случая выражается формулой $A_1 = 33 \left(\frac{c}{c_0} \right)^2 \times \frac{S \cdot c}{\lambda}$ (1) а для второго $A_2 = K_{a_2}^{аэ} \times \frac{c \cdot v}{\lambda}$, где c - концентрация паров ртути в мккюри/см³, c/c_0 - относительная упругость паров ртути, S - поверхность тела человека в см², λ - постоянная выведения ртути из организма в мин⁻¹, v - скорость дыхания в см³/мин.

Отсюда $\frac{A_2}{A_1 + A_2} = \left[33 \frac{S}{v K_{a_2}^{аэ}} \left(\frac{c}{c_0} \right)^2 + 1 \right]^{-1}$. (2) Анализ показывает, что для стандартного человека через кожу поступает от 2% до 65% всей ртути при изменении относительной упругости от 1% до 10%.

Анализ радиационной обстановки при загрязнении воздушной среды радиоактивными аэрозолями показал, что распределение их концентраций во времени и пространстве подчиняется логарифмически нормальному закону. Последнее представляется объяснимым, если, в частности, предположить, что распределение интенсивности источников загрязнения описывается усеченным гиперболическим законом, когда вероятность появления концентрации "с" обратно пропорциональна ее величине. Тогда распределение логарифмов концентраций будет равновероятным, а композиция таких распределений приводит к логарифмически нормальному закону.

Величина такого важного параметра, как стандартное геометрическое отклонение концентраций ($\beta_g \geq 1$) является при прочих равных условиях хорошим показателем степени радиационной надежности технологического процесса, а также определяет и вероятную величину дозовой нагрузки и ее разброс. Значение β_g для эмпирических распределений, как видно из таблицы лежит в пределах 2-13. Поскольку дозу внутреннего облучения можно приближенно представить как произведение трех случайных величин загрязнение воздушной среды помещения, времени пребывания человека в них

и проскока аэрозолей через средства индивидуальной защиты, то при условии, что каждая из них независима и подчиняется лог-нормальному закону с дисперсиями $\lg^2 \beta_{\text{зс}}$, $\lg^2 \beta_{\text{зт}}$, $\lg^2 \beta_{\text{зк}}$, соответственно, дисперсия дозы будет равна:

$$\lg^2 \beta_{\text{зд}} = \lg^2 \beta_{\text{зс}} + \lg^2 \beta_{\text{зт}} + \lg^2 \beta_{\text{зк}}. \quad (3)$$

Если величина загрязнения воздушной среды помещения и время пребывания в нем человека скоррелированы (коэффициент корреляции Γ), то

$$\lg^2 \beta_{\text{зд}} = \lg^2 \beta_{\text{зс}} + \lg^2 \beta_{\text{зт}} + 2 \lg \beta_{\text{зс}} \lg \beta_{\text{зт}} \Gamma. \quad (4)$$

Поскольку обычно $\beta_{\text{зд}} < \beta_{\text{зс}}$, то $\Gamma < 0$, что отражает известное положение: чем больше загрязненность, тем обычно меньше время пребывания в ней человека.

Реальные дисперсии первичных дозиметрических характеристик приведены в таблице.

Таблица

Стандартное геометрическое отклонение радиационных характеристик ($\beta_{\text{з}}$).

№№	$\beta_{\text{з}}$ внеш. для индивидуальных доз гамма-нейтронного облучения		$\beta_{\text{зн}} \approx \beta_{\text{зд}}$ для величин накопления радиоактивных веществ в организме	$\beta_{\text{зн}}$ для величин поступления радиоактивных веществ в организм	$\beta_{\text{зс}}$ для концентраций радиоактивных аэрозолей	
	γ	n			обычная	при рем. эксплуат. работах
1	1,8	1,9	2,8	2,8	5,7	3,9
2	2,5	1,7	2,3	2,3	4,2	2,8
3	2,3	2,4	2,3	1,8	2,5	4,1
4	2,3	2,1	2,3	5,1	6,6	3,2
5	2,1		2,3	1,7	2,4	10,8
6	2,6		5,0	2,1	2,0	12,9
7	2,3		1,8			
8	1,6					
9	1,8					
10	1,7					
Среднее значение	$2,1 \pm 0,3$	$2,0 \pm 0,2$	$2,7 \pm 0,7$	$2,4 \pm 0,8$	$3,9 \pm 1,6$	$6,2 \pm 3,7$

Таким образом, специфика работы заключается в том, что существует разброс показателей загрязнения воздуха и соответствующее ему размытие первичных дозиметрических характеристик.^{2,3}

Кроме того, величины накопления изотопов в организме людей, работающих в "одинаковых" условиях, имеют значительно больший разброс, чем биологические константы.

Этот факт требует введения допустимых рабочих пределов загрязнения воздуха, абсолютная величина которых меняется каждый раз в зависимости от дисперсии величин накопления изотопов в организме и отличается от СДК на коэффициент запаса.

В качестве примера рассмотрим вариант, когда распределение величин первичных дозиметрических характеристик подчиняется логарифмически нормальному закону с $\beta_{\text{з}} = 5$. В этом случае 21%

работающих будут иметь нагрузки значительно выше средней. Это означает, что при среднем загрязнении воздуха в пределах СДК и обычном режиме труда у 1/5 работающих дозовая нагрузка на организм будет превышать СДН, что нельзя считать удовлетворительным. Поэтому для оценки радиационной обстановки по усредненным значениям дозиметрических характеристик необходимо ввести новый критерий, восполняющий отсутствие у средней величины индивидуальных черт. Этот критерий может быть сформулирован следующим образом: безопасной средней величиной дозиметрической характеристики можно считать такую величину, при которой вероятность появления радиационной нагрузки, превышающей допустимую, будет приемлемо мала.

Исходя из всего изложенного, для определения допустимого рабочего предела коллективных показателей радиационной обстановки в численное значение среднегодовой допустимой величины (СДВ) или СДК в случае аэрозолей следует ввести коэффициент запаса для индивидуума $K_{и}$.

Очевидно, $K_{и}$ определяется величиной $\beta_{зд}$ и принимаемым коэффициентом риска $\beta = \frac{100}{a}$ (а - % людей, имеющих радиационную нагрузку выше допустимой).

Выражение для вычисления $K_{и}$ может быть получено следующим образом.

Доля людей ($\frac{a}{100} = \frac{1}{\beta}$) с радиационной нагрузкой больше какой-либо величины и имеющей интегральное распределение, подчиняющееся лог-нормальному закону, равна

$$1/\beta = 1 - F(m) = 0,5 [1 - \Phi(\xi)],$$

где $\Phi(\xi)$ - интеграл вероятности.

Отсюда $1 - 2/\beta = \Phi(\xi)$ или выражая ξ через обратную функцию, $\xi = \Phi^{-1}(1 - 2/\beta)$.

Коэффициент запаса $K_{и}$ определяется нами как отношение двух значений m $K_{и} = \frac{m_1}{m_2}$, одно из которых является среднеарифметической величиной (\bar{m}_1) наблюдаемого распределения первичной дозиметрической характеристики $F(m_1)$ с параметрами m_1, β_1 и β_1 , равной среднегодовой допустимой величине ($\bar{m}_1 = \text{СДВ}$), определенной по старому критерию безопасности, делитель же соответствует среднеарифметической величине распределения $F(m_2)$ со старым стандартным геометрическим отклонением β_2 , но сдвинутым относительно $F(m_1)$ таким образом, что $F(m_2 = \bar{m}_1) = 1 - \frac{1}{\beta}$, т.е. доля людей с радиационной нагрузкой больше предельно допустимой величины ($\text{ПДВ} = \bar{m}_1$), становится равной α . Вспоминая,

что $\xi = \frac{\lg m - \lg m_2}{\lg \beta_2}$ получим $\Phi^{-1}(1 - \frac{2}{\beta}) = \frac{\lg \bar{m}_1 - \lg m_2}{\lg \beta_2}$

или $m_2 = \bar{m}_1 \beta_2^{-\Phi^{-1}(1 - 2/\beta)}$, но $m_2 = m_{2g} \beta_2^{0,5 \ln \beta_2}$

и окончательно $\bar{m}_2 = \bar{m}_1 \beta_2^{0,5 \ln \beta_2 - \Phi^{-1}(1 - 2/\beta)}$

соответствует по определению коэффициента запаса $K_{и} = \frac{\bar{m}_1}{\bar{m}_2}$, что

уравнению
$$K_{и} = \frac{\beta_{зд}}{K} \quad (5)$$

1) вторичные доз. характеристики.

В настоящем докладе авторы предлагают по аналогии с существующим термином "стандартный человек" ввести понятие "стандартных" условий труда, характеризуемых коэффициентом запаса равным 5. Тогда допустимый рабочий предел загрязнения воздуха, соответствующий "стандартным" условиям, может трактоваться как новая СДК, которая будет жестче общепринятой в 5 раз.

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ОБОСНОВАНИЕ ОСНОВНЫХ РАДИОБИОЛОГИЧЕСКИХ ХАРАКТЕРИСТИК
ТОРИЯ-232 И ПРОДУКТОВ ЕГО РАСПАДА И ГИГИЕНИЧЕСКИХ НОРМ
(СДК и ПДП) В ВОЗДУХЕ ПРОИЗВОДСТВЕННЫХ ПОМЕЩЕНИЙ.

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Summary

Rats were administered, intratracheally or by inhalation, with soluble ($^{228}\text{ThCl}_4$), insoluble ($^{232}\text{ThO}_2$) thorium compounds together with its decay products as well as thorium-232 present in ores. Distribution and retention of Th-232 and its daughters were established for lung, lymph nodes, liver, bone, spleen, kidney and others.

The maximum admissible dose (MAD) was determined for small quantities of thorium inserted in ores.

The hygienic norms of thorium-232 and natural thorium (maximum admissible concentration) in atmospheres of workrooms were estimated.

Введение.

Радиотоксическое действие тория тесно связано с особенностями распределения радионуклидов ряда тория-232, уровнями их накопления в органах и тканях, скоростями выведения из организма.

Данные о метаболизме дочерних радиоэлементов ряда тория-232 в организме при поступлении их в органы дыхания совместно с материнским элементом или возникновении в результате распада материнского радионуклида в месте локализации последнего крайне ограничены. 1, 2, 3

Сведений о выведении тория-232 из легких при поступлении радионуклида в кристаллической решетке руд и концентратов в литературе не найдено. Данные о сочетанном действии микроколичеств тория и элементов, входящих в состав редкометаллических руд, отсутствуют.

Материалы и методы.

Исследования проведены на белых крысах, которым однократно интратрахеально вводили нерастворимое ($^{232}\text{ThO}_2$) и растворимое ($^{228}\text{ThCl}_4$) соединения тория совместно с дочерними радионуклидами.

Окись тория метили радиоторием. ^{228}Th находился в равновесии с продуктами распада. Вводили $4,8 \cdot 10^{-6}$ кюри $^{232}\text{ThO}_2$ и $3,4 \cdot 10^{-7}$ кюри $^{228}\text{ThCl}_4$ на килограмм веса.

Счетный медианный диаметр (СМД) $^{232}\text{ThO}_2$ был равен 1,03 ммк, массмедианный диаметр (ММД) - 4,9 ммк.

Хлорид радиотория вводили в изотоническом растворе при pH=7. 95% тория-228 находилось в мономерной форме.

Крысы забивали через 3, 12, 24, 72 часа и 30, 90, 180, 270 суток после введения.

В органах и тканях количественно определяли ^{232}Th , ^{228}Th , ^{224}Ra , ^{212}Pb , ^{212}Bi .

Результаты.

В результате проведенных последований установлено, что распределение тория-232 и его дочерних продуктов в основном зависит от растворимости соединений.

В почки и печень поступает преимущественно свинец-212. Радий-224 накапливается в 3-4 раза меньших количествах.

При введении растворимого соединения тория в равновесии с дочерними радионуклидами, радий-224, свинец-212, висмут-212 находятся, в основном, в скелете. Максимальное количество радия-224 и свинца-212 накапливается в скелете через 24-72 часа после введения и достигает 78 и 75% соответственно от общего содержания их в организме.

Торий-228 в ранние сроки после введения находится преимущественно в легких. Постепенно количество тория-228 в легких снижается и через 90 суток содержание радионуклида в скелете становится выше, чем в легких. Спустя 270 суток после введения количество тория-228 в скелете достигает 66% и в легких - 22,8% от общего содержания радионуклида в организме. Максимальное содержание свинца-212 в печени и почках достигает 6,7 и 8,8% соответственно. Содержание радионуклидов ряда тория-232 в почках, печени, селезенке значительно ниже, чем в скелете. Наибольшее количество радия-224, находящееся в почках, равно 4,1% и в печени - 2,76% от содержания в организме. В перибронхиальных лимфатических узлах находится не более 2,68% тория-228 и 1,29% радия-224 и практически отсутствует свинец-212.

При расчетах поглощенных доз, установлении токсического действия и выборе критического органа существенное значение имеет не только общее содержание радионуклида в органе, но и его концентрация. При однократном интратрахеальном введении окиси тория наиболее высокая концентрация тория-232 и -228, радия-224 и свинца-212 почти во все сроки наблюдения отмечается в легких. Лишь спустя 6 месяцев после поступления препарата концентрация радионуклидов ряда тория в перибронхиальных лимфатических узлах становится выше, чем в легких. При поступлении в органы дыхания хлорида тория наиболее высокая удельная активность тория-228 и радия-224 почти в течение всего времени наблюдения находится в легких. Лишь через 90 суток после введения препарата удельная активность тория-228 и радия-224 в перибронхиальных лимфоузлах становится в 2,5 раза выше, чем в легких. Удельная активность свинца-212 в легких выше, чем в скелете лишь в течение первых суток после введения. Уже через 3 суток удельная активность этого изотопа в почках, печени, костной ткани становится выше, чем в легких.

Скорости выведения радионуклидов ряда тория из легких зависят от их растворимости.

При поступлении двуокиси тория совместно с дочерними радионуклидами выведение тория-232, тория-228, радия-224, свинца-212,

висмута-212 осуществляется в две стадии. Около 40% поступивших в органы дыхания радионуклидов выводится с эффективным периодом полувыведения ($T_{эфф.}$) около 3-х часов, эффективный период полувыведения остальной части тория-232 и 228 равен 180-200 суток. Изменение содержания радия-224, свинца-212, висмута-212 в легких при введении двуокиси тория определяется биологическим выведением, радиоактивным распадом и накоплением в результате распада материнского радионуклида и характеризуется следующими значениями полупериодов удержания: $T_{224Ra} = 160$ суток, $T_{212Pb} = 160$ суток, $T_{212Bi} = 116$ суток. (Рис. 1).

При введении в органы дыхания хлорида радиотория динамика выведения радионуклидов ряда тория-228 существенно отлична от наблюдаемой при поступлении двуокиси тория (рис. 2). Выведение тория-228 из легких осуществляется с 3-мя периодами полувыведения: $T_{эфф.} \leq 3$ часа, $T''_{эфф.} = 2,25$ суток и $T'''_{эфф.} = 60$ суток. Изменение содержания радия-224 и свинца-212 в легких в ранние сроки после введения осуществляется быстрее, чем тория-228. Уже через 3 часа после введения хлорида тория содержание тория-228, радия-224, свинца-212 в легких составляет 65,7, 22,4 и 46,7% от введенного количества. Изменение содержания радия-224 спустя первые 3 часа после введения хлорида радиотория осуществляется с двумя периодами полувыведения: $T_{1/2} = 1,18$ суток и $T''_{1/2} = 60$ суток. Проведенные нами расчеты показали, что периоды биологического выведения радия при этом равны $T_6 = 0,93$ суток /от 3 до 72 часов/ и $T''_6 = 1,19$ суток /от 3-х до 270 суток/.

Активность легких, обусловленная свинцом-212 и висмутом-212, снижается очень быстро и через 72 часа после введения составляет всего 0,2% от введенного количества. Очень быстрый спад активности свинца-212 постепенно прекращается и в период от 3 до 90 суток активность его остается почти без изменений. Затем активность свинца-212 и висмута-212 в легких вновь начинает снижаться. В результате разных скоростей выведения радионуклидов ряда тория при поступлении хлорида тория в легких наблюдается значительное нарушение радиоактивного равновесия в ряду тория-228.

Тенденция к более быстрому выведению дочерних радионуклидов по сравнению с материнским может быть обусловлена большей скоростью перехода их в кровь. Доли радионуклидов, переходящих во внутренние органы и ткани из мест поступления соединений тория, определяется величиной всасывания их в кровь и тропностью к определяемому органу.

При поступлении соединений тория в органы дыхания скорости резорбции радионуклидов ряда тория-232 в кровь зависят от химической природы элементов, растворимости соединений тория и времени с момента его поступления (табл. 1).

Таблица 1

Скорости резорбции в кровь крыс тория-228, радия-224, свинца-212 в % от содержания в организме /сутки при интратрахеальном введении соединений тория.

Радионуклид	Время после введения					
	3ч.	24ч.	72ч.	30 сут.	90 сут.	180 сут.
	<u>вводили двуокись тория</u>					
Торий-232	0,1	0,01	0,001	0,001	0	0
Радий-224	2,2	15,9	40,3	4,2	4,3	3,4
Свинец-212	-	11,0	10,3	5,6	6,3	4,3
	<u>вводили хлорид тория</u>					
Торий-228	0,3	0,22	0,24	0,36	0,02	0
Радий-224	-	85	27	16,2	11,3	22,5
Свинец-212	-	24	27,6	29,6	31	24,6

Как следует из табл. 1 резорбция радия-224 и свинца-212 в кровь при введении в органы дыхания как растворимых, так и нерастворимых соединений тория выше, чем материнского радионуклида. Незначительный переход тория в кровь при введении двуокиси тория может быть обусловлен крайне плохой растворимостью его в воде и тканевых жидкостях. Более высокий переход радия-224 и свинца-212, находящихся в кристаллической решетке практически нерастворимого в воде соединения двуокиси тория, по-видимому, обусловлен повышенной выщелачиваемостью этих радионуклидов.

Из крови радионуклиды ряда тория переходят преимущественно в костную ткань. При поступлении ThO_2 в органы дыхания в скелет переходит до 0,12% тория-232, 17%- радия-224, 13% - свинца-212 и 7% - висмута-212. В печень и почки переходят свинец-212 и висмут-212. Радиоактивное равновесие в ряду тория-232 в скелете и внутренних органах нарушено в значительной степени как при введении нерастворимого, так и растворимого соединений тория.

Проведены исследования скорости выведения тория-232 из легких крыс в том случае, если радионуклид поступает в составе руды или концентрата. Установлено, что в случае хронического ингаляционного поступления аэрозолей, содержащих труднорастворимые редкоземельные минералы типа циркона и лопарита, эффективный период полувыведения Th^{232} из легких белых крыс в случае лопарита был равен 440 ± 50 суткам, в случае циркона 470 ± 140 суткам. При экстраполяции этих данных на человека $T_{\text{эфф.}}$ для тория, входящего в состав труднорастворимых природных соединений, составит 4,8-5,2 года.

Исследования, проведенные на крысах, которые подвергались хроническому ингаляционному воздействию пыли циркона и лопарита, позволили установить, что минимально эффективная доза равна ~ 200 бэр. Для установления величины минимально-эффективной дозы были использованы такие критерии, как средняя продолжительность жизни, оценивавшаяся по ET_{50} - эффективному времени выживаемости 50% взятых в опыт крыс, по blastomagenному эффекту и по ряду биохимических, морфологических и других показателей, характеризующих не только состояние легочной ткани, но и организма в целом. Так, производилась оценка весовых показателей легких, содержания в них растворимых и нерастворимых белков, изучалась интенсивность обмена белков легочной ткани (с помощью аминокислот лизина и глицина, меченых по ^{14}C), степень склерозирования легочной ткани, а также состояние организма в целом по ряду гематологических, иммунологических и других физиологических показателей.

Итак, полученные в эксперименте данные позволили уточнить периоды полувыведения радионуклидов ряда тория ($T_{\text{эфф.}}$), доли (f возд.), доли радионуклидов от количества во всем теле (f_2), эффективные энергии радионуклидов ряда тория ($E_{\text{эфф.}}$) и др., а также установить величину ПДМД при поступлении в организм природного тория в составе труднорастворимых руд и концентратов.

На основании этих данных проведен расчет гигиенических норм тория-232, тория естественного, а также природного тория, входящего в состав труднорастворимых руд и концентратов.

Расчет проводили для частиц пыли с массмедианным аэродинамическим диаметром (ММАД) равным 0,06 мкм, 1 мкм, 10 мкм. Использовали формулу, основанную на экспоненциальном законе выведения. В результате расчета установлены следующие значения:

СДН Th^{232} от $0,37 \cdot 10^{-14}$ до $2,6 \cdot 10^{-14}$ кюри/л

СДН Th -ест. от $0,8 \cdot 10^{-5}$ до $6,2 \cdot 10^{-5}$ мг/л

СДН Th в составе $2,5 \cdot 10^{-15}$ кюри/л
нерастворимых руд

Из расчетов следует, что гигиенические нормы варьируют в

зависимости от дисперсности пыли.

В НРБ⁴ в настоящее время принятой величиной считается среднее значение, полученное для ММАД-1 мкм и равное:

СДН Th²³² = $1,16 \cdot 10^{-14}$ кюри/л

СДН Th_{вст.} = $3,7 \cdot 10^{-5}$ мг/л.

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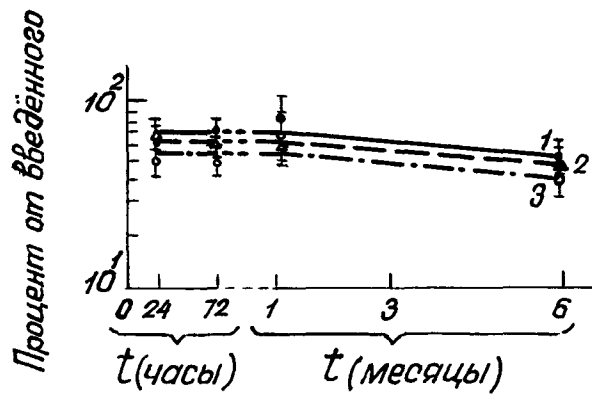


Рис. 1. Удержание тория-228(1), радия-224(2) и свинца-212(3) в легких крыс при интратрахеальном поступлении двуокиси тория.

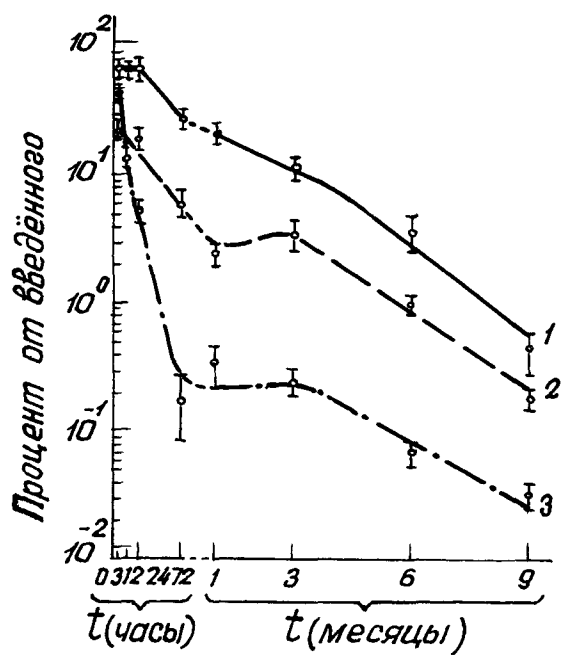


Рис. 2. Удержание тория-228(1), радия-224, свинца-212(3) в легких крыс при интратрахеальном поступлении хлорида радиотория.

INHALATION STUDY OF A SUBMICRON SIZE LEAD-212 AEROSOL

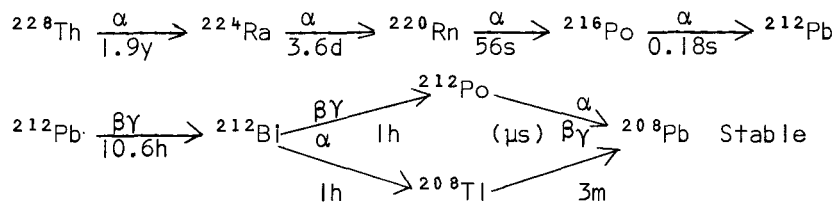
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Abstract

Dogs (purebred female beagles ~ 10 kg) were briefly exposed to a submicron aerosol of ^{212}Pb having an effective diffusion diameter between 105-120 Å, (σ_g 1.2) produced in a chamber of new design. The resulting lead burden was followed for up to 72 hours postexposure by external counting of the 240 keV gamma ray. The concentration of ^{212}Pb in the arterial and mixed venous blood were regularly measured and no significant A-V difference could be found. The blood concentration data were used to differentiate between the ^{212}Pb level actually in the lungs and that within the thoracic blood volume. This approach was facilitated by the intravenous injection of ^{212}Pb so that an increment in ^{212}Pb concentration in the blood could be correlated with the increment in thoracic radioactivity. Seven canine studies indicated that the submicron aerosol of ^{212}Pb was cleared from the lungs with a single exponential process having a biological half-time of ~ 12 hours. This mean value corresponds closely to the biological half-times in human lungs reported by other investigators for ^{212}Pb and ^{210}Pb labelled aerosols of diverse physical and chemical forms, viz. 8-12 hours, suggesting that the clearance value can be generalized for atmospheric lead. Evidence is presented that the blood is a principal clearance pathway and that external assessment of radioactive lead in the thorax must also consider the lead content of the thoracic wall structures.

Introduction

An aerosol of ^{212}Pb is formed by the decay of airborne ^{220}Rn , an alpha-emitter having a half-time of 56 seconds. This aerosol has been the subject of several recent investigations because: (a) the aerosol produced is of submicron dimensions and provides an interesting physical system for investigating the production of aerosol by radioactive emissions¹⁻⁵, and (b) of its short half-life, and physical and chemical nature, which make it of pertinence and value for biological studies including those in man⁶⁻⁹. The radioactive schema for the production of ^{212}Pb and its decay are as follows:



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Methods and Materials

The experimental animals used in the study were purebred, female, beagle dogs of approximately 10 kg body weight. Individual exposures to ^{212}Pb aerosols were accomplished through an endotracheal tube connected to a special aerosol chamber (Figure 1) while the canine subject was anesthetized with pentobarbital sodium (ca 28 mg/kg i.v.).

^{212}Pb AEROSOL GENERATION SYSTEM

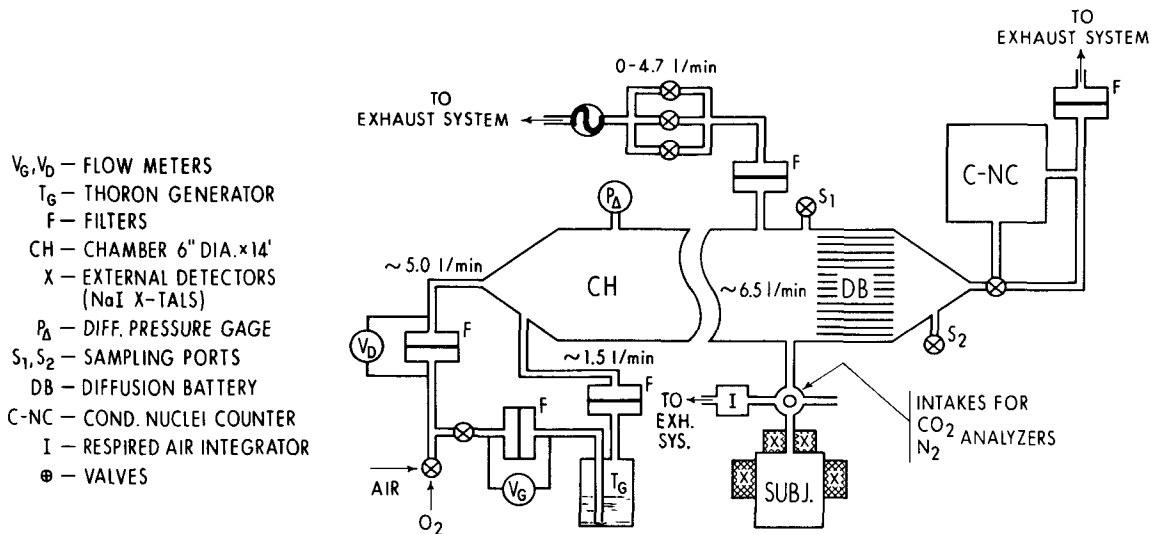


Figure 1. Schematic of the Aerosol Generation and Exposure System.

The linear chamber, CH, is complemented by four sub-units: a thoron source and aerosol generation unit (left); an aerosol analysis unit (right) consisting of sampling ports S_1 , S_2 , the diffusion battery, DB, the condensation nuclei counter, C-NC, and a variable exhaust to control the flow through DB; the exposure section (rt. center) with its associated physiological monitors and the exhaust (right) for the disposal of unused aerosol.

The ^{212}Pb aerosol was produced as the result of passing a clean air stream (1.5 l/min) through a thoriumnitrate solution enriched with ^{228}Th and extracting the ^{220}Rn gas. The ^{220}Rn -air mixture was then passed through a coarse filter and a molecular filter before being diluted further by filtered clean air (5.0 l/min). The resulting radon mixture was conducted through a linear chamber 15 cm diameter by ~ 430 cm long. At this point, connections were located which provided for (a) exposing the subject; (b) analyzing the aerosol; and (c) exhausting the unused aerosol. The volumetric flow in the chamber determined the extent to which the ^{220}Rn decayed to ^{212}Pb at the point of use, i.e. determined the transit time. At 6.5 l/min, the chamber transit time was 10 minutes which is equivalent to approximately 11 physical half-lives for the radon gas. This implies that at the point of use, the ^{220}Rn had undergone 99.95% decay.

In the linear-chamber system, the aerosol can be produced continuously under highly controlled conditions so that the characteristics of the aerosol

are quite constant and highly reproducible from day to day. The dog exposure studies utilized a submicron aerosol with the following average characteristics: an effective diffusion diameter between 105-120 Å; geometric standard deviation ~ 1.2 ; a mean ^{212}Pb activity concentration of $0.7 \mu\text{Ci } ^{212}\text{Pb}/\text{l}$ air corresponding to slightly less than 1 picogram of Pb/l. The exposures were generally of about 5 minutes duration and the initial respiratory burdens in the dog lungs averaged $\sim 5 \mu\text{Ci}$ of ^{212}Pb .

The analyses of the ^{212}Pb aerosol depended upon the determination of the mass and numerical penetration values in a diffusion battery (Fig 1). Methods of estimating the distributional parameters of a diffusive, heterodisperse aerosol had been described by Sinclair¹⁰, Hursh and Mercer⁷, and by Fuchs¹¹.

After the brief aerosol exposure, each dog was positioned in a body holder and placed within a shielded thoracic counting chamber¹². Two 0.5" by 6.0" scintillation counters (NaI) were collimated so as to view the lateral aspects of the dog's chest between the first and fifth intercostal spaces thereby avoiding activity from subdiaphragmatic structures and the supraglottic airways. The outputs of the two counters were added in the multichannel analyzer and obtained by high-speed digital print-out. Analyzer channels corresponding to the energy range 210-270 kev were used for the ^{212}Pb measurements. For ^{212}Bi , the 560-620 kev range was utilized. Dogs were serially measured by external counting for as long as was practicable, normally 2 days postexposure.

The contribution of the ^{212}Bi daughter activity to the in vivo and in vitro measurements of ^{212}Pb was determined by studying freshly-acquired filter samples of ^{212}Pb either as a distributed source, e.g. in the lung fields of a beagle phantom, or in the same geometry as other samples were analyzed, i.e. blood. Serial blood samples were required in order to correct the thoracic counts for the contribution of ^{212}Pb which was bloodborne. A sealed thorium standard (in equilibrium with its daughter products) was also utilized in the correction and calibration procedures. The $^{212}\text{Pb}:^{212}\text{Bi}$ ratios were determined as a function of time and evaluated in terms of the excessive ^{212}Pb counts measured. The appropriate relationships were then used to account for the daughter contribution to all experimentally determined ^{212}Pb activities. The counting efficiency for measuring ^{212}Pb within the canine thorax was found to be approximately 0.7% whereas for ^{212}Pb in blood samples, the efficiency was $\sim 10\%$ for the geometry used.

It was known from previous experiments^{6,7,9} that external measurements of the chest soon after an exposure to a ^{212}Pb aerosol depicted the combined processes of lead build-up in the blood and of lead removal from the lungs. Consequently, a knowledge of the blood build-up rate and a "blood correction factor" were required in order to reveal the actual time-course of ^{212}Pb removal from the lungs. This "correction" procedure⁷ in essence, assumes that at least during most of the lung clearance (0-48 hr), there is no other important extrapulmonary compartment within the thoracic field except the thoracic blood volume. This point will be dealt with more fully in the Discussion section of this paper.

Results

In seven dog studies, measurements of ^{212}Pb activity as directly obtained by external counting during the first 24-48 hr postexposure gave an apparent effective half-life for the thorax of 8.3 hr implying a simple exponential clearance for lead with a biological half-time of approximately 40 hours ($\lambda = .0173$). However, when the raw activity data were corrected for the contribution of daughter activity, viz. ^{212}Bi , then the average thoracic clearance half-time (effective) and the estimate for the biological

half-time decreased

(Figure 2).

When the daughter-corrected, thoracic activity of ^{212}Pb was also adjusted for the contribution of the bloodborne ^{212}Pb , the average, effective clearance half-time for the thorax further decreased to 5.6 hours and the corresponding biological clearance half-time decreased to 11.9 hours ($\lambda = 0.054$). In Figure 3, therefore, the average curve can be depicted by the simple exponential equation: $R = Ae^{-\lambda t}$ where R is the fractional activity retained, A the initial activity, λ , the decay coefficient, is equal to 0.054, and t is the time in hours.

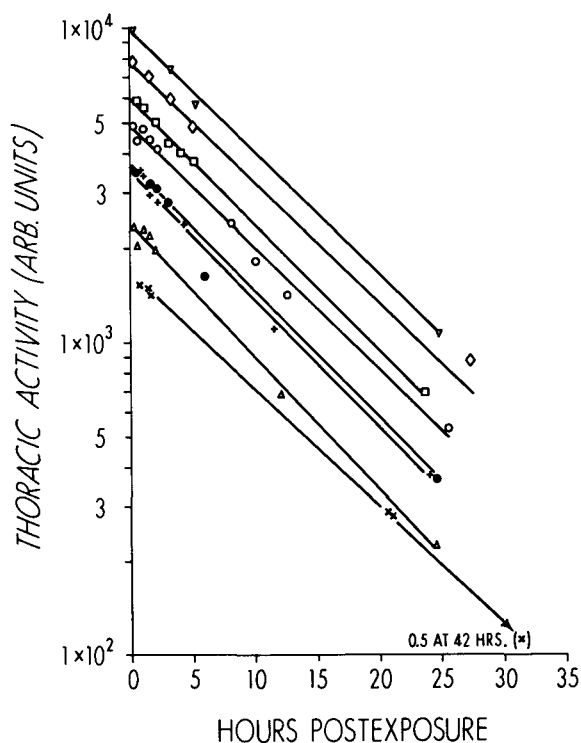


Figure 2. Graphic Summary of Thoracic Retention Data for ^{212}Pb .

These data are corrected for the Compton scattering in the ^{212}Pb energy spectrum due to ^{212}Bi activity. The intercepts of the curves were adjusted so as to avoid overlap in their presentation.

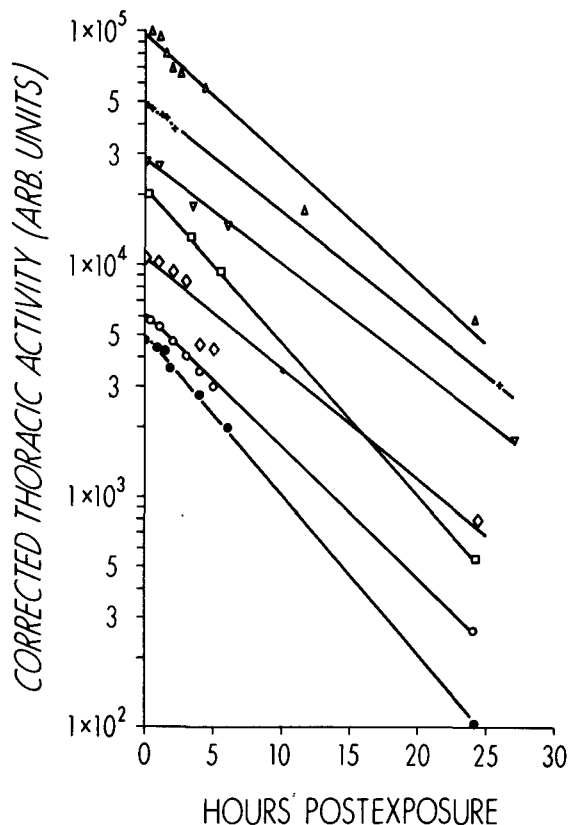


Figure 3. Corrected Thoracic Retention Data.

These curves have been corrected for bloodborne ^{212}Pb by the procedure described in the text. For this graphic presentation, the data have been arbitrarily adjusted to reduce overlaps.

Whereas, all of the clearance data can be considered exponential, the range of half-time values in the daughter-corrected data was 7.0–7.8 hours; in the data adjusted for bloodborne radioactivity, the variation increased, i.e. 4.3–6.9 hours, indicating that the blood correction procedure was largely responsible for this.

Measurements of bloodborne activity in the mixed venous and arterial blood revealed no significant ^{212}Pb concentration gradient during the first 24 hours. An A-V difference was suspected since there had been a report of an early high renal clearance of ^{212}Pb after intravenous administration⁸ which was soon

dissipated presumably due to the shift of ^{212}Pb from the plasma to red blood cells.

The blood build-up curve for ^{212}Pb peaked at approximately 10 hours post-exposure. In terms of stable or decay-corrected lead, there was no significant decrease in blood lead concentration below this peak concentration during the first 24 hours. The build-up phase in the blood appeared to be exponential and have a half-time of 4.7 hours. This is substantially faster than the rate of ^{212}Pb disappearance from the lungs. This indicated some inconsistency in the data if it is assumed that all (>95%) of the lead clearance is directly into the blood as did Hursh and Mercer⁷. This point is under further study.

One dog was given a large initial ^{212}Pb burden with the expectation that the thoracic clearance ^{212}Pb could be followed after 24 hours more accurately. At 96 hours postexposure, the thoracic level was found to be 0.18% of the initial value and far in excess of that commensurate with 11.9 hours biological half-time. The activity in the blood measured at this time was insufficient to account for more than 30% of the thoracic activity, but since the blood activity was so low, we considered this value highly speculative. Consequently the animal was sacrificed and the lungs, pulmonic nodes and blood corresponding to the blood volume of the thoracic viscera, were removed. The chest cavity was filled with a mass of wet, absorbent paper corresponding to the weight of the tissues removed and then the thorax was recounted as it was in the intact animal. Ninety-four percent of the terminal thoracic activity was found associated with the eviscerated thorax. We conclude, therefore, that the retained ^{212}Pb activity was based entirely upon the lead content of the blood and the thoracic wall structures and no lead retention could be assigned to the lungs and pulmonic lymphatics. This conclusion is entirely consistent with the biological half-time assigned to the lungs.

Discussion

Thoracic measurements in all dogs were subjected to a correction procedure to account for the bloodborne ^{212}Pb . Hursh and Mercer⁷ first reported this procedure, which entails the intravenous injection of a freshly-acquired ^{212}Pb sample and to determine the increments in systemic blood and thoracic radioactivities post-injection. Our preparation was made by sonification of a ^{212}Pb filter sample in isotonic saline followed by the injection of ~ 2 ml into a leg vein. The ratio of the increments in ^{212}Pb activity constitutes a correction factor which is derived from both geometric and blood volume considerations.

In this study, the initial thoracic measurements (0-1 hr) and the subsequent blood-corrected measurements were taken as pertaining to the removal of ^{212}Pb from the lung parenchyma. Certain corroborative evidence was cited. From other studies^{6,7,9} there is evidence of the removal of lead from the blood by biliary and urinary excretion and by redistribution to other tissues, especially skeletal. These pharmacodynamic features of blood lead are not easily studied with the ^{212}Pb isotope, but the available data indicate a very slow clearance rate for lead in the blood relative to that in the lungs.

The biological half-time of 11.9 hours for lead in the lungs is in close agreement to the 10.5-11.5 hr values reported by Hursh and Mercer⁷ for human subjects following single exposures to 0.02-0.23 μm AMAD aerosols. The clearance value is also similar to those reported by Booker, et al⁹ for ^{212}Pb labelled lead vapor in man, viz. ~ 10 hr biological half-time and by Jacobil³ for ^{212}Pb attached to atmospheric dust particles giving a biological half-time of 8 hours. Dog studies by Gibb and Hursh (unpublished), utilizing ^{210}Pb and involving ~ 0.1 μg burdens of stable lead in the form of a basic carbonate (0.2-0.6 μm AMAD), also revealed an initial lung clearance rate (after a blood-borne lead correction was applied) of 12.2 hours. The possibility that the

external counting data in these investigations were complicated by tracheo-bronchial clearance is discounted by the rate of ^{210}Pb build-up in the blood which was nearly identical to that found in this study.

These several studies with highly different aerosols of lead, both physically and chemically, give remarkably similar results, so that it appears practical to generalize the clearance of lead from the lungs of man following exposure to various atmospheric forms. In possible disagreement to this viewpoint are the data of Morken¹⁴ who reported a five minute biological half-time for ^{214}Pb on atmospheric dust from mouse lungs and the data of Albert and Arnett, who studied ^{212}Pb attached to kaolin particles¹⁵ and reported a 60 hour biological clearance half-time. It should be noted, however, that Albert and Arnett failed to account for the bloodborne ^{212}Pb .

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THE EFFECT OF BREATH-HOLDING ON THE
DEPOSITION OF HALF-MICRON AEROSOL
PARTICLES IN THE HUMAN LUNG

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ABSTRACT

Measurements of deposition of particles in the human lung during breath-holding can be made use of to calculate the diameter of the tubular passageways of the respiratory tract in life. When the inspired volume is sufficiently great for most of the aerosol to penetrate beyond the anatomical dead space, the rate of deposition during the breath-holding can be regarded as an index of the average diameter of the alveolar airways. Studies of the effect of breath-holding on deposition and recoveries of $0.5 \mu\text{m}$ particles of unit density in different lung volumes are described in this paper. Based on the results of steady-state breathing experiments, the average diameter of the airways in the alveolar region of the lung has been found to be 0.74 mms.

INTRODUCTION

Studies on the deposition of particles in human lungs show that it is possible pauses may occur in between inhalation and exhalation and also between successive breaths. The duration of these pauses should have certain effects on deposition of particles in the lung. According to Altshuler¹, breath-holding experiments are useful in assessing an average dimension of the alveolar spatial units in life. When the inspired volume is sufficiently large for most of the aerosol to penetrate beyond the anatomical dead space, the rate of deposition during breath-holding can be regarded as an index of the average dimension of the alveolar space in life.

This paper gives an account of the studies made to find the effect of breath-holding on the deposition of $0.5 \mu\text{m}$ spherical particles of unit density during the steady-state and single-breath experiments.

EXPERIMENTAL TECHNIQUES AND RESULTS

The apparatus used for the measurement of deposition is the one used by Davies, Heyder and Subba Ramu². Three different techniques were employed for measuring the effect of breath-holding on aerosol deposition. In the first case, the subject breathed the aerosol till steady-state condition was reached, with a pause for a known period of time between successive

breaths. Table 1 gives the results of the measurements for these two cases. Average breath-holding times (t) are given along with the tidal volumes (TV) expiratory reserve volumes (ERV), breathing frequencies (f) and the fraction deposited (D). It can be seen that the deposition increased by about 34% for an average breath-holding time of 1.07 secs in the first case whereas there was negligibly small difference in the second case.

Table-1

The effect of breath-holding on deposition during steady-state breathing

TV (Cm ³)	ERV (Cm ³)	f (Breaths/min)	t (secs)	(1-D) (Average)	Remarks
678	1260	16.8	0	0.906	Normal; No pause
678	1260	16.8	1.07	0.874	Pause between inhalation and exhalation
696	1200	17.8	0	0.905	Normal; No pause
696	1200	17.8	1.36	0.913	Pause between successive breaths

In the third case, the subject inhaled about 600 cm³ of aerosol-laden air, held the breath for a known period of time and exhaled more or less the same volume of air. This was followed by inhalation of clean air and maximal exhalation. The total recovery (R), recovery in tidal volume (R_t) and the recovery ($R_r/1-R_t$) of aerosol particles are calculated separately as shown in table 2. R_r is the ratio of the number of particles recovered in the

Table-2

The effect of breath-holding on the recovery of 0.5 μ m particles in the exhaled air (single-breath experiments)

TV (Cm ³)	ERV (Cm ³)	f (breaths/min)	t (secs)	R	R_t	$R_r/1-R_t$
660	980	16.5	0.0	0.96	0.90	0.72
690	820	15.4	3.0	0.86	0.76	0.54
665	941	14.4	5.7	0.79	0.67	0.42
615	950	14.2	7.6	0.73	0.62	0.33
585	1010	17.0	15.3	0.57	0.46	0.18
635	1170	15.3	24.0	0.45	0.36	0.13

reserve air to that in the inhaled air. ($R_r/1-R_t$) is the recovery of particles

in the reserve air expressed as a fraction of particles lost from the tidal air into the reserve air. Figure 1 gives the values of R , R_t and $(R_r/1-R_t)$ for different breath-holding times.

The build-up of aerosol particles in the lung before steady-state is reached is shown in figure 2 for normal breathing and breathing with pauses. Breath-holding seems to have no effect on the time needed for attaining the steady-state condition. The plot of R_n/R_{AV} against the number of breaths shows that, in all cases, steady-state is reached in four breaths. R_n is given by

$$R_n = E_n/I_n \quad \dots(1)$$

where I_n is the amount of aerosol inhaled in the n^{th} breath during build-up, and E_n the amount of aerosol exhaled in the same breath. R_{AV} is the average fraction recovered during steady-state breathing.

CALCULATION OF PASSAGEWAY DIAMETER

Landahl³ gives the following equation for the fraction deposited (D) in the human lung during breath-holding time 't':

$$-R_p \ln(1-D) = (1.8 \times 10^5 d^2 Ct) + (4 \times 10^{-6} \sqrt{Ct/d}) \quad \dots 2$$

where,

d = diameter of the particle (cm)

R_p = radius of the tubular passage (cm)

and $C = 1+1.8 \times 10^{-5}/d$, a size correction factor.

Table 3 gives the average diameters of the tidal volume (D_{TV}) and the expiratory reserve volume (D_{ERV}), calculated using equation (2) and table 2. $D(TV+ERV)$ is obtained from the results of the total recoveries for different breath-holding periods. The diameter of the tidal volume varies from 0.33 to 0.38 mms and that of the expiratory reserve volume from 0.16 to 0.20 mms.

The diameter of the airways in the alveolated region has been calculated using the deposition measured during steady-state breathing. The deposition increased from 9.4%, when there was no pause, to 12.6% when the pause between inhalation and exhalation was, on an average, 1.07 secs (Table 1). Using equation (2), the diameter of the passageway was calculated to be 0.74 mms. The diameter of the passageway calculated in the case of steady-state breathing works out to be more than that calculated in the case of the single-breath experiments.

Table-3

Average diameters of the tidal (D_{TV}) and the expiratory reserve volumes (D_{ERV}) in the lung, calculated using equation (2) and table (2)

t (secs)	TV (cm ³)	D_{TV} (mms)	ERV (cm ³)	D_{ERV} (mms)	(TV+ERV)	$D(TV+ERV)$ (mms)	$D_{TV}+D_{ERV}$
3.0	690	0.33	820	0.19	1510	0.51	0.52
5.7	665	0.34	941	0.19	1606	0.52	0.53
7.6	615	0.34	950	0.16	1565	0.50	0.50
15.3	585	0.35	1010	0.17	1595	0.46	0.52
24.0	635	0.38	1170	0.20	1805	0.48	0.58
Average	638	0.35	980	0.18	1618	0.50	0.53

DISCUSSION

Experiments using 0.5 μm particles, conducted by Palmes, Altshuler and Nelson⁴, showed that the passageway diameter varied from 0.3 to 0.4 mms. The results of similar experiments given in table 3 show that the diameter is about 0.5 mms. The diameter of the passageways calculated in the case of steady-state breathing works out to be 0.74 mms (Table 1). The difference in the diameters obtained for these two cases indicate that the most of the aerosol particles lost from the tidal air reach the walls of the lung after passing through the expiratory reserve volume and the residual volume.

Let us consider that the shape of the reserve volume is same as that of residual volume, then the radius (0.09 mms) and the corresponding expiratory reserve volume (980 cm^3) are related by

$$0.09 \propto \sqrt[3]{980} \quad \dots(3)$$

and if the radius of the residual volume is denoted by r_{RV} and the residual volume is 2040 cm^3 which is the value for the subject under consideration, then we have,

$$r_{RV} \propto \sqrt[3]{2040} \quad \dots(4)$$

Dividing (4) by (3), we get

$$r_{RV} = 0.09 \times \sqrt[3]{2040/980}$$

Thus, $r_{RV} = 0.12$ mms

Therefore the diameter of the residual volume is 0.24 mms.

Now if we add the diameters of the tidal, reserve and residual volumes, we get 0.76 mms which is in close agreement with the diameter (0.74 mms) calculated for the steady-state breathing experiment.

The calculated diameter (0.74 mms) of the alveolar airways works out to be greater than the values given by weibel⁵ in his regular dichotomy model of the human lung at 3/4 maximum inflation. If the diameters of the airways are large, it means it takes a longer time for the particles to travel from the main stream towards the walls and if, meanwhile, the subject exhales out, the fraction deposited would be the lowest and in some cases, all the 0.5 μm particles inhaled would be exhaled out as in the case of single-breath experiments.⁶

The theoretical curves (figure 1) calculated by equation (2) do not vary much from the experimental ones, showing that the relationship given by Landahl³ can be used for obtaining the diameter of the airways in the human lung using 0.5 μm particles as tracers. Better agreement between the theory and experiments would perhaps be possible if a correction is made to account for the inertial movement caused during the breathing cycle in the region of functional residual volume. Another discrepancy pointed out by Palmes, Altshuler and Nelson⁴ is the relatively small contribution of the Brownian motion term to the calculated deposition. As has been pointed out by Landahl⁷ the difference becomes serious only for particles 0.1 μm or less.

An important implication of these studies is that the dose delivered to the tissues by breathing radioactive particles of different sizes reduces considerably if the airway diameter, especially in the alveolated region of

the lung, is larger. This would result in appreciable changes in the m.p.l. values to the advantage of the progress of nuclear industry. The method of measuring the diameter of the airways by breath-holding technique is also useful for diagnostic purposes. Persons suffering from constrictive diseases will have a higher deposition of particles in the lung. Also the breath-holding technique would show the extent of constriction when compared with the airway diameter of a normal lung.

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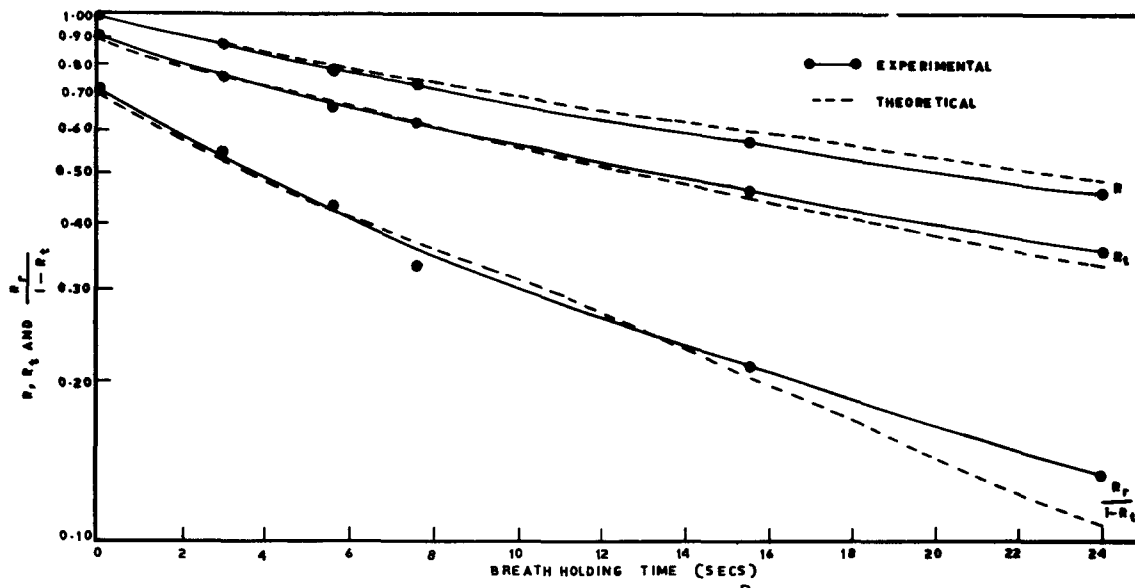


FIGURE:1. THE EFFECT OF BREATH HOLDING ON R, R_t AND $\frac{R_r}{1-R_t}$ (SINGLE BREATH EXPERIMENTS)

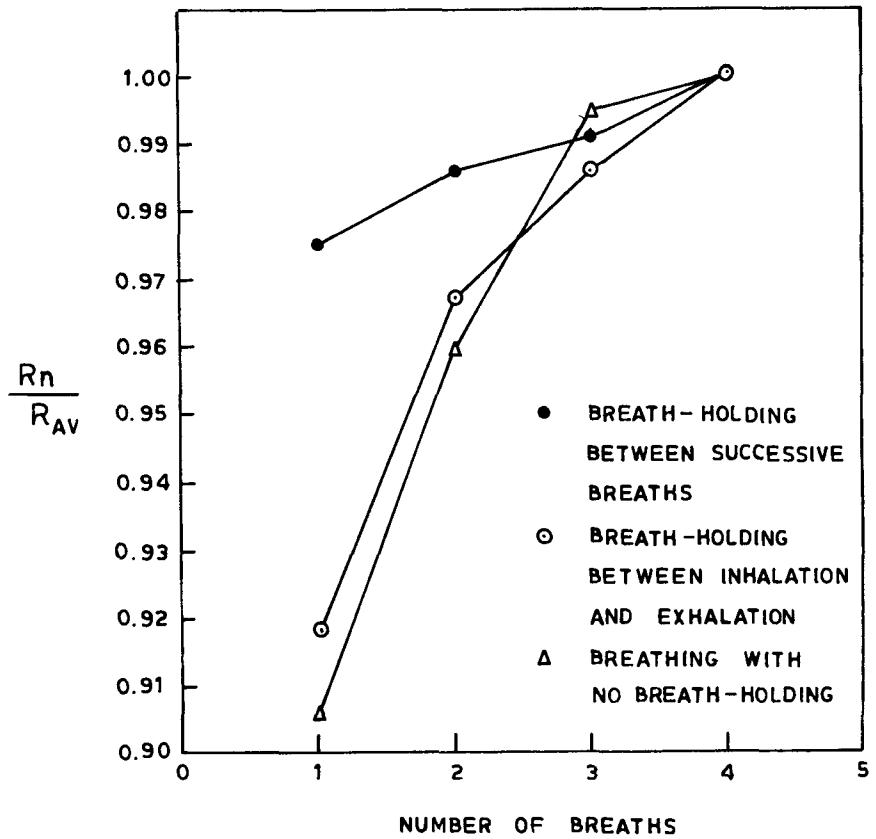


FIGURE:2. BUILD-UP OF AEROSOL PARTICLES IN THE LUNG DURING THE STEADY-STATE BREATHING.

HASL CYCLONE AS AN INSTRUMENT FOR MEASURING
AEROSOL PARAMETERS FOR NEW LUNG MODEL

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Abstract

Recently proposed ICRP Lung Model stipulates that a knowledge of an aerosol parameter, activity median aerodynamic diameter (AMAD) is essential to predict the fractional depositions in various parts of the respiratory tract. It is shown that this method of determining AMAD does not reflect significant error in the estimation of pulmonary deposition over a size range of 1.5 to 8.0 μm . Results of a large number of measurements made in Trombay Fuel Reprocessing Plant are reported. Results compare well with similar measurements made by other techniques such as Centripeter and Autoradiography. Significance of the measured AMAD is discussed with respect to maximum permissible concentrations (MPC) in air.

Introduction

Currently used MPC¹ values in air for radioactive nuclides assume that 25% of whatever inhaled goes to pulmonary region of the lung and 12½% undergoes long term retention for insoluble aerosols. This is assumed with no regard to the particle size distribution of the inhaled aerosol. In reality this assumption is not valid and hence ICRP constituted a Task Group² to evolve deposition and clearance models taking into account among other parameters, the particle size distribution of the inhaled aerosol. After considerable deliberation Task Group came to the conclusion that the deposition fractions in various parts of the respiratory tract can be predicted fairly accurately, if one knows a particle size parameter, activity median aerodynamic diameter (AMAD). Task Group further states that the spread in particle size distribution (the geometric standard deviation) does not have significant effect on the deposition fractions.

The popular instruments among others³ available for the measurement of AMAD are cascade impactor⁴ and cascade centripeter⁵, which have severe limitations with the loading and wall losses respectively. In the present work an approach of using the fraction penetrating the HASL cyclone⁶ at a particular flow rate as a measure of AMAD is presented.

Penetration Characteristics of HASL Cyclones

Health and Safety Laboratory of USAEC developed miniature cyclones⁶. These have penetration characteristics matching the so called Los Alamos Respirable Curves, when operated at a particular flow rate. Half inch cyclone⁷ is operated at 9 litres/min and one inch cyclones⁸ at 68 litres/min. Therefore penetration characteristics of these cyclones can be taken identical to Los Alamos Respirable Curve. Lynch⁹ fitted an analytical expression for

this penetration curve and worked out mass penetrations for different particle size distributions. He gives a table in which mass penetrations are listed for a given count median aerodynamic diameter and a given geometric standard deviation, assuming that the distribution is log-normal. We have taken this data and computed mass penetration fractions with respect to AMAD for various geometric standard deviations using Hatch and Choate equation¹⁰. The data thus calculated was fitted to a least squares line on a semilog graph paper. Fig.1 gives such lines for geometric standard deviations between 1.5 and 3.0. A master line given in the figure is a computer fitted least squares line taking into account the entire set of data points used for drawing various lines.

Moss and Ettinger¹¹ have plotted penetration curves on a linear graph sheet for slightly different penetration curves (ACGIH Criterion).

Mass Penetration Fractions and AMAD

If one measures mass penetration fractions through a cyclone at proper flow rates, one can find AMAD using the master line in Fig.1. As can be seen, the value of AMAD depends to some extent on the geometric standard deviation. Aerosol normally encountered in field conditions has a GSD between 1.5 and 3.0 and the situations giving rise to distributions outside these limits are rare (see Table-III). In the absence of the knowledge of GSD, lines corresponding to GSD of 1.5 and 3.0 enveloping the master line are taken to provide uncertainty in AMAD. For example, if penetration fraction is 0.70, AMAD is $2.3 \mu\text{m} + 0.12 \mu\text{m}$
 $- 0.30 \mu\text{m}$.

Pulmonary Deposition Fractions and AMAD

The object of measurement of AMAD is to predict the pulmonary deposition fractions. We can now examine whether the possible errors in cyclone measurement of AMAD have significant influence on the estimated pulmonary deposition fractions. ICRP Task Group² gives curve between AMAD and the pulmonary deposition fractions. A part of it is reproduced on the left half of Fig.1 for the size range of $1.5 \mu\text{m}$ and $10.0 \mu\text{m}$. Table-I gives for various AMADs, the errors involved in the estimation of pulmonary deposition fractions because GSD is not known. It is also indicated by error bands on the curve in Fig.1. It can be seen that the error is minimum in the size range of $4 \mu\text{m}$ (AMAD) and is less than 10% for the size range of 1.5 to $7.0 \mu\text{m}$. It is therefore concluded that for the size range of interest, the error in estimating the pulmonary deposition fraction is not significant.

Table-I

Errors Associated with the Estimation of Pulmonary Deposition by Cyclone Method for Various Particle Size (AMAD)

Particle size (AMAD) μm	1.5	2.0	3.0	4.0	5.0	6.0	7.0	8.0
% Pulmonary Deposition	19.7	17.6	14.9	13.0	11.4	10.3	9.2	8.2
with errors	+ 1.5 - 1.2	+ 1.3 - 0.6	+ 0.1 - 0.1	+ 0.5 - 0.6	+ 1.1 - 0.9	+ 1.2 - 1.5	+ 1.7 - 1.9	+ 2.0 - 2.1

Field Measurements of AMAD Using Cyclones

Measurements

Fig.2 shows experimental arrangement for cyclone air sampling. Flow rates were set at 9 litres/min for a half inch cyclone using a wet-test meter and also a soap bubble flow meter. Whatever penetrates the cyclone gets collected on an air sampling filter paper (Whatman GF/A glass fibre paper). Other sampling head collects a gross sample. The ratio of the activity on the filter paper following the cyclone and the filter paper from gross sample, provides fractional penetrations. Samples are generally taken over a period of 6 to 8 hours at a location close to breathing zone of workers. Cyclone is thoroughly cleaned before using for a subsequent measurements. Such determinations are made in different operating areas of Trombay Fuel Reprocessing Plant.

Discussion of Results

Number of measurements are made at each of the locations mentioned in Table-II. These measurements are grouped as shown in Table-II. Mean per cent penetrations and the corresponding AMAD are also listed in the table. It can be seen that AMAD does not stay constant from day to day or from one operation to the other. However a mean AMAD can be associated with each location. First three areas are the areas normally used for handling Plutonium and the other areas mostly fission products. It is of interest to compare our results with the AMAD reported in the literature. Table-III lists the range of values obtained by various investigators, using different techniques. Our values compare well with the values shown in Table-III.

Table-II

Results of Measurements of AMAD by Cyclone Method Measurements in Various Areas of Trombay Fuel Reprocessing Plant

Sampled Areas	Number of Measurements					Weighted mean per cent penetrations	AMAD (μm)
	10 to 20	21 to 30	31 to 40	41 to 60	61 to 80		
Crane Space	12	3	5	1	-	22.9	7.5
Pu-Lab	4	4	3	2	-	28.1	6.8
Pu-Lab Maintenance	1	4	1	6	-	37.5	5.2
Operating Gallery	-	1	5	6	7	52.1	3.6
Control Lab	3	3	2	10	2	41.5	4.7
Service Corridor	1	-	2	3	1	43.6	4.4

Note: First three areas give aerosol data for Pu and the subsequent four for fission products.

Revision of MPC_a Values

In light of the actual field measurements described above, we can examine the currently used MPC_a values. Currently used MPC_a values assume that 12½% of the inhaled undergoes long term retention in the pulmonary region for relatively insoluble isotopes. According to the proposed Lung Model, this percentage depends upon AMAD. Table-IV gives the per cent undergoing long term retention with respect to AMAD. It is seen that we are under-estimating the hazard if AMAD is greater than 1.5 μm . Therefore

for Fuel Reprocessing Plant, we are over-estimating the hazard by a factor of 2.0 in using the current MPC_a values for insoluble aerosols.

Table-III
AMAD Measurements Available in Literature

Author	AMAD (μm)	GSD	Comments
Andersen ¹²	3.4 to 7.2	2.0 to 2.3	Autoradiographic method. Pu aerosols covering different operations.
Sundararajan ¹³	2 to 6	1.7 to 2.3	-do-
Stevens ¹⁴	3.5 to 6.0	2.0 to 3.4	Centripeter technique for Pu-aerosols.
	4.7 to 7.0	2.5 to 3.4	Centripeter technique for fission product aerosols.
Langmead ¹⁵	4.3	1.9	UF ₄ aerosol. Centripeter technique.
	5.33 \pm 1.39	2.46 \pm 0.62	Pu-aerosols. Centripeter technique.
	2.5 to 11.0	-	-do- (Windscale)
	5 to 6.0	-	-do- (Springfield)

Table-IV
Ratios of MPC_a (for Insoluble Aerosols) for Old and New Lung Models

Particle size, AMAD (μm)	1.0	1.5	2.0	3.0	4.0	5.0	6.0	7.0	8.0
% long term pulmonary retention	14.4	11.8	10.6	9.0	7.8	6.9	6.2	5.5	4.9
$\frac{(MPC)_a \text{ (Old model)}}{(MPC)_a \text{ (New model)}}$	1.15	0.94	0.85	0.72	0.62	0.55	0.50	0.41	0.39

Conclusion

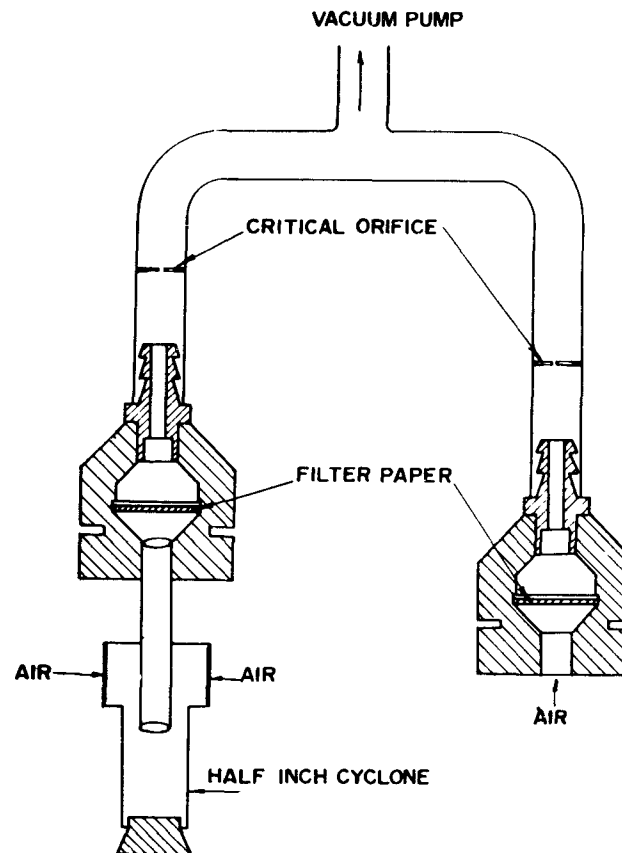
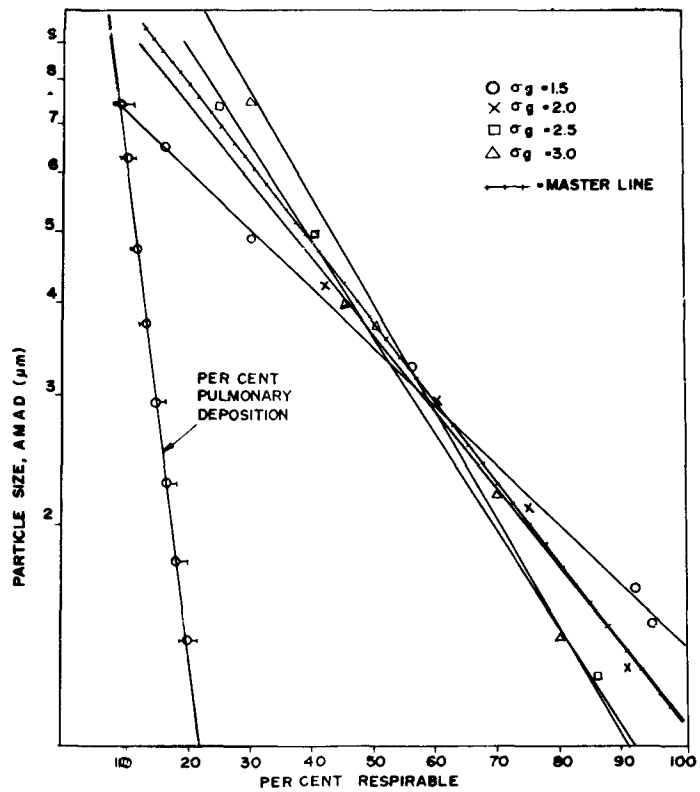
Cyclone method of measuring AMAD provides an acceptable method as long as we intend to use the information for estimating inhalation hazards. The method does not provide geometric standard deviation. The basic assumption used in this method, viz. that the penetration characteristics are identical to the Los Alamos Respirable Curve, should be borne in mind. Proper flow rates must be employed. The method is simple and operational health physicists can adopt this method without needing additional skills. The measurement incidentally provides respirable fractions. Recent trend in defining TLV⁷ (Threshold Limit values) for Silica and such other particulates, is to define TLV in terms of the respirable fractions. The method has a much wider applications.

Acknowledgements

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CYCLONE SAMPLING ASSEMBLY

DOSE COMMITMENT TO THE LUNG FROM INHALATION
OF RADIOCOBALT IN POWER REACTOR OPERATIONS

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Abstract

Personnel working in the operational and maintenance areas of Tarapur Atomic Power Plant (BWR Type) have shown detectable amounts of ^{60}Co in their system, notably in the lung area. Large number of gross and respirable air samples taken in the working areas provided the air activity levels of ^{60}Co and also the aerosol parameter, activity median aerodynamic diameter (AMAD). These data in conjunction with the long term clearance half-life available in literature are used to compute the quantity of ^{60}Co in lungs at any time due to chronic inhalation. Parameters of the new ICRP lung model are used in the computation. Radiation dose commitment to the lung due to the retained ^{60}Co is computed.

Introduction

Personnel working in Tarapur Atomic Power Plant, a 400 MWe BWR Type Power Reactor, have shown detectable amount of ^{60}Co in their system, notably in chest area^{1,2}. Air samples collected in the working environment also indicated that the major portion of the long lived component is due to ^{60}Co . Chest clearance studies have shown that ^{60}Co behaves like a class Y radionuclide as defined by the ICRP Task Group on Lung Dynamics³. Airborne concentration and the particle size parameter, activity median aerodynamic diameter (AMAD) provide the data to calculate both the material and dose commitments to the lung⁴.

Airborne Radiocobalt

The gamma spectra of air samples collected shows predominantly ^{60}Co , ^{54}Mn and ^{137}Cs . Nearly 80 to 90% of the entire airborne activity is found to be due to ^{60}Co coming in air, probably as insoluble cobalt oxide aerosol. Origin of this activation product is traced to the material of the piston (stellite) used in the water circulating pumps. It has been reported¹ that during July 1970-July 1971, 182 cases were detected with chest burdens less than 10 nCi and 8 cases with chest burdens more than 10 nCi within a year of the operation of the TAPP.

Clearance of ^{60}Co from lungs has been studied by several investigators. Most of these studies relate to accidental inhalation of ^{60}Co oxide and a few relate to the occupational exposure in reactors and hot cell areas. Table-I gives the results of these studies. It is seen that effective clearance half-life varies considerably. 500 to 1000 days may be taken as long term effective clearance half-life. It is typically a class Y radionuclide as per the classification of the ICRP Task Group on Lung Dynamics³. Using this data in conjunction with the rate of inhalation and the particle size

parameter (AMAD), it is possible to calculate the lung burden and dose commitment to the lungs making use of the equations given by Kotrappa⁴.

Table-I

Long Term Clearance Half-Life for ⁶⁰Co from Chest Area

Reference	Effective Half-life (days)
Sill ² et al, (1964)	172 to 700
Rundo ⁶ et al, (1966)	90*; 720 **
ICRP ⁷ -10 (1968)	115 - 400 40 -1000
Edvardsson ⁸ (1970)	180
Newton and Rundo ⁹ (1971)	935; 1460; 1210; 400; 750
Gupton and Brown ¹⁰ (1972)	225

*Single exposure

** Multiple exposure

Measurement of Airborne Concentration and AMAD

Technique of measuring the particle size parameter, AMAD, using a cyclone has been described in greater detail by Kotrappa⁵ et al. In brief, it consists of measuring the respirable fraction of the activity which simply is the fraction of the activity penetrating the cyclone at a proper flow rate. Making use of a graph between the fractional penetration and AMAD, corresponding AMAD can be read out. Sampling arrangement consists of taking two simultaneous air samples in close proximity to each other. One sample is normal air sample through a filter paper and another is through a filter paper following the cyclone. Ratio of the activities of the latter to the former provides respirable fraction or the fraction penetrating the cyclone at a proper flow rate. Samples are counted in 512 channel gamma spectrometer, and the ratio of the peak counts are used for calculating the respirable fractions. Absolute activities of ⁶⁰Co on these papers are also determined by standard calibration techniques.

Results of Measurements

Table-II gives a summary of the measurements carried out in different areas of TAPP. Rad waste conveyor area and Rad waste air receiver area are normally restricted. These areas show high airborne concentration, at times more than the presently adopted maximum permissible concentration of 10000 pCi/m³. Areas at 103 and 125 feet are the operating areas from where the operators control the rod movements. 200 feet level area is generally an operating area. Fuel pond is located here and general maintenance is carried out in this area. High airborne activities are observed only during special maintenance works. AMAD of 4 μm can be taken to represent an average value for all the areas of TAPP.

Computation of Lung Commitments

Method of calculating the burden and dose to different respiratory compartments from continuous inhalation of a radioactive aerosol, making use of the proposed ICRP lung model has been described in detail elsewhere⁴. Fig.1 gives the computed burdens in pulmonary region and in lymph node

regions after chronic inhalation. These are general curves for the inhalation rate of 1 pCi/day, for an aerosol of 1 μm (AMAD). To obtain results for ^{60}Co for an aerosol of 4 μm (AMAD), retention should be multiplied by 0.5. Fig.2 gives the integrated dose commitments. Again these are general curves. To obtain results for ^{60}Co one has to multiply the doses by 0.36 (0.5, correction for AMAD of 4 μm and 0.72, correction for effective energy). A few examples of such computations for two different effective half-lives are given in Table-III.

Table-II
Activity and particle size data in various areas of TAPP

Area	No. of samples	RANGE OF VALUES			Median AMAD (μm)	σ_g	
		^{60}Co (pCi/m ³)		Per cent respirable			
		Total	Respirable		AMAD (μm)		
Rad Waste	10	575-175463	245-84388	37-69	2.3-5.2	3.9	1.23
Air receiver room							
Rad Waste conveyor area	10	344-188826	132-59299	11-74	2.05-10.0	4.7	1.74
103 & 125 ft elevations including interior of Dry well	8	25-3062	19-908	30-76	2.0-6.2	3.8	1.42
200 ft. elevation	14	33-3921	23-1371	33-93	1.3-5.8	3.7	1.43

Table-III
Cumulative Retention and Dose for Continuous Inhalation of 4 μm (AMAD) Aerosols at the rate of 1 pCi/day.

Inhalation Time (days)	Retention in Pulmonary region (pCi)	Retention in Lymph region (pCi)	Dose to Pulmonary region (mrems)	Dose to Lymph Region (mrems)
<u>Effective Half-life = 500 days</u>				
100	7.1	0.13	0.03	0.01
500	27.1	2.57	0.55	1.10
1000	40.7	8.30	1.73	7.55
5000	54.0	73.5	17.0	400.0
10000	54.0	158.0	24.8	1820.0
<u>Effective Half-life = 1000 days</u>				
100	7.3	0.064	0.03	0.005
500	31.8	1.44	0.62	0.610
1000	54.0	5.2	2.24	4.45
5000	105.0	63.0	28.6	322.0
10000	108.0	148.0	66.0	1610.0

Computation for Typical Inhalation Case(Pulmonary Region)

It is seen that the concentration of ^{60}Co is of the order of 200 pCi/m^3 in continuously occupied operating areas such as 200 feet level. During a working day a standard man inhales 10 m^3 and therefore daily intake is about 2000 pCi/day . From Table-III, the retention after 500 days in pulmonary region will be $(27.1 \text{ pCi} \times 2000)$ of the order of 54 nCi . Saturation burden (after 5000 days) will be of the order of 108 nCi . Therefore it is not surprising to find lung burdens of the order of 50 nCi , if they work for 500 days in the atmosphere that has airborne concentration of $200 \text{ pCi}/\text{m}^3$. Corresponding dose to pulmonary region will be 1.10 rem in 500 days. At saturation (that is after 5000 days), there will be a steady dose rate of $\int (24.8-17.0) \times 2000/5000 \int 3.1 \text{ mrem}/\text{day}$, which is about 6% of the daily allowed dose of 50 mrem/day for lungs. Probably 10% of the allowed exposure to a person should be kept aside to take care of the internal exposure due to ^{60}Co even at saturation.

Computation for Lymph Node Region

Calculations can be made for lymph node regions on the same line as is done for pulmonary region. Currently used lung model³ assumes that 70% of the material reaching the lymph node region is retained permanently and only 10% undergoes biological elimination. Recently Morgan¹¹ has stated that experimental data supports the view that 90% undergoes biological elimination with a 1000 days half-life and only 10% undergoes permanent retention. Further Morgan states that for calculating the doses to lymph nodes, controversy still exists whether one should use the mass of pulmonary lymph nodes or whether one should use the entire mass of circulating lymphocytes. He states again that lymph node tissue is known to be highly radiation resistant and the tissue is not likely to be a critical organ. Based on these facts, calculations made on the basis of the currently used lung model³ may not be right. Pulmonary exposures would be controlling the hazard. Therefore we have not attempted to calculate the results for typical inhalation case and to draw any conclusion regarding the dose commitments.

Conclusion

Detectable amount of ^{60}Co is expected in the lungs of the workers in operational areas of TAPP. Probably 10% of the annual allowed dose should be left as the dose commitments for these workers due to inhalation of ^{60}Co alone. Routine chest monitoring programme is essential. It is recommended that further investigation be done to see whether this problem can be eliminated once for all by stopping the source of cobalt.

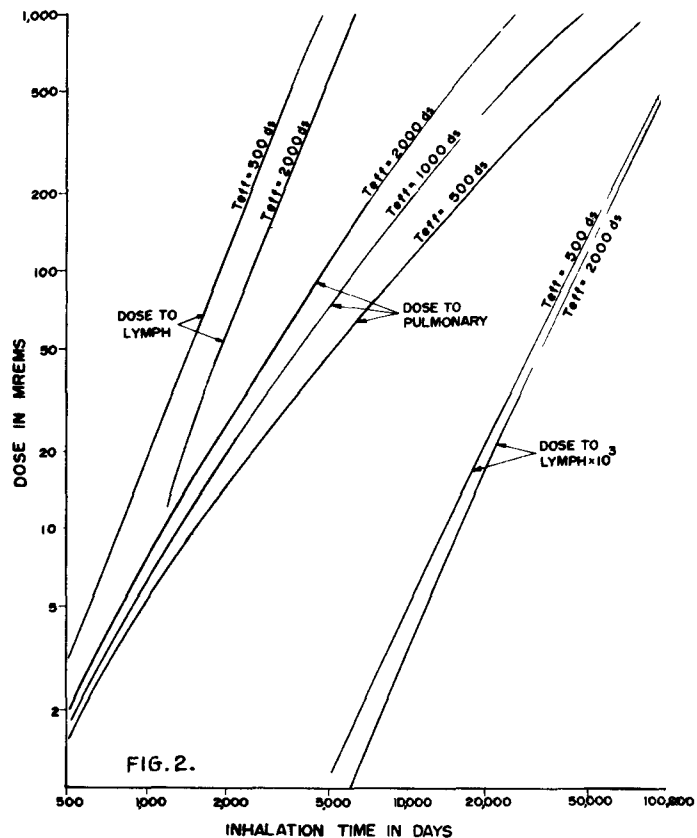
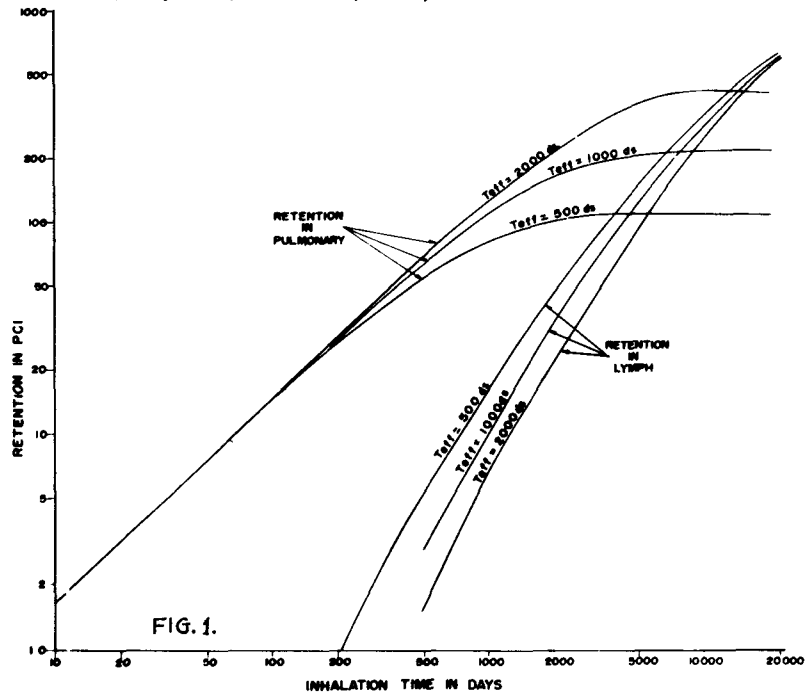
Acknowledgements

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A METHOD FOR DETERMINING THE DISSOLUTION CHARACTERISTICS OF ACCIDENTALLY RELEASED RADIOACTIVE AEROSOLS*

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Abstract

Early assessment of the dissolution characteristics of radioactive aerosol particles involved in inhalation accidents is important for hazard evaluation and selection of proper therapeutic procedures. Since accidentally produced aerosols are usually of undetermined physicochemical properties, prediction of their dissolution characteristics is difficult. Therefore, a rapid experimental evaluation of their dissolution behavior is desirable. An in vitro method is described which provides both early and long-term measurements of dissolution behavior of aerosol samples in a parallel-flow system which simulates in vivo dissolution as in the lung after inhalation deposition. Health physics applications of dissolution data are discussed.

Introduction

The biological hazards of an inhaled radioactive material depend upon the kinetics of deposition, retention, distribution and absorption of the inspired aerosol. The dissolution pattern of the aerosol particles is especially important in determining the retention and absorption from the site of deposition. Also, for estimations of lung burdens of radionuclides by urine analysis, in vivo dissolution rates of the lung-deposited aerosol particles are important.^{1,2} Further, the effectiveness of chelation therapy for inhaled actinide and lanthanide nuclides will depend on the degree of solubility of the aerosol.³

The in vivo dissolution behavior of an accidentally released aerosol may not be readily predicted because its physicochemical nature may be unknown and because the in vivo dissolution is a highly complex process.^{4,5} Due to particle size effect alone, rates of dissolution of inhaled radioactive aerosol particles may vary several orders of magnitude. Further, the chemical forms of the aerosol may greatly influence its rate of dissolution. Therefore, rapid experimental evaluation of the dissolution characteristics of radioactive aerosols involved in accidental inhalation is highly desirable. In vitro dissolution rates of aerosol particles comparable to in vivo rates of dissolution may be measured experimentally with flowing solvents.⁴ In this report a parallel-flow system is described which provides both early and long-term measurement of in vitro rates of dissolution of aerosol samples. The results of in vitro dissolution experiments on a variety of test aerosols, and health physics application of in vitro rates of dissolution are discussed.

Materials and Methods

After measurement of the radioactivity of samples of aerosol particles collected on filters, the filters were sandwiched between two 47 mm diameter 100 μm pore size cellulose acetate membrane filters and held together

* Research performed under AEC Contract AT(29-2)-1013.

tightly with plastic holders for elution with a flowing solvent, a simulant of biological fluid.⁴ The experimental arrangement with a cross sectional view of the filter holder is shown in Fig. 1. Two cavities were formed when the filter was placed in this holder. The solvent passed into the lower cavity (0.6 ml) and eluted any dissolved material diffusing into this region from the filter. Tests with colored compounds showed that the upper static layer of "solution" (0.2 ml) could be eluted out within a few hours. Thus, in this system the dissolution and removal of dissolved material from the aerosol particles occurred by the indirect contact of a flowing solvent through filters. One can envision a general analogy between this dissolution system and the dissolution of particles and transport of the dissolved material from the pulmonary region to the blood capillaries.

The solvent from a large reservoir (20 l) was introduced into the secondary reservoir (200 ml) at 1 ml/min by gravity flow and controlled with a valve. The secondary reservoir, the filter holder and the associated tubing, all non-corrosive plastics, were immersed in water bath at 37°C. The secondary reservoir allowed temperature equilibration and served as a bubble trap. The filter holder was placed in the bath at an angle to prevent reduction in solvent flow rate by gas bubbles. Aliquots of eluates on planchets were counted with a low-background proportional counter for beta emitters and alpha emitters were counted with a liquid scintillation counter using Aquasol. Low levels of plutonium were analyzed by alpha spectroscopy. Fractional dissolutions were calculated for each period of observation as the fraction of activity remaining on the filter dissolved during that period. After each dissolution, the activity remaining on the sample was also determined.

The solvent was prepared with the composition similar to that of blood serum by dissolving analytical grade chemicals in distilled water (Table 1). The basis for the choice of this composition for the solvent was discussed earlier.⁴ An antibacterial agent, 50 ppm of alkyl benzyl dimethyl ammonium chloride was also used in this solvent. In some studies a strong chelating agent, either ethylenediaminetetraacetic acid (EDTA) or diethylenetriaminepentaacetic acid (DTPA), was also added to the solvents. The pH of the solvent was kept at 7.3 to 7.4 by equilibrating with 5% CO₂ in N₂ contained in a weather balloon (Fig. 1). In this study all experiments were done with a solvent flow rate of 1 ml/min and temperature of 37°C.

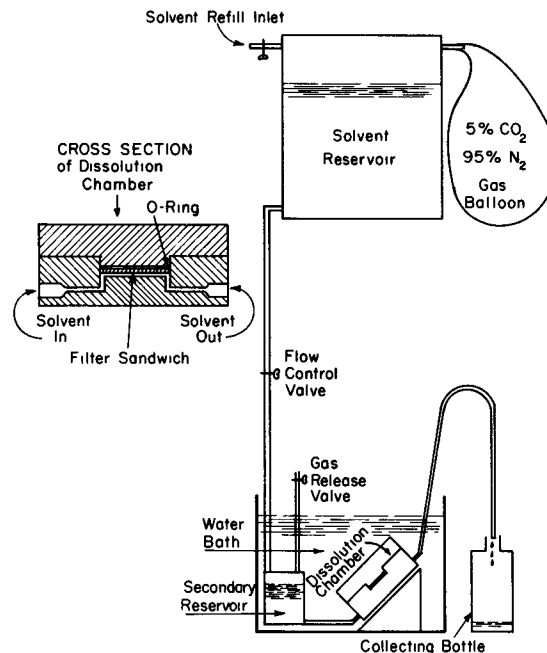


Figure 1. Schematic of parallel-flow dissolution system and filter holder assembly.

Table 1
Preparation of Serum Simulant for *In Vitro* Elutions

Chemical Compound	m moles/liter
NaCl	116
NH ₄ Cl	10
NaHCO ₃	27
NaH ₂ PO ₄	1.2
Na ₃ Citrate	0.2
Glycine	5.0
L-Cysteine	1.0
H ₂ SO ₄	0.5
CaCl ₂	0.2

The methods of production and characterization of polydisperse aerosols of ^{95}Zr - ^{95}Nb labeled zirconium oxalate, carbonate and dioxide and ^{90}Sr in fused montmorillonite clay have been described earlier.⁶ Samples of monodisperse particles of $^{238}\text{PuO}_2$ and $^{239}\text{PuO}_2$ were prepared under identical conditions with 1100°C heat treatment by the method described by Kotrappa et al.⁷ To demonstrate the usefulness of *in vitro* dissolution studies, a filter sample containing a ^{239}Pu aerosol accidentally released at another laboratory and involved in a human exposure was also used in this study.

Results and Discussion

In vitro rates of dissolution of ^{90}Sr fused clay and ^{95}Zr - ^{95}Nb labeled aerosol particles (zirconium oxalate, zirconium carbonate and zirconium dioxide) were found comparable to the corresponding dissolution in the lungs of animals.⁴ The dissolutions in the fractions collected during the first two minutes were 85%, 0.2% and 0.1%, respectively, for the oxalate, carbonate and dioxide aerosols. Both the dioxide and carbonate particles were comparatively insoluble; only 0.6% and 2%, respectively, dissolved during 150 minutes. The oxalate particles dissolved 93% during 100 minutes. The fractional dissolution of ^{90}Sr in fused clay particles during the 5th to 11th day of continuous elution was a relatively constant value of $(1.3 \pm 0.1 \text{ S. D.}) \times 10^{-3}$ per day.⁵ The *in vitro* rate constant of dissolution of ^{90}Sr -fused clay particles was about $3.3 \times 10^{-8} \text{ g. cm}^{-2} \cdot \text{day}^{-1}$.⁴ Good agreement between the *in vitro* rates of dissolution of monodisperse ^{137}Cs -fused clay particles in Beagle dog lungs, after inhalation deposition, has also been observed.⁸

The dissolution of monodisperse ($\sigma_g < 1.2$) PuO_2 aerosols of both $^{239}\text{PuO}_2$ and $^{238}\text{PuO}_2$ (80% ^{238}Pu + 20% ^{239}Pu) was studied for 3 to 4 weeks (Fig. 2). Serum simulant containing $2 \times 10^{-4} \text{ M EDTA}$ was used as the solvent for the study with $0.36 \mu\text{m}$ $^{238}\text{PuO}_2$ and during the later parts of the $^{239}\text{PuO}_2$ study. The PuO_2 aerosol particles (density, $\rho \approx 7 \text{ g. cm}^{-3}$) were relatively insoluble. The average dissolution rates of PuO_2 and other pertinent data are summarized in Table 2. If the rate of dissolution of a particle is proportional to its surface area, for spherical particles the rate constant $k = \rho Df/6$ when D is the diameter of the particle and f is the fractional dissolution per unit time.⁴ The dimensions of k are $\text{g. cm}^{-2} \cdot \text{day}^{-1}$ when the units of ρ , D and f are g. cm^{-3} , cm and fraction per day, respectively.

During the first day, about 3% of $^{238}\text{PuO}_2$ ($D = 0.36 \mu\text{m}$) was dissolved in serum simulant containing EDTA and higher dissolution of this sample,

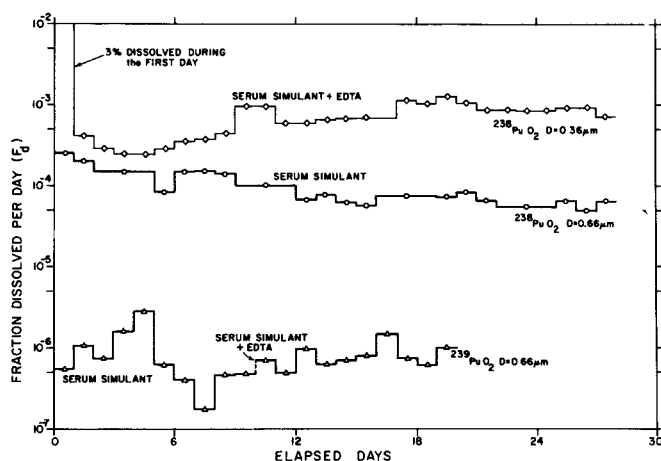


Figure 2. Dissolution data histogram for monodisperse particles of $^{238}\text{PuO}_2$ and $^{239}\text{PuO}_2$ as determined in a parallel-flow dissolution system.

Table 2
Summary of Dissolution of PuO₂ at 37°C

Sample Form	Original sample activity μCi	Physical diameter, D μm	Solvent	Span of study Days	Average fraction dissolved Per Day	Dissolution rate constant, k $\text{g. cm}^{-2} \cdot \text{day}^{-1}$
²³⁹ PuO ₂	6.8	0.66	Serum Simulant and Serum Simulant + EDTA	0 to 20	$(8.5 \pm 3.4) \times 10^{-7}$	$(6.5 \pm 2.6) \times 10^{-11}$
²³⁸ PuO ₂	4.6	0.66	Serum Simulant	0 to 28	$(9.7 \pm 5.2) \times 10^{-5}$	$(7.5 \pm 4.0) \times 10^{-9}$
²³⁸ PuO ₂	4.9	0.36	Serum Simulant + EDTA	1 to 28	$(7.1 \pm 3.0) \times 10^{-4}$	$(4.1 \pm 1.7) \times 10^{-8}$

compared to that of 0.66 μm ²³⁸PuO₂ particles in serum simulant, persisted. Increased dissolution of ²³⁸PuO₂ due to the presence of EDTA is indicated by the higher rate constant (Table 2). The presence of EDTA in the solvent did not increase the rate of dissolution of ²³⁹PuO₂ particles. It is clearly shown that the ²³⁸PuO₂ particles are much more soluble than ²³⁹PuO₂ particles.

Generally the *in vivo* dissolution of PuO₂ is considered to be extremely small.⁹⁻¹¹ Higher absorption of ²³⁸Pu compared to ²³⁹Pu from the lungs of Beagle dogs to the systemic burden, after inhalation deposition of ²³⁸PuO₂ and ²³⁹PuO₂, has also been reported.¹² Ineffectiveness of chelates to enhance ²³⁹Pu removal from ²³⁹PuO₂ particles has also been observed in DTPA chelation therapy treatments.^{3, 11} These *in vivo* observations are in qualitative agreement with the *in vitro* studies. Increased *in vitro* dissolution of ²³⁸PuO₂ in the presence of EDTA suggests the usefulness of chelation therapy in ²³⁸PuO₂ inhalation exposure.

Two dissolution experiments with an accidentally released ²³⁹Pu aerosol sample (97.5% ²³⁹Pu and 2.5% ²⁴¹Am), one with serum simulant (1.7 μCi) and another (1.2 μCi) with serum simulant containing 1×10^{-3} M DTPA were conducted. The dissolution experiment with serum simulant as the solvent consisted of 8 days of continuous elution and then elution with serum simulant containing 1×10^{-3} M DTPA for an additional four days. The early dissolution data obtained from these studies, during the first five hours, are shown in Fig. 3. The data obtained during the entire dissolution experiment are summarized in Fig. 4. Higher rates of dissolution in serum simulant containing DTPA, compared to that in serum simulant alone, were seen between 2 hours and up to about 5 days of elapsed time. When the sample eluted with serum simulant was subsequently eluted with serum simulant containing DTPA, increased rates of dissolution were observed. The total dissolutions were 29.0% in serum simulant containing DTPA during 7 days and 31.5% from the second sample during 12 days. The dissolution in serum simulant during the first 8 days was 17.9% and in serum simulant containing DTPA, during the next 4 days, 13.6%. About 30% of the sample was in a relatively soluble form and exhibited higher dissolution in the presence of DTPA.

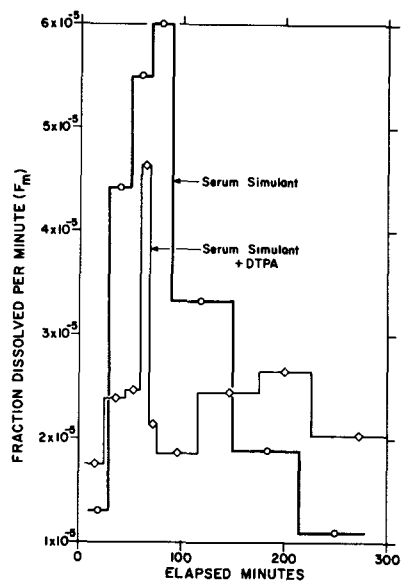


Figure 3. Early dissolution data histogram of an accidental ²³⁹Pu aerosol sample as determined in a parallel-flow dissolution system with serum simulant and serum simulant containing DTPA.

The usefulness of *in vitro* dissolution studies on accidentally released aerosols may be illustrated with the data presented. The aerosol was originally considered to be of insoluble $^{239}\text{PuO}_2$. An individual who inspired this aerosol had an estimated lung burden of about 450 nCi of ^{239}Pu and underwent treatments consisting of bronchopulmonary lavage and DTPA therapy beginning on the eighth day after exposure.^{13, 14} The urinary excretion on the seventh day was measured as 0.112 nCi. The *in vitro* dissolution data for the aerosol sample may be used to estimate the initial lung burden (after the first phase clearance), Q_0 , by Healy's method.¹ Urinary excretion, E_u , at the R th day after inhalation deposition is given as:¹

$$E_u = 0.002 \lambda_s Q_0 \int_0^R e^{-\lambda t} (R-t)^{-0.74} dt$$

with t the time in days after inhalation deposition, λ_s the daily fractional rate of dissolution and transfer to the blood (assumed constant by Healy), and λ , the total daily fractional removal rate from lower respiratory tract due to both dissolution, λ_s , and ciliary clearance, λ_c . The *in vitro* fractional dissolution of the aerosol sample in serum simulant varied from 5.2×10^{-2} to 4.5×10^{-3} per day (Fig. 4). However, the total dissolution during the first 7 days was 17.1% with an average fractional dissolution of 2.44×10^{-2} per day. This value may be used as λ_s in the Healy equation. The numerical values for the integral $\int_0^R e^{-\lambda t} (R-t)^{-0.74} dt$ when $R = 7$, are relatively insensitive to λ values; these values are 5.58 and 4.86 for λ of $2.44 \times 10^{-2}/\text{day}$ ($\lambda = \lambda_s$) and $5.0 \times 10^{-2}/\text{day}$ ($\lambda \approx 2\lambda_s$), respectively.² Also, after the rapid clearance phase, the ciliary clearance rate from the pulmonary region, is relatively slow, $< 8 \times 10^{-3}$ per day.¹⁵ If $\lambda = \lambda_s = 2.44 \times 10^{-2}/\text{day}$ then $Q_0 = 0.112 / 0.002 \times 2.44 \times 10^{-2} \times 5.58 = 411$ nCi. Similarly if $\lambda = 5.0 \times 10^{-2}$ then $Q_0 = 472$ nCi. These values are in general agreement with the estimate of Q_0 , 450 nCi obtained by whole body counting.¹³

Conclusions and Recommendations

When human exposure to an accidental aerosol is involved, an *in vitro* dissolution study should be done on a representative sample collected on a filter as soon as practicable. If size selective sampling is available only the respirable size particles should be used in the *in vitro* dissolution. Glass fiber filters for the collection of the aerosol sample should be avoided, because the elution of polyvalent nuclides from this type of filter is less than quantitative. For rapid solubility range finding, however, any filter sample containing a known activity of the accidental aerosol may be used. The method of choice for the determination of *in vitro* dissolution is continuous elution with the parallel flow system (Fig. 1) with serum simulant, 37°C , at a flow rate of 1 ml/min. Direct assaying of eluted radioactivity during the first hour of elution should indicate the range of solubility of the aerosol. If significant fractional dissolution is shown, DTPA treatment of exposed individuals may be useful. *In vitro* dissolution studies of the aerosol sample with DTPA added to the serum simulant may provide useful additional information on the efficacy of DTPA therapy. The rates of dissolution of a relatively insoluble aerosol

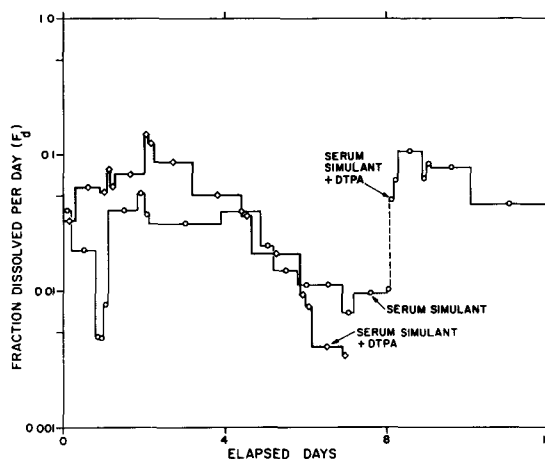


Figure 4. Dissolution data histogram of an accidental ^{239}Pu aerosol sample during several days as determined in a parallel-flow dissolution system. The solvents used are indicated in the figure.

should be determined for longer periods, several weeks, so that relatively constant fractional dissolution may be obtained. Such data then may be used for the evaluation of biological disposition of the inhaled material and for the estimation of lung burden from urine and blood radioactivity levels.¹

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BIOLOGICAL MODELING FOR PREDICTING RETENTION PATTERNS OF INHALED CONTAMINANTS*

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Abstract

Biomathematical models were developed for simulating the tissue distribution and clearance of inhaled Ba and Ce aerosols in Beagle dogs. Incorporating physical size characteristics of particles for predicting deposition and chemical solubility for estimating systemic absorption, application of these models was studied over a broad range of aerosol forms. The results illustrate the importance of determining these aerosol characteristics to aid in developing well founded predictions for its biological behavior. Procedures developed to date were more reliable in describing Ba metabolism and illustrate the more complex biochemical nature of Ce retention.

Introduction

Retention of inhaled particulate material after deposition in the respiratory tract is of vital interest to those concerned with radiation protection in nuclear industries and those concerned with other areas of industrial and environmental health. Inhaled particles are retained for minutes to hundreds of days depending upon physical size, chemical composition, solubility and respiratory patterns during inhalation. Inhalation exposures of humans to toxic materials may result from single acute, intermittent or chronic environmental contamination at relatively constant or highly variable air concentrations. All of these factors profoundly influence the doses received by the respiratory tract and other organs. Thus, some rapid accounting system which uses physical, chemical and biological factors is necessary for managing inhalation exposure data to provide time integrated tissue dosimetry information.

Dosimetry problems are simplified when whole body or tissue accumulations of the toxic materials can be measured with confidence. Whole-body counting and isotope distribution scanning after inhalations of radioactive materials with relatively high energy gamma or beta emissions are routine, however, difficulties arise when nonradioactive or very weak beta-gamma or alpha emitting isotopes are inhaled. Then, the only certain information which can be easily obtained relates to the air concentration, general chemical form of the aerosol and material recovered in excreta. All data can add to the evaluation of possible consequences of an exposure if a versatile framework is developed for relating information. One method is the use of mathematical models of the biological behavior of inhaled materials and their solution with readily available techniques. These models can be developed from experimental animal studies; however, since aerosols inhaled in accidents are likely to differ chemically and physically, simple adjustments must be provided.

Models of radionuclide metabolism have been studied in Beagle dogs to simulate inhaled radioactivity cleared from the respiratory tract to other organs as first order compartmental exchanges. The initial conditions include

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aerosol particle size, air concentration, respiratory rates and an estimation of the solubility of the aerosol. Applying the models to human exposures requires adjusting transfer rates for species differences, variations in chemical form and deposition pattern of the particular exposure. Adjustments can be made during early stages of an evaluation and refined as data become available. In this way, a unified picture of an exposure can be developed and long-term evaluation facilitated especially with regard to prospective therapy.

Biomathematical Model

A biomathematical model used in analyses of inhalation studies of ^{140}Ba , ^{140}La and ^{144}Ce in Beagle dogs is in Fig. 1. The respiratory tract is divided into the nasopharynx, tracheobronchial tree and pulmonary regions as defined by the Task Group on Lung Dynamics.¹ Aerosol deposition in these compartments depends on its physical characteristics. These regions as well as the stomach, small intestine and tracheobronchial lymph nodes are divided into two or more parallel compartments leading into a compartment in series. The parallel compartments represent inhaled material in a particular state which may be converted by dissolution or other change in chemical form and pass into a transformed state, represented by the series compartment, from which absorption into the circulation may occur. The number of parallel compartments in these organs corresponds to the number of first-order exponential functions required to represent the dissolution mathematically. The rate constants of the exponential functions were used for the corresponding compartmental transfer rates, and coefficients of the exponential functions were used to apportion the material deposited in each region among the parallel compartments. Mechanical clearance of deposited material is shown by solid arrows in Fig. 1 to avoid confusion and represents transfers between corresponding subunits of each organ configuration. All internal organ exchanges of ionic or molecular forms with blood and excretory pathways, shown as individual arrows were used with a constant fraction of the compartmental content transferring per unit time interval. Transfer of material from blood to small intestine indicates a possible association with liver and biliary secretion, however, sufficient information on this is not available.² Analog and digital computer programs were developed for simulation of the model.

Retention of Inhaled Barium Aerosols

In previously reported experiments, Beagle dogs inhaled differing chemical forms of barium: $^{133}\text{BaCl}_2$, $^{133}\text{BaSO}_4$, heat treated $^{133}\text{BaSO}_4$ and ^{133}Ba in fused clay.³ Respiratory tract retention of ^{133}Ba was measured for 16 days and then the dogs were sacrificed for tissue analyses. An additional group of dogs inhaled aerosols of ^{133}Ba in fused clay and was studied for 512 days. Samples of the aerosols from the exposures were enclosed between 25 μm cellulose acetate membrane filters and dissolved in a simulated serum solvent.⁴ The aerosol samples were placed in 100 ml of solvent which was refreshed periodically to measure filter retention of ^{133}Ba as

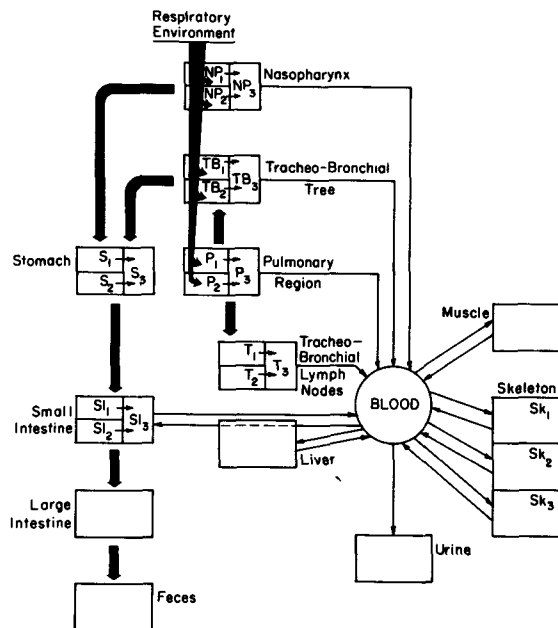


Figure 1. Biomathematical model for describing the retention of inhaled aerosols. Intercompartmental transfers of material as indicated by arrows were represented by first order rate constants.

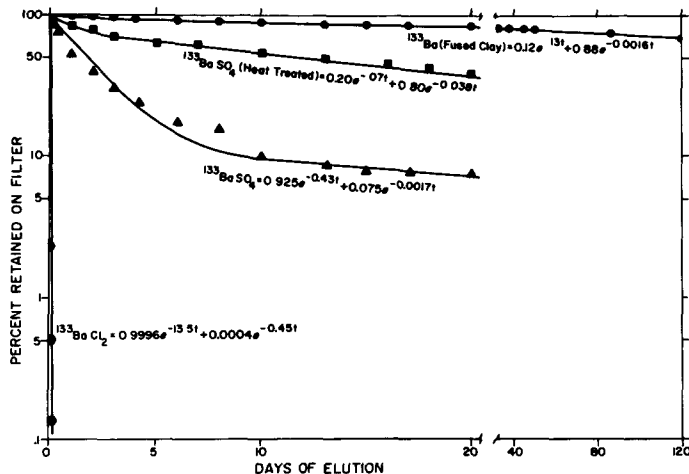


Figure 2. Retention of ^{133}Ba on aerosol filter samples subjected to dissolution in static contact with serum simulant.

shown in Fig. 2. Retention half-lives ranged from 1.2 hr for $^{133}\text{BaCl}_2$ to more than 430 days for ^{133}Ba in fused clay. Respiratory tract clearance and skeletal accumulation of ^{133}Ba following inhalation of $^{133}\text{BaSO}_4$ is shown in Fig. 3. Solid lines are the predictions of the model using transfer constants in Table 1 and initial deposition fractions in Table 2. Comparisons of the observed and predicted total body, lung, skeletal and tracheobronchial lymph node activities at sacrifice are also in Table 2 for all aerosols of ^{133}Ba . In general, the predicted tissue activities at sacrifice were sufficiently similar to observed activities to be useful in dosimetric projections for barium aerosols.

Retention of Inhaled Cerium Aerosols

The relationship between *in vivo* and *in vitro* solubility was also studied with aerosols of cerium: $^{144}\text{CeCl}_3$, $^{144}\text{CeCl}_3$ in CsCl ($\text{Ce}:\text{Cs} \approx 1:10$ by mass) and ^{144}Ce in fused montmorillonite clay. Particle sizes were about $2 \mu\text{m}$ AMAD for all aerosols. Dissolution was studied in different solvents including 0.001 M citric acid (pH 3.3), 0.001 M acetic acid (pH 3.9), saline (pH 6.9) and serum simulant (pH 7.3 to 7.9) under static solvent conditions as described previously. A parallel flowing solvent system⁴ was also used with serum simulant at pH 7.3 stabilized in an atmosphere of 5% CO_2 and 95% N_2 . All dissolution studies were conducted at 37°C . Typical retention patterns of ^{144}Ce on the filter samples are in Fig. 4 along with the respiratory tract retention of ^{144}Ce in Beagle dogs after inhalation of $^{144}\text{CeCl}_3$ in CsCl with $\text{Ce}:\text{Cs} \approx 1:10$ by mass.⁵ Dissolution of Ce aerosols was greatly influenced by solvent composition. Filter retention after 4 days ranged between 0.5% in citric acid and 80 to 99% in serum simulant. The higher pH and presence of phosphate, sulfate and carbonate species in serum simulant were likely responsible for slower aerosol dissolution. Clearance of $^{144}\text{CeCl}_3$ from the respiratory tracts of Beagle dogs had an intermediate

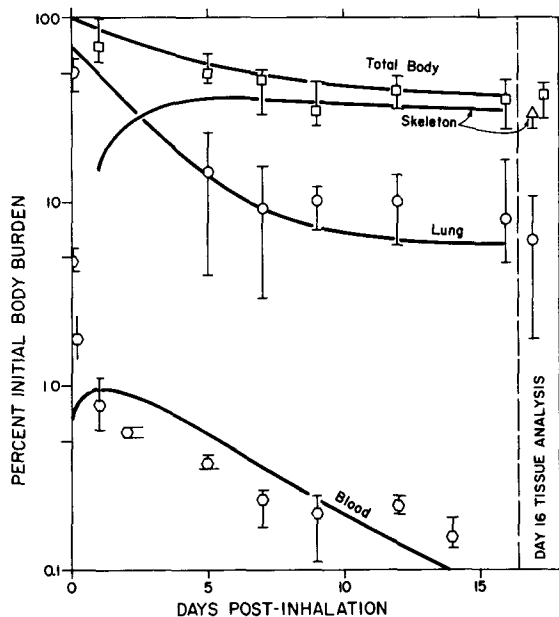


Figure 3. Retention and tissue distribution of ^{133}Ba following inhalation of $^{133}\text{BaSO}_4$ by Beagle dogs.

Table 1
 First Order Kinetic Constants Applied to Model Shown in Fig. 1
 for the Description of Ba and Ce Retention Following Inhalation

Origin	Compartment Of Destination	Transfer Rates (fraction/day)	
		Ba	Ce
Nasopharynx	Stomach	25	25
Nasopharynx*	Blood	100	100
Trachea and Bronch.	Stomach	0.9	0.9
Trachea and Bronch.*	Blood	100	100
Pulmonary	Stomach	≈ 0	.0009
Pulmonary*	Blood	100	100
Stomach	Sm. Intestine	15	15
Sm. Intestine	Lg. Intestine	5	5
Lg. Intestine	Feces	0.7	0.7
Pulmonary	TB Lymph N.	0.0001	0.0001
TB Lymph N.*	Blood	100	100
Sm. Intestine	Blood	0.35	≈ 0
Blood	Sm. Intestine	4	8.5
Blood	Urine	2.5	7
Blood	Liver	1.3	11.5
Liver	Blood	3.2	≈ 0
Blood	Muscle	40	≈ 0
Muscle	Blood	10	≈ 0
Blood	Skeleton (Sk ₁)	16.5	9
	(Sk ₂)	2.4	---
	(Sk ₃)	2.4	---
Skeleton (Sk ₁)	Blood	0.65	≈ 0
(Sk ₂)		0.014	---
(Sk ₃)		0.0007	---

Origin	Destination	Aerosol	Transfer Rates
NP ₁	NP ₃	BaCl ₂	13.5
TB ₁	TB ₃	BaSO ₄	0.43
P ₁	P ₃	BaSO ₄ (heat treated)	0.70
S ₁	S ₃	Ba-fused clay	0.13
SI ₁	SI ₃	Ce-fused clay	0.34
T ₁	T ₃		
NP ₂	NP ₃	BaCl ₂	0.45
TB ₂	TB ₃	BaSO ₄	0.0017
P ₂	P ₃	BaSO ₄ (heat treated)	0.0038
S ₂	S ₃	Ba-fused clay	0.0016
SI ₂	SI ₃	Ce-fused clay	0.0005
T ₂	T ₃		
P ₁	P ₄ **	CeCl ₃ in CsCl	0.65
P ₂	P ₄		0.018
P ₃	P ₄		0.001

* Represents transfer from dissolved or transformed state to blood only.

** Three parallel compartments were used for CeCl₃ in CsCl and the same rate constants were used for NP, TB, S, SI and T compartment configurations.

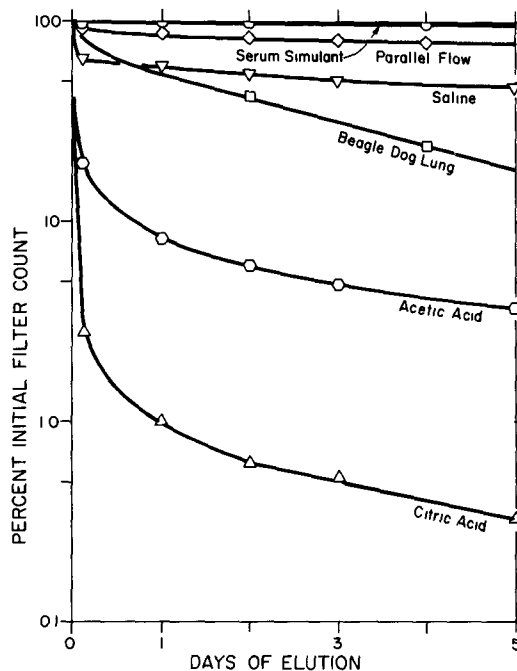
Table 2

Physical Characteristics and Deposition Fractions of Aerosols Used in Inhalation Studies with Beagle Dog Tissues Activities at Time of Sacrifice

Aerosol	BaCl ₂	BaSO ₄	Heated BaSO ₄	Ba in Fused Clay	CeCl ₃ in CsCl	Ce in Fused Clay
AMAD	2.3	1.0	0.9	2.2	2.2	1.4 - 2.7
<u>Regional Deposition as Fractions of Total Body Deposition</u>						
NP ₁	.45	.14	.03	.05	.05	44
NP ₂	0	.01	.12	.40	.40	4
NP ₃	---	---	---	---	---	2
TB ₁	.10	.09	.02	.01	.01	8.8
TB ₂	0	.01	.08	.09	.09	.8
TB ₃	---	---	---	---	---	.4
P ₁	.45	.69	.15	.05	.05	35
P ₂	0	.06	.60	.40	.40	3
P ₃	---	---	---	---	---	2
<u>Tissue Activities and Fractions of Initial Body Burdens</u>						
Sacrifice Time (days)	16	16	16	16	512	
<u>Observed</u>						
Lungs	---	.06	.20	.37	.19	See Fig. 5 for sacrifice times and tissue activities
Skeleton	.45	.30	.30	.008	.036	
TBLN	---	---	---	---	.01	
Total Body	.45	.36	.50	.38	.24	
<u>Predicted</u>						
Lungs	---	.06	.32	.40	.16	
Skeleton	.30	.31	.20	.025	.06	
TBLN	---	---	---	---	.009	
Total Body	.30	.37	.52	.43	.23	

Activity Median Aerodynamic Diameter

Figure 4. ¹⁴⁴Ce retention in ¹⁴⁴CeCl₃ aerosol samples in static contact with various solvents. Data for filter retention of ¹⁴⁴Ce in a system with parallel solvent flow and respiratory tract retention of inhaled ¹⁴⁴Ce by Beagle dogs were studied with aerosols of ¹⁴⁴CeCl₃ in CsCl.



clearance rate, however, the exact relationship of the aerosols used for the in vitro and in vivo studies is not known.

The biomathematical model was used to simulate data from the studies of the retention of inhaled $^{144}\text{CeCl}_3$ and ^{144}Ce in fused clay in Beagle dogs using transfer rates in Table 1. A comparison is in Fig. 5. A reasonable fit was obtained, however, dissolution of chloride aerosols in the respiratory tract as used in these simulations was considerably more than observed in vitro except with dilute solutions of citric or acetic acid.

Dissolution of ^{144}Ce in fused clay aerosols had a half-life of 450 days in saline during the first 25 days and a half-life of 200 days in serum simulant. Both of these dissolution rates include more rapidly soluble fractions of the aerosol samples normally observed at early times. Dissolution rates would probably have declined after longer times, however, the half-lives for clearance from the respiratory tracts of dogs were 1440 days for 90% and 2 days for 10% of the deposited aerosol.

Summary

In general, mathematical models incorporating in vitro measurements of aerosol solubility facilitate complex analyses of inhalation exposures, however, exact correspondence between dissolution rates of specific aerosols in vivo and in vitro should not be accepted without other supporting evidence. Such methods are likely to aid in distinguishing very soluble from moderately and insoluble aerosols with further study in the selection of solvents with aerosols for trivalent and tetravalent elements. Studies with serum simulant solvent predicted very long respiratory tract retention of $^{144}\text{CeCl}_3$ whereas elution of ^{144}Ce in dilute solutions of acetic and citric acid or saline more closely resembled the clearance of ^{144}Ce in Beagle dogs. This discrepancy was not observed with inhaled aerosols of barium, perhaps due to the more soluble nature of barium in most aqueous solutions.

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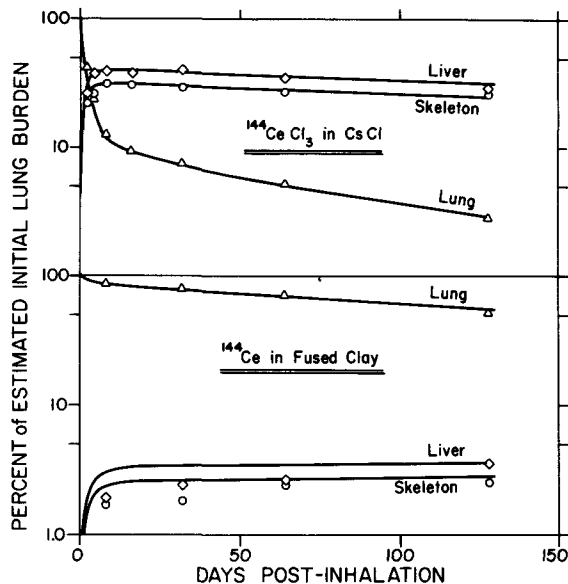


Figure 5. Tissue retention of ^{144}Ce in Beagle dogs after inhalation of aerosols of $^{144}\text{CeCl}_3$ and ^{144}Ce in fused clay.

DEPOSITION AND ELIMINATION OF IRON OXIDE
AEROSOL FROM THE LUNG OF RATS:
COMPARISON WITH ICRP PREDICTIONS FOR MAN

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Abstract

Male albino rats have been exposed to iron oxide aerosol (MAD 0.3 μm) at iron concentrations up to 700 mg/m^3 for continuous periods up to 235 min, and for intermediate periods of 30 min over a ten day period. From 0.3-3.0 mg of deposited iron were found in the rat lungs removed one day post exposure which corresponds to initial pulmonary fractional depositions of between 0.3 and 0.7 - which is comparable to that proposed for man by the ICRP Lung Dynamics Task Group (0.35). The elimination of deposited iron from the rat lung can be described by a two component exponential expression with parameters similar to those of ICRP for man except that the phase two clearance in rats was found to be shorter ($T_{1/2} = 33\text{d}$).

Introduction

The ICRP Lung Dynamics Task Group derived curves for man relating the aerodynamic diameter of particulates to the fractional deposition factor for the nasal, tracheo bronchial and pulmonary regions¹. Opportunities for testing these curves with human subjects are restricted due to the practical difficulties involved in long term exposures to high concentrations of aerosol, and the various analytical limitations. Nevertheless some useful data has been gained following relatively short term exposures using radio-tracer purposefully administered or resulting from unplanned exposure². Rats have been used in this present study, which could be exposed for up to five hours in a controlled fashion to relatively high concentrations of a series of metal oxides of interest.

The research programme is directed to the assessment of the hazard to health of industrial workers exposed to metallic fumes having a median aerodynamic diameter (MAD) below 1 μm . Metallic particulates of this size range are generated in a number of common industrial processes, such as welding and metal grinding, and are believed to be toxicologically significant³. The ICRP deposition curve for man indicates that about a quarter of airborne particulates of 1 μm MAD will be initially deposited in the pulmonary region (fractional deposition ~ 0.25) decreasing to a fractional deposition of ~ 0.1 at 5 μm MAD, as the larger particulates are deposited in the nasal and tracheo bronchial region. Below 1 μm diameter the pulmonary curve shows a steep rise so that at 0.3 μm MAD for example fractional deposition of 0.35 are predicted. Experimental verification of this portion of the curve is clearly of considerable interest in the assessment of the health hazard of metallic fumes. Of equal importance are investigations into the clearance kinetics of particulates initially deposited in the pulmonary region.

Exposure to Iron Oxide Particulate

Male albino rats were exposed to iron (MAD 0.3 μm geometric standard deviation 1.8) in a specially constructed chamber using experimental techniques described elsewhere⁴. The concentration of fume throughout each exposure period was determined by collection of the total particulate flowing through the chamber and measuring the air volume throughput. Electron microscopy was used for the particle size measurement.

Both continuous and intermittent exposures were used (Table 1). Rats were killed one day post each exposure and also at several predetermined periods up to 100 d post exposure in order to follow the elimination of deposited iron from the lung. Control rats were housed alongside and then killed at the same time as those exposed; the total lung being then removed and iron was determined by instrumental (Ge/Li) neutron activation analysis. A number of blood and body tissue samples were also analysed.

Table 1. Exposure Conditions and Deposition Factors for Rats exposed to Iron Oxide Aerosol

Exp	Duration (min)	Fe conc of inhaled air (mg/m^3)	Fe deposited in lung 1d post exp (mg)	Fractional deposition at t=0 (D)
1	16	392	0.35	0.50
2	30	250	0.60	0.72
3	Intermittent (Table 3)		3.21 3.12	0.35
4	235	143	1.45 1.21	0.46 0.38

Results and Discussion

The lung burden was calculated using the total lung weight and the iron concentrations of the exposed rat's lung corrected for iron in control rat lung. The shape of the pulmonary elimination curve can be expressed by an exponential expression of the form

$$R = A_s e^{-\frac{0.693t}{T_{s\frac{1}{2}}}} + A_l e^{-\frac{0.693t}{T_{l\frac{1}{2}}}}$$

where R is the % of the initial lung burden retained at time t, $T_{s\frac{1}{2}}$ and $T_{l\frac{1}{2}}$ are short term and longer term elimination half times, A_s, A_l are constants.

Substituting values for A_s, A_l and the corresponding elimination half times $T_{s\frac{1}{2}}$ and $T_{l\frac{1}{2}}$ taken from ICRP data¹ the elimination curve for man² can be compared with that determined for the rat (Fig 1).

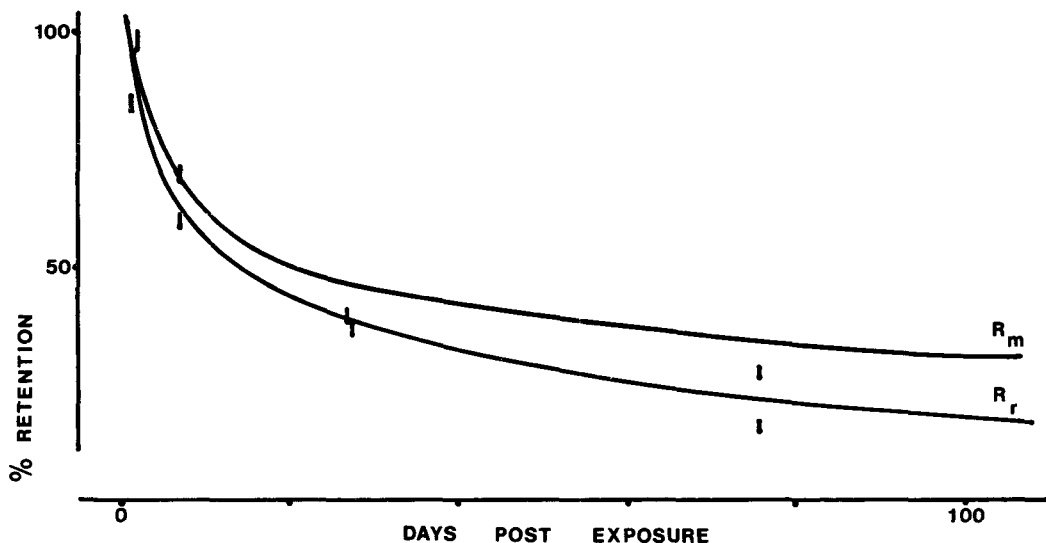


Fig 1 RETENTION CURVES FOR MAN (R_m) AND RAT (R_r)

The experimental data obtained in this present study are insufficient to be definitive, but are compatible with a similar short term ($T_{1/2}=1d$) elimination in both man and the rat, although the longer term elimination $T_{1/2}$ for the rat is shorter ($\sim 33d$) than that proposed by ICRP for man ($T_{1/2}=70d$). The ICRP curve is based upon a two phase exponential clearance model, comprising a short phase I ($T_{1/2}=24h$) and a phase II clearance of $T_{1/2}=70d$). The slower clearance represents that fraction ($\sim 40\%$) of the initial pulmonary deposited iron oxide translocated to the gastro-intestinal tract, through a process which depends upon endocytosis and ciliary mucus transport, and in addition the fraction that is translocated to the blood - according to the ICRP model some 15% of the initial pulmonary iron oxide deposit would be taken into the systemic blood directly whilst a fraction ($\sim 5\%$) would follow a lymphatic drainage route.

A contribution to the blood may also be postulated following gastro-intestinal tract absorption, but in the case of iron oxide this will be negligible. In order to test this, blood, liver and kidney samples were removed from pairs of rats one day post exposure in both experiments 3 and 4, and also from an equivalent number of control rats, and all samples were analysed for iron. The contribution of inhaled iron oxide to the blood and to organs other than lung tested proved to be insignificant. This confirms the observations of Albert and Arnett⁵ on man and also Morrow et al⁶ on dogs, who found no detectable radioactivity in blood following inhalation studies with ^{59}Fe labelled iron oxide, Morrow concluding that iron oxide behaves as an 'insoluble' material being efficiently eliminated from the gastro-intestinal tract in the faeces.

The very long term fate of pulmonary deposited iron oxide is probably inadequately described by a two component exponential expression: the rats were studied in this work for an insufficiently long period post exposure to define such components, and other

workers^{5,6} terminated their measurements 70d post exposure. However the retention of iron and other elements occupationally ingested many years previously is clearly demonstrated in the results of human lung analysis (Table 2). The 'hard metal' workers were engaged in a process involving the risk of inhalation of cobalt, tantalum and tungsten, but clearly the iron concentration in the boiler maker's lung is of particular interest, being clearly indicative of long term retention since the sample was taken five years post exposure.

Table 2. Analysis Results on Lung of Controls and Industrially Exposed Metalworkers*

	Hard Metal Workers				Boiler/ Welder	Controls	
	1	2	3	4		1	2
Co	1.0	2.6	2.0	19.2	3.9	1.0	0.9
Cr	1.0	1.7	1.7	10.6	1.6	0.4	0.3
Fe	1.0	1.1	0.7	1.7	16.6	0.7	0.8
Ta	1.0	-	0.5	1.8	-	-	-
W	1.0	26.0	48.0	130.0	-	-	-

* All results are compared to HM 1, taking the concentration of each element as unity in that sample.

Fractional deposition factors can be calculated from the expression

$$D = \frac{\text{wgt of deposited particulate in lung at } t = 0}{(\text{air conc})(\text{duration})(\text{respiration rate})(\text{tidal vol})}$$

Casarett⁷ reports deposition factors of 0.49-0.67 (mean 0.60) for iron oxide particulates (0.068 CMD σ_g 1.62), these calculations being based on minute volumes of 75cm³. In general the rats used in this present study had minute volumes of 120cm³ and the calculated deposition factors for the 0.3 μm iron oxide were lower (Table 1), being very similar to the 0.35 predicted from ICRP for man. Because of the dimensional differences it is not unexpected that for relatively large diameter particulates (>5 μm) there should be greater deposition in the nasal and upper respiratory tract of the rat than in man, but the effect of anatomical difference on <1 μm particulates is less clear, and in this study the deposition in rats could not be differentiated from man. Thomas⁸ found that for ¹³⁷Cs in rats the total body deposition plotted against particle size followed a similar trend to that of the ICRP curve for man but at about half the deposition level - the deposition factors at the lowest particle size studied (~0.3 μm MAD) being in all cases lower than that found for iron oxide in this work.

Thomas⁹ identified in rats an intermediate clearance phase due to the anatomical structure of the upper respiratory tract

which because of its downward slope towards the mouth could be expected to enhance the clearance rate compared to man. This could partly account for the phase II elimination half time observed for rats in this study ($T_{\frac{1}{2}}$ 33d) compared to that for the dog ($T_{\frac{1}{2}}$ 62d)⁶ and man ($T_{\frac{1}{2}}$ 70d)¹.

Table 3. Daily Contribution to Lung Burden during Intermittent Exposure (Experiment 3)

Day	Duration (min)	Fe conc of inhaled air (mg/m ³)	Initial pulmonary* deposition (mg)	Remaining at ^e T = 11 (mg)
0	33	378	0.52	0.25
1	31	450	0.59	0.28
2	30	402	0.51	0.26
3	30	367	0.46	0.24
4	32	524	0.70	0.37
5				
6				
7	30	583	0.73	0.42
8	29	664	0.81	0.50
9	26	599	0.65	0.44
10	32	622	0.84	0.66

Total 3.42

cf Table 1(3.21, 3.12)

* Calculated from (air conc)(duration)(min vol)(0.35)

^e Calculated using elimination equation

Using the two component elimination equation and the deposition factor which for the purpose of illustration is taken as that of the ICRP (0.35), the daily contributions of iron to the rat's lungs are easily calculated, and the total can then be compared with the actual iron content of the lungs found at autopsy (Table 3).

Conclusion

This preliminary study of the iron oxide inhalation and elimination has indicated the value of the ICRP Task Group approach. For the particle size examined, the deposition factors found for the rat were similar to those proposed by ICRP for man. Pulmonary clearance could be reasonably described in terms of a two component exponential but the phase II component was of shorter half time than that in man.

Acknowledgements

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INHALATION HAZARDS: THEY COULD BE WORSE*

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Abstract

The inhalation route of entrance of relatively insoluble radioactive toxic particles to the body may afford several mechanisms that act to reduce the potential radiation hazard, as compared to entrance by other possible routes. A relatively minor fraction of what is inhaled actually reaches the pulmonary spaces of the lung. In addition, those particles that do reach this region of the respiratory tract have the opportunity of being cleared by at least three methods; muco-ciliary movement, dissolution and subsequent removal by the blood, and translocation to tracheobronchial lymph nodes. With relatively insoluble particles all of these factors appear to work toward helping the individual to reduce the radiation insult. The methods proposed to bring about this reduction through these various factors are discussed in general terms.

Introduction

The inhalation route of entry of radionuclides to the body generally represents the most prevalent hazard in incidents involving accidental releases that may occur in the nuclear industry. Radioactive materials that become airborne are likely to do so in a variety of physico-chemical states, depending upon the industrial operation. The temperature of release, for example, may play an important role in the subsequent behavior of the nuclide-containing substance once it is deposited in the respiratory tract. The chemical or physical form of the vehicle or matrix containing the radionuclide may play a similarly important role in the subsequent metabolism of the inhaled material. Thus, the practical aspects of concern in the evaluation of potential inhalation hazards involve an explicit knowledge of the conditions under which releases may occur. This point has been particularly emphasized recently by a number of authors investigating the factors involved in the increased incidence of lung cancer in miners and workers in other dusty trades.¹

This paper explains the factors involved in assessing the relative potential hazards following human inhalation exposure and discusses many areas in which the body appears to aid in reducing the radiation risk to the body tissues.

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Lung Deposition

The Task Group on Lung Dynamics (TGLD) of Committee 2, the International Commission on Radiological Protection (ICRP) has separated the human respiratory tract into three major regions.² These are the nasopharyngeal (NP) region, beginning with the anterior nares and descending to the level of the larynx; the tracheobronchial (TB) region, continuing through the trachea and bronchial tree and including the terminal bronchioles; and the pulmonary (P) region consisting of the remainder of the respiratory tract, beginning with the respiratory bronchioles and including the alveoli. In general, larger particles deposit in the NP and TB regions and, if relatively insoluble in body fluids, soon (within 48 hours) find their way to the gastrointestinal tract to be eliminated by fecal excretion. According to the TGLD, the quantity deposited in these two areas combined might vary from a few percent up to essentially 100% of inhaled particulate material. Deposition in the TB region rarely exceeds 10% of the total inhaled aerosol. The NP region shows the greatest variation in deposition as a function of particle size, with deposition greater at the larger particle sizes and increasingly reduced as the aerodynamic particle diameters become smaller than 1 μm . Any fraction of an inhaled aerosol of an insoluble nature that deposits in these two regions is acted upon in favor of defense against the toxicant, thus protecting the individual from depositing large quantities of inhaled particles in the pulmonary spaces.

This same argument may be used in discussing the pulmonary (P) region with regard to deposition of inhaled materials. Because of the large fractions inhaled that are deposited in the upper respiratory tract, plus the sizeable fraction that is exhaled and not deposited in any part of the tract, deposition in the P region is comparatively low. Only when the particle size is very small (less than 0.5 μm aerodynamic diameter) does deposition in this region surpass 25% of the amount inhaled, as estimated by the TGLD.² Inasmuch as most practical aerosols are of a size greater than this, the body is afforded considerable protection from an inhaled, relatively insoluble substance.

The exception to these protective factors arises when inhaled particles are of a soluble nature. As an example, a recent study has shown that soluble substances deposited on the mucosa of the NP region of the Syrian hamster entered the blood at a rather rapid rate.³ These studies indicated a nasal absorption of >50% for the chlorides of strontium, barium and cesium but less than 4% for the trivalent cerium. As is often the case, trivalent cations tend to form rather insoluble ligands with proteins and other biological molecules. The report indicates that absorption from the NP region can be at least as great as from gastrointestinal absorption for a given substance. Thus, the radiobiological effects from a very soluble particulate material following inhalation can be afforded little alleviation by the body forces, in that the radionuclide may enter the circulation very rapidly. Those substances that fall between the arbitrary soluble and insoluble categories present a complicated picture that will be discussed later.

Pulmonary Retention

The major factor in considering inhalation hazards appears to be related much more to retention characteristics than to initial deposition. What is the likely fate of the inhaled fraction that is deposited in the pulmonary spaces? What lines of body defense favor the residence of a body burden of a given radionuclide to be in lung, as opposed to being initially deposited elsewhere as a consequence of a non-inhalation type of entry to the body? In one case the particle may be extremely soluble in body fluids and would act the same with

regard to localization sites and retention characteristics (metabolism) regardless of route of entry. In this instance the mode of assimilation is probably not very important. In another case, that of a relatively insoluble particle however, the particle may remain in the pulmonary spaces (alveolar region) and irradiate the surrounding tissue for a period of time dependent upon its effective half-life in that area. It may gradually dissolve and the radionuclide cation may enter the blood and either be excreted or translocated to the organ(s) most compatible with its chemical properties. The insoluble particle may also clear through the lymphatics to the regional (tracheobronchial) lymph nodes. It may also have the fortune of eventually being swept up the ciliated escalator, swallowed, and excreted in the feces. These are some of the factors that will be considered in terms of alterations, particularly reductions, in the potential radiation hazard after inhalation. The following remarks will be restricted to relatively insoluble particles deposited in the deep lung, unless otherwise noted.

Ciliary Removal:

It was mentioned that early clearance from the tracheobronchial region takes place through ciliary activity, in a matter of hours or days. This means of clearance is an important body mechanism for removing toxicant particles after accidental inhalation exposure. What of this method of clearance after the initial, rather large phase, has subsided? It is common knowledge that particles can be readily engulfed by macrophages following deposition in the alveolar region. It is also feasible and accepted that a small particle may reside on or within the surfactant lining of the lung, even perhaps prior to or following an engulfment by a phagocytic cell. What now is the fate of these particles? It makes sense that the mechanical movement of the lung alone creates some probability that the particle associated with surfactant, whether or not engulfed, will be swept upward via the muco-ciliary escalator and swallowed. In other words, it would essentially ride "piggy-back" on the normal processes of lung clearance. Such a probability may well be dependent upon the numbers of particles present that are available to be treated in this fashion, thus leading to an exponential (first order) loss with regard to the decrease in lung content with time.

This process of ciliary clearance from the pulmonary spaces is very important in clearing the lung of toxicants, including particulate material containing radionuclides. Without this process, regardless of the detailed mechanisms involved, the relative potential radiation hazard to the lung after inhalation would be considerably greater.

Dissolution of Particles:

All materials appear to be somewhat soluble in body fluids, and the fluids of the lung are no exception in the process. Mercer has emphasized the importance of solubilization of particles in removing materials (e.g., radionuclides contained in particles) from the deep lung.⁴ The rate of dissolution of a particle in an erosive active medium is a function of the total amount of available surface area on that particle. Thus, the rate of removal of particulate material from the lung by this process is a function of the total surface area available to the fluids in the lung. In most cases this surface area is made up from millions of particles of all sizes, generally accepted as being log-normally distributed according to the number occurring at a given size. There are a few particles that are very large and which carry a great deal of radioactivity, compared to a large number of small particles, each containing relatively little radioactivity. Consider the dissolution of this size distribution, assuming all chemical characteristics of the particles to be the same.

The smaller particles may, in total, represent a considerable radiation source to the lung. These will dissolve much faster however, due to the much greater surface-to-mass ratios (surface area \div volume \times density). The radionuclide cations released subsequently follow one of several pathways; (1) enter the blood and be excreted or localized in some other organ, (2) become associated into a chemical complex in the lung, (3) somehow find their way to the lymphatics or (4) somehow find their way up the muco-ciliary escalator. Of these possibilities, case 1 is the most likely route for the lone cation. In this case, the situation results in distributing the radioactivity to other organs in the body (often referred to hereafter as "internal organs"). Thus, as the dissolution process continues, more radionuclide accumulates internally or is excreted, primarily in the urine, and the initially higher dose rate to the lung is gradually reduced. The body, by this method, once again rises to the occasion by splitting the offensive lines and utilizing a means to dilute the total potential radiation dose to the body. With some other routes of entrance to the body, such as intravenous, the insult would be inflicted to two or three internal organs beginning almost immediately with no "reservoir" organ such as lung to dilute the attack. This would create a very high dose rate initially, a factor that may be very radiobiologically important. The slow migration from lung to the internal organs leads to a gradual build-up and simultaneous continuous loss from the tissues of localization, thus in most cases, never subjecting the internal organs to the larger dose rates. Such interaction between lung and radionuclides translocated to other organs has yet to be demonstrated experimentally to be less hazardous, and is no doubt dependent upon the chemical properties and physical half-lives of the materials involved. It would appear, however, that the inhalation route in this respect is somewhat favored in regard to being less hazardous for a given amount of radionuclide-containing material entering the pulmonary spaces as compared to some other route.

Translocation to Lymph Nodes:

Data are available from many sources that indicate a gradual concentration of a potentially toxic particulate substance in tracheobronchial lymph nodes following its entrance to the lung.⁵ In most instances, the concentration (quantity/gram tissue) in these regional nodes surpasses that in lung at about 100 days post-commencement of exposure, regardless of whether this be single (acute) or chronic (repeated) inhalation exposure (Fig. 1). The material migrated to the nodes appears to be in particulate form, at least as can be discerned histologically and autoradiographically, depending upon the radionuclide and quantities involved.⁶ Large accumulations of the particulate material may occur in the medullary areas, with little or no accumulation near the more peripheral germinal centers of the cortex. With alpha particle emitters, the particle appears to be localized such that the ionizing track length will not permit a release of large amounts (if any) of radiation energy to the germinal sites where lymphocyte production is manifested. Thus, large accumulations of alpha-emitting radionuclides in these nodes tend to irradiate the nutrient supply to the node (shut off the circulation), making it devoid of function after an extended period of time, but appear to act only to a minor degree directly on the cortical tissue, per se. The loss of nodal material under these conditions does not appear to be of grave consequence to the body in cases that have been experimentally observed.⁶ In a long-term study involving inhalation by Beagle dogs of the alpha-emitter ^{239}Pu , large accumulations of the nuclide were found in the pulmonary lymph nodes.⁷ In no case was there found a primary tumor in the lymphatic tissue of the nodes, but three cases of primary lesions of endothelial origin were reported. With beta particle emitters the length of the ionizing path is longer, and the extent (range) of damage is therefore greater. No primary tumors in the pulmonary

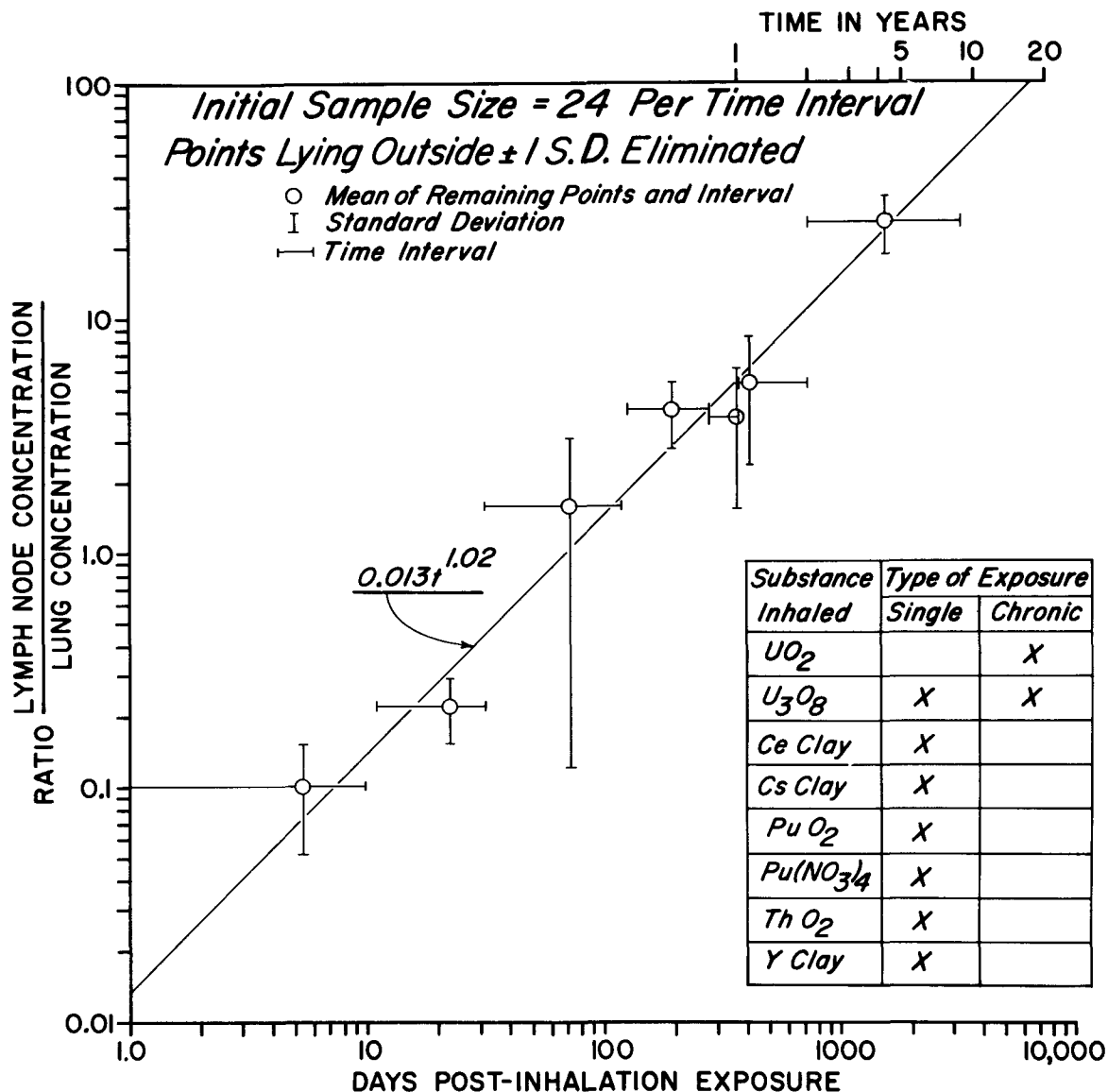


Fig. 1. Lymph node to lung concentration ratios following inhalation of various radionuclides by Beagle dogs. The total number of points (168) were segregated into groups of 24 and standard deviations (S. D.) calculated. All points outside ± 1 S. D. were eliminated from each group, a new mean and S. D. were calculated, and plotted, as shown. The time intervals spanning collection of each group of 24 points are shown, with the mean being plotted at the average time for the interval. All details concerning the individual experiments are reported elsewhere.⁵

lymph nodes have been reported, however, following inhalation of these type of emitters. It is as though these nodes were anatomically placed and physiologically devised to "clean-up" the more sensitive lung tissue and to, once again and emphatically, act as a means of effectively reducing the radiation hazard following inhalation.

Therapy

The use of chelating (and other) agents to relieve the body of deposited radionuclides has been in practice for years; an excellent review has recently been published.⁸ These chemicals have been used effectively to remove cations from bone, in particular the bone surfaces, and they act quite well for materials such as plutonium if administered soon after the nuclide reaches the bloodstream. The gross effect is one of increasing by quite sizeable amounts the quantity of radionuclide excreted in the urine; the basic fallacy is that the total reduction in internally deposited radionuclide is quite negligible. Following the inhalation route, however, there has been considerable recent evidence of significant reduction in lung burdens of radionuclide-containing particles by pulmonary lavage.⁹ Through the process of flushing out the deep lung, alternating sides at intervals of a few days, as much as 50% of an initial lung burden may be removed. This is an order of magnitude better than the use of chemical agents such as chelators, in removing materials that may have been deposited internally by another route. In addition, for that material that leaves the pulmonary spaces for deposit in internal organs, as described earlier, one can also use the chelating agents quite effectively. The combined effect of lung washing (lavage) and DTPA (administered in the lavage fluid) has been recently described following the inhalation of relatively soluble $^{144}\text{CeCl}_3$, and the combination produced a "one-two" punch for removal of the inhaled cation.¹⁰ When this combined treatment was used for an insoluble form of the same cation in fused clay particles, however, the DTPA appeared to be of little assistance in reducing the lung burden.¹¹ The lung washing technique may enhance entrance to the blood as well as performing its actual physical removal. The intravenous chelator then enhances excretion by sequestering the radionuclide as it enters the circulation.

The gross appearance from the inhalation route of entry is one of encouragement with regard to the ability to remove substantial quantities of deposited particulates. The fact that therapeutic removal of radioactive particles from the lung following inhalation can be accomplished to a degree, is an important factor in assessing relative radiological hazards as a function of route of entry to the body.

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LES PROBLEMES DE PROTECTION SOULEVES PAR LES RAYONNEMENTS NON IONISANTS

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Abstract

Considering the growing utilization of non ionizing radiation and the spreading danger from workers to the general population, the nature, sources and conditions of exposure to non-ionizing radiations, their interaction with matter, their biological and pathological effects are reviewed and the present situation of protection as to regulations, operational principles, and monitoring is discussed at the international and national levels.

Introduction

Le Comité Scientifique du congrès m'a fait l'honneur de me demander de vous exposer les problèmes de protection posés par les rayonnements non-ionisants. C'est la première fois, je crois, qu'une Organisation Internationale de Protection Radiologique inscrit ce sujet dans ses préoccupations.

Jusqu'à présent les rayonnements ionisants avaient accaparé leur attention toute entière. Cependant les rayonnements non-ionisants méritent qu'on examine attentivement les problèmes de protection qu'ils posent étant donné la puissance de certains générateurs, le développement de leur utilisation et l'extension des dangers du domaine professionnel au domaine public. Certains pays, tels les USA et l'URSS, ont notablement avancé dans ce domaine, la Communauté Economique Européenne s'en préoccupe et l'Organisation Mondiale de la Santé a créé des groupes de travail pour étudier ces problèmes. Je vais tenter de faire le point sur la situation actuelle, en m'efforçant de garder le juste milieu devant un auditoire hétérogène comprenant d'éminents spécialistes de la question, mais aussi beaucoup de personnes peu familiarisées. Je m'excuse donc auprès de certains des rappels généraux que mon exposé comportera obligatoirement. Nous examinerons successivement la nature, les sources et les modalités d'irradiation des rayonnements non ionisants, leur interaction avec la matière, leurs effets biologiques et pathologiques, les mesures de protection concernant la réglementation, la prévention et la surveillance.

Nature physique

Les rayonnements non ionisants se caractérisent par le fait que leur interaction avec la matière n'engendre pas d'ionisation.

Nous n'étudierons dans le présent rapport que les rayonnements électromagnétiques non ionisants avec parfois leur comparaison avec les rayonnements électromagnétiques ionisants.

Le spectre des ondes électromagnétiques est illustré par la figure I. Il va des rayons gamma et des rayons X ultra-durs aux ondes radio de grande longueur d'onde. Compte-tenu de l'énergie des photons et du mode d'interaction avec la matière, il est possible de distinguer plusieurs zones spectrales dont les frontières ne sont pas délimitées avec netteté.

Lasers. En outre, une autre catégorie de rayonnements électromagnétiques mérite une mention particulière : les lasers et les masers. Il s'agit de rayonnements électromagnétiques cohérents dans le domaine des longueurs d'onde de l'ultra-violet à l'infra-rouge pour les lasers, des micro-ondes pour les masers.

Les lasers sont des émetteurs de faisceaux de lumière fortement collimatés dont la densité d'énergie et la densité de puissance peuvent être considérables. Ces faisceaux lumineux sont cohérents à la fois dans l'espace et dans le temps. Ils sont monochromatiques, les gammes d'émission allant de l'ultra-violet à l'infra-rouge en passant par le domaine du visible.

Unités

Les unités utilisées en protection contre les rayonnements non ionisants sont de nature très différente de celles utilisées pour les rayonnements ionisants. Pour ces derniers on emploie des unités d'exposition (Roentgen) et des unités de dose absorbée dans la matière (rad) ; le rad est exprimé en Joule par gramme de matière. Pour les rayonnements non ionisants on emploie des densités d'énergie en J/cm^2 ou des densités de puissance en W/cm^2 ; il s'agit donc d'énergie passant au travers d'une surface et non dissipée dans un volume ou une masse. Il en résulte que les relations dose-effet, d'une part, et les normes réglementaires d'autre part, sont difficilement comparables entre rayonnements non ionisants et rayonnements ionisants.

Les sources de rayonnements non ionisants

Les sources de rayonnements non ionisants sont très diverses et doivent être étudiées séparément pour chacun d'eux.

Lumière visible - Ultra-violet - Infra-rouge. La source principale de lumière visible, d'ultra-violet et d'infra-rouge est le rayonnement solaire. Les générateurs artificiels de rayonnement ultra-violet sont essentiellement constitués par des tubes au sein desquels on produit des décharges électriques dans un gaz. Les sources artificielles de rayonnement infra-rouge sont constituées par des matériaux ayant atteint une température suffisante.

Micro-ondes. Un générateur de micro-ondes est constitué par :

- le générateur proprement dit qui crée un rayonnement électromagnétique par déplacement d'électrons dans des champs électriques et magnétiques combinés. Suivant la fréquence et la puissance désirée on utilise des tubes électroniques, des magnétrons ou des klistrons.
- un organe de transmission constitué par une ligne bifilaire, un câble coaxial ou un guide d'onde.
- une antenne d'émission (radar) ou une cavité (four).

Lasers. L'effet laser est obtenu en stimulant le retour à l'état normal d'un certain nombre d'atomes qui ont une population d'électrons préalablement inversée de leur niveau atomique. Le laser amplifie et coordonne ce retour. Pour mettre en oeuvre l'effet laser, il faut disposer :

- d'un matériel atomique dont la population électronique peut être inversée,
- d'un moyen d'inverser cette population,
- d'un moyen de stimuler le retour à l'état normal à l'aide d'un résonateur optique.

La durée de l'impulsion laser dépend essentiellement du mode de fonctionnement :

- lasers à fonctionnement continu,
- lasers à impulsion normale (durée d'impulsion d'environ 1 milliseconde),
- lasers déclenchés ou à impulsion géante (durée d'impulsion d'environ 1 nanoseconde).

Les lasers à impulsion atteignent de grandes puissances dans des temps nécessairement très courts, surtout lorsqu'ils fonctionnent " en déclenché " (exemple : le gigawatt en moins de 30 nanosecondes). Les lasers à fonctionnement continu émettent des rayonnements allant de quelques milliwatts à 500 watts.

Du point de vue technologique on distingue : les lasers à cristal, les lasers à gaz, les lasers à semi-conducteurs, les lasers à liquide.

Modalités d'irradiation

Les modalités d'irradiation par des rayonnements non ionisants sont extrêmement diverses tant par leur nature que par leur importance relative.

Elles tiennent tout d'abord à la production de ces rayonnements soit dans le milieu naturel, soit dans les activités humaines. Les applications des rayonnements non ionisants sont en effet scientifiques, industrielles, militaires, médicales, domestiques. Les personnes exposées peuvent être classées en trois catégories principales :

- les travailleurs,
- le public,
- les patients (irradiation médicale).

Lumière visible. Les modalités d'irradiation par la lumière visible sont trop communes pour qu'il soit nécessaire de les rappeler.

Ultra-violet. En milieu professionnel, une exposition peut survenir dans les cas suivants :

1. Rayonnements ultra-violetes solaires : travaux de maçonnerie, construction des routes, travaux agricoles, travaux en mer (pêche), travaux en haute montagne.

2. Rayonnements ultra-violetes artificiels : fabrication du verre, sidérurgie et coulée, métallurgie (laminage), contrôle des moules métalliques, travaux de soudage et de découpage à l'arc, fabrication de manchons à incandescence, fabrication et contrôle de lampes à ultra-violetes et de tubes à décharges, lithographie, irradiation des denrées alimentaires, des médicaments et du tabac.

Dans le domaine public, l'irradiation solaire est essentiellement liée aux loisirs, l'irradiation artificielle au bronzage esthétique.

Dans le domaine médical, l'irradiation ultra-violette est due à un certain nombre de traitements photothérapeutiques.

Infra-rouge. La surexposition professionnelle aux rayonnements infra-rouges menace les personnes qui travaillent dans le voisinage des fours ou sur des corps ou objets portés à haute température.

L'irradiation du public est essentiellement liée à l'utilisation des appareils de chauffage, l'irradiation médicale à certaines méthodes de thermo-thérapie.

Micro-ondes. Un risque d'exposition à des doses considérées comme supérieures pour l'absorption d'une certaine dose d'énergie aux niveaux tolérables pour l'organisme humain ou partielle est à signaler dans les professions suivantes : personnel volant ou personnel au sol des aérodromes; personnel maritime; mécanicien radar et personnel d'exploitation des stations radar; personnel travaillant dans les stations aérospatiales; personnel exposé autour de fours à haute fréquence; personnel travaillant avec des masers; personnes utilisant des appareils médicaux à micro-ondes; personnes affectées au contrôle des champs de micro-ondes; personnes utilisant des fours à micro-ondes (cuisines); personnes préposées à la stérilisation des denrées alimentaires et des produits pharmaceutiques; travailleurs du bois contre-plaqué.

L'irradiation du public revêt deux modalités essentielles :

- un certain nombre de personnes peuvent être exposées soit du fait de leur logement, soit du fait de leur passage aux rayonnements émis par des sources puissantes (radar);
- d'autre part, un nombre de plus en plus grand de foyers domestiques sont équipés avec des fours culinaires à micro-ondes dont les risques d'irradiation en fonctionnement normal ou anormal ne sont pas négligeables.

L'irradiation médicale est essentiellement due aux traitements diathermiques.

Ondes radio. Dans le domaine professionnel, les personnes exposées appartiennent aux techniciens de la radio, de la télévision et de certaines applications industrielles.

Le public d'une façon générale est soumis au champ d'ondes radio à des niveaux faibles sauf au voisinage des émetteurs puissants.

Dans le domaine médical, l'irradiation par ondes courtes est une modalité de la diathermie.

Lasers. Le laser permet de concentrer dans un très petit volume une puissance électromagnétique considérable et d'obtenir des températures extrêmement élevées capables de faire fondre tous les matériaux même les plus réfractaires ou de détruire les tissus. C'est pourquoi différents types de rayonnement laser trouvent d'ores et déjà de nombreuses utilisations :

- industrielle et technique : micro-soudage, micro-usinage des métaux, forage et découpage, détermination de niveaux à l'aide de rayons conducteurs dans le bâtiment, photochimie, optique appliquée (holographie), métrologie (précision extraordinaire des techniques de mesure), télémétrie (lidar : appareil fonctionnant sur le même principe que le radar et qui permet des mesures extrêmement précises), télécommunications.
- recherche scientifique. Le laser, en effet, continuera de plus en plus à faire l'objet de nombreuses recherches dans les universités et les laboratoires.

Dans le domaine public on commence à trouver des gadgets utilisant des rayonnements lasers et même des jouets.

Dans le domaine médical existent des applications médico-chirurgicales : interventions sur la rétine, destruction de tumeurs, etc ... Le rayonnement laser lorsqu'il est utilisé, assure une cautérisation instantanée et réalise une excellente hémostase.

Interaction des rayonnements électromagnétiques avec la matière

La compréhension des mécanismes d'action biologique des rayonnements non ionisants, et d'une façon générale des rayonnements électromagnétiques, est directement liée à la pénétration de ces rayonnements dans la matière vivante et à leur interaction avec elle.

La pénétration des rayonnements électromagnétiques dans la matière vivante est fonction de leur énergie et passe par un minimum pour les rayonnements visibles et voisins ainsi que le montre la figure 2. Certes, des différences existent selon la nature des tissus et les interfaces des organes. De même, des phénomènes de diffusion ou de résonance peuvent interférer avec les simples lois d'absorption. Mais d'une façon générale des interactions auront toujours lieu dans la peau, alors que les organes profonds du corps ne seront intéressés que par les rayonnements de grande énergie (ionisants) ou de grande longueur d'onde (micro-ondes et ondes radio). L'oeil constitue un cas particulier intéressant et la figure 3 montre qu'il est entièrement traversé par les rayonnements ionisants et les micro-ondes qu'il est opaque aux ultraviolets courts et aux infrarouges longs et qu'il concentre sur la rétine la lumière visible, les UV et IR voisins de celle-ci.

Les interactions des rayonnements électromagnétiques avec la matière peuvent intervenir à différents niveaux des édifices atomiques et moléculaires. De façon schématique et caricaturale, on peut admettre en première

approximation que les rayonnements ionisants perturbent les édifices atomiques et les rayonnements non ionisants les édifices moléculaires. Les rayonnements ionisants sont seuls capables, par absorption dans la matière, de former des électrons secondaires hautement énergétiques dont l'énergie cinétique s'épuise par ionisation et excitation. Les rayonnements non ionisants peuvent agir à trois niveaux différents : au niveau des électrons orbitaux externes atomiques et moléculaires avec corrélativement des réactions photochimiques; au niveau des atomes constitutifs des molécules considérées comme un tout avec des phénomènes de rotation. Les phénomènes de vibration, de rotation et de diverses déformations aboutissent à l'agitation thermique dans la matière.

On peut ajouter aux généralités précédentes les particularités suivantes concernant les effets biologiques des rayonnements électromagnétiques.

Effets biologiques

1. Rayons X et rayons gamma (rayonnements ionisants). L'une des caractéristiques des rayonnements ionisants est la distribution stochastique des atomes et molécules excités et ionisés dans la matière irradiée. Les excitations électroniques ainsi que les ionisations conduisent à des réactions radiochimiques non spécifiques. Ces réactions non spécifiques sont à l'origine de réactions biologiques secondaires de caractères généralement lésionnel. La chaleur dégagée par l'absorption des rayonnements, ainsi que les épiphénomènes thermiques des réactions radiochimiques sont sans importance sur le plan biologique.

2. Rayons ultra-violets. Les effets photobiologiques des ultra-violets sont une conséquence des réactions photochimiques. La chaleur produite par l'absorption des rayons ultra-violets, ainsi que les effets thermiques des réactions photochimiques sont d'une importance très secondaire sur le plan biologique.

3. Lumière visible. Les réactions photochimiques sont de faible importance et de caractère très spécifique (vision, photosynthèse). Chez l'homme, une grande partie de l'énergie lumineuse absorbée par la peau se transforme en chaleur. C'est la raison pour laquelle, dans le spectre de la lumière visible, il faut compter aussi bien avec des effets biologiques de caractère spécifique (tels que l'éblouissement et la photosensibilisation) qu'avec des effets thermiques de type pur (brûlures de la peau ou de la rétine).

4. Rayonnement infra-rouge. L'absorption du rayonnement infra-rouge par un tissu biologique se traduit non par une excitation électronique mais simplement par une excitation des niveaux vibrationnels et rotationnels. Au cours de ce processus d'absorption, seule l'énergie thermique de la substance irradiée augmente. Il s'ensuit que la production de chaleur est le seul effet biologique qui procède de cette absorption.

5. Micro-ondes. L'absorption des micro-ondes peut se traduire par l'excitation de quelques niveaux vibrationnels, mais ce sont essentiellement les niveaux rotationnels qui se trouvent excités. De plus, ces champs de haute fréquence engendrent des courants électriques. L'un et l'autre de ces effets se traduisent par une production de chaleur. L'effet biologique essentiel de

l'absorption des micro-ondes est une production de chaleur. Les effets non thermiques ne semblent jouer qu'un rôle accessoire.

6. Ondes radio. L'absorption a pour effet de créer dans les cellules et tissus des courants électriques qui se traduisent par de la chaleur. A des fréquences inférieures à environ 300 kHz, et pour certaines puissances de champ électrique, les cellules et les tissus peuvent subir une certaine excitation électrique, sans que cette excitation s'accompagne d'autres effets.

7. Lasers. Le rayonnement laser a une triple action biologique :

- effet thermique. L'effet thermique dépend du type et de la puissance du laser et de la structure de l'échantillon biologique exposé. En particulier, l'hétérogénéité des milieux biologiques peut entraîner une distribution thermique irrégulière et certaines différences d'action selon la nature du tissu.
- effet électrique. Le faisceau laser perturbe le cortège électronique des atomes de la matière irradiée. Il s'ensuit des perturbations qui peuvent être à l'origine de divers effets chimiques qui, à leur tour, pourront avoir des conséquences biologiques.
- effet mécanique. Le faisceau laser induit dans le milieu qu'il traverse des phonons (le phonon est la quantité élémentaire d'énergie élastique) stimulés transportant une énergie très grande qu'on appelle hypersons. Ces hypersons seraient à l'origine des effets lésionnels observés non pas au point d'impact du rayonnement laser sur le tissu mais à son point d'émergence.

Les dommages radiopathologiques

Après avoir étudié les mécanismes d'action biologique des rayonnements non ionisants, il convient d'envisager les dommages qu'ils peuvent engendrer.

1. Ultra-violets. Une surexposition aux rayons ultra-violet se traduit par des lésions à caractère aigu ou chronique. Parmi les lésions aiguës, citons les coups de soleil ainsi que l'érythème actinique. La surexposition aux rayons ultra-violet est susceptible de provoquer une kérato-conjonctivite douloureuse dont la guérison est cependant rapide et sans séquelles. Après une exposition prolongée ou répétée, des modifications cutanées irréversibles sont susceptibles de se produire. La peau devient brune et sèche, elle se ride et perd de son élasticité. En soi, il ne s'agit que d'affection bénigne. Cependant, sur cette dermatose peuvent se greffer des épithéliomas et des kératoses séniles. Il est connu que les rayonnements ultra-violet peuvent engendrer, à long terme, des cancers cutanés. Mais les relations "dose-effet" sont mal établies en cette matière.

2. Lumière visible. Les effets de la lumière visible sur l'organisme sont analogues à ceux des rayonnements ultra-violet. En outre, une exposition prolongée équivaut à une exposition à la chaleur. Enfin, les effets cataractogènes d'une telle exposition sont loin d'être négligeables.

3. Infra-rouge. Etant donné la faible pénétration, les seuls organes à pouvoir être atteints chez l'homme sont la peau et les yeux. Selon la densité du flux énergétique du rayonnement infra-rouge absorbé par la peau, on constate soit une légère hyperthermie locale, soit une hyperthermie généralisée plus ou moins grave, soit des brûlures. La lésion classique de l'oeil, en dehors des

brûlures de caractère aigu, est la cataracte.

4. Micro-ondes. Les micro-ondes sont absorbées par l'organisme. L'énergie absorbée est entièrement convertie en chaleur. L'augmentation de la température en un point donné de l'organisme humain dépend :

- de la quantité d'énergie des micro-ondes absorbée,
- de la conductibilité thermique du tissu ainsi que du transport thermique par l'intermédiaire du courant sanguin.

Les effets physiologiques et pathologiques sont une conséquence de l'augmentation locale de la température et de la charge thermique de l'ensemble de l'organisme (effet thermique). C'est la raison pour laquelle, pour de fortes densités de flux, il peut se produire localement des échauffements ou des brûlures, par exemple au niveau de la peau ou des tissus sous-jacents. Etant donné l'absence de vascularisation du cristallin, l'augmentation de température dans le cristallin peut devenir suffisamment grande pour provoquer le plus souvent et après un certain délai, l'opacification du cristallin. Une augmentation de la température dans de grandes régions de l'organisme humain conduit à une hyperthermie qui peut être létale.

En dehors des effets thermiques qui entraînent des lésions anatomiques, certains (école russe en particulier) envisagent des effets non thermiques. L'interprétation pathogénique est délicate et complexe; elle met en cause l'action directe des champs électromagnétiques et en particulier, des champs magnétiques variables sur la matière vivante. Des sujets exposés à de faibles intensités, ont présenté des troubles de type fonctionnel :

- troubles neuro-végétatifs : fatigabilité; hypersudation; somnolence (ou insomnie); céphalée; troubles sensoriels (visuels ou auditifs); instabilité émotionnelle; hyperexcitabilité neuromusculaire; perte d'appétit, nausées; troubles du rythme cardiaque, crises vagotoniques; état lipothymique, instabilité tensionnelle.
- troubles endocriniens : déficit surrénalien; hyperfonctionnement thyroïdien.
- modifications sanguines : ioniques; électrophorétiques; cytologiques - anémie, lymphopénie, polynucléose - coagulabilité sanguine et résistance globulaire, rarement touchées.

5. Ondes radio. En général, dans les utilisations professionnelles, les ondes radio induisent dans l'organisme des intensités tellement faibles qu'il n'en résulte pas d'effets biologiques. Cependant on aurait observé, pour certaines utilisations professionnelles, des manifestations neuro-végétatives, de la fatigue, de l'asthénie. Ces symptômes disparaissent en même temps que l'exposition. On n'a pas observé de séquelles.

6. Lasers. Les dommages corporels susceptibles d'être provoqués par le rayonnement laser peuvent résulter du rayonnement direct aussi bien que des rayonnements réfléchis diffusés. Les organes critiques sont l'oeil et la peau.

-Effets oculaires. La totalité du globe oculaire peut être atteinte, les dommages pouvant aller de la lésion minimale jusqu'à la déchirure de type explosif. Alors que les petites lésions de la cornée évoluent en général vers la guérison, les lésions de la rétine sont irréversibles. Les atteintes rétiniennes varient dans leur gravité en fonction de la topographie des lésions. Quelle que soit la topographie des lésions rétiniennes, il y a lieu de souligner le

caractère fréquemment indolore de celles-ci. Les facteurs aggravants des lésions sont les suivants :

Focalisation sur la rétine. Les rayons lasers qui traversent l'oeil se trouvent focalisés sur la rétine par le cristallin, ce qui se traduit par une forte augmentation de la densité énergétique. Pour l'établissement des consignes de sécurité, il faut faire entrer en ligne de compte le cas le plus défavorable où un faisceau de rayons parallèles est focalisé au niveau de la rétine sur une surface ayant le diamètre d'un disque de diffraction.

Transmission à travers l'oeil. La région de l'oeil située en avant de la rétine est transparente à la gamme des longueurs d'ondes, allant de 0,4 à 1,4 μm à quoi viennent s'ajouter deux autres gammes dans l'infrarouge. Si le rayonnement est absorbé dans la partie de l'oeil située en avant de la rétine, la densité énergétique sur la rétine peut se trouver diminuée. Mais alors, les tissus antérieurs sont exposés à un danger plus grand. Dans ce cas cependant vu l'absence de focalisation, le seuil critique des lésions est nettement plus élevé.

Durée d'irradiation. Dans le cas des lasers pulsés ou de lasers à impulsions géantes, on a constaté sur le plan expérimental que la densité énergétique admissible diminuait en même temps que la durée des impulsions. Cela se comprend aisément si l'on considère qu'avec un apport énergétique lent, l'énergie calorifique produite peut partiellement se dissiper, relevant d'autant le seuil lésionnel critique. Par ailleurs, un certain rôle est joué à cet égard par la fréquence de la répétition des impulsions.

Grosseur de l'image. Les travaux expérimentaux montrent que la densité d'énergie nécessaire à l'apparition d'une lésion augmente lorsque la surface de la région irradiée devient plus petite.

Il demeure, en matière d'effets oculaires, un certain nombre d'inconnues relatives :

- à l'effet de sommation des faisceaux de basse énergie;
- à l'effet de sommation des effets punctiformes périphériques;
- aux effets à long terme du rayonnement laser après cessation de l'exposition au risque;
- à l'effet en profondeur (effet Brillouin).

- Effets cutanés. L'incidence sur la peau d'un rayonnement laser peut provoquer immédiatement une brûlure avec coagulation locale, ce qui permet une cicatrisation rapide. Néanmoins la réaction cutanée dépend de plusieurs facteurs qui sont :

- la longueur d'onde du faisceau,
- la durée d'exposition,
- les "qualités optiques" de la peau en matière d'absorption, de transmission et de propriétés réfléchissantes. C'est ainsi que l'absorption de l'énergie laser se trouve facilitée par une densité locale plus importante de la pigmentation.

On ne sait encore que peu de choses en ce qui concerne l'exposition cutanée chronique au rayonnement laser.

Réglementation

En matière de protection, la réglementation constitue l'une des pièces maîtresses. En ce qui concerne les rayonnements électromagnétiques, un contraste saisissant existe entre l'état de la réglementation pour les rayonnements ionisants et pour les rayonnements non ionisants. Pour les rayonnements ionisants, un système cohérent et universellement adopté a été mis au point par une Commission Internationale compétente. Pour les rayonnements non ionisants, l'inventaire des réglementations fait apparaître une hétérogénéité, des lacunes, des contradictions.

Rayonnements ionisants

La protection contre les rayonnements ionisants a été codifiée par la Commission Internationale de Protection Radiologique. L'I.C.R.P. a établi un corps de doctrine qui est représenté par des recommandations générales et des recommandations particulières aux différentes modalités d'irradiation.

De fait, les recommandations ont été transcrites de façon appropriée dans les réglementations nationales de la quasi totalité des pays. Elles sont par ailleurs reconnues de façon officielle par les grandes organisations internationales (O.M.S., B.I.T., F.A.O., A.I.E.A., Comité Scientifique des Radiations des Nations Unies) ou régionales (Euratom, O.C.D.E.). Il résulte de cet état de fait une homogénéité et une cohérence exemplaires en matière de protection contre les rayonnements ionisants.

Rayonnements non ionisants

Dans le domaine des rayonnements non ionisants, il n'existe malheureusement pas d'organisme international comparable à la Commission Internationale de Protection Radiologique.

Il en résulte qu'aucune réglementation universellement acceptée n'existe, ni pour l'ensemble des rayonnements non ionisants, ni même pour certains d'entre eux. En outre, les textes réglementaires nationaux ou particuliers sont disparates avec de nombreuses lacunes. Enfin, il existe pour certains rayonnements tels que les micro-ondes des différences très sensibles entre les limites d'irradiation pouvant atteindre un facteur 1000.

Généralités

Normes fondamentales. Quelques recherches ont été faites pour tenter de déterminer une limite fondée sur un seuil de lésion pour quelques types de rayonnements avec des longueurs d'ondes déterminées et un effet biologique donné, par exemple : érythème et kérate-conjonctivite pour UV de différentes longueurs d'onde ; lésion de la rétine et de la peau pour les lasers. Mais les conditions expérimentales et les résultats disparates rendent la comparaison difficile.

L'exposition limite de l'organe est généralement expérimentée en $J.cm^{-2}$. Lorsqu'il s'agit de tissus internes cette norme est quelquefois déduite de

l'exposition des tissus externes par application d'un facteur de transmission ou de concentration.

Limites dérivées. Les recommandations, réglementations ou codes de pratique que l'on trouve pour certains types de rayonnements (UV, micro-ondes, lasers) concernent des limites dérivées. Ces dernières sont exprimées soit en densité d'énergie pour une impulsion de durée déterminée (surtout lorsqu'il s'agit d'impulsions relativement courtes) soit en densité de puissance $W \cdot cm^{-2}$.

Les valeurs recommandées sont très variables non seulement selon les pays, mais souvent dans un même pays selon l'organisme qui les propose. L'absence de méthodes de détermination, de conditions expérimentales et de moyens de mesure uniformes rend souvent leur comparaison difficile.

Etat actuel de la réglementation

UV. Des recherches existent pour déterminer un seuil de lésion au niveau de la peau ou de l'oeil en fonction de la longueur d'onde pour servir de base à une norme fondamentale. Mais, selon l'O.M.S., il est urgent de déterminer la relation dose-effet pour cancer de la peau. La seule recommandation officielle que l'on trouve est celle mentionnée par l'O.M.S., qui semble provenir d'une recommandation déjà assez ancienne de l'American Medical Association (1948): l'exposition ne doit pas dépasser :

0,5 μW par cm^2 pour une exposition ≤ 7 heures
et 0,1 μW par cm^2 pour une exposition continue de 24 h par jour.

Infra-rouges. Les connaissances sur les effets possibles à long terme sont insuffisantes, et en particulier celles sur la relation dose-effet. Les seules données sur lesquelles on pourrait actuellement fonder des normes sont les seuils de lésion connus pour la cornée (O.M.S.) :

7,6 J/cm^2 aux longueurs d'onde 0,88 - 1,1 μm
2,8 J/cm^2 aux longueurs d'onde 1,2 - 1,7 μm

Microondes

Normes fondamentales. Les bases sur lesquelles elles devraient être fondées, c'est-à-dire les effets biologiques, sont encore mal connues. Les données quantitative, lorsqu'il y en a, sont difficiles à interpréter. La question de l'existence d'un seuil n'est pas résolue et on ne peut exclure complètement la possibilité d'effets cumulatifs. D'autre part, une telle norme devrait être fondée sur l'énergie absorbée dans les tissus. Or, celle-ci ne peut être déterminée à l'heure actuelle avec le minimum de précision qui serait nécessaire pour en évaluer les risques. C'est pourquoi, il paraît difficile à l'heure actuelle de déterminer des normes fondamentales. Dunster cependant, propose comme norme fondamentale, une densité de puissance de 10 mW/cm^2 .

Limites dérivées. Les normes dérivées pour le domaine des microondes sont généralement exprimées en densité de puissance (mW/cm^2) pour une durée d'exposition déterminée. Un grand nombre de pays et d'organismes ont promulgué ou recommandé des limites d'exposition (tableau 1). On constate des variations d'un organisme à l'autre, mais il se dégage essentiellement deux grands courants :

- Aux Etats-Unis tout d'abord, dans certains pays européens ensuite, ces limites sont fondées sur l'aptitude de l'organisme à supporter une charge thermique :

10 mW/cm² supportable pendant un temps relativement long (ex. journée de travail),
1 mW/cm² recommandé par certains organismes lorsqu'il s'agit d'une exposition permanente (ex. marins dormant à proximité d'une antenne sur un bateau) et pour la population (limite de fuite à 5 cm proposée pour les nouveaux fours à microondes).

Pour des expositions intermittentes très courtes inférieures à l'heure, deux formules :

1 - Committee of the American Standards Association : 1 mW.h/cm² par fraction de 0,1 heure, ce qui donne 10 mW/cm² pour 0,1 heure.

2 - La plupart des organismes militaires aux Etats-Unis comme en Europe : la durée d'exposition en minute :

$$t = 60 \times \left(\frac{10}{W}\right)^2$$

W devant rester inférieur à 100 ou 55 mW/cm² selon des pays.
W étant la densité de puissance moyenne sur le temps t.

- La C.E.E. a créé un groupe de travail dont le rapporteur J. Dunster propose les limites simplifiées suivantes :

- exposition continue, densité de puissance dans le champ : 10 mW/cm²

- exposition intermittente, densité de puissance : $D \leq 100$ mW/cm²

durée d'exposition : $t = 60 \times \left(\frac{DWL}{D}\right)^2$

- En U.R.S.S. et certains pays d'Europe Centrale, les limites sont fondées sur des troubles fonctionnels liés à un déséquilibre neuro-végétatif plus ou moins accentué. Aussi, sont-elles beaucoup plus sévères :

1 mW/cm² pour moins de 20 minutes de séjour et port de lunettes de protection obligatoire,

0,1 mW/cm² pour une exposition de deux heures par jour

0,01 mW/cm² pour plus de deux heures par jour.

Lasers

Normes fondamentales. Organes sensibles : oeil et peau. L'effet sur l'oeil dépend de la longueur d'onde, ainsi que des conditions d'éclairage extérieur qui déterminent le diamètre de la pupille. On a cherché à établir des seuils de lésion pour la rétine.

Ici encore pas de moyens de mesure de l'énergie absorbée. Aussi toutes les réglementations portent-elles sur des limites dérivées.

Limites dérivées. Elles s'expriment différemment selon le mode de fonctionnement du laser :

- en densité d'énergie par unité de surface (J/cm^2) par impulsion pour les impulsions très courtes et les trains d'impulsions très courtes;
- en densité de puissance par unité de surface pour les rayonnements continus (W/cm^2).

Le code de pratique Britanique indique les limites suivantes pour l'exposition de la cornée (tableau II). J. Dunster propose pour la C.E.E. les limites suivantes pour l'oeil et la peau pour les lasers émettant dans l'ultra-violet, le visible et l'infra-rouge (tableau III, IV, V).

Prévention

La protection contre les rayonnements non ionisants doit comprendre la réglementation que nous venons de voir, la prévention et la surveillance. La prévention constitue le 2ème volet du triptyque de la protection radiologique. Dans le domaine des rayonnements ionisants, cette prévention a fait l'objet d'études très poussées qui ont contribué à faire de l'énergie atomique l'une des activités humaines où les risques sont les plus faibles. Elle est essentiellement basée sur la sécurité des installations, les protections liées aux appareils, les protections liées aux travailleurs, les consignes d'exploitation et les habitudes de travail.

Pour les rayonnements non ionisants de nombreuses méthodes de prévention ont été mises en oeuvre, essentiellement par les équipes scientifiques, militaires ou industrielles utilisant des sources relativement importantes. Les constructeurs également ont prévu des dispositifs de protection qu'il conviendrait cependant, dans un certain nombre de cas, d'améliorer de façon sensible.

D'une façon générale, on peut envisager des mesures de prévention au stade de la construction, au stade de l'utilisation sur un plan collectif ou sur un plan individuel.

Au stade de la construction, dans toute la mesure du possible, les sources de rayonnements non ionisants devraient posséder des écrans pour éviter tous les rayonnements parasites autres que le faisceau utile. Les appareils générateurs devraient faire l'objet d'une homologation officielle, en particulier pour ceux qui sont utilisés par le public. Il devrait être interdit d'apporter des modifications quelconques au système de protection. Peut-être conviendrait-il d'envisager un contrôle périodique des appareillages susceptibles de montrer des défauts au cours du temps.

Dans l'utilisation des sources de rayonnements non ionisants les mesures collectives devraient comprendre notamment les suivantes :

- les installations devraient être conçues de façon telle que dans tous les cas où cela est possible, elles constituent un système fermé sans rayonnement de fuite;
- pour le cas où les installations seraient en système ouvert, des zones interdites à séjour réglementé ou contrôlé devraient être établies;
- le nombre de personnes utilisant les installations devrait être réduit au minimum;

- des mesures du rayonnement ambiant devraient être effectuées afin de s'assurer que les limites d'irradiation sont bien respectées;
- des consignes strictes devraient être édictées pour le personnel.

A ces mesures collectives, des mesures individuelles peuvent s'ajouter lorsqu'il est impossible d'abaisser les niveaux d'irradiation au-dessous des limites réglementaires. Ce n'est que dans ce cas, qui devrait rester exceptionnel, que l'on peut envisager des moyens de protection individuelle portés par les travailleurs. Les organes les plus sensibles étant d'une façon générale les yeux, la peau et le corps entier selon le rayonnement, les moyens de protection consisteront soit en lunette, soit en vêtement protecteur.

Surveillance

La surveillance constitue le 3ème volet de la protection radiologique. On distingue une surveillance physique et une surveillance médicale.

Surveillance physique.

1. La surveillance physique des rayonnements ionisants a fait l'objet au cours des dernières décades d'un nombre considérable de travaux et de réalisations. On est actuellement capable de déceler nettement au-dessous des limites d'irradiation les doses reçues par les différentes catégories de personnes exposées. Les méthodes utilisées peuvent être directes ou indirectes :

- Les méthodes directes portent sur la mesure de l'irradiation au niveau des personnes elles-mêmes au moyen de détecteurs individuels (films dosimètres, stylos électromètres etc...).
- Les méthodes indirectes consistent à évaluer l'irradiation à partir de mesures portant sur l'environnement professionnel ou publique (champ d'irradiation ou de contamination).

La dosimétrie des rayonnements ionisants est devenue une discipline en soi. Elle fait appel à des méthodes de mesures extrêmement diverses. Elle a à sa disposition une grande variété de dosimètres permettant des mesures instantanées ou cumulées avec des gammes d'utilisation extrêmement larges, des sensibilités très poussées et des précisions largement suffisantes.

2. En ce qui concerne les rayonnements non ionisants, la situation est toute différente. La dosimétrie de ces rayonnements est difficile et elle n'a pas fait l'objet d'études aussi étendues et poussées qu'en ce qui concerne les rayonnements ionisants. Les méthodes utilisées ont un caractère spécifique qui limite leurs possibilités pratiques. Les difficultés sont déjà grandes pour obtenir des informations précises sur les champs de rayonnements non ionisants. Il n'existe pratiquement pas de détecteurs individuels. Il en résulte que la surveillance pour les rayonnements non ionisants ne peut être actuellement que collective.

Certaines estimations sont faites à partir de mesures portant sur les faisceaux au voisinage immédiat des sources, d'autres estimations sont faites à distance, c'est à dire au voisinage des personnes exposées dans les champs de rayonnements non ionisants.

A titre d'exemple, nous allons passer en revue quelques méthodes dosimétriques relatives aux deux catégories importantes de rayonnements non ionisants : les micro-ondes et les lasers.

Micro-ondes

Principe de mesure. Les micro-ondes sont constituées par un champ électrique et un champ magnétique ayant des composantes perpendiculaires et égales. Il est possible de mesurer l'une ou l'autre de ces composantes (le champ électrique est plus facile à mesurer) ou bien la densité totale de puissance.

Loin de l'émetteur, le faisceau est bien constitué. Dans la zone proche les deux composantes ne sont ni égales ni perpendiculaires et peuvent varier très rapidement d'un point à un autre. Il faudrait alors mesurer les deux composantes simultanément. Ces mesures sont particulièrement difficiles lorsqu'il y a plusieurs rayonnements de fréquence et de polarisation différentes et que viennent s'ajouter des phénomènes de réflexion (cas des navires de guerre et des bases aériennes).

En réalité, pratiquement tous les systèmes mesurent la densité de puissance par l'échauffement d'un capteur sensible.

Appareils de mesure :

- Pour les densités de puissance supérieures à 1 watt/cm^2 : calorimètre.
- Pour les valeurs inférieures à 1 watt/cm^2 : bolomètre.

Lasers

Domaine de mesure. On peut distinguer 3 types de lasers :

- émission continue,
- laser relaxé,
- laser déclenché.

La difficulté des mesures dépend des facteurs suivants :

- diversité des longueurs d'onde
- diversité des durées d'impulsions
- origine de l'irradiation qui peut être due au rayonnement direct, réfléchi ou diffusé (importance de la diffusion atmosphérique)
- dimension des faisceaux - les faisceaux focalisés ont des diamètres qui peuvent être inférieurs à $50 \mu\text{m}$.
- densité d'énergie ou de puissance très élevées dans les faisceaux focalisés (pouvant atteindre 100 MW/cm^2).

Unités de mesure. C'est le joule ou le joule/cm^2 pour les lasers pulsés, le watt ou le watt/cm^2 pour les lasers continus.

Principe des mesures. On peut distinguer trois types de dosimétrie :

- la mesure de la puissance du faisceau ou de l'énergie émise par pulse, dont la valeur est utile en cas d'irradiation par le faisceau direct,

- la mesure de l'intensité instantanée en un point, utile pour les cas d'irradiation par des faisceaux réfléchis,
- la mesure de l'exposition cumulée en divers points des laboratoires pour connaître les effets à long terme des rayonnements diffusés.

A ces mesures doit s'ajouter dans certains cas la détermination de la dimension du faisceau.

Appareils de mesure de la densité d'énergie ou de puissance :

- détecteurs photoémissifs
- détecteurs photoconducteurs
- détecteurs thermiques

Surveillance médicale

Les pratiques de surveillance médicale sont fort différentes d'un pays à l'autre et ceci est vrai non seulement pour les rayonnements non ionisants mais également pour les rayonnements ionisants.

En fait, il est bon que les travailleurs exposés aux rayonnements soient soumis à une surveillance médicale analogue à celle des autres travailleurs lorsque les risques d'irradiation sont suffisamment faibles. Par contre, lorsque les risques d'irradiation sont élevés, il convient de mettre en oeuvre une surveillance médicale spécifique portant sur les tissus et organes critiques pour le rayonnement considéré.

C'est ainsi que pour les rayonnements ionisants la surveillance doit porter sur la peau, l'oeil, les tissus sanguiformateurs, les gonades etc...

En ce qui concerne les rayonnements non ionisants, et compte tenu des dommages radiopathologiques éventuels, la surveillance médicale doit porter essentiellement sur l'oeil et la peau.

Cette surveillance médicale devrait comprendre des examens à l'embauche pour juger de l'aptitude du travailleur, des examens périodiques en cas de risques suffisants, des examens après emploi quand des séquelles à long terme sont possibles.

A titre d'exemple, nous présentons la surveillance médicale envisagée pour le personnel exposé aux rayonnements laser :

1. Un examen médical d'embauche et des examens médicaux périodiques au cours desquels un ophtalmologiste compétent et entraîné fera un examen oculaire complet comportant :
 - un examen de l'acuité visuelle
 - un examen des annexes extérieures de l'oeil (paupières, conjonctives, etc..)
 - un examen des différents milieux réfringents
 - un examen du fond d'oeil après dilatation de la pupille
 - un examen de la vision binoculaire
 - un examen de la vision des couleurs
 - un examen du champ visuel.

Un examen identique devra être effectué lorsque le travailleur cessera d'être exposé au rayonnement laser.

Il est souhaitable, en raison des manipulations dangereuses que doit effectuer le personnel exposé, que celui-ci ait un équilibre psychocaractériel satisfaisant et qu'il soit informé de l'existence et de la nature des risques. De même, il y aura lieu de procéder à un examen des téguments.

2. L'établissement d'une fiche de dommages.

Le rythme et la périodicité des examens systématiques doit être fonction de l'importance du risque et de la nature du travail effectué.

Les accidents cutanés se manifestent rapidement et peuvent aisément être rapportés à leur cause.

Les accidents oculaires ne sont quelquefois rapportés à leur cause que tardivement. Les lésions périphériques de la rétine peuvent n'être remarquées qu'à l'occasion d'un examen systématique ou par la victime après un laps de temps plus ou moins long, l'accident causal étant passé inaperçu et la lésion initiale étant généralement indolore.

Conclusion

Au terme de cet exposé nous sommes arrivés à conclure que les problèmes posés par la protection contre les rayonnements non ionisants sont nombreux, importants et difficiles. La situation actuelle apparaît entièrement différente de celle existante pour les rayonnements ionisants. En matière de rayonnements non ionisants des recherches importantes doivent être poursuivies dans le domaine de leurs interactions avec la matière, de leurs effets biologiques, des dommages radiopathologiques. Un effort international d'harmonisation doit être entrepris pour les systèmes d'unités, les normes réglementaires, l'homologation des appareils. Des études technologiques doivent être développées sur la sûreté des installations, les équipements de protection et les méthodes de dosimétrie pratique. Ceci rejoint les vœux émis par le groupe de travail de l'O.M.S. en 1971 que j'ai résumés dans le tableau VI. La Commission International de Protection Radiologique en 1971 a reconnu que des contrôles adéquats devraient être établis sur les sources de rayonnements non ionisants et qu'il était à présent nécessaire d'avoir des discussions internationales sur les critères biologiques servant de base aux normes. Cependant, l'I.C.R.P. a considéré que ce sujet était en dehors de ses préoccupations courantes. L'I.C.R.P. espérait que sa déclaration faciliterait une action internationale en ce domaine. Aussi je pense que le moment est peut être venu d'organiser cette action internationale. L'I.R.P.A. peut sans doute jouer un rôle important en cette matière.

En terminant, je remercie le 3ème Congrès de l'I.R.P.A. d'avoir donné une place de choix à ces problèmes et je souhaite que les participants se fassent, dans leurs pays respectifs, les propagandistes des actions à mener pour parfaire la protection contre les rayonnements non ionisants.

Tableau I
LIMITES D'EXPOSITION AUX MICROONDES

PAYS	EXPOSITION PERMANENTE	EXPOSITION PENDANT LES HEURES DE TRAVAIL	
		Durée d'exposition $t > 1h$	Durée d'exposition $t < 1h$
	Densité de puissance W en mW/cm^2	W en mW/cm^2	W en mW/cm^2 t en minutes
Etats-Unis	militaires	10	$t = 60 \times \left(\frac{10}{W}\right)^2$ avec $W \leq 100$
	civils	1	si $t > 6$ min. si $t \leq 6$ min. $W = 10$ $1 mW \cdot h/cm^2$ par 6 min.
U.R.S.S.		si $t > 2h/jour$ 0,01 si $t \leq 2h/jour$ 0,1	$W = 1$ avec $t \leq 20$ min. /jour
FRANCE (militaires)	1	10	$t = 60 \times \left(\frac{10}{W}\right)^2$ avec $W \leq 55$
BRITISH MEDICAL COUNCIL		10	

Tableau II

CORNEAL MAXIMUM PERMISSIBLE EXPOSURE LEVELS FOR LASER RADIATION
 DIRECT ILLUMINATION OR SPECULAR REFLECTION (7 mm pupil)

Lasers Type	Q-Switched 1 ns - 1 μ s Pulsed PRF < 10 per second Energy per pulse Jcm^{-2}	Long Pulsed 1 μ s - 0.1s PRF < 10 per second Energy per pulse Jcm^{-2}	Continuous Wave Long-Term Exposure Wcm^{-2}
Ruby (0.69 μ m)	3×10^{-8}	1×10^{-6}	4×10^{-7}
Neodymium (1.06 μ m)	2×10^{-7}	3×10^{-6}	2×10^{-6}
Helium-Neon (0.63 μ m)	-	-	3×10^{-7}
Argon (0.51 μ m) (0.48 μ m)	-	-	3×10^{-7}

Laser systems - Code of practice, UK, 1969

Tableau III

Derived Working Limits for Energy Surface Density
Ultra-violet Radiation (Single pulse)

Wavelength (nm)	Pulse length, t (s)	DWL (skin or eye) (J/cm ²)
200 - 315	$10^{-2} - 3 \times 10^4$	3×10^{-3}
315 - 400	$10^{-2} - 10^3$ $10^{-3} - 3 \times 10^4$	1 $10^{-3} \times t$ (10^{-3} W/cm ²)
The total power surface density over both wavelength regions must not exceed 1 W/cm ² .		

Tableau IV

Derived Working Limits for Energy Surface Density
Visible and near infra-red radiation (400 - 1400 nm) (Single pulse)

Eye (7 mm pupil)		Skin (average area 1 mm diameter)	
Pulse length, t (s)	DWL (J/cm ²)	Pulse length, t (s)	DWL (J/cm ²)
$10^{-9} - 2 \times 10^{-5}$	5×10^{-7}	$10^{-9} - 10^{-7}$	2×10^{-2}
$2 \times 10^{-5} - 10$	$1.8 \times 10^{-3} \times t^{\frac{3}{4}}$	$10^{-7} - 10$	$1.1 \times t^{\frac{1}{4}}$
Examples		Examples	
10^{-4}	1.8×10^{-6}	10^{-6}	3.6×10^{-2}
10^{-3}	1×10^{-5}	10^{-5}	6.4×10^{-2}
10^{-2}	5.7×10^{-5}	10^{-4}	1.1×10^{-1}
10^{-1}	3.2×10^{-4}	10^{-3}	2×10^{-1}
1	1.8×10^{-3}	10^{-2}	3.6×10^{-1}
10	1×10^{-2}	10^{-1}	6.4×10^{-1}
$10 - 10^4$	1×10^{-2}	1	1.1
$10^4 - 3 \times 10^4$	$10^{-6} \times t$	10	2
	(10^{-6} W/cm^2)	$10 - 3 \times 10^4$	$2 \times 10^{-1} \times t$
			$(2 \times 10^{-1} \text{ W/cm}^2)$

Tableau V

Derived Working Limits for Energy Surface Density
 Infra-red Radiation (1400 nm to 1 mm) (Single pulse)

Pulse length, t (s)	DWL (eye and skin) (J/m ²)
10 ⁻⁹ - 10 ⁻⁷	10 ⁻²
10 ⁻⁷ - 10	0.56 x t ^{$\frac{1}{4}$}
Examples	
10 ⁻⁶	1.8 x 10 ⁻²
10 ⁻⁵	3.2 x 10 ⁻²
10 ⁻⁴	5.6 x 10 ⁻²
10 ⁻³	1 x 10 ⁻¹
10 ⁻²	1.8 x 10 ⁻¹
10 ⁻¹	3.2 x 10 ⁻¹
1	5.6 x 10 ⁻¹
10	1
10 - 3 x 10 ⁴	10 ⁻¹ x t (10 ⁻¹ W/cm ²)

Tableau VI

Actions à mener

- Enquêtes permettant de déterminer effectifs et distribution des populations exposées.
- Etudes épidémiologiques sur groupes de travailleurs exposés pour détermination des effets éventuels à long terme.
- Réunir rapports sur cas d'exposition accidentelle pour meilleure description des troubles cliniques.
- Mise au point de dispositifs de mesure de l'exposition individuelle aux rayonnements non ionisants.
- Détermination de la relation énergie incidente-effet pour les divers organes sensibles aux différentes longueurs d'onde.
- Etude des moyens de protection.
- Harmonisation des grandeurs et unités utilisées pour la mesure de l'exposition et l'expression des limites.
- Harmonisation des normes de sécurité (normes fondamentales et limites dérivées).
- Harmonisation de la signalisation.
- Harmonisation des réglementations nationales (autorisations pour la fabrication, la vente ou l'utilisation).

D'où nécessité d'un organisme international
qui centralise résultats et élabore recommandations

Figure 1
SPECTRE ELECTROMAGNETIQUE

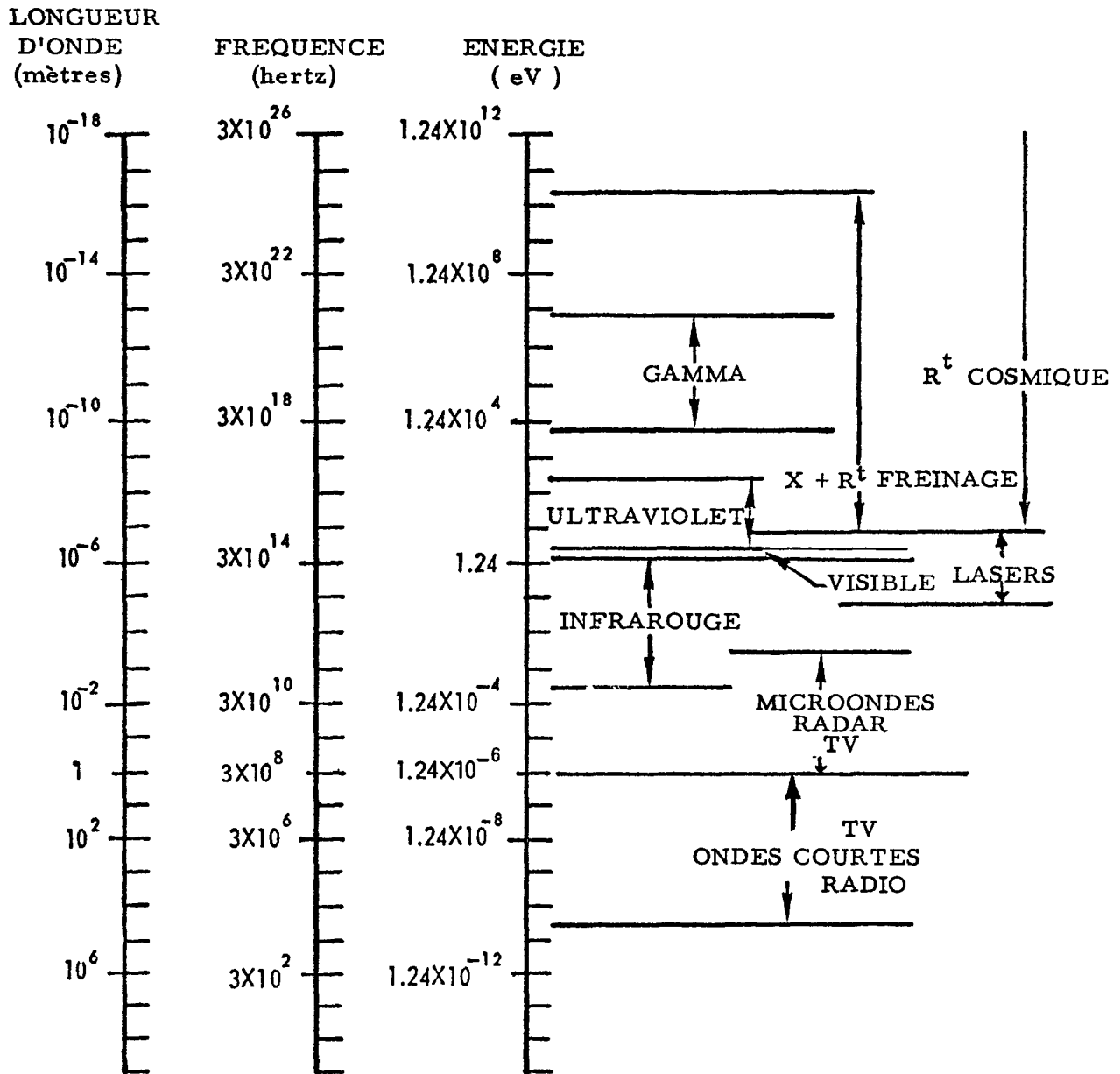


Figure 2

PENETRATION DES ONDES ELECTROMAGNETIQUES

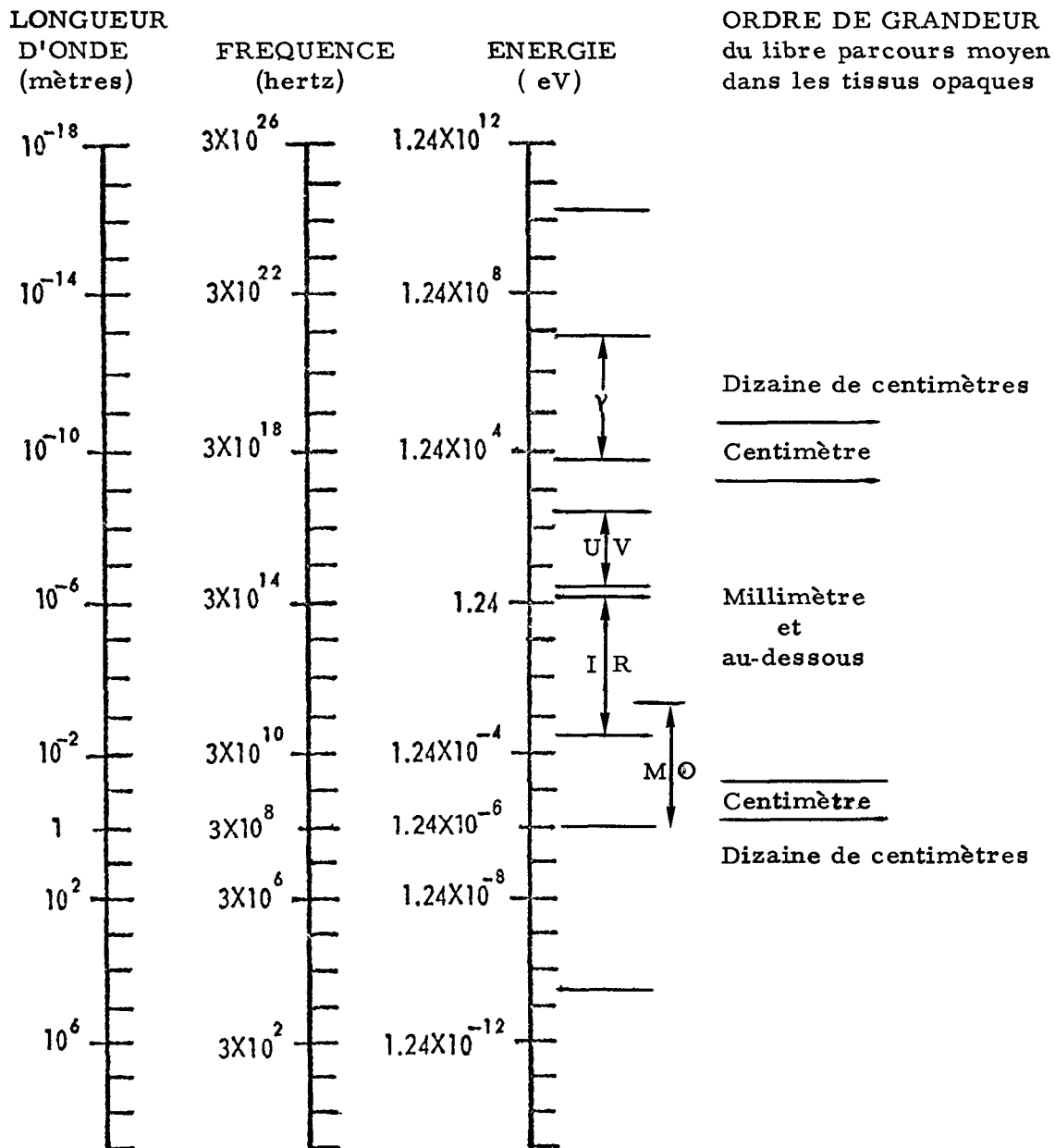
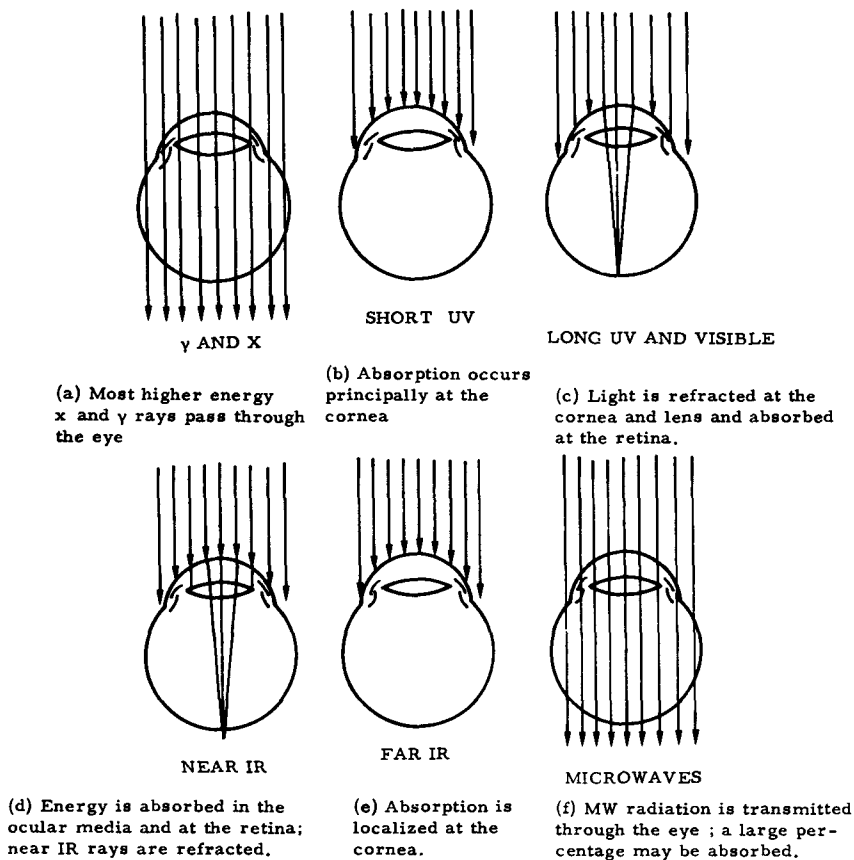


Fig.3

ABSORPTION PROPERTIES OF THE HUMAN EYE FOR ELECTROMAGNETIC RADIATION



from Laser systems - Code of practice, UK, 1969

PUBLIC INFORMATION, LEGAL ASPECTS,
EDUCATION AND TRAINING

RADIATION PROTECTION TRAINING COURSES AT E.I.R. WÜRENLINGEN, SWITZERLAND

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1. Summary

Due to the increasing applications of ionizing radiations and radioactive materials and required by radiation protection laws a growing number of professions and persons need at least a basic understanding of radiation problems and some practical instruction and training in radiation protection. The vast experience accumulated in nuclear research centers in the field of radiation protection training of employees on all levels is an excellent basis for the development of training programs for most needs. The program offered by the School for Radiation Protection of EIR (Federal Institute for Reactor Research), Würenlingen, comprises four categories :

- A) full-time health physics personnel of all levels
- B) professionally exposed personnel
- C) emergency organization personnel (fire departments, police, army etc.)
- D) students and pupils

Some typical or new courses are described in detail. The importance of such programs for improving the information and education of the public on radiation and safety matters is stressed.

2. The needs for radiation protection training

The needs for some kind of education and training in radiation protection can be grouped as follows :

- a) The persons directly involved and at risk, i.e. the professionally exposed persons in industry, medicine, research etc. are required by law to get a sufficient training. This includes not only the workers but also the responsible supervisors and managers.
- b) Persons that might be directly affected or involved in case of incidents, accidents or catastrophes (police, fire departments, transport organizations, emergency organizations, first aid and medical personnel, authorities, army, civil defense) must have sufficient knowledge and training which enable them to carry out their functions and responsibilities in case of radiation emergencies.
- c) The general public is only indirectly affected or at risk from radiation applications. But for a proper functioning as responsible citizens, opinion- or decision-makers, due to the growing concern for qualities of life and environment, and for political and economical reasons, a wide-spread and sufficient information and education of the public is highly desirable.
- d) As the reservoir for all the categories mentioned above, pupils and students on all levels and in all fields of education, training and instruction should get adequate information and basic education about radiation and safety matters during the regular curricula.

Although radiation protection originated in medicine, in most countries the nuclear research and industrial applications of radiation sources were the first areas where these training necessities were recognized and incorporated from the very beginning in the practical work and in the legal regulations. Prophylactic measures and training, safety and protection not only of the workers but also of the population and the environment were self-evident ways of thinking and operating for nuclear specialists long before the environmental protection "boom" reached politicians and opinion-makers. Today even medicine begins to catch up again with the good examples of the industrial and nuclear fields, and the demands for training of medical personnel on all levels add to the other needs for training of professional personnel and students. Thus there exists a large "training gap" in radiation protection for which the existing educational institutions are inadequately equipped and staffed and often lack the indispensable practical experience.

At present it is most urgent to give at least the legally required minimum of training to all those professionally exposed persons which are working already with radiation sources and have finished their formal professional training long ago. But as a long-range goal the incorporation of a sufficient training into the formal professional training and educational curricula must be attempted.

3. The role of nuclear research centers

It is a general tendency in most countries that the "old" established and rather well staffed nuclear research centers have lost a good deal of their original tasks to industry and have to reorientate their programs. A good way to utilize the vast experience accumulated in these centers and their staff is to enlarge the training programs in the nuclear field. Radiation protection is especially well suited for this purpose. Not only have the nuclear centers been among the first to practice radiation protection, but their experiences usually also comprise most fields of radiation uses, possibly with the exception of some medical applications. Nuclear centers offer the unique advantage that the teachers and instructors can work part-time in practical radiation protection or in research and development and thus keep up-to-date in the field and abreast with the latest developments, but also in close contact to the field requirements of practical health physics. Teachers in the purely educational institutions are much more liable to easily lose the contact with practice. The training programs provide also a refreshing and stimulating feedback on the research and development programs of a research center's health physics division and help to direct those towards the real needs which otherwise might be more difficult to recognize. For the individual health physicist who in his part-time function as instructor or teacher has to present his specialty in an easily understandable way to laymen, the healthy effects of such experiences should be stressed, for he is each time challenged to reconsider, explain and defend what he does and why.

4. History of radiation protection training at EIR

The Health Physics Division had to deal with training problems from its very beginning in 1957 because its entire staff had to be trained on site or by participating in the few international courses available at that time which usually did not concentrate on health physics alone but on nuclear science in general. The first health physics technicians got an informal and individual training from the health physicists, but after a few years, at the beginning of the Sixties, a training group with a full-time instructor was created. Members of the division collaborated also with outside course activities by authorities and educational institutions. By principle even the "full-time" instructor was required to do part-time work either in routine or development in order to keep in contact with practice. With increasing training activities he was assisted by part-time instructors for practical exercises, recruited from the experienced health physics supervisors and technicians. The advent of nuclear energy created additional needs for training of health physics and reactor operating personnel. Between 1958 and 1971 some

2700 persons of various professions and levels received a short or extended radiation protection training from or with the cooperation of our division.

In connection with the modification of the institute's program which particularly emphasized training as one of the three main pillars, the training program in radiation protection was reshaped and intensified, creating a frame within which most of the present and future needs for radiation protection training in our country can be fitted or supported. This led in 1972 to the reorganization of the training group under the new name of "EIR School for Radiation Protection", still as a part of the Health Physics Division. A large variety of training courses are in various stages of realization, some operational for a long time, others in the experimental phase, still others in the preparatory stages. The number of participants in the training program increased to about 670 in 1972 and further to about 700 in the first half of 1973.

5. Organization of the EIR School for Radiation Protection

The head of the school is a full-time instructor with a previous training as a health physics technician and an engineering school degree equivalent to a B.A. According to our principles he has also a function in routine health physics as head of the instrumentation and calibration group. He is responsible for the program of the school, the preparation of new courses and he teaches part of the theoretical topics. As assistant instructors for some theoretical teaching and most of the practical training he disposes of part of the working capacity of our health physics supervisors and some suitable technicians. This guarantees a practical instruction as close as possible to our practical experiences. The same supervisors and technicians spend part of their time assisting Swiss radiation protection authorities in the inspection of radiation protection conditions in hospitals, research laboratories, educational institutions and nuclear installations, including power stations, and cooperate in special protection problems outside the institute. Thus they have a broad experience and view to draw from for the training, but they get also useful feedbacks from the training and from discussions with course participants for their own work. Last but not least this brings our division in close, mutually interesting and beneficial contacts to almost all authorities, industries, institutions etc. which have any kind of radiation problems in our country. This will facilitate the introduction of similar viewpoints, philosophies and practices in all health physics applications.

6. Preparations for new courses

Before a new course can be given, many months of preparations are necessary. As a first step a concept is worked out giving the goals of the course, the topics to be treated and instructed, possible examinations, qualifications of participants, duration etc. This concept is submitted to authorities and interested parties for detailed discussion. Sometimes this requires that the radiation protection philosophy for a certain profession or application must first be developed. If possible we ask the responsible authority to approve and recognize the course officially as a sufficient introduction or training for professionally exposed persons or for the competence in radiation protection required by radiation protection regulation. Next the detailed material for the course is prepared, such as course texts, exercises, experiments, demonstrations, practical training, visual aids etc. and all participating instructors are familiarized with the course. The course is announced and propagated as early as possible. Up to now the response rather surpassed our most optimistic expectations. Once a course has been given for the first time, the propaganda made by the participants creates additional requests for repetition of the course or for new or modified types.

7. Execution of the courses

The theoretical instruction for the course types of similar levels has been standardized as far as feasible and split up into "building blocks" or packages

of topics which can be combined into any kind of course schedule or combination of subjects as desired, without having to work out each course entirely from the beginning. Only special topics required for certain courses need to be prepared individually, mostly for advanced courses that are not given all too frequently. The course texts are printed in large numbers, but on loose sheets, and are assembled into the requested combination and number of course textbooks for each course given. Changes and amendments of the texts are easy to make without delay or waste of outdated complete textbooks. Economics and flexibility are both optimized with this method.

In order to give all participants an efficient training in a minimum of time, courses with practical instruction are limited to 15 participants (with the only exception of the one-day monitoring course for army and civil defense where up to eight groups of ten men each can be handled in parallel). For the practical training the participants are further divided into two smaller groups.

Up-to-date methods are applied for instruction. The purely audiovisual method has been replaced by "simulation", which stands for "listen - look - touch - practice". The only purely audiovisual course is the short general introduction for new employees of the institute (D.1 Type D). Modern aids such as TV cameras and video recorder, overhead projectors, movies and slides are used. Detailed experiments, also simple, fundamental ones, and standard practical exercises facilitate the understanding of the often rather abstract subjects. We follow the development of modern didactic methods very closely and are ready to apply whatever will improve our courses. We cooperate with a new working group of the Fachverband für Strahlenschutz which has begun to treat problems of radiation protection training on all levels and which, as a first result, has collected an inventory of textbooks, training aids and training programs.

8. The program of the school

Four categories of training courses are offered :

- A) for full-time health physics personnel on all levels, from assistant or technician to supervisor, health physics engineer or health physicist. These are people from other technical professions which have to be retrained for health physics.
- B) radiation protection training for professionally exposed persons which have already completed their professional training or are further trained for a nuclear occupation such as reactor operator, radiochemistry laboratory technician etc.
- C) Personnel of emergency organizations, police, firemen, army, civil defense.
- D) Introductions to radiation protection fundamentals and practice for pupils, students and trainees in various types of schools and training programs.

All the above categories are further differentiated according to the level and previous training or experience of the participants, the type of their present or future occupation, the degree of competence or responsibility required etc. Appendix A lists the course program offered at present, while in the next section and in Appendix B four typical or novel courses are described in more detail.

9. Some examples of courses

9.1. Training of health physics technicians (A.1, A.2)

If candidates for health physics technicians have had no previous training and experience in radiation protection, they have to begin at the bottom of the program with an introductory course. An extended version of this course is also given to health physics assistants, a category of auxiliary personnel, usually without other formal professional training, which we employ for simpler routine tasks such as wipe tests, decontaminations etc. The candidates for technicians, at least those for our institute, have been selected in cooperation with the Institute for Applied Psychology, Zürich. There they undergo a special aptitude test developed by a psychologist who had been one of our first health physics tech-

nicians before he began to study psychology.

The introductory course gives the candidates a first practical idea of the type of work while it allows us to judge the aptitude of the candidates and to employ them as assistants for some time, if for other reasons the following parts of the training must be delayed for some weeks. The 2 weeks' introductory course A.1 is followed by the 12 weeks' basic course A.2, divided into six basic and practical parts (details see Appendix B). The course A.2 is terminated by examinations, written and oral ones in theoretical knowledge, and a practical examination consisting of the preparation and execution of a routine health physics operation. The successful completion of these examinations qualifies the man for employment as health physics technician and for the advanced course A.2 of 4 weeks duration. He will usually take the advanced course after some weeks or months of practical experience in routine operations, and the course familiarizes him further with special monitoring techniques and the interpretation and application of the radiation protection regulations.

How far a technician will climb on the ladder of standardized careers in health physics, provided he is employed by EIR, depends on the results of the practical experience, additional training and examinations, and of course his personal qualifications, professional accomplishments etc. The chances to follow an interesting career that in some cases may lead up to the level of an engineer is of great help in finding and retaining good personnel. The flexibility of our system allows to make optimal use of each man's qualities and to offer him a job which fits his interests and capabilities without becoming boring or stagnant.

9.2. Radiation protection training for mechanics and workmen (B.3)

This course, whose details are given in Appendix B, was originally started for mechanics and other workshop personnel who only infrequently had to do with contaminated or activated components from reactors or laboratories. The start of the first nuclear power stations in the country and the experiences during the first shut-downs quickly showed the necessity for training of all the operating and maintenance personnel in this type of course, and its content was accordingly adjusted. The course lasts one week and aims at giving the participants a practical understanding of radiation protection, the ability to work properly in controlled zones and under elevated levels of external radiation and contamination in such a way that they can protect themselves and do not endanger their coworkers. The use of protective clothing, of simple instruments for working place monitoring, and the reaction in case of incidents are instructed, thus enabling them to carry out routine tasks and repairs without direct control by a health physics technician.

9.3. Radiation monitoring course for police and firemen (C.1 Type B)

This course type originated from concerns that grew within the fire department of Zürich airport after a crash landing of a foreign passenger plane, reports on the Palomares incident, and after the crash of another plane in Eastern Europe whereby a radioisotope cargo got lost. The firemen's problem was whether rescue operations for passengers might have to be stopped or delayed and could present an unacceptable risk for the firemen if radioactive cargos were on board. The more general problem of what to do in case of a transport accident or a fire involving radioactive material applies also to police, ordinary fire departments of cities or industries, transport personnel etc. The first set of courses was given with excellent results for the fire department of Zürich airport, where the one week course will be followed at least once a year by some practical exercise and repetition, supplemented by internal repetition as part of the routine training of the firemen. The first part of the course aimed at giving an idea and a "feeling" for radiation dangers and relative risks. The institute's isotope production division provides excellent and realistic illustrative material and demonstrations. Monitoring of radiation and contamination and emergency measures form the main practical part of the course. From the first participants rumors spread quickly to other

fire departments and police, and it looks as if this course might become a frequent feature of our program.

9.4. Radiation monitoring exercise for army and civil defense (C.2)

Army and civil defense have excellent and efficient training programs for part-time and full-time specialists in nuclear defense (AC) and monitoring. The training of AC officers is handled in central courses by professional instructors, while the soldiers and corporals (several in each unit) are trained in field courses by the AC protection officers of the regiments and divisions. Civil defense operates on similar principles. Training programs and material are excellent and efficient, but the army lacked possibilities for practical training with real radiation sources under simulated field conditions. Following a request and developing ideas of an AC officer of a division we improvised a first exercise with good success. In close cooperation with army instructors and AC officers we created a one day monitoring exercise with practical demonstrations on radiation levels and shielding, contamination and decontamination, and monitoring of a "fallout" area under field conditions. The four exercises of about one hour each are executed under the direction of the AC officer of a regiment and his staff and only supervised by our technicians.

The second part consists of a visit to the institute, a movie on nuclear energy, informations and discussions on radiation protection in civil applications, environmental effects of nuclear energy etc., thus providing some basic information useful for civil life and an opportunity to discuss problems that play quite some role in politics and newsmedia at present. As we give this course to several hundred army and civil defense personnel each year, with an increasing frequency, this may develop into an effective contribution to a better information of the public, because all these people return to civil life after some days or weeks of military or civil defense service and come from virtually all regions, social and professional groups of the country.

The individual doses received during the exercise are registered by direct reading dosimeters and amount in the average to a few millirem. During the information part the significance of these doses is explained and related to doses causing acute effects, to the natural background doses and the doses expected from radiation applications and nuclear energy. Such a personal experience helps to put radiation in proper perspective for the participants of the courses, they have also got the experimental proof for the efficiency of the protective measures. Results are an increased confidence, diminished anxieties about radiation hazards, and a better basis for rational judgements.

10. Conclusions

Duration, topics and content of our courses are not yet in all cases what we judge optimal, sufficient or necessary, but they show what can be realized and required under present circumstances in various fields and professions with the acceptance and support of authorities, employers and participants, and on which lines a further development and improvement will be possible.

We hope to have shown how nuclear research centers with relatively limited means can provide valuable training opportunities in various fields, cover a good part of the training needs in radiation protection, and contribute efficiently to a better information and understanding of radiation protection and nuclear safety problems in professional groups and larger segments of the public. This may improve the chances for survival of such centers and their health physics staff and provide interesting and satisfactory opportunities for health physicists and the future development of health physics.

Appendix A

List of courses offered by the EIR School for Radiation Protection

A) Full-time health physics personnel

A.1. Radiation protection course for health physics assistants

duration : 4 weeks for operations assistants, 2 weeks for technician candidates.
participants : full-time auxiliary health physics personnel
remarks : required introductory course (2 weeks) for candidates for health physics technicians who will continue with course A.2.

A.2. Radiation protection course for health physics technicians

Basic course A.2 :

duration : 12 weeks
requirements: Course A.1 passed, formal professional training in technical fields (mechanics etc).
participants: candidates for full-time health physics technicians
examinations: theory written and oral, practice.

Advanced course A.2 :

duration : 4 weeks
requirements: courses A.1 and Basic A.2 + examinations passed
participants: health physics technicians
remarks : for details see section 9.1 and Appendix B.

A.3. Radiation protection course for health physics supervisors

duration : 12 weeks
requirements: examined health physics technician with several years of practical experience, qualifications for chief function,
participants: health physics technicians
examinations: theory written and oral. Practice: complete evaluation, preparation and execution of a large health physics operation.

A.4. Radiation protection course for health physics engineers

duration : 8 weeks
requirements: diploma of a higher technical institution (engineering school) + autodidactic study of courses A.2 and A.3, if possible during stage at our health physics division.
participants: future heads of operational health physics sections in nuclear power stations, laboratories or authority inspection groups
examinations : theory written and oral, practice.

A.5. Radiation protection repetition course for health physics technicians

duration : 1 week

A.6. Radiation protection repetition course for health physics supervisors

duration : 1 week

B) Professionally exposed persons

B.1. Radiation protection course for radiochemistry laboratory technicians

Type A : Basic course (short course)

duration : 1 week

participants: laboratory technicians and auxiliary personnel

Type B : Advanced course

duration : 2 weeks

participants: laboratory supervisors and technicians

B.2. Radiation protection course for factory-inspectors

duration : 3 weeks

participants: cantonal and federal factory-inspectors or persons responsible for radiation protection and industrial hygiene in industry

B.3. Radiation protection course for mechanics and workmen

duration : 1 week

participants: professionally exposed workers employed for assembly, maintenance, repairs, shut-down operations of nuclear components and installations.

remarks : for details see section 9.2 and Appendix B

B.4. Radiation protection course for technical X-ray personnel

duration : 1 week

requirements: fundamentals (physics and technology) of X-ray equipment

participants: sales-, repair-, assembly- and operating personnel of X-ray equipment for industrial or medical applications

C) Emergency organization personnel

C.1. Radiation protection monitoring course

Type A : EIR emergency organization

duration : 2 days

participants: EIR emergency organization members

Type B : Professional emergency teams

duration : 1 week

participants: fire department chiefs and officers, professional firemen of airports and cities, police etc.

remarks : for details see section 9.3 and Appendix B.

Type C : Enclosed radiation sources (handling of incidents)

duration : 2 days

participants: workers who operate or are responsible for enclosed radiation sources

Type D : fire-fighting in radiochemistry laboratories

duration : 1 day

participants: voluntary and industrial firemen

C.2. Radiation monitoring exercise for army and civil defense

duration : 1 day (8 hrs)

requirements: formal military or CD training in AC defense

participants: radiation monitoring and protection personnel and officers of army and civil defense

remarks : for details see section 9.4 and Appendix B

D) Introductory courses of general nature

D.1. Radiation protection introduction

Type A : professionally exposed personnel

duration : 2 days

participants: new employees to become professionally exposed

Type B : Educational institutions

duration : 3 hrs

participants: students of higher educational institutions

Type C : Hospital personnel

duration : 3 hrs

participants: professionally exposed personnel of hospitals (including M.Ds)

Type D : new EIR employees

duration : 1 hr

participants: all new employees of EIR, whether professionally exposed or not, including all auxiliary or temporary personnel

E) Radiation protection as part of nuclear training courses

E.1. Radiation protection course for reactor operators

duration : 2 weeks

requirements: complete training as reactor operator in EIR Reactor School

participants: candidates for operators licence

E.2. Radiation protection course for reactor shift supervisors

duration : 1 week

requirements: licensed and experienced reactor operator

participants: candidates for shift supervisor examination (EIR Reactor School)

remarks : E.1 and E.2. are limited to practical instruction, the theoretical training is given by the Reactor School

E.3. Radiation protection course for nuclear engineers

duration : 1 week

requirements: nuclear engineering lecture program

participants: students and graduates of engineering schools

Additional radiation protection courses for incorporation into other curricula of higher education or professional training in science, technology and in the medical field are being discussed and prepared.

Appendix B

Examples and details of some courses

1) Courses A.1 and A.2 : Training of health physics technicians

A.1. Introductory course (2 weeks)

aim : Review of the fundamentals and basic philosophy of radiation protection and of the routine tasks of a HP technician

organization: 14 lectures with short preparatory theories and main emphasis on practical demonstration and exercise of the routine tasks

topics: - fundamentals: radioactivity / radiation, sources, hazards / basic principles of radiation protection / radiation monitoring, doses, dosimetry / contamination, decontamination / waste problems.

- practical training: marking of controlled zones / radiation monitoring

instruments and dosimeters / protective clothing / contamination monitoring and decontamination / waste collection in controlled zones / behavior in case of incidents.

A.2. Basic course (12 weeks)

aim : complete training of health physics technicians. Knowledge of fundamental physics and technology required for health physics. Mastery of all routine tasks and methods for working place control

organization: 6 parts: 3 basic, 3 practical. The fundamental lectures are illustrated by experiments. Exercise of simple calculations. Main emphasis on mastery of radiation monitoring methods.

topics: Basic instruction 1 : fundamentals of nuclear physics (2 weeks)
composition of matter / radioactivity / radiations / radiation interactions with matter / dose definitions / shielding

Basic instruction 2 : radiation monitoring methods (2 weeks)
electrical and electronic fundamentals / principles of measuring/ principles of radiation detection / ionization chambers / proportional counters / GM counters / solid state detectors / scintillation detectors / chemical detectors / detection and monitoring of α , β , X, γ , n.

Basic instruction 3 : hazards of ionizing radiations to man (3 weeks)
radiation exposures of man / biological effects of ionizing radiations / external exposures / internal exposures / maximum permissible doses and derived working levels .

Practical instruction 1 : health physics at nuclear reactors (1 week)
physics, technology of nuclear reactors / reactors as radiation sources and their hazards / risks of nuclear reactors.

Practical instruction 2 : HP in radiochemistry laboratories (1 week)
fundamentals of radiochemistry / installations and organization of radiochemistry labs / types of work in a laboratory / radiation hazards and risks in radiochemistry laboratories.

Practical instruction 3 : operational health physics methods (3 weeks)
routine methods (monitoring etc) / methods as specified by HP operational manuals / individual handling of routine operations and problems at reactors and in laboratories, under supervision of an experienced HP technician.

examinations : Theory and fundamentals : written and oral
Practice : complete health physics control of a routine operation in a controlled zone. Several experts from the school, the HP division or other EIR divisions or from authorities are present.
The examinations are formally acknowledged with a diploma.

A.2. Advanced course (4 weeks)

aim : repetition and advanced treatment of basic course A.2 topics. Special monitoring methods. Interpretation and application of the federal radiation protection regulations for special cases.

organization : introduction of new topics followed by discussion of practical situations, aimed at integration of theory and practice.

topics : those of basic course A.2 / air monitoring / background compensation / activity measurements / radionuclide identification and analysis / spectrometry / DWL derivation / practical problems.

remarks : for the technicians of the SIN accelerator the basic course A.2 was supplemented by an additional practical course on accelerator problems.

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2) Course B.3. Radiation protection for mechanics and workmen (1 week)

aim : See section 9.2. Ability to handle practical radiation protection problems when working in controlled zones

organization: as course A.1.

topics : - fundamentals : short version of topics of course A.1. / evaluation of hazards and protective measures when handling and machining contaminated or activated objects.

- practical : monitoring instruments and methods for external radiation and surface contamination / behavior in controlled zones / installation of temporary zones / protective clothing / active working methods / decontamination of persons and material / waste handling/ transport regulations / practical exercises in active workshop.

3) Course C.1 Type B : Radiation protection monitoring for emergency teams(1 week)

aim : See section 9.3. Mastery of handling the first emergency actions at the site of a transport or other radiation incident. Fencing off the site, cooperation with special teams, minimizing of risks for public and environment, self-protection during rescue etc.

organization: as course A.1.

topics : fundamentals : as course B.3 with main emphasis on emergency situation

practical : monitoring instruments and methods / behavior under high radiation and contamination levels / protective clothing / practical exercises of simulated accident situations.

4) Course C.2: Radiation monitoring exercise for army and civil defense (1 day)

aim : Demonstration and realistic, but separate exercises of problems occurring in a fallout situation. Information on civil radiation protection and nuclear safety.

organization: groups of max. 10 men, rotate between 4 posts for 4 hours, second part (visit etc) four or all groups together.

topics : - post A : decontamination as a "household" problem. Contamination and coarse decontamination of military clothing with simulated fallout (Tc-99m labelled Al_2O_3 grains with diameters of 70 - 150 μm).

- post B : liquid contamination and decontamination of shoes and hand. Spreading of liquid contaminant (Tc-99m labelled glycol), decontamination with water, soap and brush.
- post C : Shielding factors and inverse square law. A Cs-137 source in a rectangle of walls of earth, wood and concrete, fourth side open, is monitored from all directions at various distances.
- post D : Radiation monitoring of a simulated fallout field. On an area of 2000 m^2 a reduced scale model (1:80) of routes for monitoring teams is laid out, spiked with camouflaged Cs-137 sources of various activities, simulating an extended contamination. The 2 man teams with ordnance equipment have to monitor a total of 41 points, calculate H+1 hr normalized dose rates and draw the contamination map on the real topographical map, control personnel doses etc.
- Part II : Introduction to civil radiation protection and regulations, movie on nuclear energy, visit to EIR reactors with demonstration of operational health physics, environmental monitoring, waste treatment, protective clothing etc. Discussions according to particular interests.

PROJECT TRAINING FOR GRADUATE STUDENTS IN HEALTH PHYSICS

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ABSTRACT

The Royal Naval College has conducted graduate training in health physics for over 10 years and has developed a system of project training which gives students practical experience in the context of the formal theoretical course. The purpose of project training is to provide realistic radiological protection problem solving at a time when the student is consolidating his theoretical knowledge.

This paper describes the procedures used to introduce, supervise, and assess these projects.

The projects are initiated early in the course by presentation of a package which includes a statement of the aims of the project, copies of relevant papers, apparatus manuals, radioactive source calibrations and guide lines for progression of the project. The student is required to write a report on his project which is presented orally and staff appraisal takes account of all aspects of the students project work.

The paper evaluates the success of this approach against the cost in staff and equipment resources required to achieve realistic and effective training.

INTRODUCTION

The Royal Naval College, Greenwich, has conducted graduate courses in health physics for over 10 years in association with other specialist post-graduate courses. The Department of Nuclear Science and Technology, which is responsible for these courses, was founded in 1959 to provide a centre for Naval Nuclear Education and Training of officers and now offers over 18 courses per year to a total of about 250 students. All these courses contain an element of health physics. The necessity for health physics in the training of reactor engineers is already well recognised² since they must understand the hazards of radiation and, as reactor operating personnel, they will receive the largest doses during reactor plant down time for repair or preventative maintenance. The experience on which this paper is based was gained over the last 10 years in two specific graduate courses, one of 12 weeks duration and the other 24 weeks.

GRADUATE COURSES

The first course, the Nuclear Radiation Protection Course (NRPC) has

already been reported³ but more recently another graduate course, the Nuclear Reactor Course (NRC), has been extended to include radiological protection as an essential qualification. The NRPC is recognised by the British Institute of Physics and the successful NRC candidate also receives the Postgraduate Diploma of the Council for National Academic Awards.

DEPARTMENT FACILITIES

The facilities available for training in the Department of Nuclear Science and Technology include the 10 kW training and research reactor JASON⁴ which was the first to be installed in any educational establishment in the United Kingdom. This has given the staff considerable first hand experience and set a pattern in terms of safety documentation, procedures and applications to other Universities. The Reactor is used to bridge the gap between simulator training and full power reactor training and provides a source of radiation which is used in many supporting training experiments and projects. In particular it provides a realistic environment for students to learn how to handle, survey and control sources of ionising radiation. The reactor is used for 49% of the time for student training and diverges to power over 480 times per year. Over 2000 students have been trained and currently the Department provides 8 different types of courses. The research utilisation of the reactor includes several applications of activation analysis and reactor dynamics in addition to specific health physics studies such as reactor shielding⁵ and the study of radioactive aerosols⁶ and some dosimetry studies.

PROJECT TRAINING

The interdisciplinary nature of environmental health engineering has been emphasised by the World Health Organisation⁷ in their booklet on The Education and Training of Environmental Health Engineers, which states that many disciplines may be required for the solution of complex environmental health problems.

This generalisation is specially true in health physics; team operation is the rule and the individual member must be familiar with the vocabulary, techniques and goals of other members of the team.

This paper describes an approach to graduate training in health physics which is designed to develop the student's awareness of other disciplines, and to give him practice at problem solving under realistic conditions. The two courses on which the experience was gained are being reviewed by objective training analysis which requires the definition of an Operational Performance Standard. This is translated into a Training Performance Standard which provides the basis for the detailed course design. All this demands effective feedback on performance of past students and also of the reactor plants on which they have worked. The replacement of conventional set practical sessions by a smaller number of set practicals and project work has emphasised the need for careful attention to the objectives of the course. The projects themselves must be well organised to ensure full benefit for the student and effective appraisal of his success.

PURPOSE OF PROJECT TRAINING

DEFINITION OF A TRAINING PROJECT

A training project is a supervised task set by the students' tutor to meet some objectives of the course. A good project must be interesting to the student but limited in scope so that it can be completed within the time allocated. The project must be realistic to permit the student to recognise

a 'real problem'; novelty and relevance provides the necessary motivation. To achieve these conflicting features the development of training projects relies to a large extent on feedback from operational establishments and spin off from departmental research projects.

The purpose of project training must be examined in relation to the overall objectives of the course. A common feature in the objectives of health physics courses is the achievement of three important attributes which the successful student must possess. The student must:

- a. have a sound knowledge of the process which produces the hazard,
- b. understand the hazards of radiation,
- c. have sufficient appreciation of practical problems to be rapidly accepted into the operating team.

The development of these attributes is an important objective of the course but it can not be met entirely by classroom instruction, especially when the students themselves are practical men. Conventional practical work can be designed to re-inforce classroom instruction or to help students familiarise themselves with equipment and techniques, but it may stultify the students interest. When practical work has to be allocated in short periods of a few hours at one time, the student will rarely have the opportunity to use an interdisciplinary approach and the stereotyped exercise limits his scope for problem solving. On the other hand, set practicals are straight forward to administrate, and it is comparatively easy to assess the students performance against that of his colleagues because the work expected is identical.

PROJECT OBJECTIVES

The approach described in this paper replaces most of the set practical sessions by more broadly specified projects designed for the following purposes:

- (1) To apply the student's theoretical knowledge acquired from the course work to problems associated with the operational situation.
- (2) To give direct experience of relevant health physics practice.
- (3) To emphasise the interdisciplinary nature of health physics.
- (4) To assess the student's ability to solve problems under realistic conditions and his ability to communicate his observations and recommendations.

The first two objectives replace the set practical but added motivation can be imparted when the project contains an element of novelty. The third objective requires an input from other disciplines such as reactor physics, reactor engineering, chemistry, metallurgy and radiobiology. This is important to ensure that the student is made aware of the relevance of his work to overall plant safety. The last objective reveals the special advantage of project training and, to achieve it, the class have to share experience gained on the individual projects by participation in a formal presentation of the project reports.

PROJECT MANAGEMENT

The introduction of project work and its subsequent extension to become a significant proportion of Course time necessitates the designation of a Project Manager. The Project Manager is responsible for coordinating individual project supervisors to ensure the satisfactory progress of the projects. These supervisors are required to carry out the following tasks:-

- (1) The production of the project outline (in association with the Project Manager).
- (2) The day to day supervision of the project.
- (3) The timely presentation by the student of a project report and the provision of guidance as to the required standard.
- (4) The assessment of the report in conjunction with at least one other member of staff.

PROJECT PACKAGE

The project work described in this paper is given to students undergoing relatively intensive training in which the duration of projects is strictly controlled. It is therefore essential that the objective and project outline are clearly defined. The use of a 'project package' has been developed in an attempt to maximise the benefit to the student.

The project package is required to:

- (i) Provide the student with sufficient information on which to make his choice of project.
- (ii) Ensure that sufficient staff work has been undertaken to permit the completion of a worthwhile project.
- (iii) Enable a comparison of the project proposals to be made in terms of the level of the work involved.
- (iv) Provide the Project Supervisor with the basis for monitoring the progress of the project.

The project package includes:

- (i) The background information necessary to place the project in context.
- (ii) The objective of the project.
- (iii) The schedule of apparatus to be made available and the apparatus manuals.
- (iv) The guide lines for initiation of the project.
- (v) Selected reference material.

Having made his choice, the student is provided with sufficient information to commence some basic reading and planning before any further discussions are held with the supervisor. In this way the student is able to take some part in the initiation of a project. A typical project package is summarised in Appendix 1.

PROJECT SELECTION

Three factors are considered in the selection of the individual projects:

- (1) The previous experience of the individual student;
- (2) the future task in which student will be engaged;
- (3) the student's performance.

It is sometimes possible to remedy obvious deficiency in the student's past experience by the selection of an appropriate project. Alternatively the student's best interests may be served by completing a project relevant to his

future commitment. The student is encouraged to discuss the various projects with the supervisors so that the selection also reflects his personal interests.

PROJECT SUPERVISION

The responsibilities of the project supervisor would appear to diminish once the project is launched since the student has sufficient background information. Experience has shown that the supervisor must act in an advisory capacity, reviewing progress at intervals determined by the duration of the project. In this context the student-supervisor relationship is important, and an informal approach has been found to provide the ease of communication which is an essential part of project work.

In the event of a major equipment failure, or a particularly interesting unforeseen development, the supervisor may redirect the project to capitalise on the situation. The project work culminates in a formal report and the supervisor is required to review the draft report and make constructive suggestions and query any doubtful aspect of the student's work.

PROJECT APPRAISAL

Within one course all the projects may have different main topics and therefore different supervisors. Although the expected work content can be assessed by the Projects Manager, the problems arising in the execution of the project may require a different work content and the demands of the individual supervisors cannot be standardised. This situation may generate some difficulties in achieving a fair comparison of each project. The supervisor is therefore made responsible for the preparation of a written summary of the report which takes account of these variable factors.

The project which carries up to one third of the course marks is finally evaluated under the following three headings:-

- a. Methodology and Practical Work. The orderliness of the approach adopted by the student and the design and execution of the project is examined to establish his penetration of the problem.
- b. Written Report. This is examined to provide further appreciation of the student's reasoning powers and orderliness in presentation.
- c. Oral Presentation. This reveals the student's ability to exercise judgement in the selection of the important aspects of the work and his performance in answering questions on the project demonstrates the depth of knowledge and comprehension. Since all the Course members and the examiners are present at the oral presentation, it serves to acquaint the other students with the subject and helps to share particular lessons that have been learnt.

EVALUATION OF PROJECT WORK

STAFF RESOURCES

At first sight the project approach to training appears to be more costly in both time and resources than formal practical work. This depends on the size of the course and the degree of utilisation of the laboratory equipment. For example, 6 groups of students could perform a set practical either (a) simultaneously and therefore requiring 6 sets of equipment, or (b) in three separate sessions requiring 2 sets. The solution adopted is usually constrained by the overall course programme, which determines the timing of the practical sessions. Since the RNC courses involve practical work in several topics it

has been policy to arrange several different practical sessions in parallel, the students completing each experiment in the various laboratories. Hence the laboratory utilisation achieved is dependent on the size of the Course, but on the average is equivalent to 3 sessions per experiment per course. This policy has ensured that, with the exception of the very low cost equipment, only a limited number of sets of equipment are required.

The development, documentation and updating of the conventional practical training also requires considerable resources and Tables 1 and 2 compare for the same allocation of practical time, the total man-hours of staff time for formal practical and project work. The Tables are based on 8 students on the course with the breakdown of laboratory work being typical, rather than specific to any one course: development costs are calculated on the assumption of a review of all experiments after every 2 to 3 courses.

TABLE 1. CONVENTIONAL PRACTICAL

TASK	STAFF HOURS	SESSIONS	NUMBER OF PRACTICALS	TOTAL TIME HOURS
Administration	20	1	-	20
Development	3	20	-	60
Basic Radiation Protection	6	2	10	120
Basic Radiation Physics	6	2	4	48
Basic Reactor Physics	8	2	4	64
Simulator	4	2	2	16
Examination of Practical work	1/5	8	20	32
TOTAL TIME:				360 hours

The calculation for the project work is based on 8 students each undertaking a different project, with the support of some basic introductory experiments and is chosen to illustrate the maximum staff effort. Some formal practical work must be included to ensure that the student is familiar with the basic radiation laboratory procedures and techniques. These costs could be reduced by combining students into groups of 2 or more for each project but this removes some of the advantage of project training.

TABLE 2. PROJECT WORK

TASK	STAFF HOURS	SESSIONS	NUMBER OF PRACTICALS	TOTAL TIME HOURS
Project Administration	3	1	8	24
Development and Consultation	2	1	6	12
Planning and Preparation	10	1	8	80
Supervision	15	1	8	120
Assessment	4	1	8	32
Presentation	3	1	8	24
Basic Radiation Protection	6	2	2	24
Basic Radiation Physics	6	2	2	24
Basic Reactor Physics	8	2	2	32
Examination of Practical work	1/5	2	6	3
TOTAL TIME:				375 hours

These figures which are based on several years experience demonstrate that the cost in terms of man hours differs little between the two alternative schemes. In the planning and preparation of projects the specialised experience of the supervisor is used and this is more stimulating for him than the

development of set practicals to achieve more limited objectives. A survey of the equipment resources required for mounting the work described in Table 2 has shown that as a result of the extensive use of project equipment in research and the flexibility in planning a reduction of 25% in the capital equipment cost was possible compared to conventional practicals.

COMMENTS ON THE STUDENTS RESPONSE

STUDENT REACTION

The total time allocated for participation in practical work in set practicals and in projects is the same. However, the student reaction to the laboratory work in each scheme is noticeably different.

The students on intensive courses will quickly form opinions on the relevance and necessity of the practical work and will reject unnecessary duplication in the presentation of the material. Set practicals with the usual close relation to the lecture material are frequently rejected as repetitive. In contrast projects have stimulated student interest and participation and help to ensure that the objectivity of the course stands up to close scrutiny by the students. One measure of the success of the project is the amount of additional time a student may be prepared to devote to the work; in fact it is common for the supervisor to have to ensure that the student does not spend an excessive amount of time on the project.

STUDENT ATTAINMENT

The effectiveness of any instructional technique in attaining some part of the course objective cannot easily be objectively assessed and subjective assessments tend to vary widely. The ultimate test is to follow up the students when they have moved on to their operational role. If the staff effort is available the students are interviewed in their work area at least one year after completing the course. Students do appear to move smoothly into their operational task, in some cases continuing to follow up the project topic as a centre of interest in their new job. This itself is a convincing demonstration of the value of this approach to the student.

CONCLUSIONS

The procedures described in this paper are offered as a product of systematic course design which could be applied to other graduate courses in health physics. The two courses on which this work has been developed are relatively long - 12 weeks and 24 weeks respectively - but it is considered that the method can be applied with success to shorter courses. The reliability of the apparatus used and the provision of guidelines for the project becomes important if the student is not to waste valuable time coping with instrument faults or re-discovering relatively unimportant information. In all cases the supervisor must ensure that there is sufficient scope for problem solving and that the project does not relapse into meaningless collection of data. The experience and judgement of training staff are taxed more heavily in the project supervision than the more passive role of monitoring a set practical. In many cases the student gains his first rigorous experience in scientific communication when he prepares his project report and presents it to his colleagues and the examiner. It is probably this increased demand on the supervisor and student which ensures the success of this approach to health physics training.

PROJECT PACKAGE

The package contains the following main items:-

1. Objective of Project
2. Background information
3. Equipment and Services supplied
4. Guide lines for progress
5. Literature references and Instrument Manuals

To illustrate this the following is an abbreviated package for a Thermoluminescent Dosimetry Project:-

1. Objective: To investigate factors affecting the precision of TLD-700 for dosimetry in low level photon and neutron fields.

2. Background: The operation of a nuclear reactor involves the staff in radiation exposure to mixed photon and neutron radiation fields. Whilst the radiation levels may be low compared to the legal limits there are both ethical and practical reasons for aiming at high precision in these measurements. For example, the success of a shield design may be judged by the man rem accumulated by the operating staff or a new reactor plant may be surveyed by short duration exposures of TL dosimeters.

3. Equipment and Services Supplied:

- i. Access to the research reactor
- ii. One Dynatron TLD reader modified to give a graphical display of glow curves
- iii. Annealing ovens
- iv. Dispensing equipment for powder, extruded chip and disc forms of TLD

4. Guidelines:

- i. Delineate radiation fields to be studied taking account of operational conditions on a power reactor against the closest approach available on the research reactor.
- ii. Gain familiarisation with TLD equipment by trial runs on irradiated samples
- iii. Assess the number of tests feasible in the time available and schedule test points
- iv. Execute selected measurements, read and analyse data for precision
- v. Run subsidiary experiments after discussing (iv) with Supervisor
- vi. Write draft report

5. Literature References and Instrument Manuals:

References and manuals are revised and amended for each project.

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THE LAW AND LOW LEVEL RADIATION
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Abstract

One of the responsibilities of the health physicist is to recommend and enforce radiation protection standards based upon recognized occupational radiation exposure guides. It follows, therefore, that the health physicist, because of his background and technical knowledge, will play a key role in evaluating a claim allegedly arising out of exposure to ionizing radiation.

It is the purpose of this paper to examine a number of latent radiation injury cases with particular emphasis on the kinds of radiation records offered in evidence, the nature of the expert testimony by both the health physicist and the medical expert and the conclusions of the court or Board in the final adjudication of the claim. The paper will also explore the views of those who hold that the present legal system in the United States is not appropriate for the handling of latent injury claims.

Introduction

It is well known that in spite of the highly successful efforts of those engaged in the field of radiological health, radiation workers run a risk of being exposed to some degree of radiation in the course of their employment-- however small that risk may be. Radiation workers may also develop certain diseases that are known to be caused by radiation but which also develop spontaneously in the absence of radiation exposure. What happens in the United States when a radiation worker sues for compensation on the basis that the disease from which he is suffering was incurred in the course of his employment?

In cases involving substantial exposure, recovery is almost always assured. However, in cases involving delayed injury, the claimant rarely recovers. Should he?

The purpose of this paper is to examine this question.

In order to understand the reasons why so few cases involving low exposures and delayed occupational injuries are compensated, one must appreciate that it is first necessary under our legal system to determine whether the exposure "caused" the injury. It is also necessary to draw a distinction between the two categories of radiation injury cases: (1) Claims involving acute effects which appear immediately or within a short period after a very large exposure; and (2) Claims involving latent effects which do not manifest themselves until years later. In the first instance they offer the litigant, the expert

witnesses and the courts little difficulty. Dosimetry can be reconstructed, and the symptoms or illnesses are usually characteristic. Because the nexus between the pathology and the disease is obvious, causal relation is relatively simple to establish. It is the second category of cases that pose problems of causation. Why?

Causation

Simply stated, it is because the proof of causal connection in a claim for injury, if we accept the medical notion of the meaning of the term, is, in most instances, extremely difficult to establish.

In a radiation claim both the physician and the court must confront the problem of causation. When the medical expert is asked to testify concerning the cause of the claimant's pathology it frequently develops that his testimony is so couched with reservation that the court or board has no alternative but to make the medical determination on its own. The reason for this hesitancy or inability on the part of the medical profession to find causation lies in its notion of the meaning of causal connection. Doctors define causation in a special sense. They prefer to base their conclusions on statistical studies of the relationship between a suspected causative factor and the disease.¹ In 1969 an outstanding authority on workmen's compensation pointed out that longitudinal studies, using an exposed population group and one or more control groups are a principal device for testing causal theories involving human pathology and that longitudinal studies yield information concerning probabilities of causal nexus in a population. But the legal problem, always, is to determine causal nexus in each individual case. "The unspoken medical assumption is that causation in a legal proceeding is a pure question of scientific fact!"² However, in the absence of a suitable test which could be used to establish the dose-response relationship at low range, the medical expert is willing to assume for purposes of conservatism that no threshold exists and that linear build-up may possibly result in adverse biological response. But when the etiology of a disease remains unknown, the medical expert will not under most circumstances say that the exposure "probably caused the disease." The reasons are crystal clear. At the present state of scientific knowledge it is simply not possible to relate individual response to disease, nor does the solution appear imminent. Some fourteen years ago a view was expressed before a Congressional Committee that "The more that is discovered about the complex etiology of disease . . . the less it appears possible to identify causality, and the more we grow dependent upon vague and arbitrary interpretations, with inevitable inequities . . ."³

When an employee develops a disease which conceivably could have resulted from occupational exposure, should the economic loss fall on the employee or should it be shifted to the employer? The courts in the United States in many instances have taken a rather juristic view in response to this question. It is clear that the courts need not be bound by medical notions of causation for medical evidence indicating a distinct possibility of a relationship between the job and the disease, while insufficient to support a finding of causation in the medical sense, may warrant a finding of causation in the legal sense. Our workmen's compensation laws in the United States typically include a mandate that they should be liberally construed to protect the employee. This being the case, decisions not to compensate where the exposures are small but, nevertheless, the employee is suffering from a radiation connected disease, may very well be inconsistent with the policy and purpose of our workmen's compensation laws.

Unfortunately most courts take the position that they cannot deviate from the requirement of medical probability. For example, in a recent Texas Supreme Court case⁴ in which the claimant was denied compensation, the court found

that the evidence did not indicate the existence of "a reasonable medical probability" of causal connection between petitioner's cancer and radiation but merely "the possibility" of such a connection. In drawing what it termed a logical distinction between a "reasonable medical probability" and a "medical possibility" the court said, ". . . a possibility becomes 'probable' when in the absence of other reasonable causal explanations it becomes more likely than not that the injury was a result of its action."

However, the opinion of the dissenting judge is worthy of note. He found that the expert medical testimony indicated that the etiology of cancer is really unknown, that the claimant's cancer could have been caused by radiation, but that there was no way to determine the cause of a particular cancer; that it is possible for a person exposed to radiation over a long period of time to develop cancer, but that it could not be stated how much exposure would be required; that any radioactive material can conceivably cause cancer on prolonged exposure; that anyone exposed to certain amounts of radiation has a higher than normal risk of developing malignant changes in the body tissues but that in this particular situation a diagnosis of probability either way could not be made.

Further, he rejected the weight the court placed upon the medical opinion evidence of experts who refused to testify that the cancer was "probably" caused by the radioactivity to which the petitioner was exposed. In his dissenting opinion he stated, "We are not to isolate the testimony of the doctors, but must determine the effect of such testimony upon other evidentiary proof in the case."

This statement was followed up with a reference to the spirit of workmen's compensation statutes and the need for liberal interpretation. He admonished the court for apparently forgetting "for the moment" the purpose of the Texas Workmen's Compensation Act. He stated that to hold, as the court held, that the evidence as a whole, which the jury considered in reaching its conclusion, did not meet the standard of proximate causation with sufficient certainty to impose liability upon the insurance carrier of claimant's employer, "is to effectively remove injuries which require medical testimony to substantiate causation from the common law of tort."

Health Physics Testimony

Now when it comes to dealing with roentgens, rads and rems and the recommendations of the various standard making bodies, we find that this is the field of expertise of the Health Physicist. It thus follows that because of his background and technical knowledge and the complicated array of terminology which finds its way into a radiation claim, the health physicist will play a key role in evaluating an individual's radiation exposure. The courts cannot adjudicate and the medical expert cannot opine until the source, duration and amount of exposure is known. In fact, the health physics experience of the injured claimant is one of the most important factors in any claim involving radiation injury and the health physics testimony and evidence is of extreme importance because it protects medical experts by assuring them that their testimony and opinions are predicated on the most probable exposure.⁵ Furthermore, the health physicist supplies and interprets available radiation records of claimants' external and internal occupational exposure (e.g., film badge and other types of dosimeter records, whole body counter records, records of bioassay data and interpretation, etc.) and he supplies estimates of exposure in the absence of radiation records and interprets other records relating to the claimant's exposure (e.g., records of work orientation and training, radiation and contamination survey reports, records relating to the radiation status of the claimant's work area and records relating to the employer's radiation protection program.)

Although it is the considered view of many lawyers familiar with radiation litigation that "records are the most important item in establishing dose from a legal standpoint,"⁶ there are still members of the health physics establishment who, I regret to say, maintain that the record developed on a day-to-day basis to assure the safety of the radiation worker has little value 15 to 20 years later when that very same record is introduced in a workmen's compensation proceeding as evidence of exposure. However, it is apparent from a reading of the cases that not all health physicists agree with this position for they have on many occasions furnished courts and boards with convincing evidence of the claimant's exposure from the radiation exposure records in their possession.

For example, in almost all of the claims containing allegations of radiation injury, radiation records (e.g., dosimetry, bioassay and other records related to the claimant's exposure) are not only supplied by the health physicist but are examined and referred to by the health physicist and the medical expert in deciding causation.

By way of illustration, in a case involving a blood disorder a health physicist testified that records showed the premises where the claimant worked were found to meet the standards of radiation protection as recommended by the National Committee on Radiation Protection. He also testified that film badge reports indicated all exposures were below permissible dose. The radiation expert in the case noted that after reviewing the case file, film badge readings and the physicist's survey of the environment, it seemed apparent that claimant's exposure had been at a relatively low level which would not be expected to give rise to incapacitating bodily injury. Compensation was denied.⁷

In another case involving exposure to radioactive tracers for a five-year period, a medical expert stated, after review of the records:

"Beginning with the exposures received, we find that these are well documented and do not appear excessive Dosimetry appears to have been reliable and film badge and monitoring reports indicate that the decedent's exposures were well below those considered maximum permissible. . . ." Compensation denied.⁸

The most popular argument advanced by those who discount the value of radiation records is the unreliability of monitoring devices to record low exposures. Yet from my own experience I know that a properly organized health protection program can and does furnish data which provides a pretty good estimate of the maximum exposure which the individual worker could have received in the course of his employment. In fact, there is testimony from the medical establishment that negates the view that records have no value. In a recent case in which the claimant wore no film badge a medical doctor noted: ". . . [claimant] wore no protective badge which would have adequately monitored his X-ray exposures." Compensation denied.⁹

While recorded exposures are valuable evidence in a radiation claim, I believe you will agree that it is important that the courts not give undo weight to evidence of exposure in recorded form at the expense of other evidence of exposure. For example, in the Texas case which I cited earlier it appears that the court may have relied too heavily on the film badge analysis alone in denying the claim of a radiation worker while disregarding other estimated evidence of exposure.

In this case the decedent was engaged in handling, assembling and disassembling nuclear materials and weapons for approximately four years. For a two-year period while "handling" the materials, he was not issued a film badge or

or protective clothing. For the other two-year periods, he was issued film badges and protective clothing. Badge analysis revealed exposure on two occasions as 36 millirems, although this was determined to be only a fraction of his total exposure since the badge was being worn under protective clothing. The amount of exposure was not known but estimated to be greatly in excess of 36 mr. In addition, evidence showed that petitioner was on one occasion in an "incident" area but the proximity of petitioner to the location of the "incident" or number of rems to which he was, in fact, subjected was not known. The protective badge worn by a fellow worker also in the "incident" area showed 6,500 millirems of radiation. Evidence further showed that, for two years petitioner was exposed to "radiation leaks" from material handled, but the amount of radiation to which he was subjected was not known since he was issued no measuring device.

In addressing itself to the petitioner's contention that "the whole evidence" of this case did create a reasonable medical probability, the court agreed that reasonable medical probability can be based upon "the whole evidence." However, the court could not agree that such evidence was before them inasmuch as the extent of any radiation beyond the relatively safe dosage of 30 mr was unknown.¹⁰

In another case it was the absence of recorded evidence which appears to have influenced the U. S. Veterans Administration in denying compensation to a veteran who was assigned as an X-ray technician from 1953 to 1954 and developed acute lymphocytic leukemia in 1969. He wore no film badge and there were no records of his work environment. In spite of health physics testimony that there was a 50% to 80% chance that occupational exposure caused his death, the Board pointed out that the evidence of record did not indicate that the veteran received "excessive radiation" during service.¹¹

In a 1961 Federal Workmen's Compensation Appeals Board Decision, the Board stated that because of the absence of a film badge during one period of the claimant's exposure, it was impossible to determine whether there was a significant exposure to radiation. Compensation denied.¹²

Total Evidence

From the point of view of the lawyer, the radiation record can never be too extensive. Accordingly, he will want to know of his client's total environmental exposure--occupational and non-occupational.

In a radiation claim a statistical game of possibilities and probabilities can, and in many cases does, greatly influence the result in a workmen's compensation case and, accordingly, the availability, accuracy and adequacy of exposure data, including prior medical as well as industrial exposure, takes on great legal significance.¹³

By way of example, take the case of a young man who developed acute leukemia after an exposure of a little more than 5 rem during a four-month period. A physician, knowledgeable in the effects of radiation, became acquainted with the case and noted that the man had received an indeterminate but apparently large amount of therapeutic radiation as a child. Keeping this in mind, and the fact that there is usually no decrease in the potential to induce leukemia by a long interval from the time of a first dose to the time of a second additional dose, the doctor felt that the worker's leukemia, if not caused, was at least aggravated and precipitated by his low occupational exposure. Compensation was granted.¹⁴

The Aggravation or Acceleration Theory

The concept of aggravation appears to offer an alternative for the medical expert who seeks to establish medical probability in the face of low occupational exposures. In one case, a veteran who had been involved in nuclear testing while in the service was then employed teaching radiological safety as a civilian. He claimed that his leukemia was a result of exposure received during six years of civilian employment. Service connected exposures were unrecorded and civilian exposures were low. However, it was noted that there were certain instances of exposures which could well have been "over permissible limits." A radiologist found that the final monitored exposure could well have been an aggravating factor and, although the degree of claimant's exposures were conjectural, the leukemia could be considered the probable result of his occupational exposure. Compensation was granted.¹⁵

In another case, the claimant had a history of working in a microwave environment. After a latent period of many years he worked intimately with a weak ionizing radiation source. A board certified radiologist found that the low exposure to ionizing radiation resulted in a reactivation and/or acceleration of a dormant cataract and pointed out that without prior sensitization of the lens by exposure to microwave radiation, the radiation from the electron microscope would not have adversely affected the claimant. Compensation was granted.¹⁶

In yet another case, claimant was employed as a medical radiology and X-ray technician for approximately eight years. He developed leukopenia. Evidence showed that radiation protection practices were good and records showed exposures were low. The Bureau's Medical Director supported a causal relationship by aggravation from chloromycetin, a potent antibiotic with a known side effect of bone marrow depression.¹⁷

In a 1971 decision a civilian X-ray technician was granted compensation for chronic myelogenous leukemia. Evidence showed he had been exposed to low cumulative exposures for 20 years in the course of employment; that radiation protection was good; that he had service-connected exposure for a period of one year at the age of 18; that during three months of training while in the service he was constantly exposed to X-ray without benefit of safety equipment or protective measures to avoid exposure. The medical opinion indicated that there was aggravation of previous pathology. The claim was allowed for leukemia due to radiation exposure.¹⁸

Occasionally an award is made even when occupational exposure is low and with no need to resort to the theory of aggravation. For example, in one case a medical radiology technician employed in that capacity from 1957 until 1961 was isolated from further ionizing radiation in 1961 as the result of blood tests and the industrial medical officer's opinion that claimant had apparently reached his "personal level of tolerance." He developed leukopenia in 1966. Radiation records revealed no excessive exposure on film badge and personal pocket dosimeter. Work was performed using the accepted precautions of lead screens and aprons. It was established that claimant used reasonable care and had not been exposed to the direct X-ray beam at any time. However a radiology specialist attributed claimant's blood disorder to "incidental radiation effects." Compensation granted.¹⁹

In another case a 36 year old physicist at a radiation laboratory developed cataracts in both eyes. In his work around accelerators from 1950 until 1962 film badge exposure showed only 0.61 R. An ophthalmologist testified that claimant had radiation cataracts. Another doctor stated that claimant's cataracts were of the location and appearance associated with radiation cataracts; that while these cataracts can occur without radiation and while

claimant's record of exposure was very low, in view of claimant's work and age group the situation was "highly suggestive." Compensation granted.²⁰

Radiation Protection Standards

The health physics profession readily admits that there is no such thing as known radiation safety; by that I mean some level of radiation exposure below which there is no biological effect whatever.²¹ In short, frank admission is made that total protection against harm from man-made radiation would require a health standard of zero exposure; that radiation protection standards are not merely technical, that they are established through a balancing of risk versus economic and social benefit. Safety standards do not take into account the physical difference among individuals. Even though for safety guide purposes use is made of a "standard man" concept to determine the mass and effective radius of the critical organs of the body, when it comes to an individual radiation claim, the claimant's dose-response can hardly be considered standard. Yet a review of the cases shows that in a substantial number of claims permissible levels of exposure are used as indices of safety when deciding the issue of causation.

For example, a radiologist noted that the claimant's exposures "were in fact considerably in excess of the maximum permissible dose." He concluded the claimant's exposure probably caused his death from lymphosarcoma. Compensation granted.²²

In still another case the medical expert noted that there was no contamination of the claimant's working environment "above permissible limits." Compensation denied.²³

In another case a health physicist testified that the premises where claimant worked were found to meet the standards of radiation protection as recommended by the National Committee on Radiation Protection. The health physicist also testified that the film badge reports indicated that all exposures were "well below the maximum permissible dose." Compensation denied.²⁴

In all of the claims referred to available film badge and other radiation records relating to claimant's exposure were introduced into evidence. Health physicists referred to records of exposure and related them to protection standards. What was the purpose of such testimony if not to imply safety or lack thereof? It has been said that there is a general tendency among laymen to assume that any exposure in excess of the various permissible levels and standards for any period whatever can be equated with proof of medical causation,²⁵ but since protection standards were never intended as indicators of absolute safety their use in the courtroom should be carefully scrutinized.

Alternative Proposals

At this point I believe that the problems inherent in our present legal system, as it is applied to low-level radiation claims, are abundantly clear. However, it is still the majority view of the legal establishment that the established principles of common law torts should continue to be employed in cases of delayed injury from radiation exposure.

Is there another route?

Professor Samuel B. Estep of the University of Michigan Law School has, over the years, suggested a somewhat novel approach to the problem. He would award compensation simply for the increased susceptibility to possible future disease. The uncontrollable factors which limit the accuracy of biological measurement by physical dosimeter readings seem essentially the basis for Estep to suggest

establishment of a "Contingent Liability Fund"²⁶ which would provide benefits to a radiation injured claimant regardless of his failure to show a causal relationship between the exposure and the injury. The proposed fund would consist of contributions by both the employee and the employer, the respective contributions to reflect both the "spontaneous" risk of leukemia and that due to the occupational exposure. In the event that the employee does develop leukemia, he is awarded a fixed amount of compensation without the necessity of adjudicating the causal relationship to occupational exposure. "Not only would such a scheme avoid the necessity for arbitrary adjudication, it would also avoid the expensive costs of administration. This would be of benefit to the worker, the employer and society as a whole."²⁷

The Estep approach is somewhat akin to the concept of national health insurance. There are those who point out that in Great Britain no man, woman or child need for any reason fall below a minimum standard of life. By a combination of insurance schemes, a worker who comes down with a disease, occupational or otherwise, is assured of full medical treatment and weekly benefits during the course of his illness.²⁸

Some ten years ago, Dr. Herman Somers in testifying before a committee of the United States Congress stated that:

"The evidence has been mounting for some time that the problems rising out of the scientific and technological revolution of our day are of a character which may not be capable of resolution within the traditional workmen's compensation design. The central question which we must ultimately face is whether or not, in the second half of the Twentieth Century, it will remain feasible, let alone justifiable, to operate a social insurance program on the old premise that a reasonably clear demarcation can be made between occupational and non-occupational disability."²⁹

Lastly, for those who reject the insurance approach, a statutory prima facie presumption in favor of the claimant has been suggested. The burden of proof would then be upon the employer to show that radiation exposure was not the cause of the claimant's disease. It is my guess that the employer may have just as much difficulty in proving no causal connection as the plaintiff now has in proving causal nexus. New York has adopted such a law. In a recent New York case³⁰ the employee, a theoretical physicist, died from acute myeloblastic leukemias. In affirming an award the Court said:

"The record discloses that decedent was exposed to radiation for a substantial part of two periods and also at other times in various amounts. The testimony of the medical experts is emphatic that there is really no 'threshold' or 'safe' dosage of radiation because at the present stage of scientific knowledge it cannot be ascertained exactly what effects radiation has on the human body. It is also admitted that each individual reacts differently to exposure to radiation. The award is supported by substantial evidence and by the presumptions [N.Y. Workmen's Compensation Law §§ 3(2),47] . . . especially so in view of decedent's good health prior to his employment."

Another example of the presumption concept can be found in the Federal Coal Mine Health and Safety Act of 1969³¹ which provides for certain presumptions in favor of the claimant in pneumoconiosis claims where it is found that the miner was employed for ten years or more in underground coal mines.

Congress has in the past considered legislation which would have instituted a Federal workmen's compensation program for employees exposed in their employment to "radioactive materials." The Price-Zelenko bill³² would have established a presumption of causation in favor of any employee who (a) received an exposure in excess of the limits set by a Federal agency and (b) developed any ordinary disease which the United States Public Health Service certified can be induced by exposure to radioactive material.

During hearings on the bill there was strong opposition and rightly so to the proposal because a presumption of medical injury would be based on some arbitrary maximum permissible dose limit.

However, it has been stated that a statutory prima facie presumption in favor of the claimant would not shift significantly the percentage of cases in which the claimant would be upheld.³³ The solution would be for the Courts to apply the laws of negligence, of product liability and of workmen's compensation in the growing field of radiation hazards in a manner which supports preference for the plaintiff when causal relationship, though not clearly established, is clearly possible. The cases involving low exposures are relatively few. If, as has been said, ionizing radiation "is the most studied, best understood and most wisely used agent,"³⁴ the cases will continue to be few. Thus compensating a few individuals who have been exposed to levels of radiation which may have "possibly" caused their disease will not establish radiation as a hazard worse than it is at the present time. If, in fact, the hazard is miniscule, it will remain miniscule except for the injured worker.

Those of you who have followed the course of this paper have reason to wonder as to the proper solution for the handling of injury claims involving low level exposures to radiation. There is no easy answer to this question. When a court of law is attempting to determine the cause of a claimant's pathological condition in a workmen's compensation case, the court is faced not only with the question of scientific etiology but with a policy problem as well; namely, whether under all the circumstances it is fair to shift the economic consequences of the pathological condition from the claimant to the employer. Some of the techniques I have described today would do this very thing, but until more research is done and we better understand biological response to radiation, a great deal of inter-disciplinary concern and effort must go into solving the problem of the worker who allegedly suffers disease and death from low exposures. IRPA, with so many qualified persons from all over the world professionally engaged and actively interested in radiation protection, can contribute significantly toward a solution of this problem.

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RADIATION PROTECTION INFORMATION AND EDUCATION FOR THE PUBLIC :
MISTAKES AND LESSONS , STRATEGIES AND TACTICS.

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1. Summary

Radiation protection and nuclear safety have become major targets for attacks by critics and opponents of many sorts. We must try to improve our methods for coping with such challenges and avoid to waste too much of our time or forces. By learning from past mistakes and analysis of the situation appropriate tactics and strategies can be developed that take care of the immediate needs and set some long-range goals for improved information and education of the public. Characteristics and problems of using or cooperating with news media are discussed and some proposals for actions by IRPA and its affiliated societies are presented.

The following ideas and proposals are the personal views of the author.

2. Problems and types of confrontations with the public

Within the last few years radiation protection and nuclear safety have suddenly become major targets for attacks by critics and opponents of many sorts. The types and motives of such attacks have been discussed a great deal and shall not be repeated here. We all agree that the reproaches are not justified, neither absolutely nor in perspective to what happens in other protection and safety areas. But there exists an obvious information gap in the public, and at least part of the blame for this falls on us. Complaining does not liberate us from these problems. The defense against the uninterrupted, often exaggerated or even stupid, but nevertheless effective attacks consumes more and more valuable time of authorities and experts without apparently leading to a quick success or visible end of such debates. The fact, that even government agencies or parliamentary committees are not ashamed of inviting self-proclaimed so-called "independent experts" and professional opponents on the same conditions as the real qualified experts shows how far the confusion about who is right or wrong has gone already. One is sometimes reminded of two sinister manifestations of the Middle Age, the Inquisition and the Crusades, which both were due to the agitation of fanatics appealing to idealistic motives and fear, and which resulted in terror, retardation of progress, defeat and failures. Some countries had to bear the consequences for centuries. Frustration, anger, resignation, exhaustion of forces and neglect of main and long-term tasks may be the undesirable results for our profession if we do not try to improve our methods of coping with such challenges or if we waste all our forces for short-lived emergency actions at the expense of a well planned long-range program. We must learn from past mistakes, analyze the situation and develop appropriate tactics and plans.

We have to deal with various characters, types and forms of confrontations, communications and contacts between experts and the public. The character of a

confrontation can be voluntary and offensive, when we have taken the initiative, set the goals and selected form, place and time, or it can be involuntary and defensive, when somebody else has invited, challenged or attacked us on his own terms.

We can assume the following coarse types of contact :

- A) Public performances: information, discussion or contradictory meetings; hearings; panel discussions; press conferences (sometimes with radio and TV coverage); seminars and symposia etc.
- B) Mass media : press; radio; television;
- C) Publications: books; specialized periodicals; laws, regulations, standards ; in the future audiovisual items such as TV cassettes;
- D) Educational programs

Some aspects of each type with regard to our problems are summarized below, and some of them are discussed in more detail in Appendix B.

A) Public performances have more frequently an involuntary character, being organized by local political parties, societies, opponent groups, utilities or authorities. The audience is restricted to some hundred persons at most (radio/TV coverage excluded) and it may often be selected or biased. The time available is always too short, be it for lectures and statements or for discussion. The atmosphere is often unfavorable, unfriendly or outright unfair. Success or failure are very much dependent on the chairman or moderator and on the show talents of the active participants. Technical troubles with microphones and other inadequacies create additional problems. Meetings are often invaded by outside organized opponent groups which try to monopolize the performance. Biased reports by mass media may present a completely distorted picture of the event to a much larger audience. Such public performances have mostly only short-lived effects and are rather negative and unsatisfactory to anybody really concerned apart from "showstars" such as politicians or professional opponents. The level of the discussions is either primitive or far above the comprehension of the audience. To sum it up : many public events are simply alibi-functions or much fuss about nothing. We should try to avoid them or restrict them to the voluntary variety.

B) Mass media have their own rules and characteristics that differ locally and nationally and that have to be known precisely if any efficiency is attempted. To some extent also many politicians and other public opinion-makers have to be handled similarly. Mass media are primarily interested in "news", i.e. new informations, sensations, stories, scandals etc. not yet published or diffused by other mass media. The main quality of such news must be to make headlines, to increase the circulation or audience. With some forms of mass media almost no limits of truth, morale or respect exist, only a sharp drop in circulation or rating can stop them. "Bad news is good news" for news media and much more interesting than dry facts. To be first to publish a sensation or information is much more important than to inquire or check the validity before publication. The correction of wrong informations is usually left to those involved or concerned, but does only rarely get the same amount of publicity or prominent place as the original uncorrect feature. The public or audience is large and of more or less unknown composition. Usually the confrontation will have an involuntary character. We have to expect biased, exaggerated or distorted presentations, even some sort of "censorship" by publishers, editors or producers with the easy excuse of lack of space or time. There is usually no long-lasting interest in a special topic with the exception of some outstanding newspapers or periodicals that have specialized editors or writers, or of some "engaged" publications or reporters. Most newsmen have to deal with a large variety of topics without having time or interest to acquire a deeper knowledge. Few discussions or direct contacts with the public are possible; only a small percentage of letters to the editor are published, and no news medium likes to admit a mistake. With some notable exceptions the effects of news media

presentations last only a few days. Voluntary, active cooperation with news media is only possible if good personal relations to editors or producers can be established. The "target" public or audience is quite different for each medium, newspaper or program feature, and a "market and media research" similar to the one done by advertising agencies is necessary for optimal efficiency.

C) Publications is used here as a collective term for all kinds of printed or otherwise duplicated material that are available and distributed to the public over a rather long or even unlimited period (e.g. through libraries). They can be used and reused at any time by laymen or experts, individuals and groups, in one or several countries at any not predetermined times. They may be copied, referred to, cited, summarized, often also discussed in mass media or special periodicals. A great advantage is ample space for detailed treatment of a topic. The size of the public is unknown. There is no direct mutual contact between author and readers and no direct, immediate discussion. Misuse is possible without chances for clarifications or corrections. But many characteristics and rules are similar to those of mass media, and equivalent precautions are necessary for good results.

D) Educational programs of all types and on all levels are probably the most efficient contacts with long-lasting effects, extended and repeated interactions between specialists and the public. But good programs need detailed, time-consuming preparations, and their efficiency depends to a great deal on the pedagogic qualities of the lecturers or teachers. Such programs allow sufficient time for a thorough presentation of the material, explanations, repetitions and discussions, but also for the creation of an atmosphere of confidence between author and public which facilitates the implantation and acceptance of the informations. The audience will be small, except for some basic courses or radio/TV educational series, but if regular teachers are selected as the first targets and the topic can be incorporated in various curricula and professional training programs, the repetition over many years will increase the size of the public and slowly build up a useful and solid foundation of basic knowledge.

The present situation challenges us mostly with involuntary confrontations of the types A and B. In order to improve our position and win back the initiative we need a long-range program for the conquest of types C and D which will produce a feedback on the mass media and the general public. Public performances should be reduced to the indispensable minimum, i.e. voluntary events such as press conferences, information meetings and symposia in a quiet, fair and matter-of-fact atmosphere with an audience that knows at least some basic facts. But most of our present activities are fire-fighting emergency actions due to unexpected attacks. We struggle to hold our positions, and many defensive counterattacks suffer from a lack of time and preparation and may make things even worse.

3. Some typical mistakes

As the mistakes during public confrontations in the nuclear field have been discussed frequently, I shall only describe two typical mistakes or "syndromes" which are causing many failures but seem to be difficult to root out.

3.1. The "prestige syndrome" or the "Peter Principle" of prominent speakers.

A favorite trick to attract the public is to feature well known personalities as principal attraction or "decoration". Apart from the fact that a Nobel Prize is no a-priori qualification for omniscience, infallibility or even just competence in a field different from the one for which the prize was awarded, many of the prominent people are also victims of the manager syndrome or the "Peter Principle". If they only have to deliver a well specified "show" such as an opening speech or an invited lecture this may work perfectly well. But there are only few such prominent persons who have time to keep so well informed on all details and latest developments that they are able to survive a battle of public discussions with a well prepared team of opponents. Fortunately there exist excellent active scien-

tists and specialists who also are fine speakers with a flair for good presentation, didactics and effective discussions. You may be congratulated if you can get some of these for a meeting. But very often you will have to select between one of two types of prominent participants. One are the top managers of government agencies, research institutions etc. who by profession have to defend certain official positions. If they have enough routine with confrontations and get a thorough briefing by a good staff, things may go well. Otherwise they can sometimes be manoeuvred into a trap and may have to improvise answers to unexpected questions. Such answers often reach their "level of incompetence" and provide excellent weapons for opponents.

The other variety of prominent people could be compared to showstars (in his latest novel Koestler names them even "callgirls"). They seek publicity, like to be in the spotlight, want to see themselves in news media, and ride, for personal satisfaction or opportunism, on any popular "bandwagon". They are smart and often have a previous solid foundation of scientific or other achievements, they are good, witty speakers, and they are willing to deliver a talk on any favorite subject such as environmental pollution, world models, futurology or birth control, provided they get well paid and publicized. They have no scruples to change the bandwagon as long as they can ride on top. But they keep mostly to the negative side in some top level protest movement, for it is much easier, less time-consuming and more colorful to criticize, to ride sharp attacks, to appeal to unconscious feelings, fears and antagonism in the public than to study a problem in detail and to try to give a balanced but much less dramatic picture, or to offer constructive critique and useful proposals that are more than fancy, futuristic but entirely unrealistic dreams.

3.2. The "babylonian or ivory tower syndrome" is well known and only too frequent in public performances of all kinds. Many speakers or panelists do not care about the intellectual level or basic knowledges of their audience. They feel obliged to prove their competence by using a disguise of specialized or sophisticated language largely incomprehensible to the layman. They embark on long monologues that drift away from the problem, or they complicate it by details, boundary conditions and reservations, that nobody anymore can understand whether the answer is yes or no. All the public does understand is that they do not understand anything, and that the experts do not know everything and seem to disagree quite a lot. No wonder that the public has less and less confidence in experts. If somebody really knows a subject he should be able to talk about it in clear simple terms and make the essential meaning of it comprehensible even to a layman. But beware of the other extreme: you may turn an audience against you also if you misjudge their level to the low side and use a baby language for an intellectual public.

4. How to improve our methods

Our forces and means are limited, much more than those of many "opponents" (this term will be used for the various partners or enemies in confrontations). We need better methods and systematic planning if we want to improve our situation. It can be compared to a small army fighting a much stronger one, and part of the solution to some of our problems lies in the application of old proven military practices in planning, decision-making and tactics. These methods have in recent years been rediscovered and "re-invented" as "modern" management techniques under fancy names, but to anybody who knows military staff methods they are old familiar practice. To summarize: we must analyze past experience, recognize the typical and main mistakes, learn from other, similar situations, draw the necessary lessons, build up a good information network and start a thorough and systematic planning of a well defined program.

It is a fundamental rule for military decision-making that before you can decide about the appropriate action, you have to analyze and evaluate the situation and its possible developments by considering the following factors :

- the task which you have to carry out or the goal that you have to reach. It must be well understood and clearly defined. Vague orders end often with failure.
- means, resources, manpower, allies that you can count on for your task;
- the "environment", "background" or "climate" in which the operations must be executed (time, place, duration, form, political, economical and psychological factors and influences etc.);
- Who is your enemy or opponent ? This includes real opponents as well as discussion partners or the audience. What is his position ? What are his motives ? What means and capabilities does he have ? What may be his plans and goals ? What support may he get from the audience ?
- What is your "firepower" ? What "weapons" are at your disposal or applicable, i.e. which arguments, documents, informations, proofs, experimental results, visual aids etc. are available for the support of your cause ? How effective are they against the probable targets ?

Once we have precisely defined the task or goal and impartially analyzed the situation, we must consider possible solutions or actions. These must be evaluated for efficiency, advantages and drawbacks, chances for success and possible reactions of the opponents. Only when we have gone through these mental exercises and can support them by some "experimental" evidence or "reconnaissance" are we ready to decide in principle how to act. Once the decision has been made, the tedious work of elaborating detailed action plans and preparing and testing all manpower, means and resources follows before we can start the action.

For our present defensive and involuntary confrontations part of that process will forcibly have to be shortened or improvised due to lack of time and freedom of action. But it would be inexcusable to omit it altogether even in "involuntary battles", and it would be a crime to start a voluntary action before all these mental and material preparations have been finished and tested. In the Appendices A and B a collection of hints and ideas for many possible situations is given.

We should also look for examples and models from other domains in everyday and public life with similar problems, where solutions have been found and tested and where we can learn a lot. Let me just name a few of them without getting into details: advertising agencies, public relations in industry or government (Atomic Forum organizations, Technical Information Division etc.), professional societies such as ANS, IEEE or medical associations, accident prevention, traffic safety, environmental protection, industrial hygiene, civil defense and army, sports, churches, charity programs, political campaigns, educational programs and methods, mass media, etc.

5. A general action program for public information

5.1. Short-range emergency program :

No doubt there is an immediate need for an emergency program in order to hold the positions and survive in the flood of opponent charges and concurrent environmental protection fashions that, though late come, now try to monopolize the field and preach the only saving faith. The details of such "crash-programs" depend very much on local circumstances and resources, but some common problems can be seen.

Part of our efforts should aim at bringing the discussion back to solid ground to a fair, matter-of-fact exchange of rational and objective arguments and facts. This may in some extreme cases require a strong, well aimed action against some fanatic and unfair opponents in order to uncover, expose and isolate them. Even when we get attacked we must make a clear distinction between honest and knowledgeable opponents who merit our consideration, and unfair fanatics without sufficient qualifications whom we must openly declare not to accept as discussion partners on an equal basis.

A second goal must be to win back the initiative from opponents and opportunists. News media, public, politicians and authorities should get the basic and any new informations and facts first and without delay from us, from the specialists, not in a distorted or delayed way from news agencies, scandal reporters or biased opponent bulletins. We must inform quickly, openly and correctly, whether the information is favorable or not (incidents etc.), and we must establish direct information channels to all concerned. All this is of course easier if we have managed to remain neutral in the nuclear dispute and can keep above economical or ideological biases. A fine example for such correct information is the recent book by Lindell and Löfveberg on "Nuclear Power, Man and Safety" which unfortunately up to now only exists in its original Swedish version.

The information handling problems play a key role here as everywhere in modern life. The flood of correct or wrong informations, arguments, statements, reports in the public discussion is even larger than in the scientific and technical areas of radiation protection and safety, and it is often much more difficult to track down the original source and form of an information or to keep up-to-date with the latest developments and publications. This problem cannot be solved by individuals but needs a well organized cooperation on national and international levels. Below I shall offer some ideas on how our societies and IRPA might help.

5.2. Long-range program

Our long-range tasks have mostly educational aspects. We must familiarize the public, the news media people, the specialists of related fields, officials and politicians with the basic facts on radiations, their effects, protection, regulations, safety of nuclear technology and its applications, and relations to other risks in modern life. Radiation must become as familiar to them as space flight or stereophonic music.

This requires educational activities on all levels and for all ages from high school to professional or academic training. The best approach is to get first the teachers on all those levels interested and trained in the subjects, then incorporate the topics in the curricula of future teachers and instructors for public schools, higher education or professional training. The last step would be the integration into the curricula of the various educational programs. Besides regular school programs we must not neglect post-graduate programs, on-the-job training in industry, adult education programs of universities or television networks, evening courses etc. In some countries basic military or civil defense training may offer additional opportunities to inform large parts of the population. If it makes things easier, our subjects can be incorporated into larger ones such as hygiene or environmental protection as suitable carriers of more general appeal or interest.

6. Some suggestions for action by IRPA and its affiliated societies

Most of the work in the information field has to be done on the national level, it is therefore primarily a challenge for our societies. A few examples from the activities of the Fachverband für Strahlenschutz show some possibilities. One of our main goals is to keep the members in close contact with the society and each other and to give them as much information as possible. A news bulletin of about ten pages is mailed to them four times a year and contains all information on our society, its working groups, coming events, interesting news, publications etc. Each year's general assembly is combined with a symposium, the proceedings of which are given to all members free of charge. The exchange of informations and experiences and the cooperation are further supported by a number of informal working groups, started by the initiatives of interested members for the discussion of problems from a certain special area such as incorporation analysis, working place monitoring etc., or, to mention two groups of special interest for the topics of this paper, on education and on public relations problems. The results

of the sessions of these groups are reported in the bulletin. When a problem has been treated, a short report, review or recommendation is issued, published and distributed to all members and, as the bulletins and proceedings, to representatives of societies, government agencies etc. On the local level the few health physicists on place must bear the whole burden of information, and it is vital for them, the society and our profession to keep all members well informed and up-to-date. If we cannot manage the information and education problems and needs of our members we have no chance at all to handle the public information problems.

How could IRPA and its societies help us? When I say "us" I mean all individual health physicists wherever they are in any way active in public information or education. We urgently need an excellent, fast and reliable information network on an international basis. Many symptoms and "viruses" of the present radiation and safety "syndromes" originate from other countries, often the USA, and infect other countries very quickly, because some opponent organizations have a good and fast information network with wide distribution. If we cannot build up something at least equivalent, we will lose the race. Certainly, in the nuclear field the Atomic Industrial Forum and its equivalents in other countries, or the American Nuclear Society, have done a fine job, but they cover only part of our field and not always in the necessary details, apart from a certain bias towards the promotion of nuclear energy. As health physics also extends into medicine, environmental protection, industrial hygiene and other areas, we need our own system which covers all these fields. To subscribe to the information services of all the other societies and organizations is far above the financial means of a health physicist, and our only international link, the "Health Physics Journal", is far too slow and too expensive to fulfill this information task adequately. One reason why many societies almost never supply informations to the "news" section of the HPJ is the delay of several months before publication which often makes the information outdated. What we would need is an "IRPA Newsletter" similar to the fine examples of the newsletter of the Health Physics Society or the new "Radiological Protection Bulletin" issued by the British National Radiological Protection Board. This should be produced by cheap means and distributed, through the societies, to all members every 2-3 months. It should contain reviews of all new informations and developments, coming events, recommendations, new reports and publications etc. In order to produce such a newsletter, IRPA would need an Information Center with a small permanent staff and offset printing facilities. This IRPA Information Center should be in close continuous contact with all IRPA societies, their working groups and members and through those with national authorities, committees etc. Through the Executive Council of IRPA and directly close connections and information exchanges should be installed to such international bodies as ICRP, ICRU, ISO, IEC etc. and to organizations such as IAEA, NEA, WHO and others. This IRPA Information Center could become a sort of clearinghouse for information on all aspects of radiation protection and even develop into an international central information service which not only publishes newsletters, reviews, handbooks etc., but also could handle individual requests for special informations. My proposal does not aim at multiplying the flood of informations, but rather at forcing it back into one or a few reliable and fast channels, reaching all of us with a minimum of delay and supplying all necessary informations in a compact form but without gaps. Of course such a project would need active cooperation and support by the societies and probably higher financial contributions to IRPA. But it could create a very useful, internationally acknowledged function of IRPA and free it from the false image of being an organization which only sponsors some congresses. It would make IRPA as useful and renowned as ICRP or certain international organizations, but without the political drawbacks and restrictions of the latter and with a more practical note than the former.

7. Some implications for our profession

I believe it is an obligation for all health physicists to be or become active in the education and information of the public. This has also some consequences and feedbacks for our profession. Health physics must remain and become even more a respected, well known and well based profession of high standards with well trained and informed personnel on all levels. This can only be done if we support our societies and IRPA and actively take part in their programs, and if the societies learn to operate as efficiently as medical associations. We must not lose the initiative or the tasks to other, more active, professional societies or promotional organizations. Close and constructive cooperation with national and international organizations, authorities etc. is very important, among other reasons also for the adequate representation of practical viewpoints. Our position should be objective and as neutral as our respective jobs allow. Correct information and the truth must be disseminated. We must continue to discuss problems openly on an international basis as we have done up to now without being afraid of publicity, abuse by opponents or pressure from interested parties.

But a good deal of self-criticism is also necessary. We must constantly improve our professional standards, eliminate mistakes, bad practices and shortcomings. All health physics tasks in industry, medicine and research must be carried out by well trained and capable health physics personnel. We should never accept an unduly reduced or unsatisfactory health physics program with the argument that economical considerations are more important than adequate and reasonable safety standards and qualified personnel in sufficient numbers. If we do not fight such bad practices we shall soon lose face and credibility. Nuclear power, nuclear medicine and other applications of radiations should be promoted, but not on the costs of reduced protection.

Appendix A

Some ideas for actions

1) "Rent-a-program"

Many professional societies, clubs and other organizations have a regular program of lectures, colloquia, information or refresher courses, excursions etc. The organizers of such programs are chronically short of topics and ideas. They welcome any reasonable proposal or offer. If our societies can offer ready "package programs" of speakers or excursions and have put those into operation a few times, a growing demand may be expected from many halls.

2) "Do-it-yourself" or "autosuggestion"

People believe best what they have found out themselves. Students, laymen or specialists from other fields who are interested or even critical of protection or safety problems could be assembled in study or working groups and be given the task of studying a well defined problem. Our societies could sponsor such a study or at least support it actively by providing the necessary subtle guidance, assistance with literature and consultants and openly and critically discuss the findings with the groups. If the results are interesting, suitable publication should be arranged and coverage by newsmedia organized. Such an experiment has of course its risks and costs a lot of time, but it will help such a group to get a better insight into our problems, philosophies, working methods and the wealth of information available as well as the amount of work and experience necessary for judging problems.

3) "Group therapy"

We cannot educate or convince the public or its opinion-makers by large public meetings of the "Billy Graham" style. Producing mass hysteria of any kind never is a lasting therapy. The "single patient treatment" on the other hand costs too much time, money and manpower. But a suitable group therapy over a weekend or a working or vacation week may offer interesting possibilities for treating carefully selected groups of prominent people, opinion-makers, politicians, newsmen. Such people who get a lot of publicity cannot be converted, convinced or neutralized in a public event where they feel obliged to give the kind of "show" the public expects. The only chance to get across any barriers is to separate them from their audience and to offer them an attractive and pleasant opportunity for free, unrestricted discussion and useful information on an exciting topic in a family-type group of interesting people, without any official obligation, function or publicity. But do not mix newsmen and prominent people, for the former could not resist the temptation of reporting on the event, and the latter would feel obliged to continue their "show". A well prepared, even exclusive program frame is important, but sufficient time for informal personal contacts, discussions and brainstorming must be available. Do not preach or try to convert, be open, informative and matter-of-fact. Present your views and problems, but let also the guests give their view of the problems and suggestions for solutions. The most important goal is to win their confidence, to break down political or other barriers, to release tensions and to get rid of resentments. The effects will not be felt immediately, but such a program will bring positive results in the future to the profit of our long-range programs. This kind of treatment could also be called the "F-treatment" where F stands for fun, friendly, food etc, but also for "feed facts, fight fiction".

4) The "domino or bandwagon effect"

Newsmedia, politicians and many other prominent people like to jump on a bandwagon, i.e. to be on the forefront of any actual happening or event getting sufficient publicity. If we therefore can get some influential people or news media interested in our problems such that they give them a lot of publicity, there is a good chance that other mass media will cover this, too and try to follow such a trend in order to get their share of the profit. Careful selection of the primary target will produce an optimal amount of spin-off.

5) "Desensibilisation" or "vaccination"

The negative public reactions and fears are very much like allergic reactions or contagious diseases, and some treatment similar to desensibilisation or vaccination might help in these cases, too. We should try to feed repeated small doses of correct informations to the public in attractive forms. We might even imitate the "syndromes" by using "anti"-headlines as eye-catchers, but followed by the correct informations and facts. Look how advertising agencies launch a new product or idea and you will see what could be done.

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Appendix B

Some remarks, lessons, rules and tricks

Public performances :

- 1) Set a clearly defined goal for the performance.
- 2) Carefully select the chairman or moderator and the other active participants.
- 3) Choose well informed, witty, quick-minded and eloquent speakers. Avoid prominent but superficially informed or narciss-minded personalities except for well prepared harmless official functions such as opening ceremonies without public discussion. For the real battle respect the "Peter Principle" and select a well informed staff member rather than the top boss.

- 4) Prepare your defense and attacks. Locate the weak points of the opponent. Study and analyze his previous actions, publications and arguments and prepare your counterattacks. Keep up-to-date on the latest developments. Imagine what moves the opponents might try (each has his typical fashion). Look at your own arguments from the opponent's viewpoint, try to guess where he might find your own weak spots.
- 5) Analyze the tactics of left wing opposition groups, radical student movements and protest groups. There are many similarities in their methods to those of nuclear opponents.
- 6) A good training in dialectics and rhetorics is very useful.
- 7) Get your supporters into the audience. Let them ask those questions which the local audience should like to ask but does not dare to or does not get a chance to ask because organized opponents may try to monopolize the discussion. Do not forget the Trojan horse trick.
- 8) If you get selected for an involuntary type meeting, try to get the best possible and most complete informations on the following points :
 - Who are the organizers, what are their goals, background, connections ?
 - Who are the other active participants, what are their views on the topic, their backgrounds, interests and connections ?
 - What audience and what intellectual level can be expected ?
 - Local, political and psychological background and environment ?
 - Available time, technical resources etc. ?
 - Are there any plans or risks that the meeting might end with the "unanimous adoption" of a biased resolution ?
- 9) An informal contact (lunch) between the speakers and other active participants before the performance may release some tensions, clear up some misunderstandings, settle some problems or disputes beforehand under four eyes. But do not get caught or misled by nice manners and words, some people change and uncover unexpectedly when on stage and before an audience.
- 10) Say clearly "yes" or "no", call a thing either black or white, use pictures that are as simple as woodcuts. You never have enough time to get into details, so why trouble the audience with things they cannot understand ? Make your statements short, clear, impressive. They should hook like a good joke and be remembered. Be witty but do not exaggerate and do not imitate a clown or showmaster.
- 11) "Steal the show" by answering some likely arguments of your opponents before they get a chance to present them, let them blast open doors. Attack is the best defense. Avoid to be caught or attacked unexpectedly.
- 12) Keep calm and matter-of-fact. Select your weapons according to the type of battle and the weapons of the opponents. Make a difference between a public meeting with its catch-as-catch-can rules and the fair-play sunday-school atmosphere of a scientific meeting.
- 13) If you are well prepared you can often refute or disprove an opponent by his own words. Give exact citations and references.
- 14) Make it clear to the audience if and why you do not accept a certain opponent or "witness" as a real specialist or expert qualified to deal with the problem, despite all titles and other merits.
- 15) Réveal to the audience the motivation, background connections, interests and real goals of some opponents who often are presented under some harmless or impressive disguise.
- 16) Avoid controversial issues or arguments that are understood by specialists only. This would only nourish the impression that the specialists disagree or that many important things are not clear or known.
- 17) If you want to gain time for finding the best answer, either smoke a pipe and look like Rodin's "Thinker" for a while, or better : explain, analyze, qualify and simplify the question or problem for the audience. This makes it easier for you to find the right answer. Explain whether it is a fundamental, impor-

tant problem or just a secondary detail. If you do not know the answer or do not want to give it, state clearly why. Explain why the problem is too complicated for a short answer, or why somebody else will be better qualified to answer, or that the exact informations are not at hand but where they can be found, say whether an answer exists or not. Never leave the impression that you got caught in a trap or that an important lack of knowledge exists when this is not the case. A possible way to react may sometimes be to show first that the questioner did not understand the problem, but then the question must be answered anyway.

Mass media (and politicians):

- 18) Some general rules for using mass media :
 - Study the characteristics of each medium and of its individual representatives (e.g. various newspapers). Experiences from one region, country or representative do not necessarily apply to another one.
 - Analyze the previous position of the medium to the problem, its public or audience, the intellectual level, style etc.
 - Contact the responsible editor or producer. How much space or time is available. Which feature, program, page or section is best suited ? What relation between text and pictures is desired ?
 - for books : select the proper editor, series, size and price category.
 - Do a good media and market research such as done by advertising agencies.
 - Get either an optimal efficiency or skip it.
- 19) In dealing with news people (or politicians) it is very important to build up good personal contacts to some carefully selected key people. Do not try a "crash-program" of convincing them, but slowly develop a basis of mutual understanding and confidence. Ask them for help and advice in public relation problems. Help them with facts, informations, consulting, news, frequent press releases or conferences, some exclusive reports or interviews. A newsman does not like to be coached or tutored, but he will welcome support and help if he gets it easily and if it is useful to him for avoiding errors or blunders.
- 20) If news media publish incorrect or biased informations, only strong, multiple and quick individual, collective and official reactions and complaints on several levels, from the responsible editor to the top management, will cause a correction.
- 21) If you get interviewed, take care of your image, keep neutral, matter-of-fact and independent. Do not get seduced to play the expert in fields other than your own. Of course you may offer your personal opinion also on other problems, but as an educated layman. Distinguish clearly between your official standpoint due to your function and your personal views as a specialist or a layman.
- 22) Do not over-simplify. If an essential element is missing in an information, it may become incorrect, miss the target or even become a bait for attacks.
- 23) Separate clearly established facts from fiction, assumptions, extrapolations, prognostics etc.
- 24) Do not get upset if even a fair and objective reporter asks you a critical or uncomfortable question. It is part of their professional technique and a way to show their independence. Do not use the same yardstick of quality for mass media as for special or scientific publications or events.
- 25) make sure that your statements are published correctly. Request to see the proofs before publication or reserve a right to reply. Submit your statements in written form, make your own tape record or get a copy of the reporter's tape.
- 26) Do not get caught unexpectedly. Give no statements which you had not time to think about. If the problem really matters to the reporter, he will give you time for preparation and tell you what he intends to do with the information.

If a reporter is not willing to do so, something is suspicious anyway.

- 27) When you contribute an article to the press, keep your language simple, explain indispensable special terms. Give it an attractive form, write a popular style, supply good illustrations. Make short paragraphs and type with double space and wide margin.
- 28) Radio programs are not very suitable for complicated topics. Their main features are news and short informations, short comments, or panel discussions.

Television :

- 29) These remarks apply to European television without commercially influenced or sponsored programs or shows, which operates on principles similar to those of newspapers or magazines.
- 30) The main information carrier in TV is the picture, it must move, not stand still. Many producers and directors prefer visual esthetics, action and gags to all other considerations such as facts, balanced information and content, truth etc. The spoken word is subordinate to the visual development and must be short and clear.
- 31) A TV program is momentarily impressive by its dynamics, some highlights and gags, but it has no long-lasting effects.
- 32) A program needs an "eye-catcher" at the very beginning or in the title which attracts the audience and prevents them from switching channels. Best carriers are regular, highly rated and well attended programs and features. They must be carefully selected according to topic, moderator, background, audience, daytime, duration, style, actuality, concurrent programs on other channels.
- 33) The larger the audience the shorter is the time available for a special topic. The duration varies from a few minutes for actualities over a preferred average of 10 - 15 minutes to the rare cases where up to an hour or more can be devoted to a single topic, but usually only in special documentary or science programs or panels on topics of high interest. Sufficient time for treating a subject in detail would only exist in educational programs, but these require long and tedious preparations and are planned years ahead.
- 34) Popular quiz programs, panel or interview series might offer a chance to introduce a problem or topic to a large audience and get additional coverage by other news media such as newspapers or magazines. Very close cooperation with producers and moderators and careful selection and preparation would be required.
- 35) If you get interviewed by TV, try not to stick to a manuscript. Inquire beforehand in what context the interview will be shown.

PUBLIC CONFIDENCE IN NUCLEAR POWER

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Abstract

Although, in the United Kingdom a healthy interest is maintained by the general public in the development of nuclear power as an energy source, concern has never been so widespread or as hostile as has been the case in the United States of America. This paper suggests reasons for this and discusses the origins of nuclear power in relation to the public and the government administrative machinery which has been built up around the subject. The method of obtaining consent to build and operate a nuclear power station is outlined and the intensive public relations exercise to which all parties contribute is put forward as a main reason why nuclear power is not only accepted by the majority but is actively encouraged.

Background

In the United Kingdom there are only two electricity utilities which operate nuclear power stations. They are the South of Scotland Electricity Board and the Central Electricity Generating Board. Both are nationalised industries and the latter is responsible for the generation and bulk distribution of electricity in England and Wales. It owns and operates 174 Generating Stations with an output capacity of 56,000 megawatts composed of a mixture of coal fired, oil fired, hydro, gas turbine and nuclear power units. The nuclear component at present consists of 8 twin reactor stations of the gas cooled magnox type whilst a further 4 twin reactor stations of the advanced gas cooled type are currently under construction. Because of the way in which the electricity supply industry is organised the general public are in direct contact with the retail distribution organizations and do not come into contact with the CEGB except on relatively rare occasions such as new generating station projects or the siting of high voltage transmission lines. With such a large organisation under unified control as the CEGB it is able to support a headquarters staff in both the planning and nuclear health and safety fields. This has enabled a uniform policy to be agreed centrally and implemented effectively. A further advantage is that single authoritative channels of communication have been established between the Board and the several Governmental regulatory authorities which are concerned with the whole field of nuclear energy.

History

The development of nuclear power in the United Kingdom started in a favourable climate in the early 1950s. Coal, the main energy source in the U.K. after the second world war was in short supply and increasing in price at a rate which was directly observable by the open coal fire burning public. Oil, then almost non-existent within the British Isles or its waters, was of foreign origin and subject to the uncertainties of foreign currency shortages and seizure of the wells mostly in the politically unsettled middle east. Gas was a rather expensive, and to many people a dangerous by-product of coal. The fanfare of publicity at the opening of the first nuclear power station at Calder Hall, dignified and given the seal of respectability by Her Majesty the Queen, did much to persuade the inhabitants of our cold and coal grimed cities that the millenium was at hand. The Windscale accident in retrospect, far from engendering hostility to the "atom", due mainly to the prompt and expert handling of the affair, did much to reassure possible antagonists that although accidents could happen, the Government and its traditional safety agencies were fully on top of the situation and no great catastrophe was likely in any way as devastating as the air raids so relatively recently endured. It is important to realize that the U.K. is so small and its communication and legislative structure so unified that attitudes and opinions are national rather than regional which the great area of the USA made almost inevitable in that country in the pre-universal television era.

With the conservative siting policy adopted for the first round of nuclear stations the plants were built in rural areas mostly on the coast and because of the influx of construction staffs, improved amenities and relatively large increases in taxes to the local authorities there was very little local opposition to the stations. Any opposition which existed was more concerned with the impact of the large structures on the visual amenities of the area and sometimes the effects of cooling water in the river or estuary.

Planning

Before a power station can be constructed irrespective of the prime heat source, planning permission is required under an Act of Parliament dealing with the supply of electricity in general. Notices giving the intent to build the station are posted in prominent places adjacent to the site and in public buildings such as the Post Office and Town Hall. Any individual, municipal authority or company business may object by writing, stating the reasons for so doing, to the Government Department concerned, which in this case is the Department of Trade and Industry, within 28 days. In practice in the interests of the public this period may be extended quite considerably.

Prior to this stage the Planning Department of the CEGB has discussed the project informally with the local government officials and their support for the proposal is solicited. If there are many objections then a public inquiry may be held at the discretion of the Secretary of State for Trade and Industry but if the local planning authority objects then the law requires

positively that such an inquiry be held. The decision as to whether to allow the station to be built rests with the Secretary of State and the inquiry merely serves to assist him. The CEGB mounts a public relations exhibition in the locality and senior Board officials including nuclear safety experts are in attendance to answer any questions in an informal atmosphere. If objections are mainly in the form of questions, public hearings are held and by discussing the various aspects of the station many of the objections are subsequently withdrawn. Quite distinct in law but coincident in time with the planning application, when a nuclear station is involved, a request for a nuclear site licence is made also to the Department of Trade and Industry and by previous informal discussion with officers of the Nuclear Installations Inspectorate, the site characteristics are derived so that argument of a lost cause is avoided. Although objections to the issue of a licence for a nuclear power station are not legislated for except by the freedom of the individual to write directly to the Chief Inspector of Nuclear Installations, questions concerning nuclear safety are asked and answered at the planning inquiry, by both CEGB and Government witnesses.

Waste Discharge

The control of discharge of radioactive waste from any source is vested in two Ministries, the Department of the Environment and the Ministry of Agriculture Fisheries and Food. Neither of these organisations has any administrative connection with the Nuclear Installations Inspectorate and each has a long tradition dating back to the 19th Century, of responsibility for controlling the disposal of various industrial wastes of all kinds. The Radioactive Substances Act 1960 in effect extended the powers of these old established ministries to include radioactive wastes and the skilled administration which over many years had won the respect of the public in looking after its interest was readily accepted as being completely impartial by both the nuclear industry and the public at large. An important part of the requirements of the Act is that before an authorisation to dispose of radioactive waste is granted the Ministries must inform and consult representatives of the local authorities in detail about the proposals. Objections can be resolved and no public inquiry is called for at any stage. Consultation at a technical level takes place between the scientists of both the CEGB and the Government Department before a formal application to discharge is lodged and thus any divergence of opinion can be discussed calmly out of the limelight of lobbyists and extremist environmentalists.

Local Liaison

Having dealt with the discussions and consultations between the licensee and the statutory bodies before and during construction there is in addition a continuing relationship with local authorities and other interested parties. One of the more interesting of these is the setting up of Local Liaison Committees whose terms of reference are

- (a) To provide information and reassurance on the manner in which radioactive materials are used at the power stations.

- (b) To explain the significance of radiological measurements which are made outside the station boundaries.
- (c) To discuss schemes for the protection of the public in the event of an accident, the schemes to be prepared by the Board in consultation with the appropriate officers of the County and local authorities.

The committee is set up by the Generating Board and representatives are invited from County and local authorities - elected members and officers together with medical officers of health - local bodies who have statutory functions such as water undertakings and river authorities, farming interests both with the National Farmers Union and Country Landowners Associations. In addition any specific organisations within 4 - 5 miles such as Trinity House (due to the lighthouse on Dungeness Head) and the Lydd Airport Authority, also at Dungeness, are invited to attend. In addition senior representatives of the authorising Ministries responsible for the control and discharge of radioactive wastes are members of the committee. The chairman is the Station Superintendent and there are representatives of the station management and Headquarters Nuclear Health and Safety Department. The press are not in attendance at the meeting but an agreed press statement is issued at the end of each meeting.

Meetings take place at the power stations once or twice each year and the results of the routine district surveys and the total curies discharged to the environment as liquid and gaseous waste are reported by the representative of the Department of the Environment; a report of station operation is given by the Station Superintendent and details of emergency plan rehearsals are given by the Station Health Physicist and discussed in detail.

The district survey which is carried out by the Station consists of measurements of airborne radioactivity and gamma dose rate from deposited radioactivity. These readings are obtained at different distances from the reactor in order to give comparative rather than absolute results. In addition any land and marine species of animals which form part of the food chain to man are assayed and the results compared with derived working levels agreed between the regulatory authority and the licensee. Particularly, in rural areas milk is collected and analysed, both for its own sake and also because the cow is a very convenient collector and averager of several radioactive isotopes which may be contaminating pasture land.

Emergency Plans

The Emergency Plan is a statutory requirement. It is very comprehensive and specifies the actions required by both CEGB personnel and outside organisations. Frequent discussions and consultations take place with local authority officers during its preparation but particularly with the police who would have the job of instituting action including distribution of stable iodine tablets to local inhabitants and in the ultimate carrying out of any necessary evacuation. Copies of the plans are given to all members of the Local Liaison Committees and they report back to their parent organisation. In all cases the recipients have

behaved in a responsible manner accepting the effort which is put into ensuring safety both of site personnel and local inhabitants rather than using the necessity for such plans as implying an impending hazard.

Regular training sessions of site personnel in emergency actions are carried out including first aid and rescue. Once each year there is a complete exercise of the plan in which a specific incident is simulated and the plan is brought into action, those taking part in the exercise having no prior knowledge of the supposed accident. The police are involved but not the general public. The exercise is witnessed by inspectors of the licensing authority and by the Board's Nuclear Health and Safety Department. Post-exercise discussions and criticism takes place in which all participants, and inspecting authorities, take part and as mentioned above the results of these exercises are discussed at the Local Liaison Committee meetings.

We believe these committees have been a most useful public relations experiment, people have been treated like intelligent and rational human beings and they have behaved like it, at no time has any attempt been made to cover up any incident no matter how minor and on all occasions the committee members have reacted in a helpful manner. Some of these elected members who have been members of these committees for some years have become remarkably knowledgeable.

Public Interest

The Board encourage visitors to all its sites both during construction and operation. Special low radiation routes through the station are planned including a visitors gallery on pile cap. Guides consisting of the wives of staff employed on the station are provided with uniforms and given instruction about the power station and they attend as and when required. A typical nuclear power station may have as many as 20,000 visitors each year including parties of children from local schools.

Two projects which are both interesting and important relate to fish farming and trout fishing.

The possibility of using power station discharges for rearing marine fish and shellfish was first discussed with the White Fish Authority about 10 years ago. The objective was to maintain the growth of these animals over the winter months, thereby bringing them to marketable size much sooner than occurs in the sea.

Initial studies were undertaken at the South of Scotland Electricity Board's Hunterston Nuclear Power Station and a conventional CEGB power station. These preliminary investigations using flatfish showed that increased growth occurred. It was decided then, that nuclear stations with their high load factors would be the most suitable sites at which to undertake future studies. Also, many of these stations are sited on open coasts where the water is unpolluted and the salinity is less variable than in estuaries.

At Hinkley Point Nuclear Power Station a private individual

in collaboration with the CEGB, is undertaking pilot scale studies on rearing crustaceans (prawns) and molluscs (oysters and clams) together. He has progressed considerably towards developing the necessary technology for a more extensive project.

The Ministry of Agriculture Fisheries & Food is planning to establish a laboratory on the Wylfa Nuclear Power Station site to investigate all aspects of rearing shellfish. They have undertaken extensive laboratory studies to develop the technology of spawning and rearing larvae. At the same time, rainbow trout have been grown in the discharge on a small pilot scale basis.

Fish farming will be a large scale development of the future, and many countries are undertaking extensive research programmes into the rearing of a variety of species. It is very probable, that in the temperate latitudes warmed discharges will make a valuable contribution to the success of such ventures. Already in the Board, provision is made at suitable new stations to build-in the necessary access to C.W. systems, so that if need be, water can be drawn for use in a fish farm.

The Trawsfynydd Nuclear Power Station, 500 MWe, has been operating successfully since 1965, it uses an inland lake as its source of cooling water. The lake was formed in 1924 to serve a small hydro-electric station to provide a local supply of electricity. An angling club, the Prysor Angling Association, was formed by local inhabitants in the early 1930's to use the natural fishing of the lake comprising brown trout coming from local streams. The lake was emptied and enlarged in 1959 prior to the commencement of the construction of the nuclear power station. At the public inquiry the Board undertook to preserve and assist the fishing rights of the angling club which has in fact continued to control the fishing of the lake. Its activities are managed by a Lake Management Committee and the Power Station Superintendent is a member of the Committee by invitation. The club realised that the lake could be developed far beyond the naturally occurring fish and since 1967 it has been trying to increase the fish population artificially. Unfortunately the brown trout, although giving good sport and good eating, is difficult to rear artificially due, amongst other things, to the predatory activities of perch. It was decided to introduce rainbow trout into the lake and the current programme consists of adding 16,000 fish per annum. Some brown trout continue to be raised on an experimental basis.

The success of the experiment is illustrated by the fact that recently the trials to choose the Welsh team for International fishing competitions were held on the lake. In addition the Board use the lake water to raise trout for stocking other lakes in the area.

The liquid radioactive effluent from the station is discharged into the lake which is also used as a source of cooling water for the turbine condensers.

The presence of small but detectable quantities of radioactive materials in the trout has not deterred the fishing and eating of the catch.

Local Involvement

Power stations employ small numbers of people, large power stations even of 2,000 MW capacity rarely employ more than 500 people. The Board's policy is to employ locally recruited labour and therefore the power station employees rapidly become part of the local community, unlike large Atomic Energy Authority establishments employing several thousand people imported into a locality and who remain in a separate group. The result is that a power station very quickly becomes "our power station" and "our nuclear power station" is even better.

Finally the activities of some of the radiological protection societies notably the Society for Radiological Protection should be given an honourable mention. This Society provides a lecturing service, free of charge, to a variety of organisations from Rotary Clubs to Mothers' Unions and this has proved a very popular and widely appreciated service to the community as a whole.

Conclusions

It is believed that the urgent need for nuclear power in the United Kingdom predisposed the public to accept the large structures comprising the power stations as necessary. The accompanying small risk of radiation exposure was also readily accepted because of the confidence which had been built up in the Government Agencies involved by virtue of their historical role in the so-called conventional safety matters such as chemical and biological control.

Whilst the future cannot be predicted with confidence it is hoped that ten years successful and safe operation now achieved will reinforce the tolerance and goodwill which has always existed between the CEGB and its neighbours.

INFORMING THE PUBLIC ABOUT NUCLEAR ENERGY

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Abstract

When the Karlsruhe Nuclear Research Center was founded in 1956, it soon became apparent that the population of the neighboring communities had to be informed about nuclear energy in an unbiased, objective way. Those discussions centered chiefly around arguments of radiation protection. The experience gained from our public relations activities in those years was used and continued by the Nuclear Engineering School of the Karlsruhe Nuclear Research Center, e.g., in special courses run for the information and education of teachers and journalists who passed this information on as opinion leaders. In a similar way it was possible to discuss the objections against siting of the first nuclear power stations in Germany with the population in the environment of those sites.

It was not until the controversy imported from the United States, which can be described simply by the names of Gofman, Tamplin and Sternglass, that organized groups of the population turned up who were against the use of nuclear power. This at the same time made the arguments more emotional, expanding them into problems of energy policy and sociological questions under the headings of "environmental protection". This different situation must be taken into account in public relations activities.

Our experience from numerous and varied discussions with action groups against nuclear power has clearly shown that big rallies and use of the mass media can result only in general information and education of the public. More complex subjects with discussions pro and con, which arise when it comes to the establishment of a nuclear plant, need early and specific approaches to homogeneous groups. These contacts should always be supported by arguments of fact and should cover only a limited subject.

Introduction

The resumption of nuclear research in the Federal Republic of Germany after the conclusion of the Paris Treaties of 1955 suffered from the severe burden of atomic energy: the bombs of Hiroshima and Nagasaki. This primarily created a sceptic attitude relative to the establishment of research facilities for the peaceful utilization of nuclear energy.

"Paleozoic" - 1955 - 1960

For the same reason the establishment of the Karlsruhe Nuclear Research Center in 1956 highlighted the necessity of factual information and education about nuclear energy of the population in the communities in the vicinity. The possibility of visiting the plants under construction, experimental lectures about health physics and nuclear technology in schools and institutions for adult education created the first contacts. Study tours of foreign nuclear research facilities, for instance Saclay near Paris in France and Mol near Brussels Belgium, were organized for special groups. This habit introduced in the early years of the Karlsruhe Nuclear Research Center of making the facilities of the Center accessible to all the interested parties at any time was retained in the years to come. At present approximately 15 000 visitors, in groups and individually visit the Karlsruhe Nuclear Research Center; they can get all the information they want, and the staff of the Center are available for discussions with them.

When the Karlsruhe Nuclear Research Center was founded, most of the questions asked by the population referred to radiation protection and radiation exposure of the environment. Most of these questions indicated a genuine concern. In a few cases, however, questions with respect to radiation protection and safety were just a pretence covering up for economic interests. These opponents were afraid that the establishment of a Research Center could cause the workers employed in their small local industries to change to jobs in the Research Center which would offer better pay.

At the earlier meetings informing about nuclear energy homogeneous groups, such as teachers, members of municipal councils, members of agricultural associations, etc. were preferably approached.

"Mesozoic" - 1960 - 1968

In the early sixties the establishment and the expansion of a Nuclear Engineering School at the Karlsruhe Nuclear Research Center made it possible to pass on information about nuclear research and the peaceful uses of nuclear energy to the public through courses and information meetings. Experience gathered in the early years was thus made use of and expanded.

Besides purely technical courses in radiochemistry or reactor technology, radiobiology and health physics, special courses were organized for specialized teachers in secondary schools from all over the Federal Republic of Germany. In this way it was possible to use teachers as "opinion multipliers" and make use of their educational possibilities and capabilities in order to pass on factual information to the younger generation to be trained in an understandable way.

In those years more and more reports were found in the press which unintentionally gave wrong information. In most cases this indicated an insufficient amount of technical knowledge with many journalists. Consequently, journalists were invited to attend brief courses at the Nuclear Engineering School where experts talked about specific selfcontained subjects, such as "biological and medical problems in the utilization of nuclear power", "reactors of the future", "reprocessing of fuels", or "nuclear safeguard methods".

Both groups, teachers and journalists, greatly helped in the publicity of nuclear knowledge through their capacity as "opinion leaders". In this way problems of radiation protection and safety were discussed, thus preparing a critical public.

These same years saw the construction of the demonstration nuclear power stations of Obrigheim and Gundremmingen in Germany, which gave rise to a thorough discussion with the population in the areas of these plants about problems of site selection. Proper Commissioning then proceeded without any major interruptions.

"Neozoic" - 1969 -

Since 1969 greatly exaggerated reports have also appeared in

the press of the Federal Republic of Germany questioning the arguments of nuclear safety of nuclear power stations and thus creating unrest even among the experts. This controversy, which was imported from the USA and can be outlined by mentioning the names of Gofman, Tamplin and Sternglass, resulted in the organized association of a few groups of the population opposing the application of nuclear power. Although this nuclear controversy and its extension from the USA to Europe, especially to the Federal Republic of Germany, had been recognized by a few experts, the scope and the possible effects had not been correctly assessed and the rate at which this phenomenon spread had been underestimated by industry.

Two factors, most of all, influenced the generation and the extent of the controversy: A generally improving environmental consciousness among broad groups of the population coupled with a certain hostility towards technology or a reduction of faith in technology.

It was necessary to take account of the representation of the problems in a popular book written by a number of experts in which anybody could be able to find factual information on the subject. For this purpose, Deutsche Verlagsanstalt of Stuttgart in early 1970 published a book entitled "Kernenergie - Nutzen und Risiko" [1]. However, it was evident from the outset that a nonfiction book would not be sufficient. Sensational reporting had to be attacked by other means. For this purpose, almost simultaneously a "collection of arguments and counterarguments" [2] was published by the Swiss Association for Atomic Energy and a volume entitled "Kernfragen" [3] by the German Atomic Forum.

Switzerland was early to recognize the direction in which the conflict threatened to move, as a consequence of the reaction of certain groups of the public to the reactor incident at Lucens (January 1969).

At an information and discussion meeting organized in Bern in the fall of 1970 by the Swiss Association for Atomic Energy the situation was indicated. Many German observers experienced their first encounter at this meeting with groups discussing only on an emotional basis. Indeed, dealing with reactor safety and radiation protection at a public forum in this way was a successful venture

with the Swiss population which seemed to be used to democratic discussions. Afterwards, the technical questions discussed were published in a generally understandable form and made available to the interested general public.

Development of Arguments

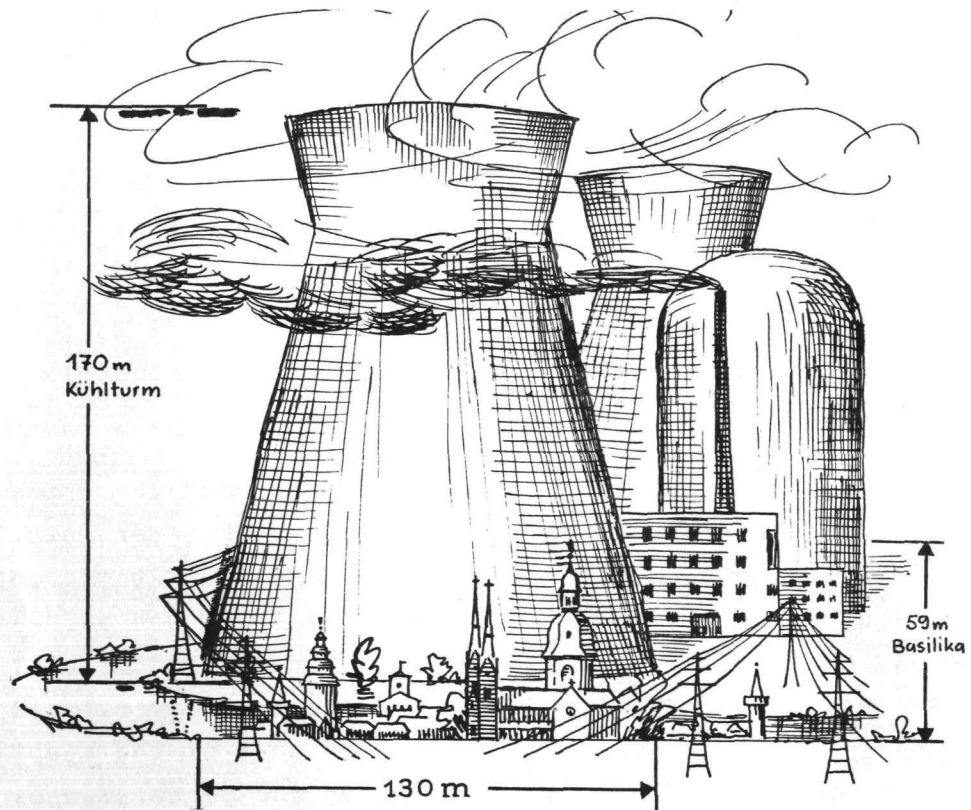
The development of arguments in discussion meetings during those years can be classified as follows:

- During an initial phase there was a generally factual, objective discussion with the interested public which mostly had a limited background knowledge.

- As a second phase organized groups expanded the arguments to other areas (emergency cooling, frequency of cancer, etc.). The discussions became more difficult. Talks with the "atomic opponents" require experts in the respective fields, such as reactor technology or medicine. This phase is the era of sensational stories, exemplified by the name of Mary H. Weik. However, scientific and technical articles are able to convert most of the sceptics from their former negative opinions about the peaceful uses of atomic energy.

- The third phase is determined essentially by the general efforts towards environmental protection. This made the arguments more emotional, extending them under the heading of "environmental protection". Their often hysteric expression leads away from problems of reactor technology and radiation protection or safety to global problems of the future such as "thermal pollution," landscaping and aesthetics, problems of energy policy and sociology. Especially the latter points are supported by political groups operating with the keywords of "changes of the system" against the profit maximization of utilities." This confronts potential reactor operators with a very complex set of questions in this third phase. One example of the emotional, distorted description presented by an action group against nuclear power stations is shown in Fig. 1. Such action-associations do not want any factually correct information, such as Fig. 2, a photomounting of the cooling tower for the nuclear power station at Gösigen, Switzerland.

Die Stadt „zur schönen Betonaussicht“



Gemeinnützige Aktion FORTSCHRITT FÜR ALLE D 8501 Feucht, Postf. 1230

Fig. 1: Cooling towers, sketch of an action against nuclear power stations

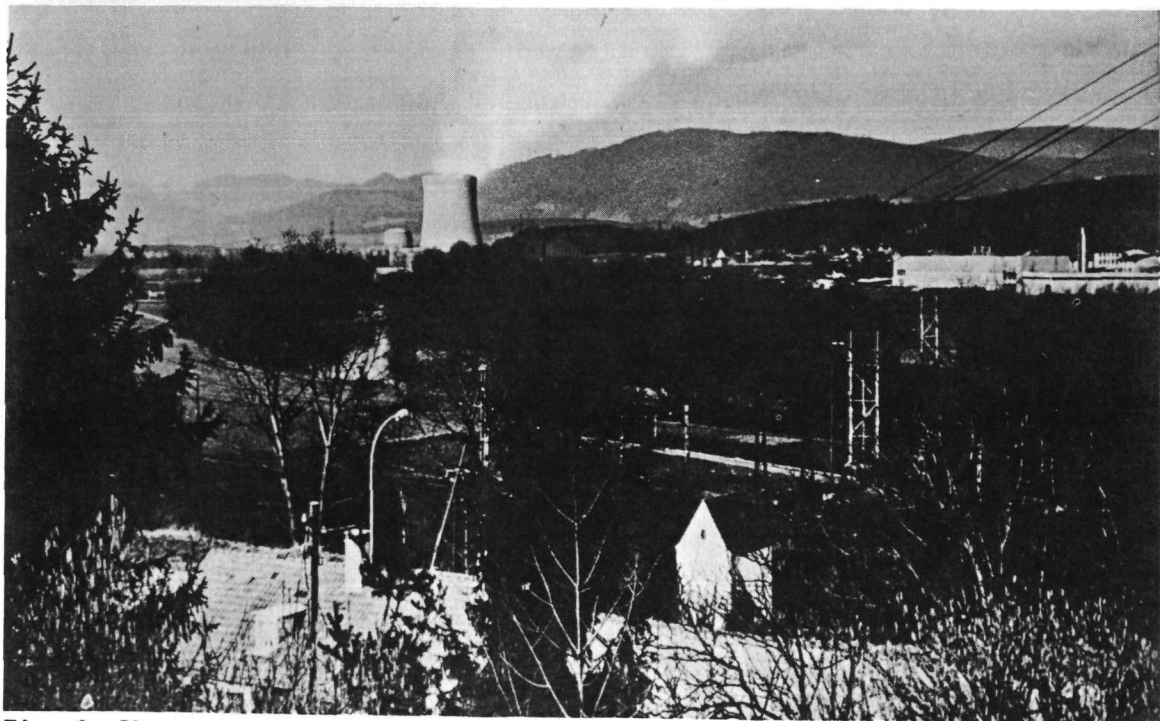


Fig. 2: Photomounting of the cooling tower for Gösgen Nuclear Power Station/ Switzerland (Courtesy Motor Columbus, Baden/Switzerland)

The success of a specific approach of the public can be well studied by the example of the nuclear power station of Neckarwestheim Germany, now under construction.

A large number of hearings were arranged within a very short period at which all the subjects could be finalized in a discussion. The villages in the community, general practitioners, journalists, etc. were invited for discussions.

Even observers not directly linked with the proposed plant, such as clerical organizations, used the opportunity and asked for a discussion between leading members of the project and opponents.

The positive outcome of this informative action in the case of the Neckarwestheim Nuclear Power Station is partly due to a native characteristic of the population of the area: they are realistic and sober people. The problems were really finalized in a discussion, and die-hard opponents were not convinced of the contrary, but the credibility of their arguments was greatly shaken in the eyes of the majority.

The situation is quite different in another siting discussion at Breisach on Kaiserstuhl, a well known winegrowing area near the Southern Black Forest.

Contrary to Neckarwestheim, which mostly covered the symptoms listed under phases 1 and 2 above, the Breisach discussion clearly highlighted the arguments of phase 3. Less specific problems of nuclear power plants were discussed rather than general problems of environmental protection: Cooling processes with wet or dry cooling towers, meteorological effects on winegrowing at a distance of a few kilometers, protection of the landscape, and in particular, the necessity of this nuclear power plant from the point of view of energy policy.

As far as the method used by the nuclear opponents is concerned, it can be said:

Local groupings like to cluster around locally well known personalities, such as the doctor, representatives of the community, chairmen of some associations, etc. who have previously shown their interest in public affairs.

The press of the organized opponent groups is well versed in the art of lending credibility to their arguments by quoting from well known experts. Objections raised in the way of stories even sometimes catch well versed experts by surprise.

Observations have shown that our opponents like to quote foreign technical literature. In Germany American literature is quoted, whilst in the United States, as far as we know, it also applies vice versa. Linguistic incompetency and the inability to follow the quotation often kill any answers that might be given, which weakness is played upon quite consciously by the opponent. In this way any quotation taken out of its context, even if it is a quotation from a well known expert - preferably Nobel prize winners are quoted here - disturbs the listener. He is ashamed of his lack of information and no longer participates in the discussion.

Experiences

This changed situation must be borne in mind in public relations work. Our experience from numerous and varied discussions with individuals and committee actions against nuclear power has shown quite clearly that large-scale meetings and the use of the mass media will be able to produce only a general information and education of the public. Events of this kind are not the right way of explaining even to an interested public more complex situations, such as the problem of the risk probability, in sufficiently accurate mathematical terms, to make the population risk conscious or to explain problems of cost benefit relations. Alternative thinking when it comes to problem solutions often verges in the well known German quotation of St. Florian: "St. Florian, pass by my house, hit others."

Good results were experienced with homogeneous groups in which one specific subject was discussed at a time. Such groups consisted for instance of physicians, teachers, students, municipal councillors and the leaders of local government groups.

In the light of personal experience gained in meetings of various kinds the authors would like to make the following recommendations:

- No excessive technical specialization.

- Simple, uncomplicated language without any sayings and without any technical terminology.
- Problems should be simplified to a permissible extent in order to meet the understanding of the respective target group.
- Meetings should consist of a brief introduction to the problem followed by a discussion.
- The subject to be discussed should be clearly defined before the meeting by mutual agreement among the groups.
- Organizing several small-scale discussions with greatly varying audiences is preferable to one large-scale meeting.

For each subject that is likely to be touched upon one well-trained expert should be available who has sufficient technical and formal knowledge of the problems.

Present Activities

In the light of the overall situation, the management of "Gesellschaft für Kernforschung" advised by the Scientific and Technical Council decided, to establish a department within the health physics division responsible for "Nuclear Power and the Public." This new department is to engage in the discussion between the public and environmental committees and nuclear power. It is to help return the controversy from the emotional, aggressive atmosphere which seems to be preferred, or even sought, by many environmental action committees back into a sober factual atmosphere which will be the only basis for fruitful work in the field of peaceful utilization of nuclear power.

The activities to be pursued by the department will be the observation and critical evaluation of public hearings and the scanning and assessment of all those publications which deal with the subjects of environmental protection in general, and nuclear power and technology in particular.

Other important duties of the department are the informing of all interested parties on the current state of discussion between nuclear technology and environmental committees, which time and again maintain that they represent the public at large; crystallize the controversies and their arguments and finally, make avail-

able to the public factual information about nuclear power and technology through publications and by actively participating in public discussions.

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METABOLISM OF RADIONUCLIDES

A STUDY OF INHALED SODIUM-22

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Abstract

This is a study of small intakes by inhalation of ^{22}Na which occurred during the machining of an irradiated target. The arrangements for handling and machining the targets are described. Measured levels of surface contamination and airborne activity, together with particle size data, are given. Whole body counting results, and measurements of the distribution of activity in the body and its variation with time are presented. The information obtained is considered in relation to predictions based on the models of the I.C.R.P. Task Group on Lung Dynamics, and the data given in I.C.R.P. Publication 10.

Introduction

^{22}Na is produced on the Nuffield Cyclotron at the University of Birmingham by bombarding magnesium targets with deuterons. At the end of the irradiation, during which about 50 mCi of ^{22}Na is normally produced, the target is removed from the cyclotron and stored for several weeks to allow short-lived activity to decay. The ^{22}Na is present in a thin layer on the surface of the target, and this active layer has to be removed so that the ^{22}Na can be recovered and processed.

This paper reports studies made to estimate the radiation dose resulting from intakes of fine dust produced during the machining of an irradiated target. The intakes were accidental and the bulk of the activity taken into the body is considered to have been inhaled due to the failure to wear a breathing mask.

Target Machining

Using long handling tongs, the irradiated target is transferred from the storage facility to a shielded enclosure within which is a scraping machine. The walls of the enclosure are made of interlocking lead bricks and a lead-glass window is included to allow the machining process to be observed. Access to the enclosure is via a two part perspex lid, one part of which can be moved. At the time of these intakes, the lead enclosure itself was not ventilated but the room in which the equipment is housed has extract ventilation. The general layout of the room and equipment is shown in Fig.1.

The active layer is removed from the surface of the target by a scraping tool which traverses its face and removes a thin strip of metal. The scraping process is carried out dry without the use of a cutting fluid. The scrapings fall down an inclined trough into a can which is capped and then manually removed using tongs. For the whole operation the operator spends a total of about an hour in the room.

Surface Contamination Levels

When this scraping facility was originally designed it was not thought that contamination outside the lead enclosure would be significant. However when the facility was brought into use, surveys revealed that loose activity was escaping from the enclosure. Typical levels of loose surface contamination at the end of a scraping run, at the positions indicated in Fig. 1., are given below i.e., much of the room was found to be contaminated to quite significant levels.

Position (See Fig.1)	Surface Activity Level ($\mu\text{Ci}/\text{cm}^2$)
S1	6×10^{-4}
S2	1×10^{-3}
S3	5×10^{-4}
S4	5×10^{-4}

Air Contamination Levels

Air samples have also been run during the target loading, machining, and can removal stages of the operation. Samples have been taken using static samplers positioned as shown in Fig.1., these positions being close to the positions occupied by the operator. Typical levels of airborne activity are shown below and these should be compared with the 40 hour week m.p.^{c} air values of $2 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$ and $9 \times 10^{-9} \mu\text{Ci}/\text{cm}^3$ for 'soluble' and 'insoluble' materials as given by I.C.R.P.¹.

Position (See Fig.1)	Air Activity Level ($\mu\text{Ci}/\text{cm}^3$)
A1	1×10^{-8}
A2	2×10^{-7}

Size Selective Sampling of Airborne Activity

In order to obtain information required for specifying the requirements of filters to remove this airborne activity, size selective sampling has been carried out using a cascade centripeter of the type described by Hounam² and calibrated by O'Connor³. Samples were taken at the positions shown in Fig. 1 during target loading, machining, and can removal, and results typical of those obtained are shown below. This distribution is very close to a log-normal distribution with an activity median aerodynamic diameter of 2.7μ and a geometric standard deviation of 1.9.

Aerodynamic diameter (microns)	Percentage of particles less than stated diameter
12.5	99.3
4.0	69.4
1.5	19.6

Whole Body Counting after Accidental Intake

On one occasion the target scraping machine operator failed to wear a breathing mask. When this was known it was realised that an inhalation intake had probably occurred, so it was decided to carry out whole body counting in order to investigate the distribution of activity and its variation with time and to estimate the dose.

Strictly speaking the intake was not a single well-defined intake but a few small intakes over a period of some hours. The first whole body count was made 2 days after the estimated mid-point of the intake and a further 7 counts were made, the last being 67 days after intake. Counting was in the 1.28 MeV photopeak, and the results obtained have been corrected for normal background and ^{40}K contribution by using data from an uncontaminated person of similar build to the person concerned.

At the first count, a total count of 22,680 was recorded, corresponding to $0.4\mu\text{Ci}$ of ^{22}Na activity in the body at that time. The results of the whole body counts were plotted to estimate the count and activity on the day of the intake ($t=0$), and the results were then normalised to the result at $t=0$ and these are plotted in Fig. 2.

Distribution of Activity in the Body

The whole body counter used for this study has four large sodium iodide detectors. Two are above the subject and positioned at the chest and pelvis, and the two below the subject are at the head and knees. For all the counts that were made, the distribution of counts among the four detectors was essentially constant and not significantly different to the distribution found after injection of ^{24}Na into a human subject.

As well as counts made using the whole body counter, checks were also made using a collimated sodium iodide detector which could be accurately positioned over various parts of the body. Counts were made 2, 11, and 18 days after intake with the detector positioned over each lung and the lower part of the trunk on each occasion. These measurements showed no significant change in the distribution of activity at these three positions over the time interval mentioned above.

Predicted Behaviour of Inhaled ^{22}Na and Comparison with Recorded Data

From the size selective air sampling data, the amounts of activity deposited at various sites can be predicted using the Deposition Model of the I.C.R.P. Task Group on Lung Dynamics⁴. This model gives the following predicted depositions.

Region	Percentage of Inhaled Activity Deposited
Nasopharynx	60 - 65
Tracheo-bronchial	8
Pulmonary	15 - 20

The I.C.R.P. Task Group on Lung Dynamics also proposes a clearance model and suggests clearance times and routes from various regions for several categories of inorganic compounds. In the case of this study, the radioactive material was sodium, but most of the original magnesium was also present and it was of interest to know how the active sodium would behave in this situation. In contact with moisture and body fluids the hydroxides of these metals would form. Sodium hydroxide is listed by the I.C.R.P. Task Group as a Class D material exhibiting rapid clearance from the lung, whereas magnesium hydroxide is a Class W material exhibiting intermediate clearance. For the radioactive sodium hydroxide, the Task Group's Clearance Model predicts clearance as below.

Region	Clearance percentage, route, and biological half-life
Nasopharynx	50% to systemic blood, 4 mins 50% to G.I. tract, 4 mins
Tracheo-bronchial	50% to systemic blood, 10 mins 50% to G.I. tract, 10 mins
Pulmonary	80% to systemic blood, 30 mins 20% to lymph, 30 mins, and then all to blood, 30 mins

Therefore if the sodium quickly separates from the magnesium, within a few hours of the inhalation all the activity will have been transferred to the G.I. tract and systemic blood. That transferred to the G.I. tract will also quickly transfer to the blood⁵ so the behaviour after that time would be expected to be identical to that of orally administered ^{22}Na . Confirmation that this is effectively so from two days after the intake is given in Fig. 2 where the normalised whole body counts are compared with the data given in Publication No. 10 of I.C.R.P.⁶, in which the clearance of ^{22}Na is described by a three component exponential derived from whole body counting studies of orally administered $^{22}\text{NaCl}$. It is seen that there is quite good agreement between the measured whole body activity and that predicted by the I.C.R.P. clearance formula, particularly bearing in mind the reported variability⁷ in biological elimination rate that can occur as the stable sodium intake is varied. Unfortunately no data is available to confirm the predicted very rapid clearance from the lung, but the data from 2 days after the exposure confirm that no translocation of activity from the lung occurred after that time. This is in accord with the findings of similar studies made from 9 to 285 days after the inhalation of another Class D material ($^{137}\text{Caesium sulphate}$) by Miller⁸.

Dose Estimate

From the I.C.R.P. Task Group on Lung Dynamics deposition model, of the total initial deposit of 0.5 μCi , the deposits in the nasal, tracheo-bronchial and pulmonary regions are expected to be 0.34, 0.046, and 0.114 μCi respectively. For the nasal and tracheo-bronchial regions it has been assumed in each case that the activity has been uniformly deposited over an area of 100 cm^2 , cleared according to the biological half-times of 4 and 10 minutes, and the resulting doses have been calculated to be 3.3 and 1.1 mrem respectively. Using the data given by I.C.R.P.¹ the average dose to the lung, assuming a biological half-time of 30 minutes, has been calculated to be 0.14 mrem, and from the data given in I.C.R.P. Publication 10, the whole body dose has been estimated to be 10 mrem.

Conclusion

From the above evidence it is concluded that the 0.5 μCi ^{22}Na inhaled was rapidly cleared from the lung and thereafter exhibited the same behaviour as does orally administered ^{22}Na . The most significant dose was the whole body dose of 10 mrem, the estimated additional doses to the nasal, tracheo-bronchial, and pulmonary regions being only 3.3, 1.1 and 0.14 mrem respectively.

Acknowledgements

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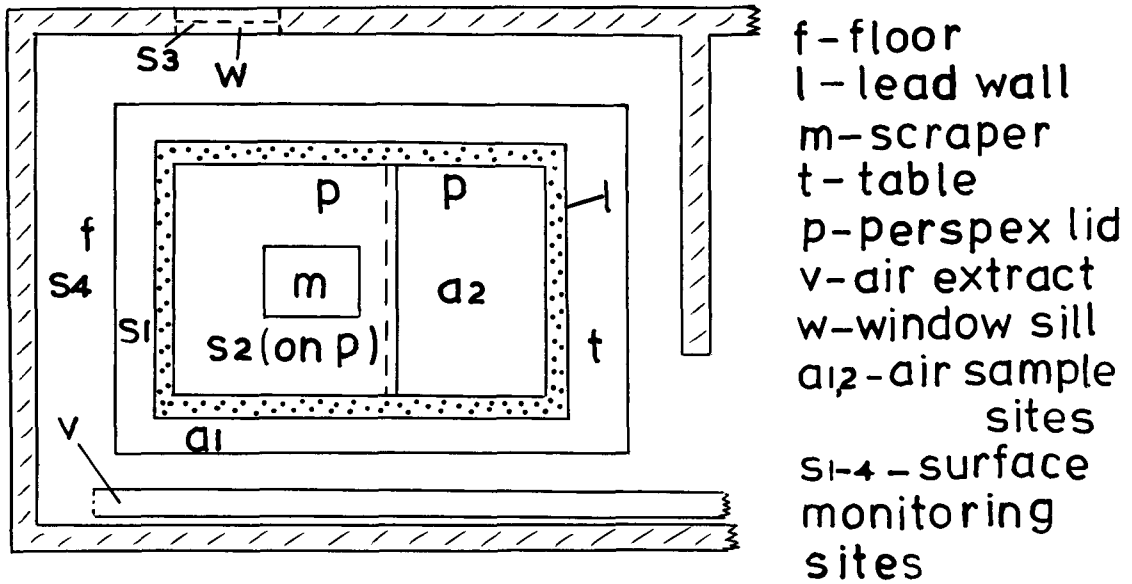


Fig.1. Layout of scraping facility.

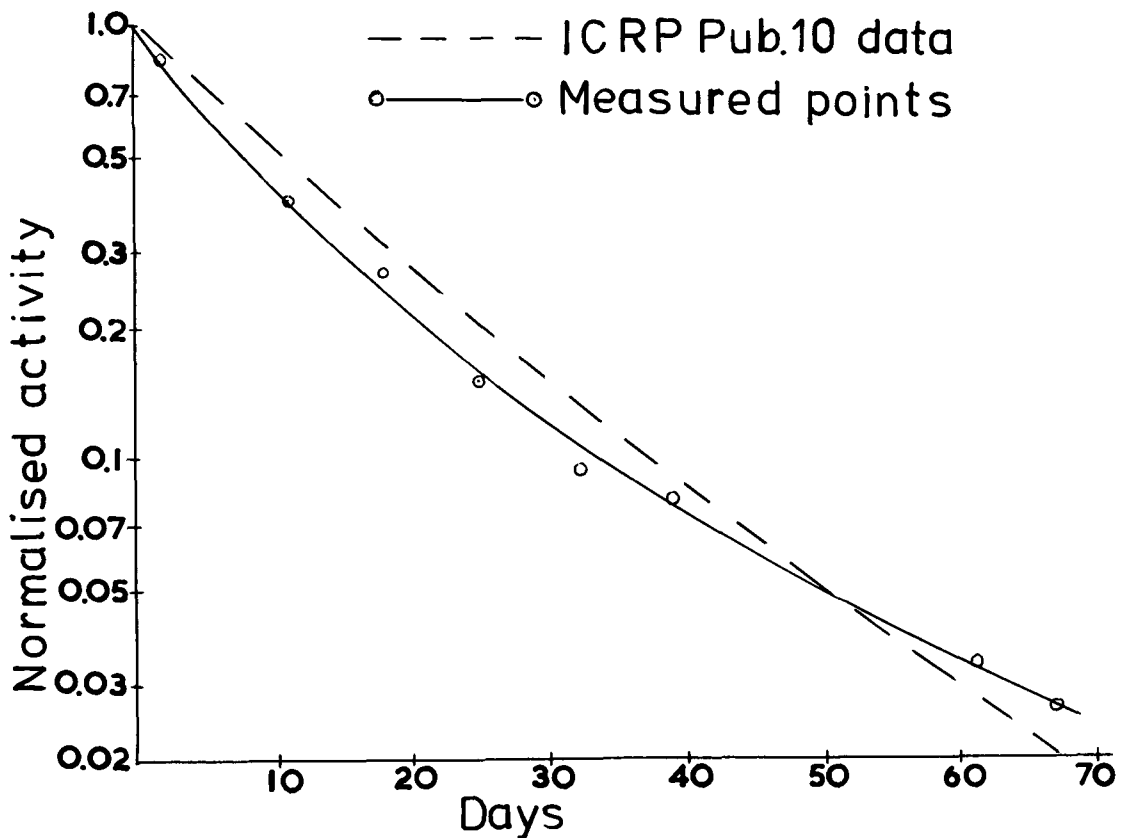


Fig. 2. Variation of whole body activity.

МЕТАБОЛИЗМ СВИНЦА-210 И ПОЛОНИЯ-210 В ОРГАНИЗМЕ

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ABSTRACT.

Metabolism parameters of Pb-210 and its product (in the body) Po-210, necessary for dosimetry and standardization are obtained in man during short-termed observations. The authors of this report have carried out their investigations on a volunteer group. These investigations deal with the specification of absorption and clearance rate of these isotopes administered per os as indicators and sufficient for many years' observations using in vivo measurements and excretion analyses. Genetically related mixture of Pb-210 and Po-210 was administered orally to the four people (the authors) whose excretions had been previously analyzed for Po-210 for a year after a single incorporation. This experiment, as the authors think, will contribute to the understanding of Po-210 metabolism. The first results of the investigations are presented in this paper and we intend to go on with these experiments in the forthcoming years.

Реферат

Параметры метаболизма свинца-210 и образующегося из него (в организме) полония-210, необходимые для дозиметрии и нормирования, в настоящее время получены у человека в основном в кратковременных наблюдениях.

Авторы данного сообщения выполнили исследование на группе добровольцев по уточнению коэффициентов всасывания и скорости выведения этих изотопов, принимавшихся через рот в индикаторных количествах, достаточных для многолетних наблюдений по прижизненным измерениям в теле и по экскретам. Четыре человека (авторы исследования), у которых предварительно в течение года исследовалась экскреция полония-210 после однократного его поступления, приняли через рот генетически связанную смесь свинца-210, висмута-210 и полония-210. Такая схема опыта по замыслу авторов позволяет выяснить обмен полония-210, который формируется в организме непосредственно из свинца-210. В докладе приводятся первые результаты исследования, которое будет продолжено в последующие годы.

Введение

Если метаболизм полония-210, первично поступающего в организм человека, изучен достаточно полно для целей дозиметрии и нормирования, то этого нельзя утверждать по отношению к свинцу-210 и, особенно, к образующемуся из него в организме полония-210. В пуб-

ликациях 10 и 10A (МКРЗ)¹ данные по метаболизму свинца- 210 не приводятся. В литературе отсутствуют также и сведения о судьбе полония- 210 , образующегося в организме из свинца- 210 , без которых расчёт поглощенных доз не может быть выполнен, поскольку основная доза создается не самим свинцом- 210 , а его дочерним полонием. Различия в обмене полония, первично поступающего в организм ("внешний" полоний) и полония, образующегося в местах фиксации свинца через висмут- 210 ("внутренний" полоний), вряд ли могут быть отвергнуты без данных эксперимента.

Принятая система параметров метаболизма свинца- 210 (публикация-2, МКРЗ)² включает: коэффициент всасывания 0,08, биологические периоды полувыведения для всего тела (в сутках)-1460, скелета-3650, печени-1947, почек-531, отношение полония- 210 к свинцу- 210 во всем теле-0,09, почках-0,18, печени-0,18, скелете-0,10 и др.

Приведенные значения в последние годы были подвергнуты проверке рядом исследователей (Харш и Суомела³, Хольтцман⁴, Кохен, Рен и Айзенбад⁵, Бланхард⁶, Хилл⁷). По нашим предыдущим исследованиям высказывалось сомнение, в частности, по коэффициенту всасывания свинца- 210 , который в отдельных случаях достигал 50%. Появились также доказательства, что отношение полония- 210 к свинцу- 210 в скелете равно 0,7-0,8, а не 0,1 как это принято в публикации 2 МКРЗ.

Все изложенные соображения дают достаточно оснований для постановки заранее запланированных исследований по изучению метаболизма свинца- 210 у человека в условиях надежного контроля поступления изотопа и его выведения в многолетних наблюдениях.

Методы и материалы исследования

Четыре практически здоровых человека (три мужчины и одна женщина) в возрасте от 45 до 50 лет приняли индикаторное количество хлористого полония- 210 однократно через рот в виде раствора при $\text{pH}=3$ в количестве 0,5 микрокюри на человека. Удельная активность раствора определялась на четырех-пийном счетчике по альфа-излучению. В течение недели ежедневно, а затем 1 раз в неделю в течение месяца и далее каждый месяц в течение 8 месяцев собирались полные суточные выделения мочи и кала. В пробах выделений определялся полоний- 210 после его осаждения на фольгу из никеля. Суточное выведение изотопа выражалось в % от введенной дозы. Этот предварительный опыт был поставлен для того, чтобы получить данные по обмену первично поступающего "внешнего" полония- 210 у каждого испытуемого и в последующем опыте учесть их при определении "внутреннего" полония- 210 . Далее те же лица, когда у них остатки изотопа стали пренебрежимо малы, приняли однократно в растворе соляной кислоты ($\text{pH}=3$) генетически связанную смесь Рв- 210 , Вл- 210 и Ро- 210 , полученную при многолетнем хранении стеклянных ампул с радоном- 222 . В смеси оказалось 1,0 микрокюри Ро- 210 , 1,9 микрокюри Вл- 210 и 3,3 микрокюри Рв- 210 . Определение принятой активности в растворе осуществлялось:

полония- 210 - по альфа-излучению на четырехпийном счетчике,
висмута- 210 - по бета-излучению на том же счетчике,
свинца- 210 - по равновесному висмуту- 210 (бета-излучение), по гамма-излучению свинца- 210 и по альфа-излучению равновесного полония- 210 .

Стандарты 100% введенной дозы готовились также путем введения раствора изотопов в чистые суточные пробы мочи и кала. Ошибка измерения дозы не превышала 1%.

После приема изотопов осуществлялся сбор суточных выделений по ранее описанной схеме. Одновременно испытуемые измерялись на

счетчике всего тела (гамма-спектрометре с датчиком из кристалла йодистого натрия толщиной 1 мм и диаметром 15 см). Детектор размещался против лобной кости. Регистрировалось излучение свинца-210 с энергией 46 кэв.

В выделениях проводились определения:

- полония-210 как по альфа-излучению высушенных проб, так и после осаждения его на фольгу из никеля,
- свинца-210 в высушенных пробах на гамма-спектрометре, по бета-излучению равновесного висмута-210 и по повторному осаждению полония-210 на никелевую фольгу после предварительного накопления.

Результаты исследования

Полный анализ всех отобранных проб еще не закончен. Он требует значительного времени. По мере отдаления от времени приема дозы и уменьшения уровней в экскретах методы суммарной альфа и бета-активности, как и гамма-спектрометрия проб оказываются неприемлемыми из-за возрастающих ошибок измерений (более 10%). Уже через 2 недели мы были вынуждены определять свинец-210 по полонию-210, повторно осаждаемому на диски никеля после накопления в течение 1-2 месяцев.

Приведенные в таблицах 1-4 материалы и вытекающие из них выводы ко времени открытия конгресса могут быть значительно пополнены. Здесь из-за ограниченности места мы хотим обратить внимание читателя лишь на явно доказуемые положения, не требующие обширных обосновывающих выкладок:

1. Среднее всасывание полония-210 при приеме его в чистом виде или в смеси со свинцом-210 превышает 18% (таблица 1) и 22% (таблица 2); среднее всасывание свинца-210 превышает 18% (таблица 3).

Таблица 1

Выведение "внешнего" полония-210 в % введенной дозы
(в предварительном опыте, без приема свинца-210)

Экскреты	Среднее выведение по 4 испытуемым в дни после приема									
	1	2	3	4	5	6	35	41	145	160
Кал	28,92	38,40	13,58	3,87	1,44	0,82	0,03	0,03	0,025	0,010
Моча	0,29		0,07	0,04	0,03			0,03	0,007	0,0014

2. Выведение "внешнего" полония-210 в интервале времени от 6 до 160 суток после приема через рот происходит по $T_{эф} = 36$ суток, что хорошо согласуется с $T_{эф} = 40$ суток, принятым в публикации Ю МКРЗ. С калом выводится полония-210 в 10-20 раз больше, чем с мочой; хотя в отдельные сутки эти различия не всегда проявлялись.

3. В начальном периоде после поступления смеси свинца-210 и полония-210 вычлнить в экскретах "внутреннего" полония-210 не представляется возможным. Судя по расчетам экскреции дочернего "внутреннего" изотопа и полагая его обмен одинаковым с "внешним" полонием, в первые 5 дней "внешний" полоний в сотню раз превышал уровень выделения "внутреннего" изотопа. И лишь через 50-150 дней можно ожидать превалирования "внутреннего" полония над "внешним" в 2-6 и более раз. Фактическую динамику выведения и ее параметров предстоит получить в дальнейших наблюдениях.

4. Выведение из организма свинца-210 происходит все с более

Таблица 2

Выведение "внешнего" и "внутреннего" полония-210 у людей в % от введенной "внешней" дозы. Изотоп принимался в смеси со свинцом-210

шифр испытуемого	Сутки после введения								
	1	2	3	4	5	6	7	14	
РПВ	кал	47,26	20,10	8,49	1,59	0,21	0,46		0,021
	моча		0,05			0,02	0,03		
ТМН	кал	44,50	26,11	14,57	4,78	1,07	1,26	0,38	0,127
	моча		0,12	0,31	0,02	0,04	0,05	0,02	0,054
НАИ	кал	36,27	46,88	7,70	0,53	0,26	0,26	0,11	0,166
	моча	0,02	0,02	0,04	0,03	0,01	0,04	0,08	0,010
ИМС	кал	28,84	11,86	17,00	0,00	4,27	0,75	2,34	0,616
	моча		0,02	0,21	0,01	0,01	0,05	0,03	0,011
Среднее	кал	39,22	26,29	11,94	2,30	0,51	0,68	0,94	0,232
	моча	0,02	0,06	0,19	0,02	0,02	0,04	0,04	0,025

Таблица 3

Выведение свинца-210 у людей в % от введенной дозы

Шифр испыту- емого	Сутки после введения										
	1	2	3	4	5	6	7	14	21	28	
РПВ	кал	55,69	15,57	7,83	1,03	0,23	0,22		0,07	0,02	0,01
	моча	0,47	0,21	0,13	0,09	0,07	0,08		0,07		
ТМН	кал	40,35	27,62	11,64	4,58	0,51	0,51	0,09	0,07	0,05	0,08
	моча	0,32	0,03	0,025	0,05	0,06	0,02	0,02	0,08		
НАИ	кал	34,94	44,51	8,68	0,32	0,12	0,06	0,05	0,06	0,05	0,06
	моча	0,18	0,08	0,07	0,03	0,02	0,03	0,03	0,03		
ИМС	кал	38,56	19,72	23,74	0,00	7,55	0,29	0,33	0,09	0,04	0,015
	моча	0,46	0,10	0,09	0,03	0,15	0,07	0,06	0,01		
Среднее	кал	42,39	26,35	12,97	1,98	0,29	0,27	0,16	0,07	0,04	0,04
	моча	0,36	0,11	0,08	0,06	0,05	0,05	0,04	0,05		

Таблица 4

Скорость счета от свинца-210 (30-56 кэв) в лобной кости (в % от первого измерения)

Шифр испытуемого	Сутки после введения							
	2	8	15	22	29	36	99	128
РПВ	100		125	90	87	80	68	
НАИ	100		94	90	80			
ИМС	100	100	155	120	110	135	65	45
ТМН	100	209	200		209	213		148
Среднее по пер- вым трем испы- туемым	100	100	127	101	96	98	67	45

замедленной скоростью и достаточно заметный спад начинается примерно через 15 дней после приема активности. С 15 по 29 сутки, если судить по измерениям лобной кости, скорость выведения свинца-210 из организма соответствует 34,6 дням, а за последующие 99 суток - 90,5 дням. Скорость выведения свинца-210 по экскретам оказалась примерно такой же как и уменьшение счетности в лобной кости.

Заключение

Полученные данные позволяют предположить, что некоторые коэффициенты метаболизма свинца-210 и полония-210, принятые в существующих рекомендациях по нормированию, не обеспечивают соответствующих гарантий. Всасывание этих изотопов (18-20%) у человека выше в 2-3 раза, чем принято. При поступлении их в составе пищевых продуктов, как было ранее нами показано⁸, достигает 50%.

В длительных многолетних опытах предстоит получить и другие константы метаболизма свинца-210 и полония-210, которые до настоящего времени не могут считаться представительными.

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USE OF A SPECIALIZED ANALOG COMPUTER FOR THE MODEL OF
THE METABOLISM OF SOME RADIONUCLIDES

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Introduction

Methods for mathematical modelling of the radionuclide metabolism in the human body and experimental animals and the method of compartment models, in particular, have been successfully employed in the solution of many theoretical¹⁰ and applied⁵ problems for a long time.

When the behavior of a radionuclide-containing system is described by a linear model, the solution is, generally, easy to obtain. The computation difficulties involved usually arise either from the amount of computing to be performed or from the incompleteness of the set of the model parameters.

However, there are a number of situations where the linear kinetic models of radionuclide transport cannot be applied. These include cases where coefficients in differential equation systems are functions of time or contain products of functions. Such systems cannot be integrated analytically, as a rule, even in quadrature solutions. Meanwhile, the solution of such problems is of considerable practical and theoretical value, since this class of models includes the interaction of radionuclides with chelate complexing agents, e.g. DTPA, the protective action of stable iodine and, finally, the models simulating the changes of metabolic "constants" with aging.

A continuous solution for non-linear models may be offered and the structure of these solutions can be studied for arbitrary input functions through the application of analog computers (AC) equipped with special devices for non-linear systems of the abovementioned types.

Although the use of analog computers for the purposes of the compartment analysis has been already discussed^{2,4}, the application of AC in the studies of non-linear models of the DTPA action, iodine metabolism and age-related changes of some parameters is a new aspect to the problem.

This paper deals with these models and some methods of solution and discusses the effectiveness of this computer analysis technique. Space does not permit a detailed discussion of each problem and an exhaustive analysis of the solutions obtained. Thus, the aim of this paper is to examine the techniques involv-

ed in the solution of these problems and to suggest suitable structural schemes to be analysed on analog computers. Therefore, specific solutions for the models under consideration and the numerical evaluation of their parameters will be outside the scope of this work.

1. Non-Linear Model of the Decorporative Effect of DTPA

It was assumed in constructing the model that:
 (a) A single dose of hepatoostetropic radionuclide, e.g. plutonium, is injected and distributed among the organs and tissues of the organism which may be represented as a system of four compartments (Fig.1). These are a compartment of blood including the pool of soft tissues X, two compartments representing the parts of the skeleton Y₁ and the liver Y₂ where exchange occurs and, finally, a general compartment Z of the radionuclide which slowly exchanges in the skeleton Z₁, and the liver Z₂ (Z₁+Z₂=Z).

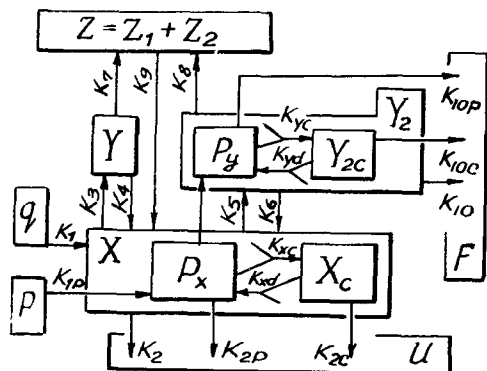


Fig.1. Nonlinear metabolic DTPA-Pu model

(b) DTPA (P) administered to the bloodstream (P_x) may be further transferred to the exchange compartment of the liver (P_y) or go to urine (U) and faeces (F).

(c) The formation and disintegration of the DTPA - radionuclide (X_c and Y_c) complex occurs in the blood compartment and the exchange portion of the liver, the excretion rates of DTPA and the complex being the same and exceeding by far that of the radionuclide itself

This mechanism underlies the decorporative effect of DTPA.

(c) The rate of complex formation is proportional to the product of concentration values of the radionuclide and chelate, and for the total volume of distribution - to the product of their amounts X P_x and Y P_y. The proportionality factor represents the effectiveness and selective properties of the chelate in the organism media.

The above model is described by the following system of differential equations:

$$\begin{aligned}
 \frac{dX}{dt} &= k_1 q - (k_2 + k_3 + k_5)X - k_{xc} P_x X + k_{xd} X_c + k_4 Y_1 + k_6 Y_2 + k_9 Z; \\
 \frac{dY_2}{dt} &= k_5 X - (k_6 + k_8 + k_{10})Y_2 - k_{yc} P_y Y_2 + k_{yd} Y_{2c}; \\
 \frac{dY_{2c}}{dt} &= k_{yc} P_y Y_2 - k_{yd} Y_{2c} - k_{10c} Y_{2c}; & \frac{dU}{dt} &= k_2 X + k_{2c} X_c; \\
 \frac{dX_c}{dt} &= k_{xc} P_x X - k_{xd} X_c - k_{2c} X_c; & \frac{dF}{dt} &= k_{10} Y_2 + k_{10c} Y_{2c}; \\
 \frac{dY_1}{dt} &= k_3 X - (k_4 + k_7)Y_1; & \frac{dP_x}{dt} &= k_{1p} P - (k_{2c} + k_{5p})P_x + k_{xd} X_c; \\
 \frac{dZ}{dt} &= k_7 Y_1 + k_8 Y_2 - k_9 Z; & \frac{dP_y}{dt} &= k_{5p} P_x - k_{10c} P_y + k_{yd} Y_c
 \end{aligned}
 \tag{Eq.1}$$

on the initial condition that:

$$q(0)=1; P(\tau)=1; x(0)=y_1(0)=y_2(0)=Z(0)=P_x(\tau)=P_y(\tau)=0 \quad (\text{Eq.2})$$

The above system (Eq.1) was studied on an analog computer, some operational amplifiers of which were used for the modelling of DTPA transport and some other - for the nuclide transport (Fig.2).

As seen from the system (Eq.1), the complexing process is included in the terms $K_{Xc}P_x X$ and $K_{Yc}P_y Y_2$ of the model. This pro-

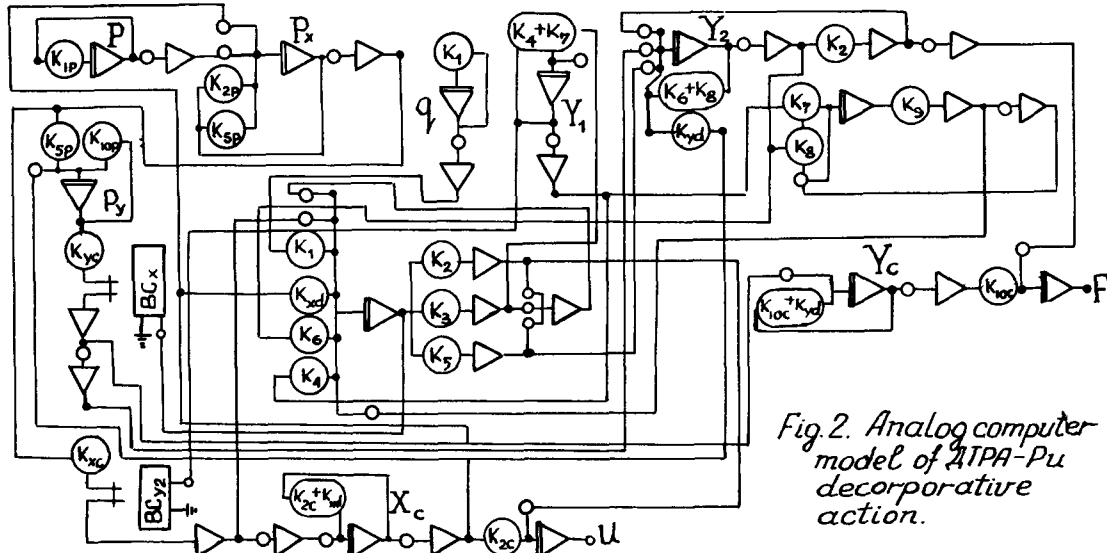


Fig. 2. Analog computer model of DTPA-Pu decorporative action.

cess was performed by special units of the computer which effected the same reaction through the scheme of $K_{Xc} P_x \delta_0(x)$ and $K_{Yc} P_y \delta_0(y_2)$. $\delta_0(x)$ and $\delta_0(y_2)$ are Kronecker's symbols.

$$\delta_0(x) = \begin{cases} 1, & x > 0 \\ 0, & x = 0 \end{cases}, \quad \delta_0(y) = \begin{cases} 1, & y > 0 \\ 0, & y = 0 \end{cases}$$

δ_0 imply the termination of complexing, when the free radio-nuclide content of the compartments X and Y_2 becomes equal to zero.

Application of multiplier units results in a considerable error and a decision bias due to the output residual voltage.

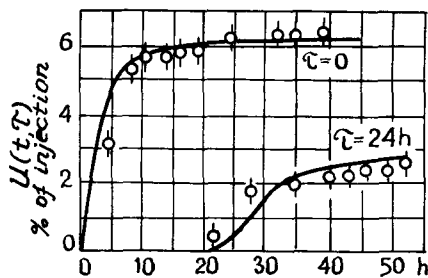


Fig. 3. The solution $U(t)$ of system (1).

To compare the predictions with the real behavior of the system, the results of our own study of the initial stage of the metabolism and excretion of plutonium citrate in the rat's organism, following a single administration of 1 ml 5% solution of DTPA, were used (Fig.3).

As the graphs show, the computed prediction for the function $U(t)$ is consistent with the experimental findings. Also, similar results were obtained for the functions $F(t)$ and X . The predic-

tions and experimental curves for citrates ^{241}Am and ^{144}Ce were also found to be in good agreement, the magnitude of constants K_i being, naturally, quite different.

To summarize, it should be noted that the application of AC offers an apparent advantage of producing multiple sets of continuous solutions for any compartment, for arbitrary functions of the q and P intakes. The model in question reproduced on the analog computer may be used in the study of different modifica-

tions of DTPA administration with a view to devising an optimal protection.

2. Non-Linear Model of Iodine Metabolism

Below follows a description of the structural scheme that may be used both in the evaluation of tactics of protection of the thyroid gland with potassium iodide and in the case of radiation destruction of the compartment "thyroid". Consider the two-compartment model of iodine metabolism (7) shown in Fig.4.

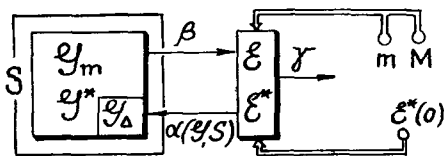


Fig.4. Two-compartment model of iodine metabolism.

The disturbance in exchange processes is induced by a change in the rate of the iodine uptake by the thyroid, i.e. by a change in the communication constant of α . Bonnel and Adams' offered a linear approximation of the α dependence on the level of the thyroid globulin depot repletion: $\alpha = R(1 - y_m/S)$, (Eq.3)

where R corresponds to the maximal iodine absorption from plasma ("full iodine insufficiency").

G_m - level of iodine pool repletion (mg iodine) due to its dietary intake (mg in 24 hrs).

S - full iodine pool - the size of the globulin depot expressed in terms of mg iodine the maximum amount of which can be taken up by the thyroid.

The uptake of large amounts of stable iodine results in a rise in the G_m level which is limited by S .

An exposure of the thyroid to doses of tens of kilorads may cause the destruction of the globulin depot S .

By substituting (Eq.3) into the set of equations which describe the model, we obtain:

$$\begin{aligned} d y / d t &= R \epsilon - R y_m \epsilon / S - \beta y, \\ d \epsilon / d t &= -(R + \gamma) \epsilon + R y_m \epsilon / S + \beta y + m + M(\tau) \end{aligned} \quad (\text{Eq.4})$$

m - dietary intake of iodine

$M(\tau)$ - quantity of stable iodine injected for the thyroid blocking at a moment τ .

A modification of the same system for radioactive iodine will be:

$$\begin{aligned} d y^* / d t &= R y_m \epsilon^* / S - \beta y^* \\ d \epsilon^* / d t &= -(R + \gamma) \epsilon^* + R y_m \epsilon^* / S + \beta y^* \end{aligned} \quad (\text{Eq.5})$$

where $S = S(D)$, while, in its turn, $D = K_D \int_0^t y^*(t) dt$.

The approximate form of this function for humans will be as shown in Fig.5.

The differential equations (4) and (5), as shown in a general form, have no analytical solutions.

To construct a model of this system on AC, a relevant structural scheme should cover:

- the state of iodine metabolism described by the equilibrium values of the pools and flows of stable iodine;
- changes occurring in the system, when large quantities of stable iodine are administered; the blockage of the thyroid;
- an adequate response to the administration of the radioactive tracer;
- the destruction of the globulin depot S caused by the radiation injury of the thyroid gland.

Fig.6. shows a structural scheme which meets all the above re-

quirements.

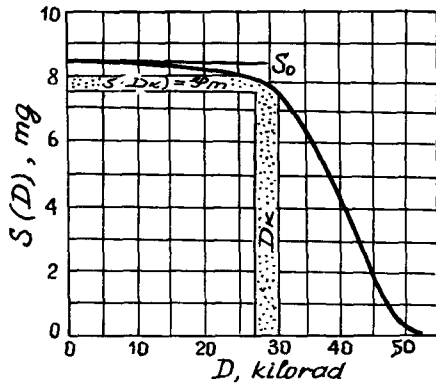


Fig. 5. The size of globuline depot as a function of irradiation dose.

The S-level changes are induced by diode elements, as the dose function is fed to their inputs. The G magnitude with respect to the level of S is limited by means of a diode limiter.

The application of dividers and multipliers imposes certain restrictions at the choice of the scale. For instance, the equilibrium values of G_m and S are 7 and 8 mg, respectively. Stable iodine doses may be as large as 500 mg. If a 100 V scale of the analog computer is used, voltages corresponding to G_m and S values will be within 10 V. Since the procedures of division and multiplication of low values of voltage involve a considerable error, the stability and reproducibility of solutions may be adversely affected.

To cause a disturbance in the analog model, the computer is supplied with doser elements which make it possible to generate voltage pulses varying in frequency and amplitude.

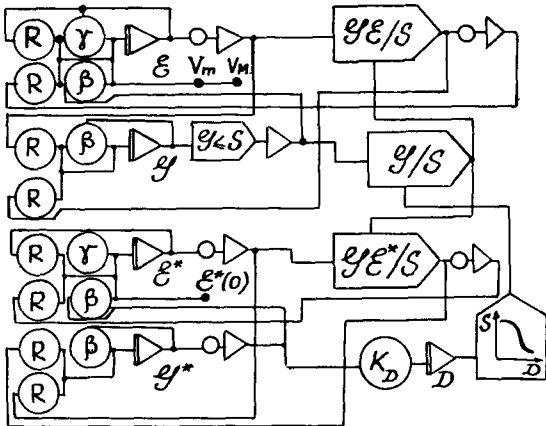


Fig. 6. The structural scheme of non-linear model of iodine metabolism. \square -non-linear transformations unit

An optimal method of protection may be developed by means of simulating different patterns of radioactive and stable iodine intakes with the aid of these doser elements.

Fig. 7 gives an illustration of the computer-predicted curve for radiiodine excretion from the thyroid in conditions of large dose burdens. The sharp bend corresponds to such magnitude of $S(D)$ that $S(D) = G_m$, while the destroyed portion of the globulin depot $G_a = S(0) - G_m$. At this moment $\alpha(G, S) = 0$. A general discussion of this problem can be found in our paper⁷.

3. Age-Related Model of Strontium and Calcium Metabolism

The dietary intake of global strontium-90 by humans commences as early as the birth, and even earlier, in prenatal life. The existing strontium-calcium metabolism models, however, are based on the parameters of adult standard man. It stands to reason that these parameters, including those of the compartments, should change with aging τ . These changes are primarily due to the build up of the skeletal calcium pool in the course of its development with time.

The changing requirement in the calcium taken with diet $q(\tau)$ is related to the skeletal pool increment $dx/d\tau$ and the size of this pool X by the following non-linear differential equation:

$$dx/d\tau = q(\tau) - \kappa(\tau)x. \quad (\text{Eq. 6})$$

This equation is a special case of a more general four-compartment model⁶ and a solution may be obtained by numerical methods.

Using the data on the age-associated requirements in calcium

found in¹¹ and the Mitchell curve⁹ which gives the rate of calcium

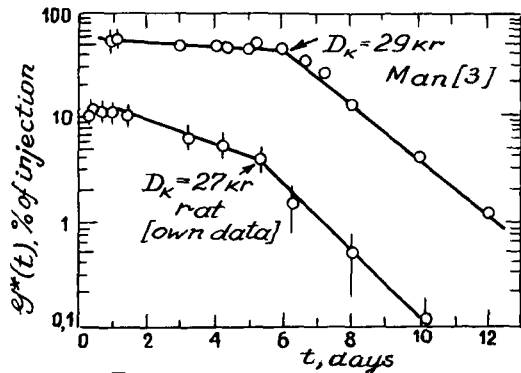


Fig.7. Computer prediction of iodine excretion from thyroid at great dose barbiturates
o - experimental data.

increment in children's skeleton, the function $K(T)$ is easily derived from Eq.6. By introducing the compartment of plasma the calcium concentration in which does not change with age, it is possible to construct a more general two-compartment model (Fig. 8a) which will correspond to the system (Eq.7).

$$\frac{dX_{pl}}{dt} = [K_u(\tau) + K_1(\tau)]X_{pl} + K(\tau)X + q(\tau), \quad (\text{Eq.7})$$

$$\frac{dX}{dt} = K_1(\tau)X_{pl} - K(\tau)X.$$

When modelling the system in analog computer $q(\tau)$ function and non-linear parameters $K(\tau)$ and $K_1(\tau)$ were simulated with

the aid of special non-linear units. AC solutions are in good agreement with calculation results.

Conclusion

Three types of non-linear problems of the compartment analysis are considered. The problems are studied and solved by means of an analog computer. To this end, a suitable structural scheme was developed for each problem. The obtained predictions are in good agreement with some experimental findings, which give every reason to consider the suggested models to be adequate.

Thus, the analog computer modelling provides an effective means for the quantitative analysis of the sufficiently complicated non-linear problems of radionuclide transport in the living organism.

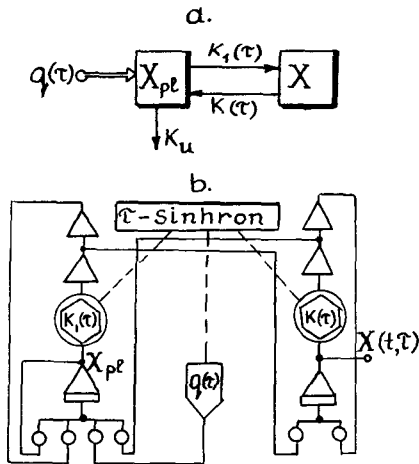


Fig.8. a. Age-related model of Sr and Ca metabolism
b. The structural scheme of AC-model.

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INTERPRETATION OF EXPERIMENTAL DATA ON POLONIUM-210
METABOLISM FOR COMPUTING ADMISSIBLE LEVELS

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Abstract

Experimental data on ^{210}Po metabolism in animals show that there are some uncertainties for the value of absorption into the blood from the gastro-intestinal tract and from the respiratory system; for the choice of the critical organ and for assessment of the nonuniformity of the internal irradiation. However there is no real basis for radical changes in the existing admissible levels of ^{210}Po intake into the human body.

^{210}Po is one of the most highly toxic radionuclides /1/. The existing ICRP recommendations are based on the admissible ^{210}Po content in the body of the professionals equal to 0.03 μCi , and the spleen is taken as a critical organ with the ^{210}Po content equal to 0.002 μCi /2/. This paper sets out to analyse experimental data on ^{210}Po metabolism in animals for calculating admissible levels of this radionuclide intake in man.

Absorption from the gastro-intestinal tract

ICRP recommendations are based on the value of ^{210}Po absorption from gastro-intestinal tract into the blood equal to 0.06. This value was obtained by administering inorganic compounds of ^{210}Po to animals /3, 4, 5/. Our data for dogs and rats confirms this value. However Morrow et al. /6/ have

shown already that absorption into the blood depends on physico-chemical properties of the ^{210}Po compound administered. According to their data for cats the rate of absorption from intestinal tract for soluble polonium citrate was 10 times higher than for colloidal polonium. It was Hill /7/, Litver /8/, Kauranen and Miettinen /9/, who first drew attention to the fact that the assessment of the natural ^{210}Po intake by people from the Arctic regions who eat reindeer meat gives a value of ^{210}Po absorption from the intestinal tract, which is much higher than was assumed from experiments. Our indirect assessment of the natural ^{210}Po absorbed into the blood, which enters the human body from the environment mainly through food, is 0.35 /10/. This high absorption into the blood may be explained by the fact that ^{210}Po which enters the body with meat or other food stuffs is in form of organic compounds, where it is bound with highly soluble aminoacids. Another reasonable explanation for this fact is that the ^{210}Po natural intake into the body is 10^5 - 10^6 times less the amounts of ^{210}Po administered to experimental animals. This difference may influence the physico-chemical state of ^{210}Po microquantities with pH in the intestinal tract, and thus the level of absorption into the blood. Johnson and Watters' latest data /11/ show that ^{210}Po entering rats in the form of organic compounds with milk from exposed cows is absorbed from the gastro-intestinal tract into the blood in much higher levels than with administration of inorganic compounds. This may serve to confirm the first proposition. Thus the level of absorption of ^{210}Po in the form of organic compounds in food stuffs is higher than that in the form of inorganic compounds. It would thus seem that there is no reason to increase the value of the ^{210}Po absorption coefficient in order to calculate the admissible intake for professionals, as they are dealing with inorganic ^{210}Po compounds. However, when calculating the ^{210}Po intake via food chain it is necessary to take into account the fact that organically bound radionuclide is more easily absorbed into the blood.

Absorption from the respiratory system

The level and rate of radionuclide absorption from the lungs into the blood are mainly determined by the degree of solution of the inhaled compound.

Berke and DiPasqua /12/ consider that ^{210}Po absorption into the rats' body after multiple inhalation amounts to 20-26% of the radionuclide inhaled. The ^{210}Po absorption coefficient for rabbits' lungs which was obtained through comparison of ^{210}Po retention after intravenous and intratracheal administration, the retention being measured by histoautoradiography and counting tracks, is 27-48% of the total amount of radionuclide retained in the lungs /13/. For rats, 29.4% of the ^{210}Po administered intratracheally is absorbed into the blood. Smith et al. /14/ obtained a value of 20.3-32.3% for the ^{210}Po absorbed by dogs inhaling polonium chloride. Little and McGandy /15/ studied the ^{210}Po absorbed into the blood through smoking. According to their assessment at least 38% of the total amount deposited in the lungs is absorbed

into the blood. Thus the probable quantity of the inhaled compound, assessed on the basis of above-mentioned data, allows us to say that about 19-34% of the ^{210}Po inhaled is absorbed into the blood.

The dynamic lung model for retention and clearance of radionuclides inhaled into the respiratory system /16/, which was developed by ICRP, is an important step forward compared with their former recommendations. According to this model polonium and its compounds belong to the "W" class (moderately soluble compounds). This model's parameters, characterizing the ^{210}Po behavior in the respiratory system, correspond to the actual process of ^{210}Po clearance from the lungs. Our data for rats and other experimental data /14/ fully confirm that the longest biological half life from lung is 50 days.

According to the ICRP model the quantity of radionuclide absorbed into the blood from respiratory system is determined by the following expression:

$$\frac{D_3 f_a \lambda_a}{\lambda_a + \lambda_r} + \frac{D_4 f_c \lambda_c}{\lambda_c + \lambda_r} + \frac{D_5 f_e \lambda_e}{\lambda_e + \lambda_r} + \frac{D_5 f_h \lambda_h f_i \lambda_i}{(\lambda_h + \lambda_r)(\lambda_i + \lambda_r)}$$

where D_3 , D_4 , D_5 - the corresponding coefficients for deposition in three regions of the respiratory system (nasopharynx, tracheobronchial region and pulmonary region) which depend on the aerodynamic diameter of the particles; f - corresponding fractions of radionuclide in each region cleared with the according constants of biological elimination - λ (f and λ do not depend on the particles' size); λ_r - the constant of radioactive decay. Substituting the corresponding figures for D , f and λ for the "W" class and $\lambda_r = 0.005$ for ^{210}Po and solving this expression, we obtain an absorption coefficient of 9.4-14.5% for the particles in size range of 0.01-10 μm . As can be seen from the above, this calculated value is substantially less than the absorption value for ^{210}Po obtained from actual experiments. This divergency can be explained by the fact that ^{210}Po in the body can not be considered a homogeneous, moderately soluble compound, because in pH in the body polonium can be found in two fractions simultaneously - aggregated (insoluble) and ionic (soluble) - which have their own rates of clearance from the system.

Analyzing the routes of ^{210}Po metabolism in the respiratory system according to the ICRP model, it can be seen that 80% of the material deposited in the pulmonary region is transported by the cilia through the tracheobronchial tree to the gastro-intestinal tract and only 15% with $T_{1/2}(\text{biol.})=50$ days is absorbed through alveolar membranes into the blood. It would seem that the value of f_e - fraction transported from the pulmonary region into the blood, for ^{210}Po amounts to 0.45 instead of 0.15. There follows a parallel reduction in the values for the f_i and f_s - fractions transported to the gastro-intestinal tract, to 0.3 and 0.2 respectively. Once the parameters have changed in this way the absorption coefficient will be equal to 15.8-32%, which corresponds more closely the actual experimental data.

Distribution in the body

^{210}Po in the blood is found in two forms - in an ionic, highly dispersed state and in an aggregated state simultaneously.

The highly dispersed ionic form of ^{210}Po probably bound with organic acids, is easily soluble and transportable; ^{210}Po in this form is distributed in the body correlating with sulphur distribution, being its analogue and possible substitute in organic compounds. ^{210}Po in this form is easily excreted through the kidneys with urine.

The other fraction is the aggregated form of ^{210}Po . Its basis is the colloidal or pseudocolloidal forms of ^{210}Po with the body pH. This form of ^{210}Po can be nonspecifically bound with protein. The aggregated ^{210}Po is not diffusible because it is in the form of large aggregates which cannot enter the membranes and the walls of blood capillaries. They are phagocitized by macrophages and the cells of the reticulo-endothelial system. That is why this form of ^{210}Po is mainly deposited in the liver, spleen, lymphatic nodes and, partly, in the adrenal glands. This form of ^{210}Po is excreted through the intestines with the bile. These states of ^{210}Po do not appear to be stable. ^{210}Po can transfer from one state to the other.

Thus, for example, aggregated ^{210}Po can be destroyed transferring to the highly dispersed form under the influence of complex-formation with organic ligands.

A part of ^{210}Po (apparently in the dispersed form) is adsorbed to the surface of the erythrocytes, and when the latter are destroyed it is deposited in the spleen and liver. The ratio of these two forms of ^{210}Po depends on many factors: the pH environment, the presence of other chemical compounds (phosphates, citrates, thiols et al) and, finally, on the concentration of polonium atoms.

Critical organ

Though at present the spleen is considered as a critical organ for ^{210}Po , there exist at least three more organs with the same radiosensitivity level, where ^{210}Po retention is 2-3 times higher than in the spleen - that is the kidneys, liver and lymphatic nodes. At the same time ^{210}Po concentration in the gonads, regarded as belonging to the first group of radiosensitivity, is only 2-3 times less than in the spleen. We observed that the relative ^{210}Po retention in the spleen decreases in the following succession: mouse-rat-rabbit-dog-man. This speaks for the fact that it is hardly reasonable to choose the spleen as a critical organ.

The earliest changes under the influence of minimal ^{210}Po quantities exceeding admissible levels 10-50 times, may be found in the function of the liver enzymatic system, in the skin and endothelial capillaries, in the blood system and in the state of enzymatic systems and bile secretion function of the liver. Those changes are as follows: transient bilirubinaemia, increased aldolase content in the blood serum, changes in the volume of renal plasma flow; displacement of adsorption exponents of ^{131}I bengal-rose by the liver cells /17/. With higher

levels of radiation after ^{210}Po administration to experimental animals the liver and kidney also show more signs of serious damage compared with other organs. However, the relatively uniform ^{210}Po distribution with prevailing retention in the reticulo-endothelial system leads to the irradiation of practically all the organs and tissues. In connection with this, damage to the neuro-endocrinal systems is characteristic for ^{210}Po ; this damage in its turn indirectly furthers the development of radiation damage in various organs stemming from the direct impact of α -irradiation /18/. The essentially adaptive reactions of the sympatho-adrenal system and the pituitary-adrenal cortex system may exceed the physiologically expedient level and aggravate the progression of radiation sickness. One should bear in mind that the direct impact of ^{210}Po on the hypothalamopituitary region and on the reproductive system is more important than its effects on the reticuloendothelium, where the radioisotope preferentially accumulates. The concept of "the critical organ" or the preferentially irradiated organ is inadequate for analysis of the pathological process following exposure to ^{210}Po . It is probably fair to say that the critical organ concept is valid only when we consider the reaction of the body as a whole that develops as a result of the direct impact of the isotope on tissues and its indirect effects. An this considerably complicates the choice of the critical organ for ^{210}Po . However calculation of admissible irradiation levels taking various systems as the "critical organ" (kidneys, liver, spleen, reproductive system, the whole body) reveals no essential divergencies for assessing admissible intake.

Non-uniformity of irradiation

In spite of the generally uniform ^{210}Po distribution in the body, the difference between the highest concentration (in kidneys) and the lowest (in the skeleton) constitutes two orders. In the homogeneous tissues the ^{210}Po distribution is fairly diffusive and relatively uniform. However in some organs (kidney, spleen) the distribution of ^{210}Po is non-uniform. Thus, for example, the ^{210}Po concentration in the renal convoluted tubules is approximately 30 times higher than in the medulla, as was obtained by the histoautoradiographic method used for rabbits /13/. Even with the regular intake of radioactivity by man at uranium mines the ^{210}Po concentration in the renal cortex is 4.2 times higher than in the medulla /19/. In rabbit spleen after a single administration of ^{210}Po the ratio of red pulp/follicle concentrations is about 5/13/. On the whole, the maximum concentrations of the radionuclide in the kidney and spleen are usually 2-3 times higher than the mean concentration for organ, which is used to calculate admissible levels. In connection with this fact one should take into account the non-uniformity of distribution while assessing the maximum levels of irradiation on the basis of the mean concentration for the organs (or the total content in the body divided by the organ's weight).

Elimination half-life from the organs

The decrease of ^{210}Po in the organs after a single administration is well described by one exponential function during a long-term observation. Only in the initial phase, which is limited to several days after administration, it is possible to determine for certain organs one more rapidly eliminated fraction with a short half-life (lymphatic nodes, liver, kidney). The rate of decrease for ^{210}Po amounts, as a result of biological elimination and radioactive decay expressed through T_{eff} , can be taken as similar for all organs and tissues. The T_{eff} differences in the experimental data can be explained by the insufficient number of measurements, by individual deviations and by the range of measurements. This speaks for a similar mechanism of ^{210}Po metabolism in tissues. The T_{eff} value ranges from 29 to 42 days /18/ as was confirmed by experimental data on dogs, rats and mice after a single ^{210}Po administration. The value of 37 days may be generally accepted. This value corresponds well to the data for man which range from 22 to 47 days /21-24/. The rate of ^{210}Po excretion from rabbits is considerably higher. T_{eff} for rabbits is about 6-19 days /25/.

Conclusion

Thus after analyzing the experimental data on ^{210}Po metabolism obtained from animals, it is possible to conclude that there are some uncertainties regarding the coefficient of absorption into the blood from the gastro-intestinal tract (organically bound compounds of ^{210}Po) and from the respiratory system; regarding the choice of the "critical organ" and the assessment of non uniform internal irradiation in organs. However at present, these uncertainties do not provide adequate ground for substantially modifying existing admissible levels of ^{210}Po intake for man.

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"REGULARITIES IN METABOLISM OF RADIOACTIVE ISOTOPES
UPON INCIDENCE ON THE SKIN"

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Abstract

This report based on the experimental data and calculated-dosimetric estimates for the first time presents a discussion on the metabolic regularities of radionuclides of the elements of various groups of the Mendeleev periodic system when their solutions are applied to the skin. Common features in the distribution of these agents in the skin irrespective of their chemical origin have been established. Essential differences in the accumulation levels of radioactive substances in the skin and in the level of percutaneous resorption have been found. It is shown that transfollicular route is the main way of radionuclide penetration into the body through the skin.

In order to develop the problem of setting standards for skin contamination with radioactive substances and to find effective decontamination means, one must have proper data on the regularities in metabolism of radioactive substances when they come in touch with the skin integument.

Three consecutive stages in the metabolism of radioactive substances are conventionally assumed as follows:

on the surface of the skin, within the skin and within the body.

1. The analysis of the kinetics of radioactive contamination of the skin and of the dynamics of its decontamination may allow us to assume that radionuclides on the surface of the skin are distributed superficially at least as three layers /1,2 /. The upper friable and easily removed layer is formed as a result of mechanical deposition of radionuclides (carriers). The second layer is formed due to physico-chemical processes that determine the interaction of a radioactive

substance with active groups (radicals) of the skin superficial structures. Formation of the third layer /2/ is stipulated by ionic exchange between the macrocomponent (the surface of the skin) and the microcomponent (the radioactive ion).

Results of hystoautoradiographic studies allow us to come to the conclusion that the main route for the penetration of the majority of radioactive substances into the skin is the transfollicular route and to a lesser extent it is the trans-epidermal route /3/.

2. Metabolism of radioactive substances within the skin layer after their solutions are deposited on the surface of the skin has been studied mainly by Soviet scientists /3-5/ as well as by Japanese specialists /6-9/. Methods of solving this complex task have been found and use the technique of radiometry of consecutive horizontal skin sections 20-40 μm thick /3/. This allowed us to have data on the levels of accumulation, the mode of the intercutaneous distribution and elimination from the skin of pigs of uranium fission fragments (Cs^{137} , Sr^{89} , Ba^{140} , Y^{91} , $\text{Ce}^{141,144}$, Nd^{147} , Pr^{143} , Te^{132} , Mo^{99} , I^{131}) /3/ as well as of Po^{210} /5/ and transuranium elements (Pu^{239} , Am^{241}) /4/.

Analysis of the experimental data has established that the character of the distribution within the skin of uranium fission products is the same despite their chemical origin. The characteristic feature is a sharp reduction in the concentration of radionuclides within the skin according to the depth of the layers down to 200 μm , a more moderate reduction in the layer from 200 to 600 μm and almost a uniform distribution in still deeper layers.

Analogous regularities have been also noted in principle for Po^{210} , Pu^{239} and Am^{241} /4,5/.

The curve showing the distribution within the skin of fragmentary radionuclides down to the depth of 600 μm is well described by the following exponential equation:

$$C_x = C_0 (a_1 e^{-K_1 x} + a_2 e^{-K_2 x} + a_3 e^{-K_3 x}), \text{ where}$$

C_x is the concentration of a radionuclide at the depth of x , $\mu\text{Ci}/(\text{cm}^2 \cdot \mu\text{m})$, C_0 is the concentration extrapolated to the depth $x=0$, x is the depth in μm ; a_1 , a_2 , a_3 are the contribution of each exponent to the general function of distribution; K_1 , K_2 , K_3 are the constants characterizing the gradient of decreased concentration for the corresponding exponents, μm (see Table 1).

Experimental data and calculated absorbed doses within the skin for β -sources of various energies prove the adequacy of the surface distribution model for estimating absorbed doses of β -radiation in the skin, beginning with the energy of 0.2 MeV. In the case of more soft β -radiation sources, account should be made of the contribution to the dose due to the effect of radioactive substances that have penetrated into the skin /3/.

Osanov and his colleagues /4/ have estimated experimentally the dose distribution function for a thin plane α -source.

While the general character of the intercutaneous distribution of radioactive substances within the skin is quite of one and the same type and does not depend considerably on the time of the radionuclide exposition on the surface of that organ, the levels of their accumulation within the skin differ considerably. According to this index, all the radionuclides that have been studied may be arranged in the following order: I^{131} , $Po^{210} > Ce^{144}$, Pr^{143} , $Nd^{147} > Mo^{99}$, $Te^{132} > Sr^{89}$, $Ba^{140} > Cs^{137} > Pu^{239}$, Am^{241} .

According to the data obtained by us, the maximum accumulation of the radionuclides within the skin is achieved in a relatively short interval after the radioactive solution is applied onto the skin (within 15 minutes to one hour). The amount of radioactive substances in the skin (the surface layer 30/mm deep being excluded) is within the range from 4-5 per cent (I^{131} , Po^{210}) to 0.1 per cent (Pu^{239}) of the amount of the radionuclide applied to the surface of the skin.

In order to study the intimate exchange mechanisms of radioactive substances in the skin it is very important to have data available pertaining to the peculiarity of their interaction with various biochemical components of that organ.

Table 2 presents the data of the distribution of the radionuclides of molybdenum, tellurium, strontium, barium, yttrium, cerium, praseodymium and neodymium between various skin fractions (soluble in lipids, soluble in water and residual).

These studies carried out in cooperation with Shvydko, N.S., /3/ have also considered the distribution of radionuclides in the above mentioned fractions after their isolation from epidermis, derma and subcutaneous connective tissue. All the experiments yielded identical results that testified to the fact that despite their chemical origin, all the radionuclides present in the skin were mainly linked with insoluble proteins and hardly solving inorganic compounds (phosphates, sulphates, etc).

A relatively small amount of radionuclides interact with lipids and proteins soluble in water. All this allows us to suppose one type of mechanism that determines parameters of time for exchange and elimination of radionuclides from the skin.

This hypothesis is to some extent corroborated by the results of preliminary studies /5,10/ aimed at estimating effective half-lives (T_{eff}) of a number of radioactive substances in the skin (see Table 3).

As one can see from Table 3, despite the origin of a radionuclide in this or that group of elements of the Mendeleev periodic system, they have a common feature which is a rapidly exchangeable fraction (with T_{eff} equal to several hours) that accounts for 65-95 per cent of the amount of radionuclides deposited in the skin. The contribution of the relatively slowly exchangeable fraction (with T_{eff} equal to several days) is of the order of 5-35 per cent.

3. The levels of percutaneous resorption of radioactive substances into the body depend on the type of the chemical compound and the aggregate state of a radionuclide. As can be seen from systematized data obtained during our studies and from the published data, the range of values of this index lies within several hundredth fractions of a per cent up to several per

cent of the amount of the radionuclide applied to the surface of the skin. This allows us to classify radioactive substances according to their low, medium and high level of percutaneous resorbtion. With due consideration to some exceptions depending on the type of the chemical compound we classify the following elements: uranium, thorium, transuranium and transplutonium elements, radionuclides of the sulphur subgroup (tellurium, polonium) as radionuclides with the low level of percutaneous resorbtion (to the tenths fractions of a per cent); radionuclides of the elements of the IInd group, uranium fission products as radionuclides with the medium level of percutaneous resorbtion (from several fractions of a per cent to one per cent) and radionuclides of the alkaline elements, the chrome subgroup, the VIIth and VIIIth groups of the periodic system as those with high level of percutaneous resorbtion (more than one per cent). At the same time the analysis of all the available data on the exchange of radioactive substances being applied onto the surface of the skin did not reveal any correlation between the levels of their accumulation in the skin and within the body.

4. Comparison of the peculiarities in the distribution within various organs and tissues of radionuclides of the elements of various groups of the Mendeleev periodic system depending on the route of their entering the body, allows one to conclude that the character of their distribution when they are applied on the skin is the same as in the case when they enter the gastro-intestinal tract.

Under the conditions of additional total body X-irradiation, the exchange of radioactive substances applied onto the skin varies insignificantly. Therefore, this factor should not be listed among those that may aggravate radiation effects on the living organism.

Table 1

The values of the parameters of the function characterizing the change in the concentration of β -radionuclides in the skin according to various depths

Parameter	a_1	a_2	a_3	K_1	K_2	K_3
Value	0.83	0.15	0.02	6.93×10^{-2}	1.73×10^{-2}	2.56×10^{-3}

Table 2

The distribution of radionuclides in various skin fractions, %

Radionuclide	Fraction		
	soluble in lipids	soluble in water	residual
Mo ⁹⁹	14.5±2.5	29.6±6.4	55.9±8.0
Tl ¹³²	11.8±2.6	6.5±1.6	81.7±11.4
Sr ⁸⁹	10.3±2.7	14.4±4.3	75.3±17.2
Ba ¹⁴⁰	11.1±2.9	7.7±2.4	81.2±11.3
Y ⁹¹	11.4±2.2	4.4±1.2	84.2±1.3
C ^{141,144}	12.5±0.5	3.8±0.2	83.7±0.2
Pr ¹⁴³ +Nd ¹⁴⁷	5.8±0.7	1.9±0.1	92.3±0.7

Table 3

The kinetics of elimination from the skin of some radioactive substances

Radio-nuclide	compound	T _{eff} 1, hours	per cent	T _{eff} 2, days	per cent	Bibliography
H ³	H ₂ O	1	95	12	5	(10)
Cs ¹³⁷	CsCl	8	65	2	35	(10)
Pu ²³⁹	Pu(NO ₃) ₄	13	80	5	20	(10)
Po ²¹⁰	Po(NO ₃) ₄	8	70	15	30	(5)

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"DETERMINATION OF THE CONTENT AND KINETICS OF THE
BEHAVIOUR OF SOME RADIONUCLIDES IN THE BODY OF
MAN BY THE IN VIVO TECHNIQUE"

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Abstract

The regularities in accumulation, distribution and elimination of krypton-85, xenon-133 and iodine-131 from the body of man have been studied on volunteers. It is shown that intake of the inert radioactive gasses and gaseous iodine into the human body is primarily through the respiratory organs. The intact skin is of little importance in this process.

Были исследованы закономерности накопления, распределения и выведения из организма человека криптона-85, ксенона-133 и йода-131. [1]

Эксперименты проводились с привлечением добровольцев-мужчин в возрасте от 27 до 50 лет. Время контакта с криптоном-85 и ксеноном-133 составляло от 0,5 до 66 часов. Концентрация этих радиоактивных изотопов в герметичной камере объемом 3,1 м³ составляла $5 \cdot 10^{-8} + 10^{-6}$ кюри/л в зависимости от цели эксперимента. Экспериментальные исследования показали, что накопление и выведение криптона-85 и ксенона-133 из отдельных органов и тканей человека достаточно точно подчиняется экспоненциальному закону.

Следовательно, для расчета дозы при внутреннем облучении радиоактивными изотопами инертных газов можно использовать уравнение (1)

$$D = 0,59 \cdot E \cdot q \cdot Q \cdot R \cdot \frac{T^P}{T^P + T_{ЭК}^\sigma} \left\{ t_{ЭК} - \left[1 - \exp - \frac{0,693(T^P + T_{ЭК}^\sigma)}{T^P + T_{ЭК}^\sigma} \cdot t_{ЭК} \right] \left[\frac{T^P \cdot T_{ЭК}^\sigma}{0,693(T^P + T_{ЭК}^\sigma)} - \frac{T^P \cdot T^\sigma}{0,693(T^P + T^\sigma)} \cdot (1 - \exp - \frac{0,693(T^P + T_{ЭК}^\sigma)}{T^P + T^\sigma} \cdot t) \right] \right\} \dots \dots \dots (1)$$

где: D - доза облучения, рад;
 E - энергия излучения, Мэв;
 q - фактор, учитывающий поглощение излучения;
 Q - концентрация нуклида в воздухе, кюри/л;
 R - коэффициент распределения нуклида в органе или ткани, л/кг;
 $t_{ЭК}$ - время экспозиции в атмосфере нуклида;
 t - время после экспозиции;
 T^P - физический период полураспада нуклида;
 $T_{ЭК}^\sigma$ и T^σ - биологические периоды полувыведения соответственно при экспозиции и после экспозиции.

Параметры, характеризующие накопление, распределение и выведение криптона и ксенона из организма человека, которые входят в уравнение (1), приведены в таблице 1.

Обращает на себя внимание тот факт, что скорость накопления инертных газов в организме человека превышает скорость их выведения. Это явление особенно четко прослеживается на криптоне-85, накопление которого в жировой ткани людей заканчивается практически через 4-5 часов, а выведение через 8-9 часов. Более высокие темпы накопления инертных газов в организме по сравнению с темпами выведения, по-видимому, можно объяснить неодинаковым перепадом концентраций инертных газов в крови и в насыщаемых тканях. В процессе поступления инертного газа в организм этот перепад концентраций всегда больше, чем при выведении. Объясняется это тем, что в начале контакта концентрация газа в насыщаемой ткани близка к нулю, тогда как в крови она достигает равновесного значения через несколько минут, вследствие интенсивного газового обмена в легких. В процессе же выведения, после того, как в насыщаемых тканях накопился газ, перепад между концентрациями будет незначительным из-за непрерывного перехода газа из ткани в кровь.

В таблице 2 приведены результаты расчетов по формуле (1) тканевых доз в теле человека при внутреннем облучении радиоактивными изотопами инертных газов. Было принято, что $q = 1$.

Таблица I

Параметры, характеризующие накопление, распределение и выведение изотопа криптона и ксенона из организма человека

Органы и ткани	Изотопы криптона			Изотопы ксенона		
	$R_{л/кг}$	$T_{эк}^{\sigma}$	T^{σ}	$R_{л/кг}$	$T_{эк}^{\sigma}$	T^{σ}
Жировая ткань	0,46	1,4 часа	2,7 часа	1,4	5 часов	6,3 часа
Мышцы и другие ткани	0,047	8 мин	8 мин	0,13	0,4 часа	0,7 часа
Кровь	0,046	30 сек	30 сек	0,17	30 сек	30 сек
Легкие	2	30 сек	30 сек	2	30 сек	30 сек

Таблица 2

Тканевые дозы в теле человека, создаваемые радиоактивными изотопами криптона и ксенона при внутреннем облучении за 36 часовую рабочую неделю

	D , рад/неделя при $a = 10^{-7}$ кюри/л			
	Легкие	Кровь	Мышцы	Жировая ткань
⁸⁹ Kz	0,27	0,006	0,007	0,6
^{85m} Kz	3,6	0,09	0,09	0,7
⁸⁵ Kz	3,7	0,084	0,08	0,9
⁸⁷ Kz	20	0,45	0,4	2,2
⁸⁹ Kz	18	0,42	0,14	0,2
⁹⁰ Kz	19	0,45	0,03	0,03
⁹¹ Kz	4,5	0,1	0,009	0,009
^{131m} Xe	2,0	0,17	0,13	1,4
^{133m} Xe	2,5	0,21	0,13	0,5
¹³³ Xe	2,1	0,18	0,14	1,5
^{135m} Xe	1,0	0,085	0,02	0,03
¹³⁵ Xe	4,6	0,4	0,26	2,0
¹³⁷ Xe	18	1,5	0,12	0,16
¹³⁸ Xe	12	1,0	0,27	0,45

В опытах по изучению поступления ксенона-133 через кожные покровы участвовали трое добровольцев.

Испытуемый, тело которого не было защищено одеждой, помещался в экспозиционную камеру. После герметизации в камеру вводили ксенон-133. Органы дыхания испытуемого были изолированы от воздуха экспозиционной камеры с помощью шлема противогаза, в

подмасочное пространство которого подавали чистый воздух для дыхания. Время экспозиции составляло 3 часа.

Установлено, что поступление ксенона-133 через кожные покровы составляет не более 0,4% по сравнению с поступлением через органы дыхания. В опытах, в которых делалась попытка определить выделение ксенона-133 из организма испытуемых через кожные покровы, количественных данных получить не удалось в силу незначительности эффекта. Проведенные опыты показывают, что поступление и выведение инертных радиоактивных газов из организма человека происходит в основном через органы дыхания. неповрежденные кожные покровы в этом процессе играют исчезающе малую роль.

Аналогичные исследования [2] были проведены с газообразным радиоактивным йодом-131. Коэффициент F характеризует суммарную скорость поступления газообразного йода через органы дыхания (F_1) и через кожные покровы (F_2), т.е.

$$F = F_1 + F_2 \dots \dots \dots (2)$$

В свою очередь $F_1 = \gamma \cdot Q \cdot V \dots \dots \dots (3)$; $F_2 = \alpha \cdot Q \cdot S \dots \dots \dots (4)$

- где: Q - концентрация йода в воздухе, кюри/л;
 V - скорость легочной вентиляции, л/час;
 S - площадь кожного покрова, м²;
 γ - доля активности, остающаяся в органах дыхания и переходящая в кровь;
 α - коэффициент, характеризующий поступление йода через кожные покровы, $\frac{\text{л}}{\text{м}^2 \cdot \text{час}}$

Определение параметров, входящих в уравнения (3) и (4), осуществлялось в опытах с привлечением добровольцев. Всего было проведено 23 опыта. Из них в 18 исследовалась резорбция йода неповрежденными кожными покровами и в 5 опытах изучалось поступление йода через органы дыхания.

Концентрация радиоактивного йода в экспозиционной камере поддерживалась с помощью генератора. Аэрозольная фаза и пары элементарного йода улавливались фильтрами. В воздушное пространство камеры попадали только парообразные соединения йода. Систематический контроль концентрации йода-131 в камере давал возможность при необходимости корректировать в ходе опыта ее значение путем изменения скорости подачи из генератора. Колебание концентрации йода-131 в воздухе экспозиционной камеры не превышало $\pm 20\%$ от среднего значения.

Результаты исследований представлены в таблице 3. Как видно из таблицы 3, коэффициент задержки газообразного йода в органах дыхания у разных испытуемых отличается на небольшую величину, тогда как резорбция газообразного йода кожными покровами колеблется в 5 раз (α меняется от 1,5 до 7,3). Разброс скорости поступления газообразного йода через кожные покровы нельзя объяснить неодинаковыми исходными значениями концентрации йода-131 в воздухе, так как ее колебание в 100 раз не привело к заметному изменению величины коэффициента α у испытуемого К.А.

Все остальные параметры в процессе эксперимента (температура, влажность, режим питания и др.) были также одинаковыми. По-видимому, колебания скорости поступления газообразного йода через кожные покровы можно объяснить в основном индивидуальными фи-

Таблица 3

Основные параметры, характеризующие поступление газообразного йода в организм человека через органы дыхания и неповрежденные кожные покровы

Испы- туе- мый	Эффективный пе- риод полувыве- дения йода- ¹³¹ из крови, T_1	Эффективный пе- риод полувыве- дения йода- ¹³¹ из щитовидной железы T	Коэффициент за- держки йода в органах ды- хания λ	Доля активности йода- ¹³¹ , пос- тупившая из крови в щитовидную же- лезу α_1	Коэффициент пос- тупления газооб- разного йода через неповрежденные кожные покровы, α_2 , л/м ² , час
С.А.	6,2 часа	6,2 дня	0,75	9,1%	5,1 4,8 [■]
П.С.	-	-	-	-	2,5 ^x 4,5 [■]
К.А.	5,3 часа	6,4 дня	0,65	5,0%	2,1 2,1 2,6
К.В.	-	-	-	-	2,1 ^x 2,7 ^x 2,9 [■]
А.В.	4,7 часа	6,0 дня	0,58	8,0%	2,7 1,5 2,0 [■]
Ю.М.	4,3 часа	6,0 дня	0,8	3,7%	7,3
С.Ю.	-	-	-	-	5,1 ^x
Ф.В.	6,0 часа	7,3	0,67	10,6%	-

$$\bar{T}_1 = 5,3 \pm 0,8 \text{ часа}; \quad \bar{T} = 6,4 \pm 0,5 \text{ дня}; \quad \bar{\lambda} = 0,7 \pm 0,08; \quad \bar{\alpha}_1 = 7,2 \pm 2,9\%; \quad \bar{\alpha}_2 = 3,8 \pm 2,1$$

ПРИМЕЧАНИЕ: x) - рассчитано при $\bar{\alpha}_1 = 7,2\%$; ■ - определено с физической нагрузкой.

физиологическими особенностями испытуемых. В опытах с нагрузкой средней тяжести значения коэффициентов α практически не отличаются от значений коэффициентов при спокойном состоянии испытуемых. Для некоторых испытуемых была оценена относительная величина скорости поступления газообразного йода через органы дыхания и через неповрежденные кожные покровы

F_1/F_2	127	47	40	80
Испытуемый	К.А.	С.А.	Ю.М.	А.В.

Из приведенных данных видно, что резорбция газообразного йода кожными покровами составляет $1 \pm 2\%$ по сравнению с резорбцией органами дыхания.

Можно отметить, что резорбтивная способность кожных покровов человека по отношению к газообразному йоду выше чем к ксенону, проникновение которого через кожные покровы составляет не более 0,4%. Качественно такое различие в проницаемости кожи по отношению к газообразному йоду и ксенону можно объяснить различием механизма проникновения этих изотопов. Несмотря на то, что в настоящее время не существует единого мнения относительно механизма кожной проницаемости, но, вместе с тем, по мнению авторов [3] поступление радиоактивных веществ характеризуется следующими тремя процессами.

1. Активный физиологический процесс всасывания.
2. Процесс диффузии через кожный барьер.
3. Способность кожи, как губки, впитывать в себя и удерживать вещества, находящиеся на ее поверхности.

Можно однозначно утверждать, что инертный ксенон проникает через кожный барьер вследствие диффузии, тогда как газообразный йод вероятнее всего поступает через кожу за счет трех выше перечисленных процессов.

В связи с тем, что исследования основных параметров, характеризующих поступление газообразного йода в организм человека, проводились в лабораторных условиях на соединениях йода неизвестного физико-химического состава, представляется очень важным оценить пригодность полученных данных к реальным условиям.

В настоящее время определение физико-химического состава парообразных соединений йода представляет самостоятельную проблему, решение которой сопряжено с большими трудностями. Однако с точки зрения радиационной опасности даже при известном физико-химическом составе газообразного йода в воздухе определяющим параметром является величина радиоактивности, накопленная в организме. Эксперименты по определению коэффициента задержки газообразного йода в реальных условиях на водо-водяном реакторе и на радиохимическом заводе представлены в таблице 4 [2].

Несколько повышенное значение коэффициента задержки в реальных условиях можно объяснить присутствием в воздухе элементарного йода и йода в виде аэрозолей, которые, по-видимому, в органах дыхания задерживаются с более высокой эффективностью, чем парообразные соединения. Коэффициент задержки в органах дыхания человека газообразного йода лежит в пределах 0,7 - 0,9, что подтверждает принятое в рекомендациях МКРЗ значение этого коэффициента, равное 0,75.

Таблица 4

Значения коэффициентов задержки газообразного радиоактивного йода в органах дыхания человека

Место эксперимента	Формы йода в воздухе, %			Коэффициент задержки
	Аэрозоли	Элементарный йод	Летучие соединения	
Лаборат.исслед.	-	-	100	0,7
Водо-водяной реактор	3-21	9-16	70-86	0,84
Радиохимический завод	3	5-10	90-95	0,9

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RETENTION AND DISTRIBUTION OF INORGANIC MERCURY (^{197}Hg , ^{203}Hg)
IN THE HUMAN BODY AFTER SINGLE INHALATION

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ABSTRACT- Inorganic mercury (^{197}Hg , ^{203}Hg) was inhaled by two workers during decontamination procedures in a hot cell. The retention and distribution of the mercury in the body were studied with a whole-body counter. The biological half-lives in the whole body were about 33 days, which was shorter than those reported for organic mercuries. The biological half-lives in the upper abdomen were about 50 days.

Scanning along the body axis and counting at several different points above the upper abdomen revealed that the mercury deposited mainly in the kidneys and in the liver. The activities of the mercury-203 in the kidneys and in the liver were determined using the counting efficiencies obtained from a REMAB phantom. The total activities in the kidneys 10 days after inhalation were almost equal to those in the liver. The high concentration in the kidneys, about 5 times as high as that in the liver, suggested that the kidneys were the critical organ for the inhalation of inorganic mercury.

INTRODUCTION

A solution of radioactive mercury-197 (2 ml, 100 mCi, $\text{Hg}(\text{NO}_3)_2$) was spilled in a hot cell and the decontamination was carried out by four workers with full face masks and protection clothes. At first, it was thought that no inhalation of the radioactive mercury-197 had occurred in the human subjects. However, to make sure of it, the subjects were monitored with a whole-body counter on the 10th day after the decontamination procedures. It was found that two out of the four subjects were contaminated with radioactive ^{197}Hg and ^{203}Hg . Therefore, to assess the radiation doses of the radioactive mercuries to the critical organ, the distribution in the body and the effective half-lives of the radioactive mercuries were studied with the whole-body counter.

METHOD

Subject, contaminant and whole-body counter

The age, height and weight of the contaminated workers are given in Table 1. Their body builds were not so different from that of a typical Japanese. The whole-body counter used in this study consisted of a NaI(Tl) crystal of 8 inch ϕ \times 4 inch and a 400 channel pulse height analyser. The monitoring room was shielded with 200 mm Fe + 3 mm Pb.¹ The measurements of the radioactivities within the body were carried out by three different geometries, i.e., 1) by standard chair geometry, 2) by placing the detector above the upper abdomen of the human subject who lay on a bed in a supine position (see Fig. 1) or in a prone position and fixing the distance from the detector surface to the bed as 22.5 cm and 3) by scanning along the body axis. In the case of scanning, the detector was provided with a collimator having a 5 cm slit, and the distance from the detector to the bed was kept as 42.5 cm.

Determination of counting efficiencies for ^{203}Hg in the kidneys and liver

Table 1. Age, height and weight of the contaminated workers

Subject	Age	Height (cm)	Weight (kg)
A	29	160	53
B	32	169	50

The counting efficiencies for ^{203}Hg were obtained using a REMAB phantom (Alderson Research Lab. U.S.A.). The volume of the right kidney of the phantom was 90 ml and that of the left was 110 ml and the total volume was 200 ml. The volume of the liver of the phantom was 1300 ml. The average weight of the kidneys of adults of Japanese is 270 g and that of the liver, 1440 g.² Therefore, there were some discrepancies in the organ sizes between the phantom and a typical Japanese.

The vessels for the kidneys and liver of the phantom were filled with a standard solution of ^{203}Hg . The solution of ^{203}Hg was prepared as follows. Mercury-203 was dissolved in a solution containing HgCl_2 (2 mg) + KClO_3 (7 mg) per ml of 3N HCl. The activity of ^{203}Hg in the solution was 15.84 $\mu\text{Ci/ml}$. One ml of this solution was divided into two kidney vessels in proportion to their volumes (i.e., right, 0.45 ml, left, 0.55 ml). The radioactive solution was diluted until 200 ml for both the vessels with a diluting solution containing NaCl (10 mg) + HgCl_2 (2 mg) per ml of 0.5N HCl. The vessel for the liver also was filled with the same radioactive solution and diluting solution as those for the kidneys.

On the condition that the NaI(Tl) detector was placed above the central part of the upper abdomen and the distance from the detector to the bed was fixed as 22.5 cm, the counting efficiencies for the ^{203}Hg in the kidneys and liver were determined in the supine and prone positions. They were as follows.

$$\begin{aligned} \eta(k)_s &= 0.013 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the kidneys in the supine position,} \\ \eta(k)_p &= 0.065 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the kidneys in the prone position,} \\ \eta(l)_s &= 0.054 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the liver in the supine position,} \\ \eta(l)_p &= 0.012 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the liver in the prone position.} \end{aligned}$$

RESULTS

Determination of effective half-life of mercury in the whole body

Since it was difficult to determine the counting efficiencies for the radioactive mercuries in the whole body, the radioactivities in μCi in the whole body were not determined. Only net counting rates in the energy ranges of $77 \pm 43 \text{ keV}$ (for ^{197}Hg) and $279 \pm 59 \text{ keV}$ (for ^{203}Hg) were measured by standard chair method from the 10th to 36th day after inhalation. To obtain the net counting rates, the contribution of the ^{40}K and ^{137}Cs in the body to the energy ranges of ^{197}Hg and ^{203}Hg was subtracted. Also, the contribution of ^{203}Hg to the counting rate in the energy range of ^{197}Hg was subtracted. The contribution was estimated to be 0.81 (for subject A) and 0.83 (for subject B) times the counting rates in the photopeak of ^{203}Hg . These values were obtained after 29th day postinhalation when the ^{197}Hg had decayed sufficiently.

The net counting rates of the ^{197}Hg and ^{203}Hg in the whole body as determined by standard chair geometry are plotted against time in Fig.2. The effective half-lives were obtained by the least square method. The effective and biological half-lives of ^{203}Hg are listed in Table 2.

Distribution of radioactive mercury in the body

The distribution of radioactive mercuries in the body was estimated by scanning along the central body axis with the detector provided with the collimator. The energy range used in this scanning was from 34 to 338 keV to include the energies of ^{197}Hg and ^{203}Hg . The result of scanning on the 10th day after inhalation for subject A is illustrated by a solid line in Fig.3. It was found that the radioactive mercuries deposited mainly in the upper abdomen, but somewhat in the chest, in the lower abdomen and in the other parts of the body. On

Table 2. Effective and biological half-lives of ^{203}Hg in the body

Subject	Effective half-life in		Biological half-life in	
	whole body (day)	upper abdomen (day)	whole body (day)	upper abdomen (day)
A	19	23	32	45
B	20	24	35	49

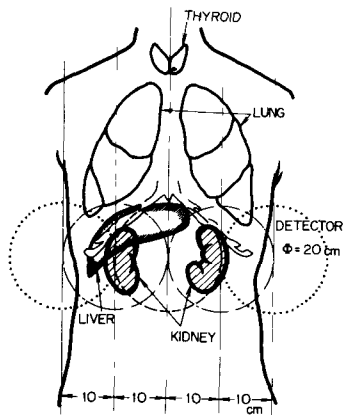


Fig.1. Relative position of detector and the kidneys and liver.

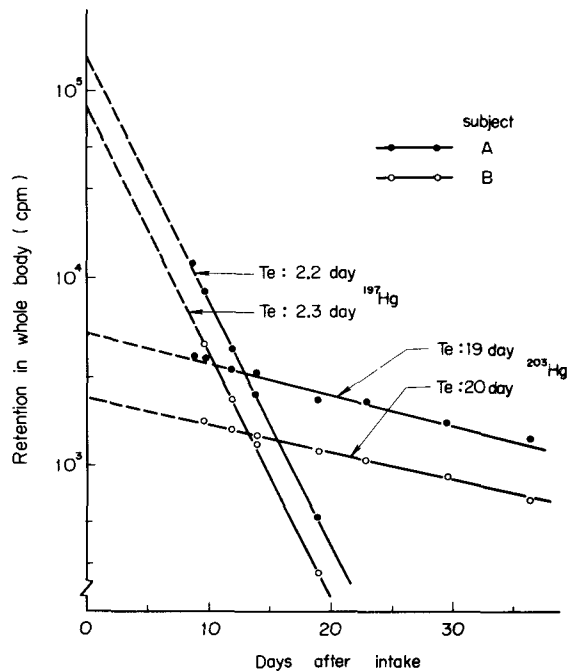


Fig.2. Retention of ^{197}Hg and ^{203}Hg in the whole body after single intake. (Te : effective half-life)

the assumption that the counting efficiencies were the same for any points along the body axis, it was calculated that about 60 per cent of the mercuries in the total body deposited in the upper abdomen. To know the organ in which the radioactive mercuries in the upper abdomen mainly deposited, the position of the detector (without collimator) was moved above the upper abdomen from the right to the left perpendicularly to the body axis at 10 cm intervals in the supine position (see Fig.1). The counting rates at each position decreased in the following order; center 10 cm right > 10 cm left > 20 cm right > 20 cm left. This result supported that the mercuries deposited mainly in the kidneys and liver.

Determination of effective half-life of mercury in the upper abdomen

The human subjects lay on the bed in a supine position and the distance between the NaI(Tl) detector and the bed was fixed as 22.5 cm as usual. The detector was placed above the center of the upper abdomen (see Fig.1). On these conditions, the mercuries deposited in the upper abdomen would be detected without much error, because the mercuries in the other parts of the body were relatively small in quantity as shown in the above paragraph for distribution.

Here, again, the net counting rates of the ^{197}Hg and ^{203}Hg were obtained by the same procedures that taken for the mercuries in the whole body. And, the

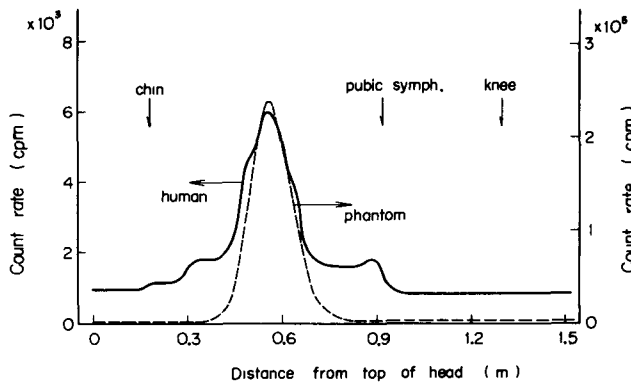


Fig. 3. Profile curves from human subject and phantom.

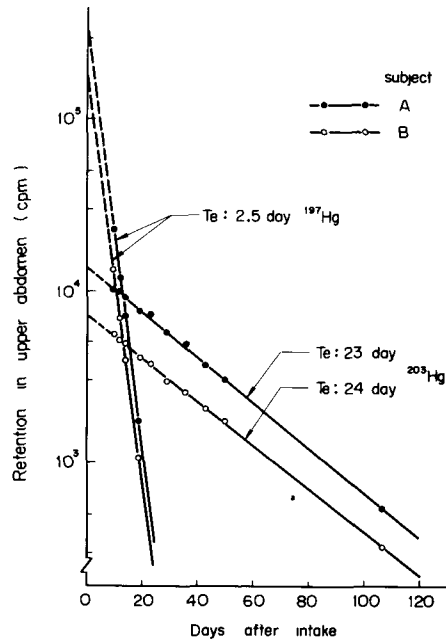


Fig. 4. Retention of ^{197}Hg and ^{203}Hg in the upper abdomen.

contribution of the ^{203}Hg to the counting rates in the energy range of ^{197}Hg were 0.72 (for subject A) and 0.68 (for subject B) times the counting rates in the ^{203}Hg photopeak.

The change with time of the net counting rates of the ^{197}Hg and ^{203}Hg in the upper abdomen is shown in Fig. 4. The effective and biological half-lives of ^{203}Hg are listed in Table 2, showing that the half-lives in the upper abdomen were longer than those in the whole body.

Determination of the radioactivities of ^{203}Hg in the kidneys and liver

It was possible to determine the radioactivities of ^{203}Hg in the kidneys and in the liver from the measurements of the human subject lying on the bed in the prone and supine positions.

If no mercury is involved in the body except for the kidneys and liver, the counting rates in the prone position, $P(\text{cpm})$, and those in the supine position, $S(\text{cpm})$, are given by the following formulas,

$$P = C E \{ \eta(k)_p K + \eta(l)_p L \}$$

$$S = C E \{ \eta(k)_s K + \eta(l)_s L \}$$

where $C : 2.22 \cdot 10^6$ (dpm/ μCi),

$E : 0.83$, emission rate of γ -rays of 279 keV per disintegration of ^{203}Hg ,

K : organ burden in μCi of ^{203}Hg in the kidneys, and

L : organ burden in μCi of ^{203}Hg in the liver.

As described already, on the condition that the distance from the detector to the bed was fixed as 22.5 cm,

$\eta(k)_p$ and $\eta(k)_s$: 0.065 and 0.013 (cpm/dpm), respectively, and

$\eta(l)_p$ and $\eta(l)_s$: 0.012 and 0.054 (cpm/dpm), respectively.

From the above formulas,

$$L (\mu\text{Ci}) = \frac{1}{CE} (19.6 S - 4.08 P)$$

$$K (\mu\text{Ci}) = \frac{S}{0.0135 CE} - 3.96 L .$$

Therefore, it was possible to evaluate the activities in the kidneys and in the liver from the counting rates in the prone and supine positions. The activities in these organs on the 10th day and 50th day after inhalation were calculated

from the counting rates on those days. The results are given in Table 3. The table shows that on the 10th day the ratio of the activities in the kidneys to those in the liver was about 1 : 1 in both the subjects. If the subjects had the kidneys of 270 g and the liver of 1440 g like a typical Japanese, the concentration of ^{203}Hg in the kidneys must be about 5 times as high as that in the liver on that date, suggesting that the kidneys were the critical organ for the inhalation of inorganic mercuries.

Table 3. Radioactivity of ^{203}Hg in the kidneys and liver on the 10th and 50th day after inhalation

Subject	Organ	Activity (μCi) on day		Ratio of activities (kidney/liver) on day	
		10th	50th	10th	50th
A	kidney	0.079	0.028	0.97	1.17
	liver	0.082	0.024		
B	kidney	0.045	0.018	1.00	1.34
	liver	0.045	0.013		

DISCUSSION

Biological half-life

Table 4 summarizes the effective and biological half-lives of ^{203}Hg reported by several workers for the whole body. ³⁻⁷ These data suggest that the biological half-lives of inorganic mercuries are shorter than those of organic ones, and the retention function of mercury should be expressed by three components.

Table 4. Comparison of effective and biological half-lives in the whole body. (Figures in parentheses show biological half-lives)

Chemical form of Hg	Route of entry	Effective and biological half-life			Author
		fast component (day)	intermediate component (day)	slow component (day)	
inorganic	inhalation	-	-	20, 19 (35) (32)	this study
inorganic	oral	-	-	22 (42±3)	Rahola et al.
^{203}Hg -methyl mercury	oral	-	-	29 (76±3)	Rahola et al.
^{203}Hg -neohydrin	oral	-	-	30 (84)	Johnson et al.
^{203}Hg -neohydrin	oral	0.22 (0.22)	7 (8.2)	-	Greenlaw et al.
monomethyl ^{203}Hg nitrate	oral	-	-	28, 27 (71) (66)	Falk et al.
			8.2 (10)		ICRP ⁷

The present study showed that the mercuries in the upper abdomen have longer half-lives than those in the whole body. This is consistent with the finding by Falk et al. that the monomethyl- ^{203}Hg nitrate deposited in the liver region decreased more slowly than those in the other regions of the body.

Distribution

Falk et al. have studied the distribution of the monomethyl- ^{203}Hg nitrate

in the body by scanning along the body axis. Their profile curves of the net counts are very similar to ours (Fig.3) as a whole. Their conclusion was that the mercury mainly accumulated in the liver region and somewhat in the cerebellum region. Our observation could not reveal special accumulation in the cerebellum, but this may be attributed to the poor resolution of the detector used in this study and/or to the difference of the chemical form of the mercury absorbed. The present study suggests that the kidneys are the most important organ where mercury deposits in the highest concentration. Falk et al. did not refer to the deposition in the kidneys.

Assessment of dose commitment due to the radioactive mercuries in the kidneys

Unfortunately, in this study the retention function of the ^{203}Hg in the kidneys could not be accurately estimated, for the measurements of the ^{203}Hg started on the 10th day after inhalation, and the fast and intermediate components of the retention were missed. However, it was probable that the dose commitment to the kidneys due to the fast and intermediate components would not be greater than that due to the slow component, as was estimated by Johnson et al. for the dose commitment to the whole body.

The dose commitment to the kidneys delivered by the slow component of the retention of ^{203}Hg were estimated as 104 and 57 mrem for subject A and B, respectively, and those to the liver, 21 and 12 mrem, respectively, on the assumption that the slow component of the retention of ^{203}Hg in the kidneys and in the liver had the same effective half-lives as those observed for the upper abdomen.

Since the radioactivity of ^{197}Hg in the body was not determined, the calculation of the dose commitment due to the ^{197}Hg was impossible, but the dose commitment to the kidneys and liver were perhaps the same order of magnitude as those due to the ^{203}Hg , inferring from the effective energies, the counting rates and their effective half-lives in the upper abdomen (see Fig.4). Anyway, the sum of the dose commitment due to the ^{197}Hg and ^{203}Hg were far less than 8 rem, the ICRP permissible dose for 3 months.⁸

Error in the determination of radioactivities in the kidneys and liver

Some error may be introduced in the estimation of the radioactivities of ^{203}Hg in the kidneys and liver, because the estimation was made on the incorrect assumption that no radioactive mercury was involved in the organs and tissues other than the kidneys and liver. However, as the deposition in the organs and tissues other than the kidneys and liver was not large as shown in Fig.3, the error would not be serious.

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A CASE STUDY OF HUMAN CONTAMINATION DUE TO
INHALED THULIUM-170 OXIDE

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Abstract

One worker incidentally inhaled submicron particles of thulium-170 oxide. Data obtained by in-vivo counting and bioassay over about 450 days after inhalation fitted to a sum of three exponential terms having effective half-lives of about 3, 23 and 90 days, respectively, though the half-life of 3 days was not distinctly determined. Attempts were made to express the daily fecal and urinary excretion by power function and to relate the excretion with the decrease of the chest burden. In addition, the distribution and transfer of thulium in the body were discussed.

1. Introduction

An incident occurred in the course of the sealing of thulium-170 sources for non destructive test in a hot laboratory at Oarai Laboratory, Japan Atomic Energy Research Institute (JAERI). The process of the sealing which involved the mounting of a neutron activated thulium oxide pellet (about 50 Ci of ^{170}Tm) in a titanium metal capsule (6 mm ϕ x 11 mm) and subsequent arc welding (3,000°C, in argon gas), was carried out in a hot cell by one worker. Following the welding process, the worker stepped in the cell and stayed there for about 5 minutes so as to carry out the sources from the cell. Leaving the cell he made a contamination check and found his hands and clothing contaminated in some measure. Then, he felt a doubt whether he had inhaled air born particles of the thulium oxide. Any air monitor was not running throughout the time of the work because it was early in the morning.

Unfortunately, four days later the worker was sent to Tokai Laboratory, JAERI, where a whole-body counter was installed. By the first measurement it was found that an appreciable amount of ^{170}Tm was deposited in his chest. Though the exposure was not itself very serious, to obtain as much information as possible the determination of ^{170}Tm by in-vivo counting and bioassay was followed until the levels of the activity were very low.

2. Methods

2. 1. In-vivo counting

Measurement was made to determine the deposition of ^{170}Tm in the chest of the contaminated subject with a whole-body counter.¹ The other series of measurements with this counter were carried out to know the distribution of ^{170}Tm in the body. The whole-body counter consisted of a 8 in. ϕ x 4 in. NaI(Tl) detector in a cubical steel room, a multichannel analyzer and associated electronics.

In chest counting, the 8 in. ϕ x 4 in. NaI(Tl) detector was placed above the chest at the distance of 22 cm from the bed on which the subject was lying on the back. In this case, the xyphoid sternum was adjusted to an edge of the crystal. Occasionally the position of the subject on bed was replaced with a prone position, to obtain the distribution of ^{170}Tm in the chest.

In profile counting, the crystal provided with a 5 cm thick lead collimator having a 5 cm slit, was moved manually along the body axis to four fixed positions, that is, above the head, chest, lower abdomen and the thigh, keeping the distance from the crystal to the bed at 42 cm.

All measurements were carried out, using the energy band from 25 to 90 keV which contains the X- and gamma-ray lines of ^{170}Tm (52, 84 keV).^{2,3,4}

In order to obtain the counting efficiency, calibrating measurement with a RANDO phantom (Alderson Research Lab., USA) was made using 149 ^{170}Tm sources in small polyethylene capsules, the total activity of which was 0.40 μCi . These capsules were inserted in hole grids within the lungs of the phantom. The arrangement of capsules simulated the uniform distribution of the contaminant in lungs. Therefore, it might differ from the truth and some systematic error might exist.

The lung size of the subject was evaluated by means of radiograph techniques, and the effective tissue thickness of the subject's chest wall was estimated according to the method which was described in detail elsewhere.^{5,6} As the result, both data of the subject were somewhat larger than those of the phantom, and it was concluded that the counting efficiencies observed on the phantom required 10 per cent corrections.

2. 2. Bioassay

After the detection of ^{170}Tm with the in-vivo counter, the subject was asked to collect the samples of urine and feces. Consequently, the sampling program was not started until 4 days after the inhalation. Both programs of the urine and fecal sampling were initiated on the 24-h sampling basis but afterward changed on the 2 or 4 consecutive day sampling basis. These procedures of sampling were continued until 134 days. After a long interval, additional samples of urine and feces were obtained for three days from 445 to 447 days.

The thulium-170 contents of the samples were determined by beta counting, for which a gas flow proportional counter was used. The counter had a detection limit of about 3 pCi. For the counting the simplest procedure was adopted as follows. In urine samples the radioactive thulium was coprecipitated with basic calcium phosphate after adding a thulium carrier. The resulting precipitates were dried, powdered and prepared for counting. Feces was ashed in a furnace before counting.

3. Results

3. 1. Retention in the chest and distribution in the whole-body

Measurement for ^{170}Tm in the chest of the subject with a whole-body counter was carried out over the period from 4 to 447 days after the inhalation. The results of twenty-four measurements are plotted in Fig. 1(a). A least squares best fit analysis by a computer was tried on the plots to fit the data to a sum of exponentials. A good fit to a sum of two exponentials was obtained as shown in Fig. 1(a) with effective half-lives of 23 ± 4 days and 90 ± 11 days.

Location of ^{170}Tm in the whole-body was roughly measured with a whole-body counter by means of the manual scan technique described above. The counting was carried out 9, 73 and 126 days after inhalation. Since it was difficult to determine the activity in the four different parts of the body, i.e., the head, chest, lower abdomen and the thigh, the deposition of ^{170}Tm in those parts of the body was expressed in terms of counting rate. The counting done on the 3 different days showed that the deposition in the chest predominated over the rest of the body, but the tendency of the decrease of the counting rates at the four different parts of the body suggested the redistribution in the body at the

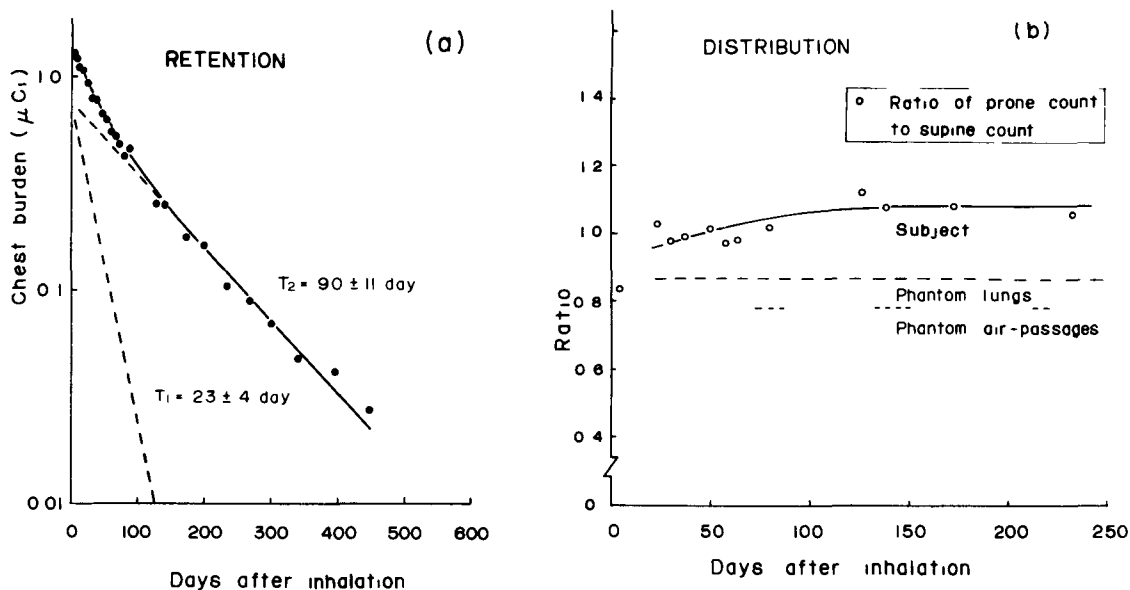


Fig.1. Retention (a) and Distribution (b) of thulium-170 in chest.

late stage of contamination (126 days).

3. 2. Distribution in the chest

To get an idea of detailed distribution of ^{170}Tm in the chest, the subject was measured both in a prone and in a supine positions by a fixed crystal-bed geometry. Twelve measurements were made over the period from 4 to 232 days after inhalation. The ratios of prone counts to supine counts were calculated. In Fig. 1(b) the ratios are plotted against time. A fitted-curve is shown by a solid line.

In a geometry similar to that taken for the human subject the phantom was measured in a prone and supine positions two times each; one, with ^{170}Tm sources in the lungs, and the other, with the sources in the lower air-passages below the throat. In the latter, the thulium-170 sources were simulated to the deposition in the trachea and the bronchia. The ratios of the counting in both positions were 0.87 and 0.78 for the sources in the lungs and in the lower air-passages, respectively, as shown in Fig. 1(b) by a broken line and by a dotted line. The ratios obtained from the phantom were lower than any ratios of the subject, which were between 1.0 and 1.1, except for 0.84 at the first measurement. The ratio for ^{170}Tm in the lungs (0.87) was higher than that for ^{170}Tm in the lower air-passages (0.78).

From these results, it is presumed that the minimum ratio of the subject at the first measurement (0.84) might indicate the initial deposition of ^{170}Tm in the tracheo-bronchial parts of the air-passages. The rather high ratio of the subject over a period of observation as compared with that of the phantom seemed to be explained by 1) the difference of geometry due to the size of the body and due to inhomogeneous deposition in the chest and by 2) the difference of the absorption and scattering of the photons through the media, i.e. the lungs, adjacent organs, surrounding soft tissue, rib cage, etc.⁷

3. 3. Urinary, fecal and total excretion

Forty-seven 24-h urine samples were subjected to analysis until 134 days after inhalation. The results obtained through beta counting are shown in Fig. 2(a) where the urinary excretion rates in $\mu\text{Ci}/\text{d}$ are plotted semi-logarithmically against time in days after inhalation with the maximum value of 1.6 nCi/d .

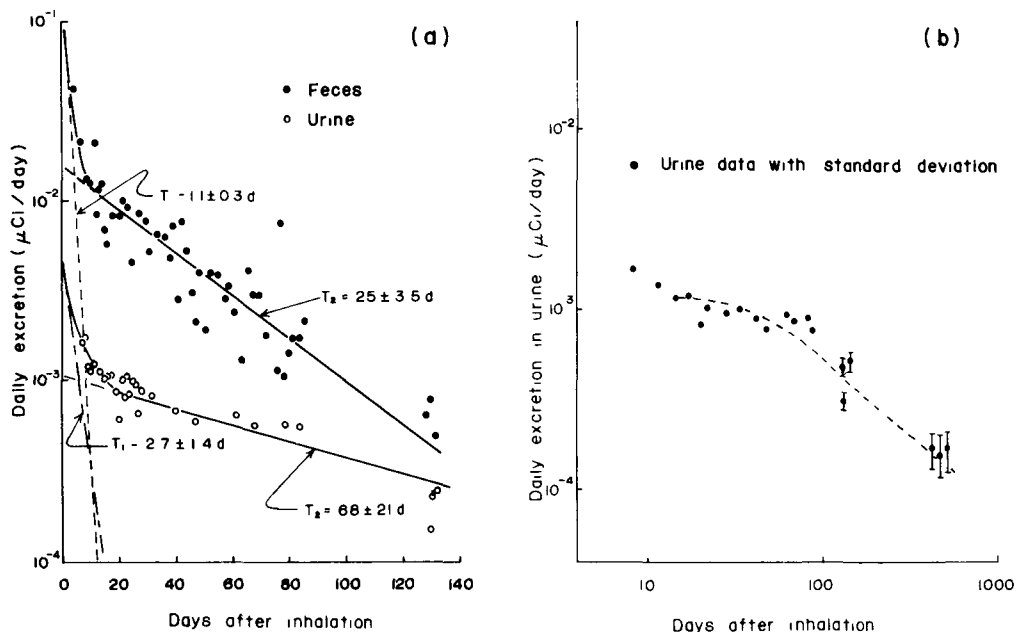


Fig.2. Excretion data of thulium-170, plotted semi-logarithmically (a) and logarithmically (b).

In this figure thirty plots from 47 measurements were taken to illustrate the excretion. With the aid of creatinine determination the samples with insufficient contents were excluded from the plots. A least squares best fit to a sum of two exponentials was obtained as shown in Fig. 2(a) with effective half-lives of 2.7 ± 1.4 days and 68 ± 21 days.

In order to see the long-term trend of urinary excretion, the data of the sample taken during the period from 445 to 447 days added to the graph of urinary excretion rates on a log-log scale. This is shown in Fig. 2(b). The plots in this figure are shown along with a curve which is arbitrarily drawn to show the long-term pattern. This pattern of urinary excretion was similar to those calculated from the radionuclide in the lungs, when the biological half-lives in the lungs were 30 and 300 days (these correspond to effective half-lives of 23 and 90 days shown in Fig. 1(a), respectively) and the exponent of the power function of urinary excretion was assumed to be $-1.0 \sim -1.7$. This calculation method is shown in the ICRP Publication 10A.⁸

During the 134 days of observation, there were 49 fecal samples in total. Over the period from 4 to 85 days all outputs of feces except one were collected. The results of determination are presented semi-logarithmically in Fig. 2(a) with the maximum value of 43 nCi/d. A good fit to a sum of two exponentials was again obtained with the effective half-lives of 1.1 ± 0.3 days and 25 ± 3.5 days.

The total activity of ^{170}Tm excreted daily in feces and urine was obtained by combining the fecal and urinary data. In this case the daily feces and the total of a single output collected on the same day as 24-h urine samples, were combined together. Here again a good fit to a sum of two exponentials was obtained. The exponentials had half-lives of about 5.4 days and about 31 days, respectively. Since there was no data for the time earlier than 7 days after inhalation, the estimated half-life for the rapid component of excretion (5.4 days) might have some ambiguity.

The daily excretion of ^{170}Tm in feces collected from 4 to 134 days after inhalation was consistently higher than that in urine collected for the same period. The ratio of urinary to total excretion changed from 0.06 to 0.3 during the above observation period. Whether this ratio changed with time was tested by

regression analysis. The coefficient of correlation obtained was 0.86 and this correlation was significant at the 1 per cent level (linear correlation was supposed until 134 days).

4. Discussion

4. 1. Half-lives

It may be interesting to compare the three sets of effective half-lives obtained from the data of urine, feces and chest. The shortest half-life appeared in the short-term component of fecal excretion (about 1.1 days). On the other hand, the longest half-life appeared in the long-term component of chest clearance (about 90 days) and the intermediate half-lives of similar length, in chest and feces (about 23 days and 25 days each). Therefore, the behavior of ^{170}Tm inhaled in the body as the oxide was presumed as follows.

(1) Within the first ten days there was a very rapid clearance, though, unfortunately, no data supporting this were obtained on the deposition and excretion for the first 4 days. The major part of deposition in the naso-pharyngeal region and in the ciliated area of the tracheo-bronchial region was excreted in feces via the gastro-intestinal tract and the minor part of the deposition was excreted in urine with a biological half-life of less than 3 days after being absorbed into the blood.

(2) In the next period of about 4 months, there was a rapid decrease with a biological half-life of about 30 days. During this period, the fecal excretion involved 1) the material initially deposited in the pulmonary region, subsequently moved up the bronchial tree and swallowed, though a certain quantity of the material was removed through the very rapid clearance phase above mentioned, and 2) the material initially deposited in the pulmonary region, absorbed in blood and afterward excreted via bile and others. These exogenous and endogenous component of excretion could not be separated by measurements.

(3) In the third period, the deposition in the chest decreased exponentially with effective half-lives of 90 ± 11 days which corresponded to the biological half-lives of 140 to 450 days with a mean of about 300 days. The activity detected in the chest would be mainly due to the non-transportable ^{170}Tm deposited in the lungs though the measurements by chest counting included the radionuclide in the lungs, lymph nodes and chest wall. In addition, as described in the results, the urinary excretion data (Fig. 2(b)) were explained by the two components which had about 23 and 90 days half-lives which were the same as those in the chest.

In short, in this case study we assumed that the retention of ^{170}Tm in the chest fitted a sum of three exponential terms having effective half-lives of about 3, 23 and 90 days, respectively, though the half-life of 3 days was not distinctly determined. On these assumptions, it was estimated that at least $1.5 \mu\text{Ci}$ of ^{170}Tm were initially deposited in the lungs of the subject.

4. 2. Distribution

The nature of the work caused the inhalation and the subsequent study concerning the aerosols strongly supported that the inhaled material was thulium-170 sesquioxide ($^{170}\text{Tm}_2\text{O}_3$) which was insoluble in water. Since it was impossible to make observation on the material inhaled, particle size investigation was carried out on the other thulium oxide pellet in which thulium-170 was previously formed by neutron activation. Thulium aerosol produced on the various conditions was measured with an electron microscope and a cascade impactor. Repeated measurements on the aerosols showed that the activity median aerodynamic diameter (AMAD) of this particle size distribution changed with the duration time of arc welding and with time after welding, and fluctuated between $0.01 \mu\text{m}$ with a geometric standard deviation, σ_g , of 5 and $0.5 \mu\text{m}$ with σ_g of 1.4. Although there was no evidence that this was the case in the incident, we estimated that submicron particles were inhaled in the incident.

Using the compartment model proposed by the Lung Dynamics Task Group,⁹ the depositions of thulium-170 in the lungs of the subject were evaluated, as a class Y compound. The percentage depositions obtained were about 1% for the

naso-pharynx region (N-P), about 8% for the tracheo-bronchial region (T-B) and about 60% for the pulmonary region (P). The dominant deposition in the pulmonary region predicted the presence of the long-term component of the retention in the chest as seen in Fig. 1(a). Moreover, the characteristics of the non-transportable submicron particle might be responsible for the rapid excretion at early stage and the subsequent slow excretion in urine as in Fig. 2(a).

Human exposure to thulium-170 oxide has been reported in literature. Eakins and Morgan,¹⁰ and Strambi and Testa¹¹ investigated different inhalation cases and studied excretion patterns, but no thulium-170 could be detected in urine.

Thomas and Kingsley¹² studied inhalation of $^{171}\text{Tm}_2\text{O}_3$ in beagle dogs and reported that 63% of the sacrificed body burden was in the skeleton, 18%, in the lungs and 11%, in the liver 128 days post exposure. However, on our observations, the depositions in the skull and in the femur were not clarified though the possibility remained.

4. 3. Urinary and fecal excretion in relation to chest retention

The urinary, fecal and total excretion rates of ^{170}Tm in $\mu\text{Ci}/\text{d}$ observed during the period from 7 to 126 days after inhalation were plotted against time in days after inhalation, on log-log paper, after being corrected for radioactive decay. Each set of values could be described by a power function, $y = At^{-B}$, where y is the activity excreted per day in $\mu\text{Ci}/\text{d}$, t , time in days and A and B , constants. The values of A and B were 0.002, 0.058, 0.085 ($\mu\text{Ci}/\text{d}$) and 0.25, 0.65, 0.71 for urinary, fecal and total excretion, respectively.

Integration of these power functions from $t = 7$ to 126 days gave the total amounts of urine, feces and total excreta during the period. They were 0.1, 0.57, 0.69 μCi , respectively. On the other hand, according to the direct chest counting, about 0.75 μCi were lost through biological routes from the chest region during the period from 7 to 126 days after inhalation. Although it was impossible to take into account the rapidly decreased component of the excretion which might appear for the first 10 days, this value, 0.75 μCi , agreed fairly well with the total amount (0.69 μCi) excreted during the same period. This attempt was also made on the exponential expression of the same data, and the similar result obtained.

Acknowledgements

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GASTRO-INTESTINAL ABSORPTION OF CERIUM IN SUCKLING MICE

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Abstract

The absorption of lanthanides from the gastro-intestinal tract is known to be extremely low in adult mammals. Our previous studies as well as some recent publications revealed that the absorption of various heavy metal ions, including lanthanides, from the G.I. tract is relatively high during the early stages of life in mice and rats, and decreases gradually during the suckling period. We investigated specifically and quantitatively the variations with age of the absorption of cerium from the G.I. tract in young mice during their suckling period.

Post partum albino mice were injected intraperitoneally with $^{144}\text{Ce}^{+3}$ citrate at different times during the lactation period. Twenty four hours after the injection to the mothers, the suckling litters were sacrificed and the ^{144}Ce content in their digestive tract, liver and carcass determined by gamma counting. The internal deposition, via gastrointestinal transfer as determined from the ratio between the ^{144}Ce activities found in the liver and carcass to that ingested with the milk during 24 hours of suckling, was about 4.5% during the first 24 hours of life and about 2% for the whole suckling period. These values, obtained under normal physiological conditions, are about two orders of magnitude above the currently accepted values for gastrointestinal absorption of lanthanides in adult mice, and confirm previous findings regarding the age dependence of the gastro-intestinal absorption of heavy metal ions during the early period of life. Various suggestions put forward to explain these findings are discussed. The assumption that the discriminatory properties of the G.I. tract in mammals are not fully developed at birth seems consistent with all recent experimental findings.

Introduction

In spite of the non-metabolic role of the rare earths, small but significant fractions of lanthanides are absorbed from the gastrointestinal tract of very young mammals.

We investigated specifically and quantitatively variations in the absorption of cerium from the gastrointestinal tract of suckling mice, under normal physiological conditions¹, and confirmed earlier findings that the absorption of ^{144}Ce during the suckling period is age dependent and much higher than in the adult mouse²⁻⁶.

The relatively high fraction of lanthanides transferred from the gastrointestinal tract into the blood stream in the very young may have some

practical consequences. The Maximum Permissible Intake values generally used are based on experiments with adult mammals whose absorption of lanthanides is extremely low. These values, though safe and acceptable for adult humans, could cause unacceptably high internal exposures when fed to infants.

Methods

Gravid albino mice were divided into seven experimental groups of four. Every mouse received an intraperitoneal injection of 5 $\mu\text{Ci}/0.2$ ml. $^{144}\text{Ce}^{+3}$ citrate during the lactation period. The seven groups were injected at different times: 3 hours after delivery, and the 2nd, 4th, 7th, 10th, 12th and the 16th day after delivery. By the 16th day the litters already consume a significant amount of solid food in addition to milk.

Twenty-four hours and 28 days after dose administration to the dams 4 neonates of each litter were sacrificed and radioassays performed on the digestive tract, liver and the remaining carcass. The first day was chosen on the assumption that during this short time there would be no excretion by the young of the contaminant ingested through the milk.

Measurements were carried out in a 3" well-type NaI crystal, connected to a single channel analyzer. For calibration of the counting system $5 \times 10^{-4} \mu\text{Ci}$ of ^{144}Ce diluted in 10 ml of water was used as a standard. The counting efficiency at 133 keV was about 1%.

Results

The transfer and internal deposition of ^{144}Ce in neonates of various ages after suckling for 24 hours from contaminated mothers is presented in Table I. Earlier experiments ^{2,3} showed that the content of the carcass could be taken as representing the retention of the skeleton. Ce transfer via milk during the first 24 h was practically equal to the activity determined in the G.I. tract while the internal deposition due to gastrointestinal transfer was calculated as the ratio between the combined ^{144}Ce activities found in the liver and carcass to that ingested during the first 24 hours.

The transfer of cerium through the intestinal wall decreases from 4.4% at birth to less than 1% at weaning. From the transfer ratios one finds that about 2.3% of the G.I. tract content is internally deposited during the entire lactation period, which corresponds to about 4.5×10^{-5} of the ^{144}Ce injected to the dam. The total fraction of contaminant transferred from the mother to her suckling offspring during lactation is so low that no radiation hazard to the breast fed infant is involved, provided lactating mothers consume food contaminated with radioactive lanthanides not exceeding the currently accepted Maximum Permissible Intake values.

The last column of Table I shows the retention of ^{144}Ce in the neonates that were allowed to suckle freely, 28 days after the dose was administered to the mother. The internal contamination of neonates is higher the earlier the injection of the dam occurred, mainly because of the longer suckling period.

Discussion

Many factors such as dietary characteristics (solid or liquid) and the chemical state (unbound or chelated) of the food, as well as the degree of peristaltic motion of the intestine are known to affect the absorption of heavy metal ions from the G.I. tract. Shiraishi and Ichikawa suggested that pinocytosis could be the main transfer mechanism of dissolved cerium ions in suckling mammals⁵. This mechanism requires the operation of chelation or polymerization processes which form aggregates or large colloids. Only in such

form could cerium ions be transported across the membrane of the G.I. tract by pinocytosis. This hypothesis, however, seems to conflict with the conclusions of Inaba and Lengemann who showed that cerium remained in the epithelial cells of the villi of the G.I. tract until eliminated by the process of sloughing⁶. Clearly the role of pinocytosis as a route of transfer cannot be regarded as proven so far. Their suggestion that the ileum might be regarded as a possible site of entry due to the previous accumulation of cerium within the epithelial cells⁶ is also not convincing. Limiting ourselves to measuring the transfer for 24 hrs suckle only, any accumulation factor is obviously eliminated without a significant reduction on the transfer of cerium into the neonates as compared to our earlier experiments².

Factors other than the formation of colloid or macromolecules may influence the absorption of polyvalent ions from the intestine. Some data indicate that the association with milk facilitates the absorption of calcium and strontium from the G.I. tract⁷⁻⁹. Taylor suggested that the high absorption of radionuclides in very young animals may be caused by the inhibition of an existing block by lactose or other factors present in the milk diet¹⁰. If this interpretation were correct one might expect a positive correlation between the amount of milk ingested and the fractional absorption from the G.I. tract. Although milk production increases steadily during the first week of lactation, the fractional absorption of cerium, (as shown in Table I) decreases continuously from the day of birth. The enhanced absorption of lanthanides from the G.I. tract in the very young cannot therefore be accounted for by the "milk effect" alone according to our experiments.

Plutonium and actinides resemble lanthanides in their extremely low absorption from the G.I. tract in adult mammals. Studies on the intestinal absorption of ingested Pu also showed an increased absorption in the very young and a gradual decrease in the G.I. transfer with age¹¹⁻¹⁴. However, it was demonstrated by Finkel¹³ that milk was not the main factor contributing to the enhanced absorption of Pu from the G.I. tract of suckling rats.

The mechanism responsible for the discrimination of the G.I. tract against heavy metal ions is thus not well understood and needs further study. It seems to take some time for an active transport mechanism favoring calcium ion absorption on the one hand, and the establishment of an effective exclusion mechanism against metabolically useless metal ions on the other, to develop after birth. This selective barrier becomes fully operative only after weaning, when the diet becomes varied and the need for a discriminatory mechanism arises.

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TABLE I. Transfer and Internal Deposition of ^{144}Ce in Mouse Neonates at Various Ages

Age of neonate at ^{144}Ce injection to dam (days)	After first 24 hr of suckling		28 days later
	Transfer via milk = body burden (a)	Internal deposition (b) (%)	Body burden ^(a)
0	8.0 ± 0.3	4.4	9.6
2	9.8 ± 0.3	3.3	9.0
4	10.8 ± 0.4	2.5	7.0
7	11.7 ± 0.5	1.9	3.4
10	12.8 ± 0.5	1.7	2.1
12	13.9 ± 0.5	1.5	0.8
16	3.8 ± 0.3	< 0.6	0.6

$$(a) \quad 10^6 \times \left[\frac{\text{Body burden of offspring}}{\text{Dose injected to dams}} \right]$$

$$(b) \quad 100 \times \left[\frac{\text{carcass} + \text{liver}}{\text{G.I. tract} + \text{carcass} + \text{liver}} \right]$$

A HUMAN METABOLIC MODEL FOR ^{14}C -LABELLED METABOLITES USEFUL IN DOSE ESTIMATION*

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Abstract

An average individual (70-kg man) takes in some 350 g of carbon per day, and his total body content of carbon is about 14 kg. Assuming uptake to blood is essentially complete, this corresponds to a biological half-time (T_b) of 28 days. Metabolic models used in ICRP Publication 10 allow only for $T_b \cong 12$ days for glycine and of less than 1 day for CO_2 . To be in agreement with the stable carbon data, one or more terms of longer half-life are needed. Such a model is defined in this paper and is more conservative than that for glycine in ICRP Publication 10 by about a factor of 3. Data of D. L. Buchanan on uptake of $^{14}\text{CO}_2$ in mice indicate that the equilibrium level approached 10^{-4} times the specific activity of $^{14}\text{CO}_2$ in air, suggesting uptake of about 1% to blood with further dilution by dietary stable carbon of ~ 0.01 . G. V. LeRoy *et al.* have determined the retention of $^{14}\text{CO}_2$ in man during the first day, but need for a compartment of long biological half-life is indicated here also. When these compartments are included in the models given in ICRP Publication 2 and ICRP Publication 10, it is found that the former model overestimates dose to the total body by a factor of ~ 30 , while the latter underestimates it by a factor indicated to be at least 16 and which might be as high as ~ 100 .

Introduction

The purpose of this report is to present a mathematical model for ^{14}C metabolism which can be used in health physics for estimating radiation dose to man from intake of ^{14}C . This model is needed for an adequate estimation of radiation dose received by research workers or others who work with ^{14}C -labelled compounds and incidentally or accidentally take ^{14}C into their bodies. In this paper, experimental data provided by others will be used and interpreted with a mathematical model.

Experimental Data

D. L. Buchanan¹ obtained data from a study of $^{14}\text{CO}_2$ inhalation by mice in stable CO_2 levels ranging from 0.03% to $\sim 5\%$ CO_2 in air. He let the mice inhale $^{14}\text{CO}_2$ for as long as 40 days, and then he studied the retention out to 50 days after the end of exposure. By serial sacrifice, both during exposure and following the cessation of inhalation exposure, he took many tissues (but not bone) and plotted the results expressed as the ratio of specific activities

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in tissues to the specific activity of air. He called this the air carbon fraction, and from his graphs, the value increased from zero at time zero up to $\sim 10^{-4}$ at 40 days or so. Thus, the equilibrium level of the specific activity was only 0.01% of the corresponding level in the air breathed by the mice.

Other data are available also, e.g., the paper by Skipper² which deals with the trapping of ^{14}C in bone following injection into blood of ^{14}C -labelled Na_2CO_3 . Here the bone was seen to retain a small fraction for a long time, but there seemed to be no chronic accumulation of C in bone.

Data are available from another experiment of Buchanan³ involving continuous feeding of a ^{14}C -labelled diet of sucrose and yeast for a period of 40 to 50 days and serial sacrifice of mice and rats from which tissues were obtained. These data show the ratios of labelled to unlabelled C in tissues and foods applied and equilibrium set in at ~ 40 days after intake began.

ICRP Publication 2⁴ refers to data of Nardi for its T_b value of 10 days for total body. A T_b value for bone based on studies on mice and a value for fat are given in Publication 2 also. Note that all of these were obtained on small animals (mice and rats).

Metabolic Model for ^{14}C

Denote by $R(t)$ the fractional retention of dietary ^{14}C in the body at time t after uptake of a unit amount into the bloodstream. Let $E(t)$ denote the cumulative excretion (via all paths), and we write

$$E(t) = 1 - R(t). \quad (1)$$

It is assumed that

$$R(0) = 1 \quad \text{and} \quad R(\infty) = 0,$$

that is, the life span of man is considered to be long enough so that retention at 50 to 70 years $\rightarrow 0$. The mean residence time (MRT) in the body is

$$\begin{aligned} \text{MRT} &= \int_0^{\infty} t \frac{dE(t)}{dt} dt \\ &= -\int_0^{\infty} t \frac{dR(t)}{dt} dt \\ &= -tR(t) \Big|_0^{\infty} + \int_0^{\infty} R(t) dt \\ &= 0 + \int_0^{\infty} R(t) dt. \end{aligned} \quad (2)$$

Thus it has been proved (and this was also noted by Bergner⁵ and probably others) that the integral of the retention function over all time is the mean of the probability distribution of residence times for an atom to be released from the body after the initial introduction of one atom at time zero. Now, one additional expression is needed to prove the MRT is given by the ratio of the body burden, $q(t)$, to the daily uptake into blood. Snyder⁶ noted that for chronic intake with $f_1 dt$ units of the isotope entering the body in dt units of time, the retention at a time t is given by, $\tau \cong t$,

$$q(t) = \int_0^t f_1 I d\tau R(t - \tau). \quad (3)$$

By changing the variables, i.e., by letting $t - \tau = T$ and $dT = -d\tau$, then (3) becomes

$$q(t) = \int_0^t f_1 I d\tau R(t - \tau) = f_1 I \int_0^t R(T) dT. \quad (4)$$

Rearranging the equation, one obtains

$$\frac{q(\infty)}{f_1 I} = \int_0^\infty R(T) dT = \text{MRT}. \quad (5)$$

Application of the Model

In applying the model to man, it is assumed that body carbon is 14 kg and the diet contains 350 g/day of carbon³ such that

$$\text{MRT} = \frac{14,000 \text{ g}}{350 \text{ g/day}} = 40 \text{ days,}$$

which corresponds to a biological half-life of $0.693 \times 40 \cong 28$ days. For mouse and rat, it is assumed that 1/5 of the body weight (and food weight per day) is carbon. Thus, for a 25-g mouse ingesting 4 g of food per day,

$$\text{MRT}_{\text{mouse}} = \frac{1/5 \times 25 \text{ g}}{1/5 \times 4 \text{ g/day}} \cong 6 \text{ days;}$$

and for a 200-g rat eating 20 g of food per day,

$$\text{MRT}_{\text{rat}} = \frac{200}{20} \text{ day} = 10 \text{ days.}$$

In the above estimates, it is assumed that complete (100%) uptake of dietary C occurs.

The above values for mouse and rat agree with Buchanan's tissue data (from the experiment using a diet labelled with ¹⁴C) in that the ratio of specific activity of organs and tissues to specific activity in diet is approximately given by

$$\frac{q^*/q}{I^*/I} = 1 - e^{-\lambda_b t}$$

which approaches 1 at large times (40 to 50 days in the case of rats and mice) where the asterisk denotes ¹⁴C. Then we can show

$$1 - \frac{q^*/q}{I^*/I} = e^{-\lambda_b t}$$

which indicates that the exponential decreases with the reciprocal mean residence time as the decay constant. Buchanan's feeding data are in approximate accord with this equation. However, in some cases (brain and muscle, for example) more than one exponential is indicated, but the MRT for most tissues is 4 to 10 days which is approximately correct. Now Buchanan's inhalation experiment can be interpreted.

New Model for $^{14}\text{CO}_2$ Inhalation

The daily intake of C in inhaled air is needed. For a mouse inhaling 0.15 cc per breath (the tidal volume) and 163 breaths per minute,⁸ 23 cc of air are taken in per minute which corresponds to an intake of 33 liters per day. Air normally has 0.03% CO_2 , and since CO_2 has 44 g/mole and 1 mole of any gas occupies 22.4 liters, then

$$3 \times 10^{-4} \times \frac{33 \text{ liters}}{\text{day}} \times (22.4)^{-1} \frac{\text{mole}}{\text{liter}} \times \frac{12 \text{ g C}}{\text{mole}} = 0.005 \text{ g C/day}$$

is inhaled by the mouse. Since he eats 4 g of food per day with 1/5 being C, he ingests

$$4 (1/5) \text{ g C/day} = 0.8 \text{ g C/day.}$$

So he inhales $\sim 10^{-2}$ as much carbon as he ingests. Thus the inhaled radioactive $^{14}\text{CO}_2$ is diluted 100 times by ingestion of uncontaminated C present in food. This accounts for 10^{-2} of the 10^{-4} fraction observed by Buchanan. Evidently only 1% of the inhaled C is taken up by the blood in a form metabolically similar to C in food. This could be related to the fact that alveolar air has 3 to 5% CO_2 concentration or 100 times the air inhaled. Thus the specific activity of the inhaled air is diluted by another factor of 100 to produce the equilibrium level in tissue.

It is believed that this is an important parameter to use in the case of inhalation intake of $^{14}\text{CO}_2$ by a man who had inhaled, accidentally or otherwise, $^{14}\text{CO}_2$.

To get μCi -days residence, we use

$$0.01 \mu\text{Ci} \int_0^{\infty} e^{-0.693t/28} dt \text{ days} = 0.4 \mu\text{Ci-days}$$

where 0.01 μCi represents uptake of 1% of 1 μCi inhaled by man. Compare this to the ICRP Publication 2 value of

$$0.75 \int_0^{\infty} e^{-0.693t/10} dt \mu\text{Ci-days} = 10.8.$$

Thus from their model, one overestimates the dose (given by

$$D = 51 \times \left(\frac{q}{m} \frac{\mu\text{Ci}}{\text{g tissue}} \right) \times \left(\frac{6 \text{ MeV}}{\text{dis}} \right) \times (\text{MRT days})$$

so dose is proportional to μCi days) by a factor of

$$\frac{10.8}{0.4} = 27.$$

When the above is compared to the model in Publication 10 of ICRP,⁷ care must be taken because Publication 10 only gives μCi -days residence for intake to the blood. There, for $\text{NaH}^{14}\text{CO}_3$, they give a value of 0.58 μCi -day for bone, but this is for uptake to bone of 1 μCi and not for inhaled ^{14}C . For the case of injection into blood, that fraction f , which has the metabolism of C taken in food, would give

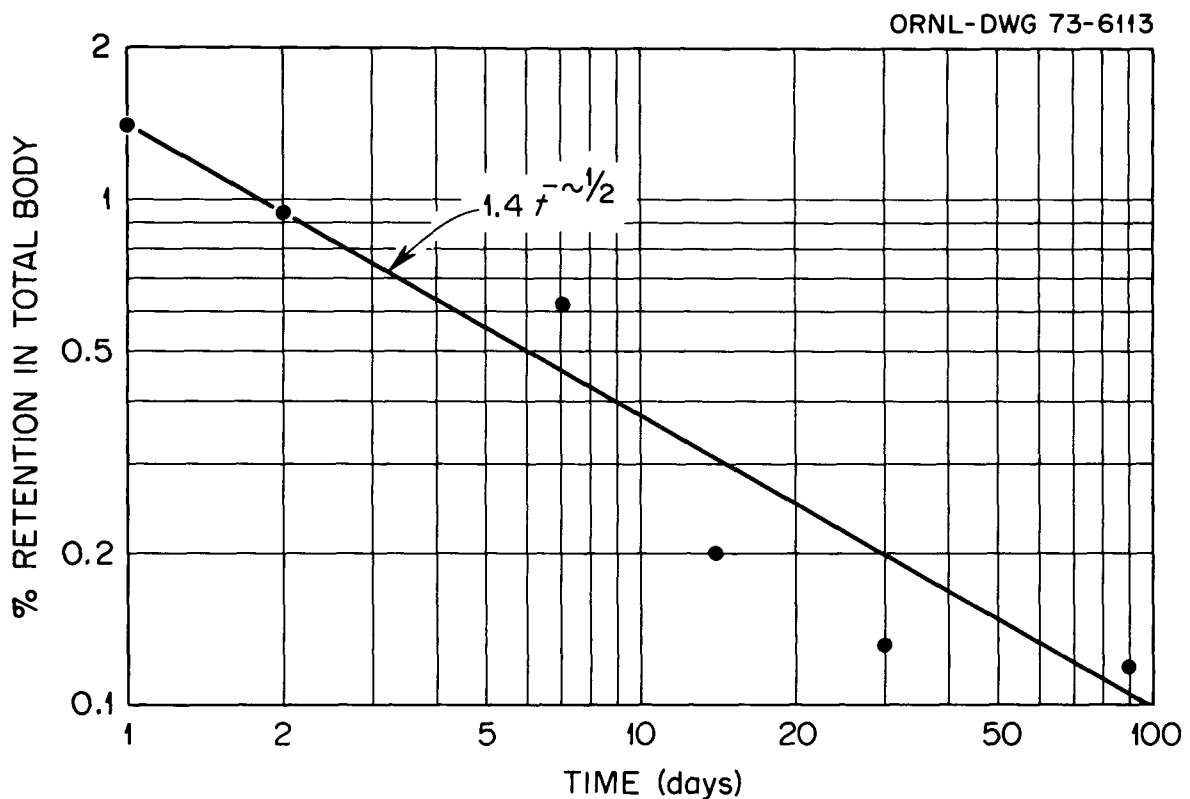
$$f \times \int_0^{\infty} R(t) dt = 40 f \mu\text{Ci-days.}$$

Included here is a logxlog graph of Skipper's data on $^{14}\text{CO}_2$ total body retention in mouse after a single injection of $\text{NaH}^{14}\text{CO}_3$ (Fig. 1). From here it can be noted that the power function will fit the data, and the MRT out to 900 days is only 1 day, somewhat lower than the earlier estimate of 6 days. This might indicate rapid exhalation of $5/6$ of the total activity injected. Also, although the power function implies some trapping, it should be noted that there is no excessive concentration of C in bone or other organs in the body. More data on larger animals are needed here. Meanwhile, the value $f = 1/6$ is suggested from the above data on mice.

Thus, from the above considerations, it can be seen that a model for $^{14}\text{CO}_2$ inhalation of a single intake has been derived. Note, however, the value of 1% uptake of ^{14}C into blood from inhalation is based on a very small animal--a mouse. The author knows of no data on a larger animal, such as a dog, from which the 1% could be verified. Note also in the above that the mean residence time factor indicates how fast the specific activities rise for the case of continuous intake. Also, when more than one exponential is involved in the retention, then the integral of the retention function gives the MRT; hence, it can be said, conversely, where the MRT is known, one can infer the approximate correctness of the retention function. For example, Publication 10 gives the retention function for injection of $\text{NaH}^{14}\text{CO}_3$ into the blood of man as

$$R(t) = 0.7 e^{-0.693t/0.05} + 0.3 e^{-0.693t/0.4}, \quad t \text{ in days,}$$

and from this



Skipper's $^{14}\text{CO}_2$ Data Injection of $\text{NaH}^{14}\text{CO}_3$ into Mice.

Fig.1.

$$\int_0^{\infty} R(t) dt = \text{MRT} = \frac{0.7 \times 0.05 + 0.3 \times 0.4}{0.693} \text{ days} = 0.22 \text{ days}$$

which is much below the value of 40 days. This suggests the existence of longer-term exponentials. If one exponential is added in the amount of 2% with a 1400-day half-life, then

$$R(t) = 0.7 e^{-0.693t/0.05} + 0.28 e^{-0.693t/0.4} + 0.02 e^{-0.693t/1400}$$

and then

$$\text{MRT} \cong 43 \text{ days.}$$

Although this MRT concept implies longer-term exponentials, it does not tell how many to add, and only experiments will indicate that.

A Model for ^{14}C -Labelled Glycine

While the above is for $^{14}\text{CO}_2$ (or $\text{NaH}^{14}\text{CO}_3$), a model is given in Publication 10 for ^{14}C -labelled glycine (an amino acid) injected into blood of man. There it is recommended that

$$R(t) = 0.2 e^{-0.693t/0.12} + 0.2 e^{-0.693t/0.9} + 0.3 e^{-0.693t/6} + 0.3 e^{-0.693t/35}.$$

From this function, the MRT can be seen to be ~ 15 days, lower by a factor of 3 than the value above of 40 days. Thus it would be recommended that a component with a coefficient of 0.02 and a T_b of ~ 1200 days be included to increase the MRT up to ~ 40 days.

Conclusions and Recommendations

From the above it is concluded that in mice, 1% of the inhaled $^{14}\text{CO}_2$ enters into long-term retention in the body. The mean residence time of C in the diet of mouse is 6 days, while for man, it is 40 days. It is recommended that the 1% uptake to blood be used rather than 75% (ICRP Publication 2 value) and the residence time of 40 days be used for metabolites (compounds which are involved in the buildup and breakdown of cells and tissues) ingested into the body of man. For a single injection of $\text{NaH}^{14}\text{CO}_3$ into blood, some studies indicate a lower residence time in mice (~ 1 day) than for food labelled with ^{14}C (6 days). When the above models are used in dose estimation for CO_2 , it is found that ICRP Publication 2⁹ overestimates the dose to the total body by a factor of ~ 30 , while the ICRP Publication 10 model underestimates it by a factor indicated to be at least 16 and which might be as high as ~ 100 . Probably the single exponential is an oversimplification and should be replaced by several exponentials of fairly long half-times, but to determine these, further data on man, or on several species of animals, would be desirable.

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METABOLIC BALANCES OF ^{210}Pb AND ^{210}Po IN UNEXPOSED MEN*

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Abstract

The metabolic balances of ^{210}Pb and ^{210}Po were measured in each of 12 men maintained on a metabolic ward. These nuclides were determined in urine and feces from each subject collected for one month or more from each. Representative diets, drinking water and atmospheric levels in the ward were also sampled. The mean levels (\pm S.E.) were for ^{210}Pb and ^{210}Po , respectively, in diets over a period of five months (1.25 ± 0.04) and (1.63 ± 0.05) pCi/day, in urine (0.275 ± 0.026) and (0.269 ± 0.033) pCi/day, and in feces (1.333 ± 0.062) and (1.89 ± 0.10) pCi/day.

The mean overall balances (the difference between diet and excreta) of (-0.235 ± 0.075) pCi ^{210}Pb /day and (-0.337 ± 0.100) pCi ^{210}Po /day show that over the collection period, larger amounts were excreted than taken in the diet. Atmospheric intake accounts for an additional ^{210}Pb intake of 0.07 pCi/day, but it contributes a ^{210}Po intake of only about 0.02 pCi/day. The contribution of cigarette smoke and dietary levels of calcium to the balances is discussed.

Introduction

The metabolic properties of ^{210}Pb and its decay product, ^{210}Po , in people exposed only to normal environmental levels of these nuclides are important because they contribute a large fraction of the dose from internally deposited nuclides.^{1,2} The ^{210}Pb produces essentially no dose, but with its 22-year physical half life, it can accumulate in the body. On the other hand, the ^{210}Po with its 5.3-Mev alpha particle contributes 90 to 95% of the dose from this series (^{210}Pb - ^{210}Bi - ^{210}Po), but its 138-day half life allows only a limited accumulation from sources other than the ^{210}Pb in the body.

Metabolic balance studies may indicate other routes of intake and excretion. Thus, if the balance is negative (based on diet and excreta), one may look for other sources of intake which may be difficult to measure directly, such as atmospheric aerosols and cigarette smoke.

Some dietary and excretion data have been reported in unexposed populations, but the studies were generally of a limited nature. Glöbel et al.³ studied the metabolic balances of these nuclides on one person over a one-month period. Magno et al.⁴ measured the ^{210}Pb in "typical" diets from several cities, and Morse and Welford⁵ determined these levels in a composite New York diet. Some urinary excretion rates have been measured in people with potentially high exposures.⁶⁻⁹

* Work performed under the auspices of the U. S. Atomic Energy Commission.

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We report here on a study of the metabolic balances of ^{210}Pb and ^{210}Po in men maintained on a metabolic ward for four weeks or more.

Experimental Methods and Materials

Twelve fully ambulatory patients in good physical condition were studied under strictly controlled conditions on the Metabolic Research Ward.^{10,11} Their ages were from 42 to 64 years with an average age (\pm S.E.) of (52.0 ± 1.7) years, and their weights were from 62.6 to 87.0 kg with a mean of (76.0 ± 2.0) kg. Each consumed a constant diet daily which weighed about 2 kg and contained 2200 calories, about 200 mg of calcium and 800 mg of phosphorus. Each subject chose his own daily consumption of water intake (average 2.8 liters/day) which was then maintained constant over the period of study. In some of the studies the calcium intake was increased to 800 or 2400 mg/day by addition of either milk, calcium lactate or calcium gluconate tablets to the above low calcium diet. Most of the patients had been maintained on the ward on the same diet for many months prior to these studies.

Complete urine and stool collections obtained for the study were usually pooled on a six-day basis, although some were combined into four-, eight-, or ten-day pools. Aliquots of representative diets were taken for analysis once a week over a period of 27 weeks. Two sets of drinking water samples of 1 liter each were measured. The atmospheric concentration of these nuclides in the ward was estimated from collections made at several periods during the study.

The ^{210}Pb and ^{210}Po in the samples were determined by the previously described procedure¹ of wet ashing and plating the ^{210}Po onto a silver disk which was then alpha counted. Calcium was determined by atomic absorption analysis on a Perkin Elmer Model 303 Atomic Absorption Spectrophotometer.¹² The accuracy of the analytical results was generally $\pm 5\%$ or better, except that those of the ^{210}Po were about 10%.

Results and Discussion

The metabolic pathways of ^{210}Pb and ^{210}Po are illustrated in Fig. 1. Intake is from food, water, air and cigarette smoke, while output is through excretion and other pathways, such as loss of hair and desquamation of skin.¹³ Both nuclides may enter the circulation from the gut and be excreted into the gut. One large contribution (and probably the major one) to the ^{210}Po pool is that from the decay of ^{210}Pb present in the body.

The largest source of intake is diet. The levels from the representative 6-day samples are shown in Fig. 2; the overall mean values (\pm S.E.) are 1.248 ± 0.029 and 1.630 ± 0.048 pCi/day for ^{210}Pb and ^{210}Po , respectively. The coefficient of correlation between the ^{210}Pb and ^{210}Po data is essentially zero ($R = -0.20$, $P \gg 0.05$). However, the mean ratio of ^{210}Po activity to that of ^{210}Pb , 1.33 ± 0.06 is more important, since correlations may be neither evident nor important in data which vary little about their means.

These data show some systematic variations of the ^{210}Pb , such as minima at the 3rd and 8th 6-day periods and maxima at the 5th and 17th periods. The variations are about 20% of the mean. The ^{210}Po shows similar structure but it lags 3 to 5 periods behind the ^{210}Pb . Some compensation for these variations is made in the balances by using dietary levels extant at the collection times. However, the means of the 27 measured diets and of the diets of the individuals were essentially identical.

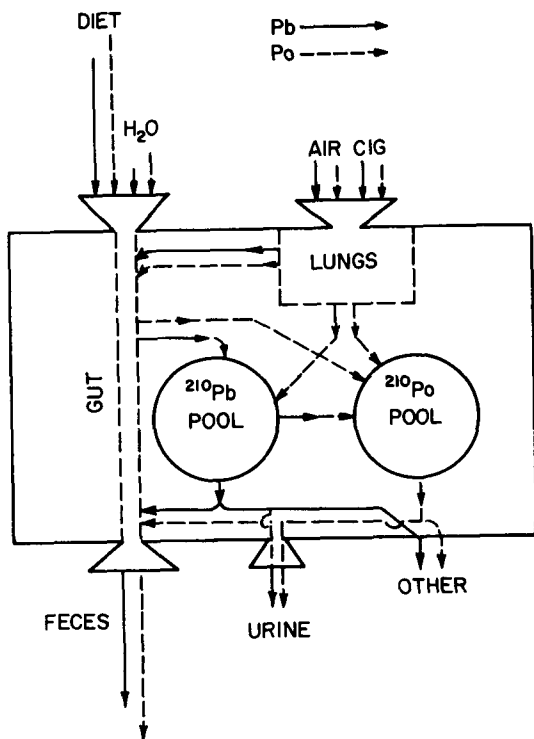


Figure 1. A model of the metabolic patterns of ^{210}Pb and ^{210}Po .

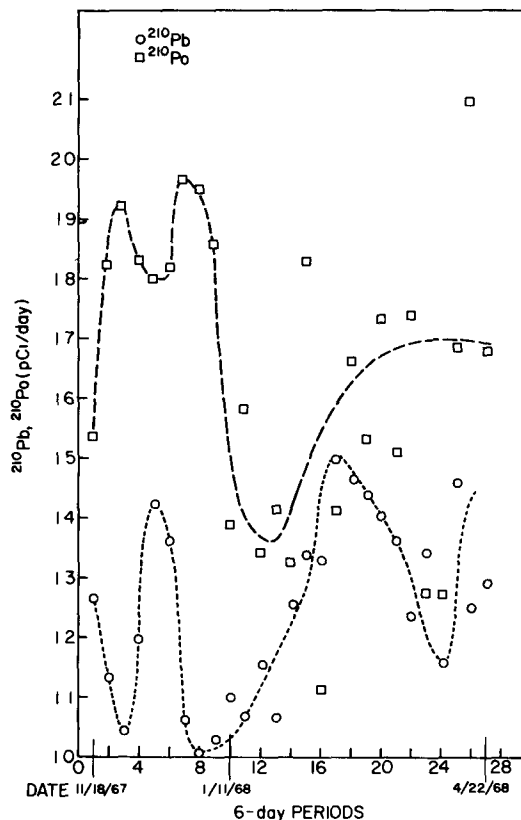


Figure 2. Contents of ^{210}Pb and ^{210}Po in the diets of the subjects measured in representative 6-day samples.

The mean ^{210}Pb content of these diets is lower than that found by Morse and Welford⁵ in New York City of 1.40 ± 0.08 pCi/2 kg and than the results of Magno et al.⁴ for four U. S. cities of about 1.7 pCi/2 kg. The differences may be due to the lack of some constituents high in ^{210}Pb present in the standard diet, but not present here, such as fresh vegetables and milk. The seasonal variations observed in these data suggest that these values may be at least as representative of a U. S. diet as the single and composite diets reported elsewhere. Drinking water contributes in addition about 0.11 and 0.10 pCi/day to the ^{210}Pb and ^{210}Po intakes, respectively.

The contributions of inhalation to intake are more difficult to assess because they were not measured directly, and they will be estimated only for an average subject. This is estimated from the breathing rate and the atmospheric concentration. Because of the more sedentary condition of these men relative to working men, we assumed the average man's daily breathing rate to be $15 \text{ m}^3/\text{day}$ rather than the $20 \text{ m}^3/\text{day}$ of the ICRP "Standard Man".¹⁴ The atmospheric concentrations of the nuclides are shown in Fig. 3. The mean specific activities were (10.9 ± 0.9) and (1.76 ± 0.16) pCi/1000 SCM (Standard Cubic Meter) for ^{210}Pb and ^{210}Po , respectively. The total intake from the atmosphere is then 0.16 pCi ^{210}Pb and 0.026 pCi ^{210}Po per day.

The mean ^{210}Pb excretion rates for each subject, along with his respective dietary and water intake and the respective balances are shown as a function of increasing balance in Fig. 4. Because of the large uncertainties and because they were not measured directly, the contribution of inhalation are not included in these figures. The ^{210}Pb values indicate that the balances depend to a large

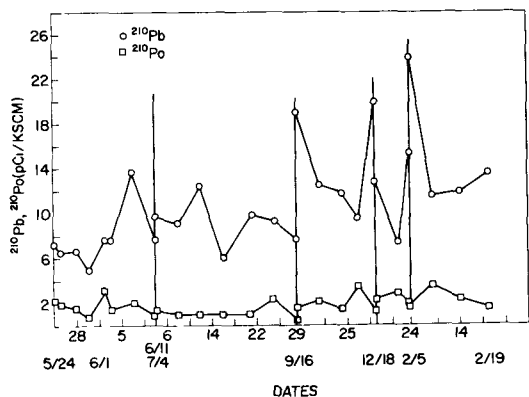


Figure 3. Concentrations of ^{210}Pb and ^{210}Po in atmospheric aerosols sampled in various periods from 5/24/68 to 2/19/69. Each date is the mean date of the sampling period.

The mean (\pm S.E.) excretion rates and ranges are shown in Table 1.

The contribution of cigarette smoke to the nuclide intake is shown in Fig. 5 by balance vs. number of cigarettes smoked per week for the seven subjects on whom we measured cigarette consumption. For ^{210}Pb the linear regression of balance (B) vs. cigarettes/week (C) is

$$B = (-0.10 \pm 0.17) - (0.0015 \pm 0.00043)C$$

and the correlation coefficient is -0.59 ($P \approx 0.10$). While neither the intercept nor the slope is statistically significant, they do give an indication of the values. The intercept is equivalent to the atmospheric intake of 0.1 pCi/day and the slope is about 0.21 pCi/day, in a person smoking one pack per day. (These seven people smoked an average of 22 cigarettes per day.) For ^{210}Po the levels of significance are even poorer, although, if the intercept is forced through zero, the slope of -0.0015 pCi/day is significant ($P \approx 0.05$). Previous studies on the nuclide content of cigarette smoke^{15,16} lead to an estimated total intake of 0.30 pCi ^{210}Pb and 0.72 pCi ^{210}Po per day.

The results in Fig. 5 are compatible with the directly measured values of intake from smoke, if one assumes that retention of inhaled nuclides in the respiratory tract is 50% as indicated by various data in the literature.^{14,17-19} Daily deposition would then be 0.23 pCi (0.08 from air and 0.15 from cigarettes) and that from ^{210}Po would be 0.37 pCi (0.01 from air and 0.36 from cigarettes) for persons smoking one pack/day.

The balance based on intake from diet and water only, is negative, but it becomes essentially zero, if we add to the intake the contribution from inhalation, as shown in Table 2. In this table the dietary intakes are the average of our representative 6-day samples, the inhaled values are those estimated for inhalation alone, and the excretion values are the overall averages. The different means, taken from the overall averages in Table 2 (diet + water + inhala-

extent on the fecal values, which in most cases are about equal to the dietary intake. The more negative balances show more fecal output than dietary input and the more positive ones less fecal output. There also appears to be a general downward trend of both fecal and urinary levels as the balance increases. The calcium balances and high calcium diets (noted by the numbers in mg/day intake next to the calcium balance points) do not appear to affect the ^{210}Pb balances.

The ^{210}Po results were similar to those of ^{210}Pb but their values scatter more and show fewer associations, such as little correlation between diet and feces. The order of the subjects by balance is different than that for ^{210}Pb .

Table 1. Mean excretion rates and balances of ^{210}Pb and ^{210}Po .

	^{210}Pb (pCi/day)	^{210}Po (pCi/day)
Urine	0.275 ± 0.026 (0.11 to 0.41)	0.269 ± 0.033 (0.079 to 0.52)
Feces	1.333 ± 0.062 (0.99 to 1.83)	1.89 ± 0.10 (1.54 to 2.46)
Balance*	-0.235 ± 0.075 (-0.73 to +0.25)	-0.337 ± 0.100 (-0.95 to +0.25)

* diet + water - excreta

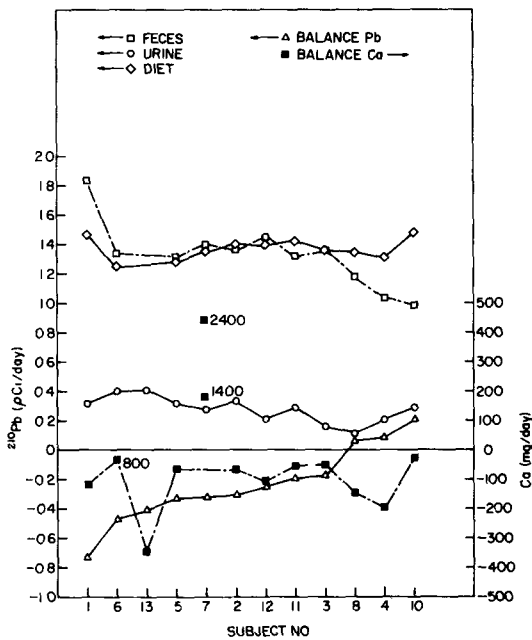


Figure 4. The mean daily levels of ^{210}Pb for each subject in diet, feces and urine and the mean daily ^{210}Pb and calcium balances. The subjects are listed in order of ^{210}Pb balance. The calcium intakes greater than the standard 200-mg per day level are given in mg/day for Subjects 6 and 7.

of 600 pCi (the ^{210}Pb body content of an average man²⁰) and with a mean balance of ^{210}Po of less than 0.1 pCi/day, the implied half life is very long and comparable to the 7-year values observed in people high in ^{226}Ra .²¹ The ^{210}Po activities are then essentially in radioactive equilibrium with the ^{210}Pb (> 90% of the ^{210}Pb activity).

Thus, the low ratios of ^{210}Po to ^{210}Pb in diet and excreta confirm the previous estimates²⁰ that only a small fraction of the ^{210}Po in the body is supported by diet when the ratio of the activity of ^{210}Po to that of ^{210}Pb is about unity in vivo. Even though the residence time of ^{210}Po is much longer than the 25 days assumed earlier,²⁰ the limiting factor would be the 138-day physical half life. With no excess of excretion over intake, the accumulation from the diet would be only about 24 pCi, compared to 500 to 600 pCi in the body (if the ^{210}Po is nearly in radioactive equilibrium with the ^{210}Pb).

tion - excreta), is -0.02 pCi/day for ^{210}Pb and -0.06 pCi/day for ^{210}Po which is essentially identical to that of individual balance taken from the mean of the balances for each individual based on his diet, water and excreta (-0.275 and -0.269 pCi of ^{210}Pb and ^{210}Po , respectively) to which has been added the estimated mean daily inhalation intake of 0.23 pCi of ^{210}Pb and 0.37 pCi of ^{210}Po . Thus, the mean balances approach zero to within 3% of the intake values.

These results are lower than those of Glöbel et al.³ in Germany. Their diet (and fecal excretion) had more than twice the nuclide content of ours, 4.65 pCi of each, and the $^{210}\text{Po}/^{210}\text{Pb}$ ratio was about unity compared to our ratio of 1.33. The urinary excretion of ^{210}Po of 0.3 pCi/day was comparable to ours, but the ^{210}Pb excretion was twice this value, in line with the higher dietary levels. The values of Glöbel et al. are probably not typical of the U.S., either because they studied a particular person or because their foods were different.

The balance of the ^{210}Po is important to estimates of radiation dose, while the amount in the body under normal intake depends mainly on the ^{210}Pb content of the body rather than on the intake. For an internal source of ^{210}Pb

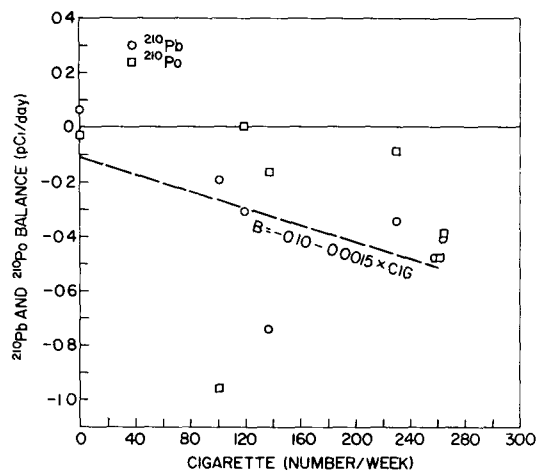


Figure 5. ^{210}Pb and ^{210}Po balances plotted against the weekly cigarette usage for each of the seven subjects for whom this quantity was determined.

Table 2. Summary of components of metabolic balances.

I. Intake		
A. Ingestion		
Diet	1.25	1.63
Water	<u>0.11</u>	<u>0.10</u>
	1.36	1.73
B. Inhalation (50% retention)		
Air	0.07	0.02
Cigarette Smoke*	<u>0.15</u>	<u>0.36</u>
	0.24	0.37
C. Total	1.60	2.10
II. Output		
Excretion		
Feces	1.33	1.89
Urine	<u>0.28</u>	<u>0.27</u>
	1.61	2.16
III. Balances		
A. This Table	-0.02	-0.06
B. Individual (see text)	0.00	+0.03

The values of $^{210}\text{Po}/^{210}\text{Pb}$ ratios of 1.07 in urine and 1.47 in feces make it possible to estimate the amounts of one nuclide given a measurement of the other. However, because of the large variability of these ratios, this estimate is reliable only to within a factor of 2 or so.

In summary, the ^{210}Pb - ^{210}Po content of the diets appear to be representative of those in the U.S. when compared to the data of others. They exhibit seasonal variations, a phenomenon not noted in other measurements.³⁻⁵ Both the diets and fecal excretions had a 30 to 40% excess of ^{210}Po over ^{210}Pb , while urinary excretion exhibited equal amounts of each. When account is taken of the contributions of inhalation to intake, this group is in material balance with respect to both nuclides. This result tends to justify the assumption on inhalation used in arriving at the intake values.

* 1 pack/day

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LONG-TERM METABOLISM OF ^{90}Sr IN RHESUS MONKEYS (*Macaca mulatta*)*
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ABSTRACT

The plasma content (P_t) and daily excretion rate (E_t) were measured, and the whole-body retention (R_t), was calculated for 7 to 15 yr after a single injection of ^{90}Sr in young (2.3-3 yr), adolescent (3.1-5 yr), and adult (> 5 yr) rhesus monkeys. Half-times of 6-term exponential equations of P_t and E_t —0.4-0.5 d, 1-2 d, 3.5-7 d, 40-80 d, 300-650 d, and 2400-3600 d—are similar within each age group and also among the 3 age groups. The half-times of the first 5 terms of equations of R_t are similar to those of P_t and E_t , but those of the 6th terms are longer, 4200-5600 d, presumably representing re-deposition of ^{90}Sr recirculated from remodeled bone. The coefficients of the metabolic equations demonstrated greater early ^{90}Sr excretion by the older monkeys and greater late retention of ^{90}Sr by the growing monkeys. When age at injection was considered, ^{90}Sr retention in monkeys agreed with retention of ^{85}Sr or ^{90}Sr in other simians, beagles, and human adults.

INTRODUCTION

Even before the first thermonuclear weapons were exploded in the Marshall Islands (March 1954), it was recognized that ^{90}Sr would be a hazardous constituent of the radioactive fallout, but there was little information on ^{90}Sr metabolism in species other than rodents.¹⁻³ In 1954, urged by Dr. Willard F. Libby, then a member of the U.S. Atomic Energy Commission, and the late Dr. Joseph G. Hamilton, director of the Crocker Laboratory, we fed or injected animals from the laboratory's small colony of rhesus monkeys with low doses of ^{90}Sr .⁴⁻⁶

As new techniques were developed,^{7,8} metabolic information began to be available from human beings directly, and interest in study of other species declined. Controlled long-term study of the alkaline earths in human beings is not possible, because there are no long-lived γ -emitting isotopes of these elements. Furthermore, the long-term data from the human ^{226}Ra cases are equivocal, because of late radiation effects and uncertainty in their ^{226}Ra exposure levels. Therefore, we maintained and gradually added to the ^{90}Sr -injected monkey colony to obtain from individual monkeys over a significant part of their 30-yr life span metabolic data that could be used to test long-term models of alkaline earth metabolism.⁹ This report summarizes our study of ^{90}Sr in monkeys and compares their long-term retention of ^{90}Sr with that in other species.

METHODS AND MATERIALS

Rhesus monkeys caught in the wild were purchased from animal dealers. Ages were determined from dentition¹⁰ and skeletal roentgenograms.¹¹ The animals have been assigned to 3 age groups: young, 2.3-3 yr, most epiphyses open, skeleton and body weights increasing rapidly, sexually immature; adolescent, 3.1-5 yr, some epiphyses open, skeleton and body weights increasing slowly, sexually mature; adult, > 5 yr, epiphyses closed and permanent teeth erupted, skeleton and body weights stable. Colony management was described earlier.⁴ A high-Ca diet (0.03-0.12 g/d/kg body weight) was fed to suppress reutilization of ^{90}Sr .

Strontium-90 diluted in 3% Na citrate, was given in a single i.v., i.m., or i.p. injection. The only differences in ^{90}Sr behavior related to route of injection were in the early plasma concentrations. Experimental protocol is shown in Table 1.

The monkeys are housed in individual noncommunicating stock cages from which, with careful cleaning, we obtain complete excreta collections. Excreta were collected frequently during the first year and thereafter in two 7-d pools every 8 wk. In some cases urine and feces were collected separately for the first few weeks. Blood samples were drawn frequently in the first weeks and at least twice yearly

Table 1. Injection and retention data for ^{90}Sr -injected monkeys

Injection data							
Monkey ^a no., sex	Age (yr)	Weight (kg)	^{90}Sr (μCi)	Injn. date	Days to death	^{90}Sr in body (%)	Cause of death or reason for euthanasia
<u>Young monkeys (2.1 to 3 yr)</u>							
37F	2.6	4.4	12.8	2/15/60	1	45.4	Test animal
36F	2.3	3.1	12.8	2/15/60	4	56.2	Test animal
52F	2.3	2.8	130.5	11/13/61	21	48.5	Test animal
53F	2.7	2.6	130.5	11/13/61	66	69.3	Test animal
29F	3.0	3.3	41.6	9/10/58	280	34.8	Positive TB test
51F	2.7	2.8	26.1	11/13/61	441	26.6	Test animal
20F	2.7	3.6	49.8	1/15/57	707	13.8	Amoebic dysentery
50F	2.7	3.1	26.1	11/13/61	1211	24.8	Test animal
34F	2.9	3.6	56.2	2/21/58	1921	15.4	Accident
35F(1) ^b	2.4	4.0	56.2	2/21/58	2037	~10	Reinjected by mistake ^b
28F	2.6	2.9	41.6	9/10/58	2087	15.5	Accident
<u>Adolescent monkeys (3.1 to 5 yr)</u>							
10F	3.3	3.4	56.3	11/10/55	94	45.1	Test animal
64F	4.3	4.2	33.1	3/27/67	427	16.9	Test animal
33F	3.2	3.8	56.2	2/21/58	2247	7.3	Leptospirosis
9M	4.3	9.1	38.0	4/21/54	2520	18.0	Cyst on neck
31F	4.1	4.1	61.1	10/27/57	2639	5.6	Intussusception
27M	3.6	3.2	41.6	9/10/58	3159	11.2	Cage paralysis
65F	4.4	4.5	33.1	3/27/67	---	5.1 ^c	Alive, 2242 days p.i.
63F	4.1	5.4	46.9	1/ 9/67	---	5.8 ^c	Alive, 2319 days p.i.
32F	4.2	4.5	61.1	10/27/59	---	7.1 ^c	Alive, 5360 days p.i.
21F	3.2	4.2	49.8	1/15/57	---	5.7 ^c	Alive, 6010 days p.i.
<u>Adult monkeys (>5 yr)</u>							
G8M	6.8	8.9	4.4	9/ 9/69	2	37.0	Test animal
G11M	~ 7	7.0	5.0	6/24/70	16	50.7	Test animal
98F	9.3	5.6	3.5	10/28/69	35	17.9	Test animal
40F	8.8	8.5	70.4	1/ 9/67	98	7.0	Pneumonia
35F(2) ^b	7.9	5.2	50.1	9/23/63	147	~ 8	Test animal ^b
7M	> 6.5	4.2	37.5	3/16/54	181	21.4	Cage paralysis
23M	> 7	5.0	49.8	1/15/57	3183	7.8	Accident
38F	5.4	3.2	50.1	9/23/63	3411	2.4	Uterine tumor
8F	~ 5	5.5	39.9	4/ 1/54	3507	2.9	Leptospirosis
39F	5.5	3.2	50.1	9/23/63	---	1.3 ^c	Alive, 3599 days p.i.
61M	6.7	6.8	117.6	2/25/63	---	3.8 ^c	Alive, 3809 days p.i.
62M	12	9.4	168.3	2/25/63	---	1.6 ^c	Alive, 3809 days p.i.

a In Ref. 4 monkeys 7M, 8F, 9M, and 10F were identified with pet names as follows: 7M, Tony; 8F, Rosy; 9M, Stupe; 10F, Pat.

b Monkey 35F was reinjected (by mistake) with ^{90}Sr 2037 d after her original injection. When she was killed 147 d after the second injection, the measured ^{90}Sr in excreta, bones, and tissues was compatible with the approximate ^{90}Sr body contents shown.

c Body count 4 to 7 mo before the closing date of this report, Aug. 1, 1973.

after the first year. After 1968 whole-body bremsstrahlung were measured periodically. Tissues and bones of dead monkeys were analyzed for ^{90}Sr .

Samples were dry-ashed, dissolved in acid, and analyzed for ^{90}Sr by one of the following methods: (a) samples of $> 10^4$ dis/min—small aliquots were dried on glass planchets; (b) samples of 10^2 - 10^4 dis/min—large aliquots were concentrated by coprecipitation with CaC_2O_4 ; ^{1,2} and (c) samples of $< 10^2$ dis/min—large aliquots or entire samples were analyzed by a commercial laboratory using a ^{90}Y -extraction process. ^{1,2}

RESULTS AND DISCUSSION

We have chosen to describe plasma activity, P_t , daily excretion rate, E_t , cumulative excretion, E_t^* , and whole-body retention, R_t , as sums of first-order exponentials, of the form

$$A_t = \sum_{i=1}^n A_i \exp(-0.693t/Ta_i). \quad (1)$$

Half-times, Ta_i , and time after injection, t , are in days. The data for P_t and E_t were plotted semi-logarithmically for each monkey. Curves were fitted by the method of least squares in 6 straight-line segments over the intervals shown in Table 2, and the coefficients of their exponential equations, P_i (%) and E_i (%/d), and the half-times, Tp_i and Te_i , were determined graphically. Cumulative excretion was calculated for each monkey by integration of its equation of E_t at the postinjection times shown by the data points in Fig. 1. Whole-body retention was calculated from the relationship

$$R_t (\%) = (100\% - E_t^*) = (100\% - \int_0^t E_t dt). \quad (2)$$

Tabulations were made of the values of P_t and E_t read from the individual monkey's curves and of their calculated values of R_t at the times shown by the data points in Figs. 1-3. Mean values of P_t , E_t , and R_t (the data points shown in the figures) were calculated for the 3 age groups. The individual values usually varied over a 2- to 4-fold range. The mean curves shown in the figures were also analyzed into 6 exponential terms, and their coefficients and half-times are collected in Tables 2-4.

Total excretory clearances of the 3 monkey groups (see curves of P_t and E_t in Figs. 2,3) ranged over 4.5-9 plasma volumes/d and tended to be lower in the younger monkeys. The best current estimate of excretory clearance of ^{90}Sr in adult man⁹ is 4.3 plasma volumes/d (13.6 liters/d for a 70-kg man with an average daily intake of 1 g Ca, 0.014 g Ca/kg body weight). The greater clearance in the monkeys is probably related to their 2- to 10-fold greater Ca intake.

The half-times of the comparable terms of the equations of P_t , E_t , and R_t (Tables 2-4) were similar within the 3 age groups, which is to be expected, because retention, plasma activity, and excretion rate are all dependent on bone metabolism. However, in the equations of R_t for the adolescent and adult monkeys, Tr_6 was 4200 to 5600 d, while in their equations of P_t and E_t , Tp_6 and Te_6 were 2400 to 3640 d. Although the data are insufficient to demonstrate a significant difference, such a finding is compatible with redeposition in the skeleton of some ^{90}Sr recirculated during bone resorption.

The coefficients, P_{1-3} , E_{1-3} , and R_{1-3} , representing ^{90}Sr uptake in bone, are smaller in the metabolic equations of the young monkeys than in those of the adults. The coefficients, P_{4-6} , E_{4-6} , and R_{4-6} , representing turnover of ^{90}Sr -labeled bone, are greater in the equations of the young monkeys than in those of the adults. The relative magnitudes of the coefficients of the equations of P_t , E_t , and R_t in the 3 age groups demonstrate again the dependence of ^{90}Sr uptake and retention on bone-growth status. ¹⁵⁻¹⁸

The half-times of 6-term exponential equations of ^{90}Sr retention in young-adult beagles¹⁴ and a 57-yr old woman⁷ were similar to those in the retention equations of the monkeys, when all retention curves were analyzed over the same postinjection intervals (Tables 2,4). The finding of common half-times in exponential equations of ^{90}Sr retention in monkeys with varying bone-growth status, young-adult beagles, and a middle-aged woman supports the view that the dynamics of skeletal metabolism are qualitatively similar among mammals. ¹⁵

If age at injection is considered, ^{90}Sr retention in our monkeys agrees with the retention of ^{85}Sr or ^{90}Sr in young and adolescent rhesus monkeys studied elsewhere,¹⁹ young and adolescent monkeys of 3 species of Cercopithecus (West African monkeys of small body size),^{20,21} 1.4- to 9-yr-old beagles,^{14,16,18} and in adult human subjects during the first 100 d after injection. For example, 15 d after injection, retention of ^{85}Sr or ^{90}Sr was $27 \pm 9\%$ and $34.3 \pm 10.2\%$, respectively, in 8 adult rhesus monkeys and 9 human beings 27-61 years old.^{7, 22-24} By 350 d, retention of ^{85}Sr in 9 human adults

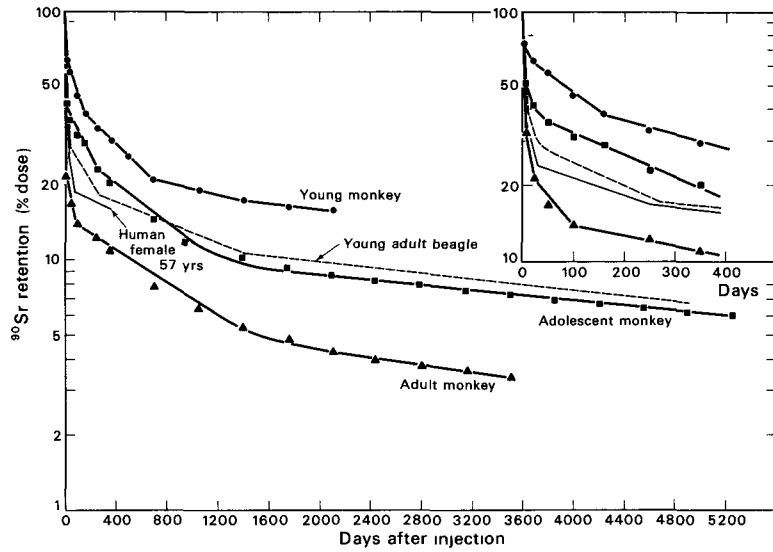


Fig. 1. Retention of a single injection of ^{90}Sr in rhesus monkeys, young-adult beagles,¹⁴ and a 57-yr-old woman.⁷ Parameters of the exponential equations appear in Table 4.

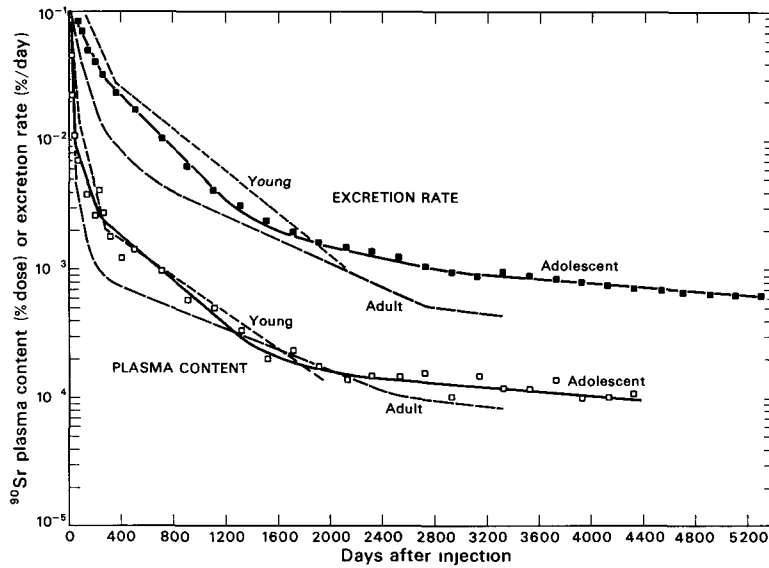


Fig. 2. Plasma content and excretion rate (urine and feces combined) after a single injection of ^{90}Sr in rhesus monkeys. Parameters of the exponential equations appear in Tables 2,3.

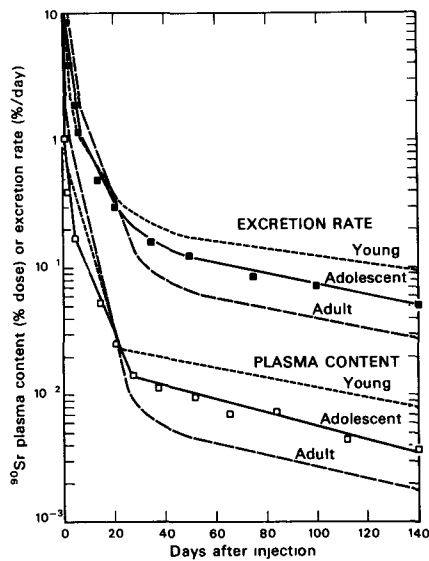


Fig. 3. Expanded time scale of Fig 2.

Table 2. Plasma content: Coefficients (P_i) and half-times (T_{p_i}) of the exponential equations that describe the ^{90}Sr content of the plasma^a after a single injection in rhesus monkeys.

Exponential term	Fitting interval t_1 (d)	t_2 (d)	Young		Adolescent		Adult	
			$t_f = 1900$ d ^b P_i (%)	T_{p_i} (d)	$t_f = 4600$ d ^b P_i (%)	T_{p_i} (d)	$t_f = 3450$ d ^b P_i (%)	T_{p_i} (d)
1 ^c	0	1	—	—	—	—	—	—
2	1	5	1.0	.65	1.2	1.1	1.7	.53
3	5	25	.88	4.4	.28	5.8	1.8	3.3
4	25	250	.026	60	.019	51	.007	40
5	250	1500	.0032	420	.0038	300	.0008	640
6	> 1500	—	—	—	.00024	3500	.0002	2400

a Total plasma ^{90}Sr was calculated from ^{90}Sr in weighed samples of whole blood and the measured hematocrit, assuming a blood density of 1.056g/ml and a plasma volume of 36.4 ml/kg body weight in rhesus monkeys.¹³

b Time of last available measurement.

c The earliest components of the blood curves have not yet been analyzed.

Table 3. Excretion: Coefficients (E_i) and half-times (T_{e_i}) of the exponential equation of the rate of ^{90}Sr excretion, and intercepts (E_{i^*}) of the equations of cumulative excretion of ^{90}Sr after a single injection in rhesus monkeys.

Exponential term	Young			Adolescent			Adult		
	$t_f = 1900$ d E_i (%/d)	T_{e_i} (d)	E_{i^*} (%)	$t_f = 5250$ d E_i (%/d)	T_{e_i} (d)	E_{i^*} (%)	$t_f = 3300$ d E_i (%/d)	T_{e_i} (d)	E_{i^*} (%)
1 ^{a,b}	16	.40	9.2	41	.46	27	31	.42	19
2	5.9	1.7	14	7.9	1.3	15	21	1.2	36
3	1.5	7.1	15	2.0	6.5	19	4.1	5.5	32
4	.19	80	22	.094	41	5.5	.091	88	12
5	.053	360	28	.058	300	25	.0086	570	7.2
6	—	—	—	.0017	3640	8.9	.0011	2400	3.8

a The parameters of the first terms of the equations of E_t were obtained from curves in which 0-1 d excretion was plotted at 0.5 d, and fecal lag was neglected. Such curves artificially depict constant renal excretion during the first 24 h, and gastrointestinal excretion as both constant and instantaneous.

b Same fitting intervals as in Table 2.

Table 4. Retention: Coefficients (R_i) and half-times (T_{r_i}) of the exponential equations of whole-body retention of ^{90}Sr after a single injection in rhesus monkeys. Parameters of ^{90}Sr retention curves of young-adult beagles and of a woman are shown for comparison.^a

Exponential term	Monkey						Beagle		Human subject	
	$t_f = 2100$ d		$t_f = 5250$ d		$t_f = 3450$ d		$t_f = 4900$ d		$t_f = 397$ d	
	R_i (%)	T_{r_i} (d)	R_i (%)	T_{r_i} (d)	R_i (%)	T_{r_i} (d)	R_i (%)	T_{r_i} (d)	R_i (%)	T_{r_i} (d)
1	11	.50	23	.38	28	.41	19	.6	22	.44
2	10	2.1	18	2.0	24	2.2	32	2.7	31	2.5
3	11	17	17	10	24	7.0	18	14	22	14
4	20	66	14	100	9.2	22	11	120	4.6	130
5	26	300	17	500	8.8	540	6.7	540	22	1000
6	21	5400	11	5600	6.0	4200	13	5300	—	—

a Data from LLOYD *et al.*¹⁴ for beagles injected with $< 30 \mu\text{Ci/kg}$ of ^{90}Sr , and data from COHN *et al.*⁷ for a 57-yr-old woman patient MOS were replotted, and the retention curves were fitted over the same intervals (Table 2) used to analyze the monkey curves.

(some ill but with apparently normal skeletons) was $15.9 \pm 4.7\%$ ^{7,23}—apparently greater than in 5 adult monkeys, $11 \pm 4.3\%$. However, by 350 d, decay and excretion of ⁸⁵Sr introduce large errors in whole-body γ -ray measurements, which, added to individual variability, render any difference insignificant. Thus, rhesus monkeys of varying ages seem to be a suitable model system in which to evaluate long-term metabolism of the alkaline earth elements.

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FOOTNOTES AND REFERENCES

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A STUDY OF THE TRANSLOCATION OF RADIOSTRONTIUM
FROM WOUNDS AND THERAPY BY LOCAL INSOLUBILIZATION

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Abstract

The translocation of ^{85}Sr was followed by a 2 h external counting of the wound and homologous bone and by measurement of the blood radioactivity. Two types of wounds were simulated on Maccacus monkeys : puncture wounds and lacerations.

The same experimental procedure was applied in order to test therapy through Sr insolubilization by rhodizonate (K), Mg SO_4 , alginate (Ca) and aluminium phosphate gel, on laceration wounds either 5 or 15 min after the contamination.

It was verified that local or IV administration of DTPA did not result in an increased absorption of Sr.

Introduction

Wounds can be classified into three classes : abrasion, laceration and puncture. The first will not result in any significant absorption after contamination ¹ ; the second one only can be treated by local insolubilization of the radionuclides. Most authors have studied wounds contaminated by untranslocable nuclides (Pu and Am), whereas our study was concerned with strontium, a translocable fission product with a high radiotoxicity. For safety purposes and easier measurement, ^{85}Sr was chosen.

Material and methods

Twenty eight Maccacus monkeys weighing 5 kg were used. Wounds were performed on the postero-external side of the thigh. Puncture wounds were simulated by IM injections ; lacerations were made by incision and ecrasement of the muscle 2 cm long and 2.5 cm wide. A catheter was introduced into the arteria femoralis of the opposite leg for blood samples. From 10 to 100 μCi of ^{85}Sr (0.1 cm^3) were deposited on the wound and the radioactivity of the wound and the homologous area on the other leg was measured by NaI (Tl) detectors with the same

counting efficiency. The radioactivity of the blood samples (1 cm³) was measured in a well-type crystal. The various treatments were applied 5 or 15 min after exposure.

Results

Effect of the type of wound on absorption.

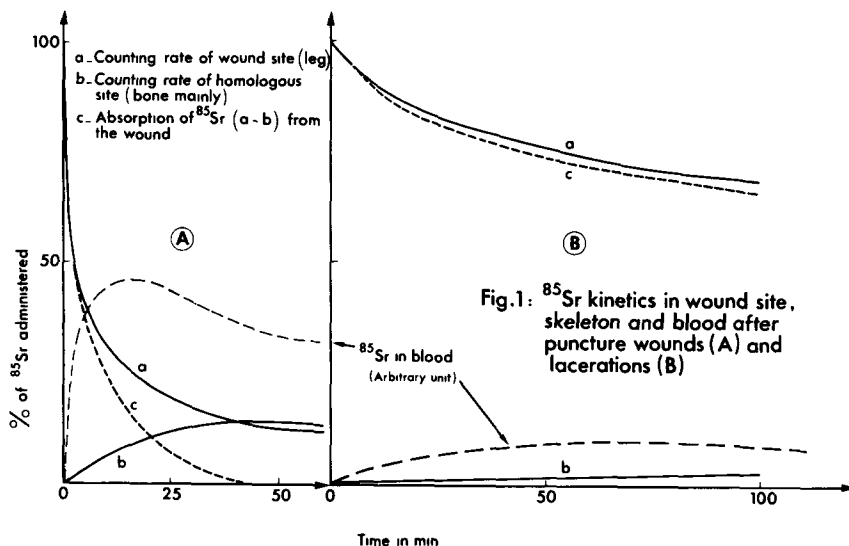


Fig. 1: ⁸⁵Sr kinetics in wound site, skeleton and blood after puncture wounds (A) and lacerations (B)

Following puncture, all the Sr was absorbed within 40 min (fig. 1 A, average on 2 animals). Blood concentration was highest within 20 min, then slowly decreased. Bone uptake seemed to stop after 50 min's time. Following laceration (fig. 1 B, average on 6 animals) these processes were slower and not so complete.

Sr insolubilization tests on lacerations. (17 animals).

As shown by fig. 2, therapy must be early : when administered 5 min after exposure, all the insolubilizing reagents resulted in a more or less significant

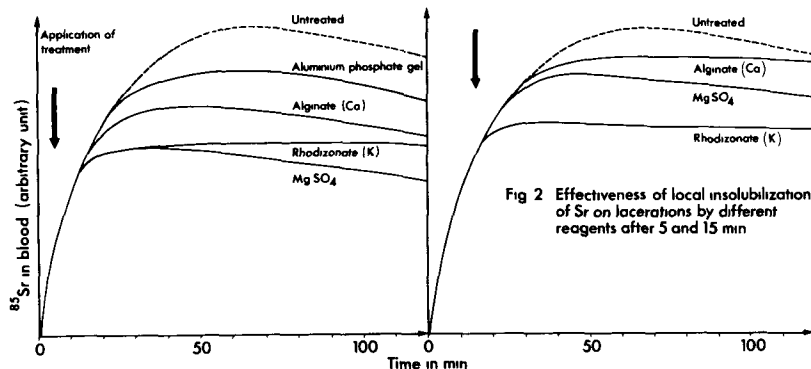
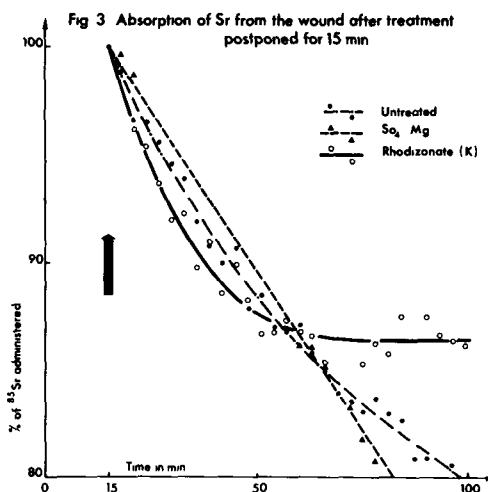


Fig 2 Effectiveness of local insolubilization of Sr on lacerations by different reagents after 5 and 15 min

decrease of absorption ; when administration occurred 15 min later, only K rhodizonate (powder) and Mg SO₄ (saturated solution) had a significant effect. Counting of the wound showed rhodizonate to be more efficient (fig. 3), which was verified by measurement of urinary excretion (table 1).



Effect of DTPA on Sr absorption. (3 animals).

In case of wounds contaminated by a mixture of fission products, DTPA (used as DTPA Ca Na₃) can be used in order to chelate the lanthanons. Table 1 shows that Sr absorption was not enhanced by DTPA whether insolubilized or not.

Treatment after 15 min	Absorption from the wound % of deposit	Urinary excretion during 24 h % of deposit
Untreated	22	7
Mg SO ₄	33	2,5
Rhodizonate	10	1,5
DTPA	23	5
Rhodizonate + DTPA	10	1
Rhodizonate + DTPA + DTPA (IV)	20	2,5

Table 1 Influence of treatment on absorption and urinary excretion of Sr deposited on laceration

Discussion and conclusion

Lacerations are very difficult to standardize and result in widely dispersed data (slide number 5).

As a conclusion, laceration wounds contaminated by Sr can be treated by insolubilization whereas puncture wounds cannot, because of access and delay. Rhodizonate seemed to be the best insolubilizing reagent. DTPA did not act on Sr absorption.

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INSTRUMENTS AND METHODS FOR DAY-TO-DAY MONITORING OF
TRITIUM AND CARBON-14 IN PRODUCTION

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Abstract

A complex of instrumentation and techniques for dosimetric monitoring at industrial enterprises using unshielded beta-emitters of T and ^{14}C is offered. Air-borne radionuclides are assayed by automatic monitors developed on the basis of gas counters. A portable device for detection of radioactive contamination of surfaces is worked out. Scintillation and gas counters have been used to measure ^{14}C and T content in personnel. The sensitivity of the methods is sufficient for monitoring of all categories of exposure.

Introduction

The problems of protection from tritium and ^{14}C radiation arise in the manufacture and application of tracer compounds, luminophores, tritium-titanium targets as well as in spheres involving the use of the energy of nuclear fission and fusion. Certain difficulties, as far as the dosimetry of these radionuclides is concerned, are due both to their specific radiation characteristics and insufficient knowledge of the radiotoxicity of their compounds^{1,2}.

This paper does not deal with the biophysical aspects of the indirect dosimetry of T and ^{14}C ; it describes instrumentation and methods for detecting low-energy beta-emitters in the human body and environment.

Monitoring of Radionuclides in Air

Gas filling counters are employed as detecting devices in all the monitors developed. The instruments also incorporate devices for passive and active shielding of the counters from external radiation, systems for intake, processing and regulation of the flows of assayed air and filling gas, electronic registering systems, control and signalling assemblies as well as high- and low-voltage power supplies for the counters and transistorized circuits, respectively. The instruments are designed as portable double-tier columns with detecting device at the bot-

tom and gas systems and electronic blocks at the top.

A mixture of aerosol-free air (0.1 l/min) and methane (0.5 - 1 l/min) is fed to the detectors of instruments A and B. The air piping and proportional counter bodies are made of Teflon. The operating volume of Counter A (Fig.1a) of 0.15 l is limited by a cylindrical cathode consisting of 22 metal wires. The de-

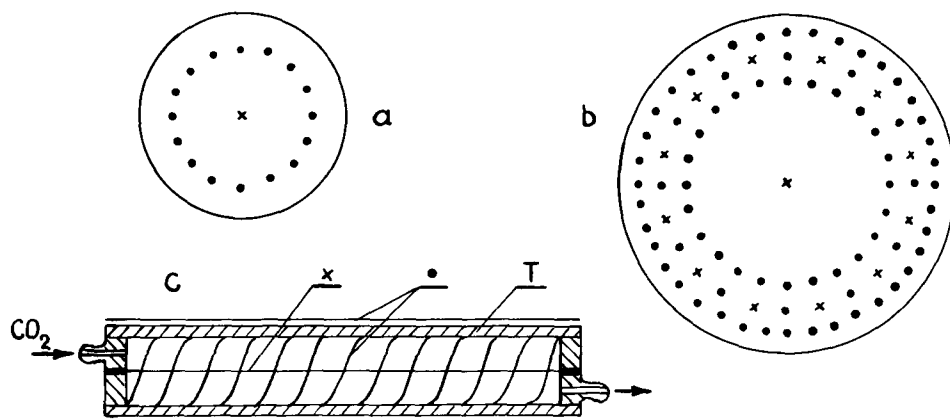


Fig.1. Schematic constructions of gas-flow counters.
x - anodes; . - cathodes; t - quartz tube.

detector and pre-amplifier are surrounded by a 5 cm thick lead shield. Pulses are registered by an integral discriminator with a sensitivity of 0.5 mV. To calibrate counters A and B, a flow of methane bubbler-saturated with T-ethanol vapours is passed in succession through the multi-wire counter and a calibration counter with known parameters. The counting efficiency is calculated with due regard to the ratio of the detector volumes and the known efficiency of the calibration counter.

The detection efficiency for T and ^{14}C beta-radiation for counter A operation in air-free methane is over 90%. A 500V - long counting plateau has no appreciable slope. Introduction of air affects the counting characteristics considerably. For a 5% admixture of air, the plateau length is reduced to 250V and its slope is 2%/100 V. The count curve for a mixture containing 10% of air in the region of 3.4 to 3.6 kV has a slope of 8%/100 V. The efficiency of beta particle detection in the working point (3.5 kV) is 85%. The count curve becomes still less stable as the air portion increases. The counter background of 150-180cpm remains stable up to 4.0 kV. The detector and gas system are cleaned from THO vapours and gaseous tritium exponentially ($\lambda = 1 \text{ min}^{-1}$), from the moment radioactivity intake has ceased.

The 0.5 l counter B with a multi-wire cathode is enclosed in a ring of protection counters located in the same volume of gas (Fig.1b). A two-channel panel selects coincidence and anticoincidence pulses.

The counter B anticoincidence background is 35 cpm in methane, 70 cpm in 10% air mixture and 85 cpm at 20% content of air, respectively. The count curves for T and ^{14}C have a plateau slope of about 5%/100V at 10% and 20% air in the mixture. The counting efficiency is about 90%.

The assay of the energy composition of air-borne radioactivity is based on the correlation between the number of coincidence counts in the measuring and protection counters and the path of

primary ionizing particles⁴. Thus, the ratio of coincidence and anticoincidence counts for T is 0.1, whereas for ¹⁴C - 1.6. The counter B measurement results may be used for estimating the ratio of nuclide concentrations in air.

Counter C (Fig.2) is intended for a selective determination

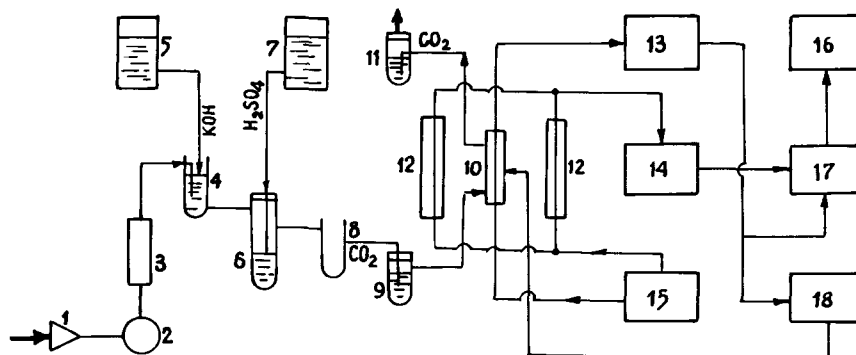


Fig.2. Block-diagram of air-borne ¹⁴C monitor.

1 - air intake; 2 - compressor; 3 - oven; 4 - bubbler; 5 - vessel with KOH; 6 - reaction vessel; 7 - vessel with H₂SO₄; 8 - dessicator; 9 - bubbler with cyclohexan; 10 - CO₂-counter; 11 - liquid seal; 12 - shielding counters; 13, 14 - input blocks; 15 - high voltage block; 16 - recorder; 17 - measuring block; 18 - electronic suppressor.

of air-borne as carbon dioxide or carbonate aerosols. Air under assay is bubbled through a flow of a 10% solution of KOH to absorb CO₂. The solution of KOH and K₂CO₃ is passed to a reaction vessel with a 25% solution H₂SO₄. Carbon dioxide resulting from the reaction: $K_2CO_3 + H_2SO_4 \rightleftharpoons K_2SO_4 + H_2CO_3$; $H_2CO_3 \rightarrow H_2O + CO_2 \uparrow$ is passed through a desiccator and a quenching agent (cyclohexan) bubbler to the counter. CO₂ acts both as an operating gas in Geiger counter and a radiocarbon carrier.

The cathode of the CO₂-counter is provided with a 0.3 mm-dia. platinum wire coil wound at a pitch of 5 mm inside the inner surface of a 18 mm-dia. quartz tube (Fig.1c). The counter volume is 30 cm³; the anode diameter 0.1 mm.

The count curve in measuring crude CO₂ is quite good due to the incorporation of an external suppressor assembly with a pulse height of 1.5 kV, duration 2 msec. The plateau length is 400 V; the slope - 2%/100 V; the working point - 3600 V. The background of the counter surrounded by a 5 cm-thick lead shield at anticoincidence with a ring of shielding Geiger counters is 4-5 cpm. The counter radiometric characteristics for air monitoring are given in the Table. The minimal detectable concentration (MDC) in the atmospheric air is defined as a level capable of inducing a count rate equal to that of the background. The simplest monitor A ensures a reliable detection of maximum permissible concentrations for personal of THO vapours (5.10⁻⁹ Ci/l⁶) and ¹⁴C compounds (3.5 x 10⁻⁹ Ci/l). The model B is capable of detecting concentrations lower than the above by an order as well as differentiating beta - radiation with respect to energy. The provision for the chemical selec-

Table of Parameters of Air-Borne Radioactivity Monitors

Counter	Gas to be assayed	Sample volume, cm ³	Background cpm	MDC, Ci/l of air	Response time min.
A	Air	15	150	5×10^{-9}	1
B	Air	100	85	4×10^{-10}	5
C	CO ₂	30	4	2×10^{-14}	30

tion of CO₂ makes counter C applicable for monitoring of air-borne ¹⁴C for all categories of exposure⁶. These instruments intended for operation under various conditions offer a still wider range of uses:

- (a) automatic recording and signalling of emergency situations;
- (b) simultaneous sampling of air from several checking points;
- (c) differentiation of gaseous tritium and THO vapours by means of the desiccator assembly;
- (d) continuous catalytic combustion of ¹⁴C and tritium-labelled organic compounds in air to form CO₂ and THO.

Monitoring of Radioactive Contamination of Surfaces

National standards treat of radioactive contamination of surfaces in terms of particle output from a unit of area regardless of radiation energy⁶. To comply with this, the efficiency of our portable instrument hardly depends on radiation energy. Detection is effected by a four-section gas-flow proportional counter with an open window (Fig.3). Anode wires are designed to run parallel to the examined surface. The outside cathode made of 0.6x0.6mm mesh brass net is separated by a 2mm clearance from the surface.

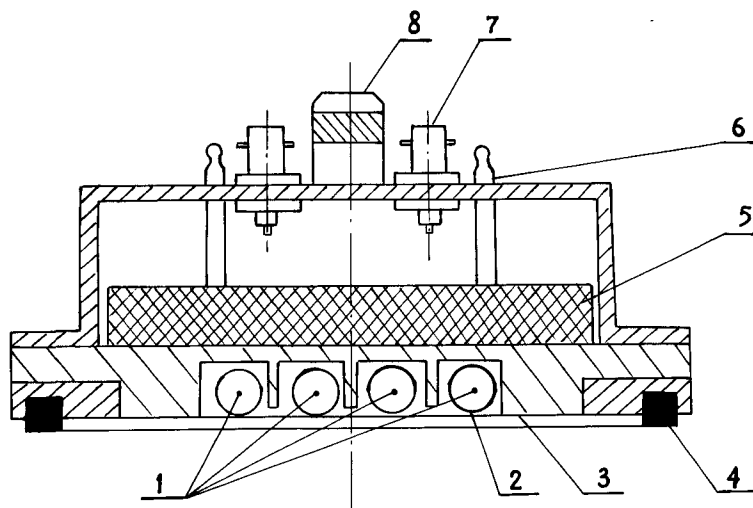


Fig.3. Surface contamination detector. 1-anodes; 2-insulators; 3-net; 4-rubber ring; 5-lead disk; 6-gas connection pipe; 7-plug connector; 8-grip.

The sensitivity area is 40 cm². The detector comprises a transistor pre-amplifier of signals. The gas (methane) flow rate is less than 0.5 l/min.

The measuring panel incorporates an integral discriminator,

a rate meter, a pointer indicator and a power supply.

Solid sources of beta radiation of tritium, ^{14}C and ^{137}Cs are used for calibration purposes. Plateaux of 200 to 300 V length with a slope of 5%/100 V have been obtained. The counting efficiencies at the point of operation are: 70% - for tritium, 80% - for ^{14}C and 90% - for ^{137}Cs . The proximity of the above values is due to the peculiarity of open-window counter operation, which detect a great portion of ionization electrons released in the gas layer bounded by the examined surface and the cathode. The counter background is 6 cps. The instrument reliably detects contamination under 0.01 of the maximum permissible levels for operation surfaces ($2.000 \text{ particle/cm}^2 \text{ min}^{-6}$).

Measurement of Tritium and ^{14}C Content in Human Body

The internal contamination of the human body induced by such beta-emitters as T and ^{14}C may be determined only indirectly on the basis of the results of biological sample assays.

T and ^{14}C concentrations in the body liquid samples and exhaled air were measured by scintillation and gas counters. Samples of urine, saliva, blood and exhaled vapour condensate were mixed with a liquid scintillator containing 8 g PPO + 0.2g POPOP + 100 g naphthalene per 1 l dioxane. Different methods of preparation of tritium-containing samples with respect to the degree of purification, such as distilled, activated coal-treated (urine only) and untreated samples, were tested. Precipitates were separated by filtration.

The beta-radiometry of the samples was carried out on a scintillation coincidence counter with two venetian-blind photomultipliers Ф3У-81А. A 30 ml cuvette for samples is made of Teflon and provided with quartz windows. The electronic recording system selects time-coinciding (within 100 nsec) pulses fed from the two photomultipliers and analyses them with respect to amplitude by means of a differential discriminator with a threshold ratio of 10:1. The measurements were carried out under balance conditions. The instrument was calibrated with the aid of a solid emitter of X-rays simulating tritium radiation⁷.

The best sensitivity is ensured in measuring samples containing 20% water or 8 to 10% urine, with the tritium counting efficiency being 12 to 15%; the counter background is 50 cpm. The sensitivity threshold to tritium in water is $3 \cdot 10^{-9}$ and in untreated human urine - $7 \cdot 10^{-9} \text{ Ci/l}$, respectively (measurement time -30 min; relative error -25%).

An analysis of the instrument background shows that 65% is contributed by internal processes occurring in the photomultipliers and generating light impulses; 30% - by radioactive and cosmic radiation, and 5% - by random coincidence of dark current pulses and the phosphorescence of samples.

The minimal detectable concentration of tritium is four orders below the "initial levels of the examination" of personnel in contact with tritium oxide⁸. The scintillation counter sensitivity to ^{14}C concentration in urine or water samples is 10^{-9} Ci/l which is quite sufficient for the purposes of industrial dosimetry.

Another modification of the indirect dosimetry method is provided by the measurement of THO vapours and $^{14}\text{CO}_2$ in exhaled air by means of gas monitors of types B and C. The air to be assayed is continuously sampled from a through-flow vessel where exhaled air comes. The CO_2 counter is capable of measuring ^{14}C concentrations up to $2 \cdot 10^{-12} \text{ Ci/l}$ in exhaled air (CO_2 content -3%). This makes it possible for ^{14}C excretion to be reliably monitor-

ed during one week after one "initial level of the examination" of $\text{NaH}^{14}\text{CO}_3$ incorporation. During the first days after exposure this level can be detected by instrument B as well. For a similar THO incorporation the body fluids tritium concentration is $35 \mu \text{Ci/l}^8$, whereas this parameters for exhaled air is $1.5 \cdot 10^{-9} \text{Ci/l}$ at the body temperature. However, when air is transferred to the detector, the air temperature is equalized to that of the device parts. This leads to the condensation of some vapours and the residual concentration of THO in the assayed air is determined by the ambient temperature. Thus, $5 \cdot 10^{-10} \text{Ci/l}$ of exhaled air corresponds to the "initial level of the examination" at 20°C . This concentration is reliably detected by counter B.

So, the above complex of instrumentation and techniques guarantees an all-round monitoring of exposure conditions of operation in contact with unshielded tritium and radiocarbon compounds. Depending on the process used and scale of production of different radioactive substances, the dosimetrical service may be supplied with some of the instruments of this complex or a suitable combination of them. The counters have been tested for a long period of time at the works of manufacturers specializing in the large-scale production of various radionuclide items, such as labelled organic and inorganic substances, luminous compounds and devices, metallic targets with tritium, etc. The test results have shown the described equipment and methods to comply with the requirements of day-to-day dosimetry.

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SAMPLING FOR TRITIATED WATER VAPOUR

by

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ABSTRACT

Collection of tritiated water vapour $(\text{HTO})_v$ from air by bubbling the air through non-tritiated water and subsequent measurement of the accumulated activity by liquid scintillation counting is often used as a simple method of monitoring for $(\text{HTO})_v$ in air.

Expressions for the collection efficiency of a model collecting device of this kind are derived here in terms of the initial mass of water in the collector, the total air flow, the humidity in the air entering and leaving the collector, the intrinsic efficiency of the collector and the ratio of the relative isotopic concentration $[T/H]$ in the vapour phase to that in the liquid phase at the effective operating temperature of the device.

Predictions from the model are compared to the measured efficiencies of collectors with and without fritted glass air dispersers, for water masses from 50 g to 200 g, air volumes 0.1 to 20 m³, air flow rates up to 160 cm³/s and water temperatures in the range 5°C–35°C. Intrinsic efficiencies greater than 95% and agreement between predictions and results to within a few percent is demonstrated for a practicable range of the variables.

Introduction

A simple method of collecting a sample of tritiated water vapour $(\text{HTO})_v$ from air is to bubble the air through water as shown in figure 1. This is, of course, a particular application of the general laboratory technique of gas washing. The comparative collection efficiencies of various kinds of gas washing bottles were investigated experimentally many years ago⁽¹⁾.

The method has been widely applied for tritium monitoring since the collected activity may be easily measured in a liquid scintillation counter⁽²⁾. Collection efficiencies observed in particular sampling systems have been reported^(3–5). In the last reference a theoretical expression ignored the sampling conditions and was of very limited applicability.

Here, the influence of relevant variables upon the collection efficiency is investigated theoretically. The experimental determinations of some of the parameters are reported and the experimentally observed dependence of the collection efficiency on some of the parameters is compared to the theoretical predictions.

Theory

Suppose that in the model bubbler shown in figure 2, air containing water (H_2O) at X g/m³ and $(\text{HTO})_v$ at C $\mu\text{Ci}/\text{m}^3$ is being bubbled through water. Let ϵ be the fraction of the air from which the ingoing tritiated water is removed and assume that this fraction is saturated by water vapour from the bubbler. Suppose that the saturated vapour density at the bubbler water/air

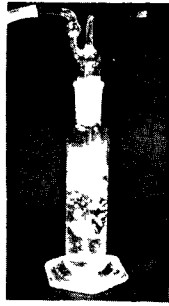
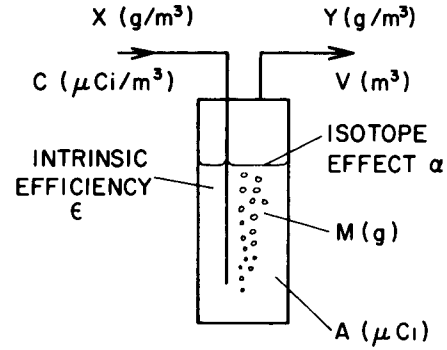


Figure 1: Laboratory and field bubbler: Nominal volume is 250 cm³.

Figure 2: Model bubbler.



interface is $Y \text{ g/m}^3$ and that the ratio of specific activity of HTO in the vapour phase to that in the liquid phase at the water/air interface under the particular dynamic conditions is α , the isotope effect coefficient.

Let the mass of water in the bubbler be $M \text{ g}$ and the activity of the tritium in it, $A \text{ } \mu\text{Ci}$.

The rate of change of mass of water in the bubbler with air volume $V \text{ m}^3$ passed through is given by

$$\frac{dM}{dV} = \epsilon (X - Y) \quad (\text{i})$$

and, assuming that the activity is uniformly distributed in the bubbler water, the rate of change of activity in the bubbler with air volume is given by

$$\frac{dA}{dV} = \epsilon \left(C - \frac{\alpha AY}{M} \right) \quad (\text{ii})$$

Equation (i) may be integrated directly to give

$$M = M_0 - \epsilon (Y - X)V \quad (\text{iii})$$

where M_0 is the initial mass of water in the bubbler.

Assuming that α is independent of V and M , if $X \neq Y$ the solution for A can be shown to be

$$A = \frac{MC}{(Y-X)W} \left[1 - \left(\frac{M}{M_0} \right)^W \right] + A_0 \left(\frac{M}{M_0} \right)^{W+1} \quad (\text{iv})$$

where A_0 is the activity initially in the bubbler.

$$W = \left(\frac{\alpha Y}{Y - X} \right) - 1$$

If $X = Y$ then

$$A = \frac{M_0 C}{\alpha X} \left[1 - \exp \left(-\frac{\epsilon \alpha X V}{M_0} \right) \right] + A_0 \exp \left(-\frac{\epsilon \alpha X V}{M_0} \right) \quad (\text{v})$$

In the trivial case of $Y = 0$,

$$A = \epsilon C V + A_0 \quad (\text{vi})$$

while in the special limiting case of $X = 0$ and $\alpha = 1$,

$$A = A_0 - \frac{CM}{Y} \ln \left(\frac{M}{M_0} \right) \quad (\text{vii})$$

The fraction of the sampled activity that is retained in the bubbler is the number of practical interest. This is the overall collection efficiency (E) and when, as is usually the case, $A_0 = 0$,

$$E = A/CV. \quad (\text{viii})$$

Figures 3–5 illustrate how variations in X , Y , ϵ and α affect the variation of collection efficiency with air volume sampled. For convenience the inefficiency $(1-E)$ is shown, expressed as a percentage. The initial mass of water M_0 is 100g in all these figures. However, since the variables X , Y and M_0 could have been combined as (X/M_0) and (Y/M_0) in the model, the curves shown can be interpreted for other values of M_0 by normalising the values of X and Y to the new M_0 ; i.e. by multiplying by $(M_0/100)$.

When the mass of water introduced into the bubbler and the mass of water lost from the bubblers are both small compared to the mass of water in the bubbler, the efficiency may be approximated by the expression

$$E = \epsilon - \frac{\epsilon^2 \alpha Y V}{2M_0} \quad (\text{ix})$$

When $W = 1$, (i.e., $X/Y = 1-(\alpha/2)$), $(1-E)$, calculated from equations (iv) and (viii), is linearly dependent upon volume V and is identical to the value calculated from equation (ix). A convenient set of conditions that produce a value of unity for W ($X = 10\text{g/m}^3$, $Y = 20\text{g/m}^3$, $\alpha = 1$) is repeated on figures 3–5. Hence in figure 3, the linear relation ($X = 10$) is also the approximation for all values of X as given by equation (ix). In figure 4, $W = 1$ for the three cases where $\alpha = 1$ so that equation (ix) predicts $(1-E)$ identical to the value from equations (iv) and (viii). In the other case ($\alpha \neq 1$) and in figure 5, linear approximations from equation (ix) are shown by the dashed lines.

Note from figures 3–5 that the inefficiency is most sensitive to the values of Y/M_0 and ϵ for the ranges of variables shown. Clearly, equation (ix) is adequate for predicting bubbler performance over a wide range of sampling conditions and values of ϵ . Determination of values of ϵ , α and the effects of the sampling conditions upon the overall collection efficiency of a particular bubbler are considered separately in the following sections.

Intrinsic efficiency (ϵ) of a bubbler

In general, ϵ will depend upon the extent of the dispersion of the two phases and their time of interaction, the air flow rate and the temperature. A high value for ϵ is desirable, from the point of view of overall collection efficiency.

Values of ϵ are difficult to predict *ab initio*. However the measured value of E is the lower limit of ϵ which might be expected to be the dominant limiting parameter determining E when

$$\frac{\alpha Y V}{2M_0} < 1 - \epsilon \text{ and } \ll 1 \quad (\text{x})$$

The intrinsic efficiency, ϵ , may be estimated therefore from the measured E without the estimate's being very dependent upon the α and Y which are difficult to measure accurately.

To determine experimentally when ϵ decreases below, say, 0.98, the sampling conditions should be chosen so that $\alpha Y V < .04 M_0$. For the conditions $M_0 > 50 \text{ g}$, $\alpha = 1$ and $Y = 20 \text{ g/m}^3$, V should therefore be less than 0.1 m^3 . Figure 6 shows the method used to measure ϵ for various kinds of bubblers, masses of water, temperatures and air flow rates. In these experiments input humidity X was 3 g/m^3 and the ambient air temperature was $\sim 23^\circ\text{C}$.

Figures 7 and 8 summarize the results of the measurements. The efficiencies predicted from the sampling conditions, using equation (ix), with Y (taken from psychrometric tables) corresponding to the temperature of the bulk of the bubbler water and values for α and ϵ of unity are also shown. Because this represents a lower limit to the efficiency that could be attributed to the sampling conditions, where the experimental points drop below this curve, the intrinsic

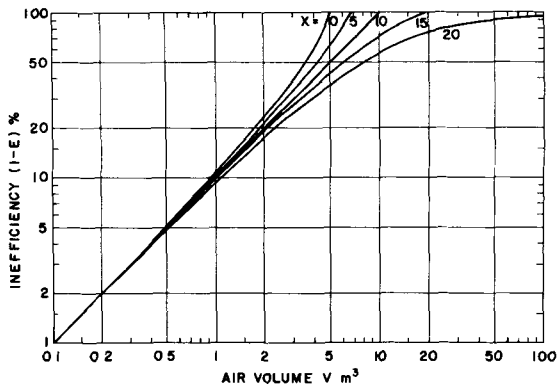


Figure 3: Calculated variations of collection inefficiencies with air volume for different values of sample humidity. Fixed parameters are $Y = 20 \text{ g/m}^3$, $M = 100\text{g}$, $\alpha = 1$, $\epsilon = 1$. Values of sample humidity ($X \text{ g/m}^3$) are given on the appropriate curves

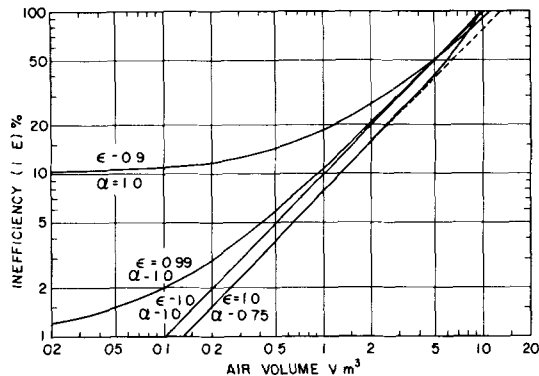


Figure 4: Calculated variations of collection efficiencies with air volume for different values of intrinsic efficiency ϵ and isotope effect coefficient α . Fixed parameters are $X = 10 \text{ g/m}^3$, $Y = 20 \text{ g/m}^3$, $M = 100\text{g}$. Values of the intrinsic efficiency ϵ and isotope effect coefficient α are given on the appropriate curves. The dashed linear extension when $\alpha \neq 1$ is calculated from equation (ix).

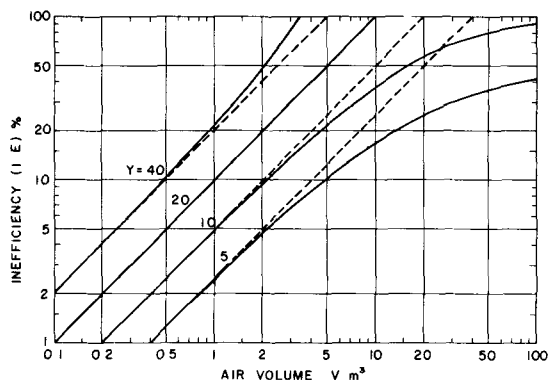


Figure 5: Calculated variations of collection efficiencies with air volume for different values of effluent air humidity. Fixed parameters are $X = 10 \text{ g/m}^3$, $M = 100\text{g}$, $\alpha = 1$, $\epsilon = 1$. Values of effluent air humidity ($Y \text{ g/m}^3$) are given on the appropriate curves. The solid lines are calculated from equations (iv), (v) or (vii) with (viii). The linear dashed extensions are from the approximate equation (ix)

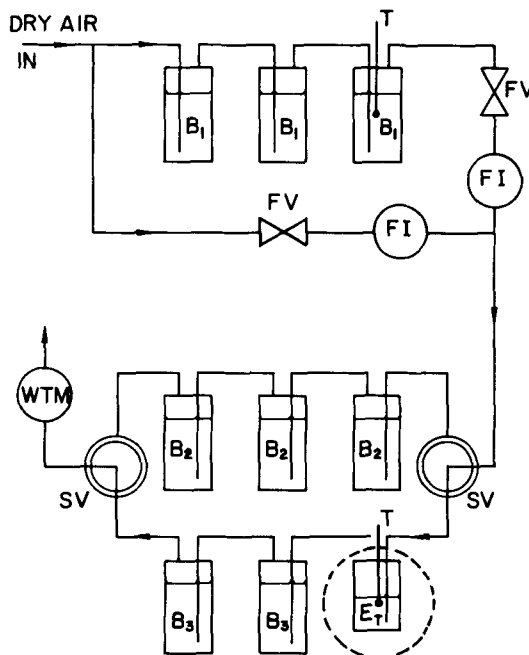


Figure 6: Outline of experimental arrangement for measuring properties of various bubblers. Tritiated water vapour was introduced into an air stream by passage through a series of bubblers (B_1) filled with tritiated water then diluted with dry air to give the requisite relative humidity. A metered flow was passed firstly, through the bubbler (E_1) under test and secondly, through a series of bubblers (B_3) to collect the activity escaping from the test bubbler. A measured volume of 0.096 m^3 was used throughout. The valves (FV) and rotameters (FI) were used for setting up the flow rates and for maintaining steady conditions. The wet-test meter (WTM) was used to measure the total volume of air passed by the test bubbler. During set up of the flows and after completion of a particular sample, the tritiated flow was diverted by valves (SV) through the second series of bubblers (B_2). The water temperature (T) in the last tritiated source bubbler and the test bubbler were measured during each sample. The activity in the test bubbler and the activities in the series of bubblers collecting the escaped activity were determined by diluting the water in each bubbler to a known volume and measuring the activity in an aliquot with a liquid scintillation counter. The experimental efficiency was then $A/(A + \sum B_3)$. Sufficient members of the B_3 series were included in the measurement to ensure that the activity not accounted for was less than 1% of the total lost. Generally, 2 were sufficient.

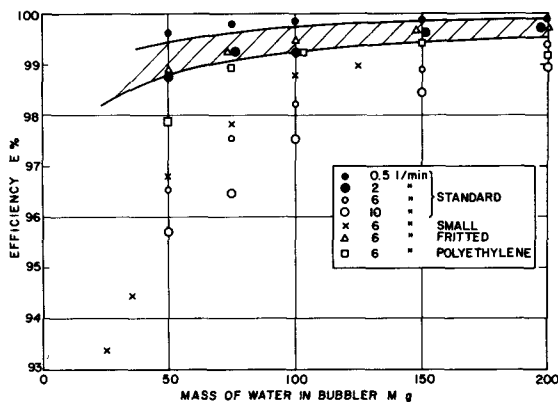


Figure 7: Measured efficiencies with different quantities of sampling water for various bubblers at a fixed air flow rate and for one bubbler at various flow rates. The band defined by the hatched area is the range of minimum efficiencies calculated from equation (ix) for all conditions used assuming an intrinsic efficiency $\epsilon = 1$. The standard bubbler is the one illustrated in figure 1. 'Small' refers to a similar one of volume 125 cm³. 'Fritted' refers to the standard type with a fritted stone air disperser. The polyethylene bubbler was similar in size to the standard with a coarse frit.

efficiency was the limiting parameter. This occurs with the smaller water masses and at the higher flow rates with the standard bubbler as might be expected. Nevertheless, even with only 25g of water ϵ was greater than 0.93. In all cases with 200g of water as collector, the intrinsic efficiency was greater than 0.99. With the fritted bubblers which more finely dispersed the air in the water a reduction in ϵ with decreasing M_0 was not observed at the flow rate used. Indeed, the total efficiency was better than predicted from the sampling conditions alone. In this case and at the lowest flow rate in the standard bubbler, agitation and mixing of the water in the bubbler was noticeably less than in the other cases. The effective temperature of the water at the air/water interfaces may therefore have been lower than that of the bulk of the water, resulting in a lower effective value for Y , and accounting, in part, for the high collection efficiency observed.

Clearly, the intrinsic efficiencies of even simple bubblers are high enough and are sufficiently independent of mass of water and flow rate for most practical purposes when used with air flow rates below 10 l/min and water masses above 50g in these types of bubbler.

Isotope effect coefficient (α)

The specific activity of $(\text{HTO})_V$ is known to be less than that of the water phase with which it is in equilibrium. The coefficient ranges from 0.86 at 0°C through 0.91 at 20°C to 0.94 at 50°C^(6,7). However equilibrium conditions are not necessarily attained in a bubbler; the high efficiencies observed above (figure 7) are not completely explained by α 's having equilibrium values. Appropriate values of α can be estimated for particular conditions from the initial rate of change of efficiency (E) with air volume (V) since, from equation ix,

$$\alpha = -\left(\frac{dE}{dV}\right) \cdot \frac{2M_0}{\epsilon^2 Y} \quad (\text{xi})$$

This is demonstrated in the next section.

Variation of collection efficiency with sampled air volume

Figure 9 shows the results of two experiments in which the efficiencies of two bubblers were measured for various sampled air volumes under different conditions using the apparatus outlined in

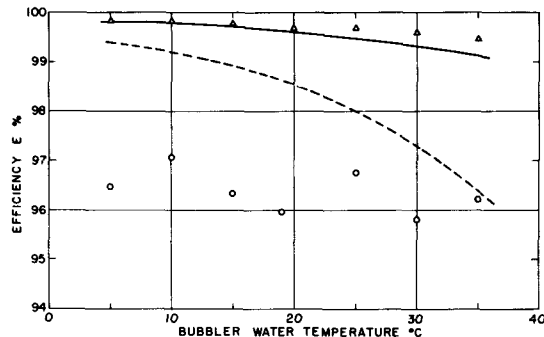


Figure 8: Measured efficiencies of bubblers at various water temperatures.

Δ – Bubbler with fritted glass disperser and $M_0 = 200\text{g}$.

\circ – Laboratory bubbler as in figure 1 with $M_0 = 50\text{g}$.

The solid and dashed curves are the lower limits to the respective efficiencies calculated assuming that $\epsilon = 1$ and α is the equilibrium value for the measured temperature.

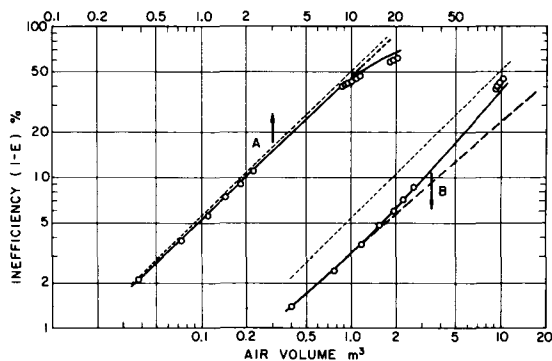


Figure 9: Measured inefficiencies of bubblers with $M = 200\text{g}$, sampling at 6 l/min for various total air volumes. Series A = $X \sim 18\text{ g/m}^3$, $Y \sim 23\text{ g/m}^3$. Series B = $X \sim 3.4\text{ g/m}^3$, $Y \sim 15\text{ g/m}^3$. Solid lines are calculated variations using values for α determined from the initial values of (dE/dV) . For series A, $\alpha = 0.85$, for series B, $\alpha = 0.71$. Dashed lines are calculated from equation (ix) using the same conditions. The dotted lines are calculated using the ambient temperature (23°C) to determine Y and equilibrium α in equation (ix).

figure 6 with the samples B3 being replaced periodically. The appropriate values of α were determined using equation (xi) from the initial slopes established by the experimental points. The curves for the complete experiment were calculated from equation (iv) and are drawn in figure 9. Note that linear approximations also on figure 9, using the ambient temperatures from which to estimate Y and the equilibrium values of α predict the efficiencies to within a few percent anyway. Of practical note here is that although carried out under similar ambient conditions, the evaporative self cooling of the series B with the drier input air results in a reduced loss rate and higher collection efficiency than in the A series. The actual bubbler bulk water temperature was about 3°C below the laboratory air temperature.

Conclusion

The most important parameters determining the overall collection efficiency for $(\text{HTO})_v$ of a bubbler are the intrinsic efficiency and the humidity of the effluent air. Deviation of the former from unity may be disregarded for a practicable range of air flow rates, temperatures and bubbler types. The dependence of the efficiency upon the humidity lost in the effluent air may be linearly related to the total air flow with a precision adequate for most monitoring purposes.

Acknowledgement

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A RUGGEDIZED ULTRASENSITIVE FIELD AIR SAMPLER
FOR DIFFERENTIALLY DETERMINING TRITIUM OXIDE
AND GAS IN AMBIENT AIR ATMOSPHERE

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Abstract

The instrument described is an operational, practical, ruggedized, ultrasensitive, tritium field air sampler assembled for the simultaneous, differential sampling of the environmental air for tritium oxide and elemental tritium. The system uses hardware assembled and packaged in such manner as to facilitate use in the field as well as in the laboratory. The sampling system occupies relatively small space and is simple to operate. The detection sensitivity approaches tritium background levels and is achieved by high volume sampling, efficient removal of tritium oxide and elemental tritium ("tritium gas"), and counting the recovered fractions by liquid scintillation spectrometry.

Introduction

The AEC standard for the maximum permissible concentration of tritium gas in uncontrolled areas is 200 times greater than for tritiated water vapor. Because of this difference, monitoring for tritium in the past at Mound, as well as at other locations, was for tritium as the oxide with the assumption that if the levels did not exceed the standard for the oxide, then the gas would be well within the standard.

When tritium in the environment or releases of tritium to the environment are measured, a measurement of oxide or even total tritium would not be sufficient to determine whether concentration guides have been exceeded; measurement of both tritium gas and tritiated water are necessary. Numerous methods for monitoring tritium oxide have been reported, but the literature is scant concerning tritium gas measurements. Mound Laboratory's goal is to determine background levels of tritium oxide and gas, both from natural and artificial sources, and to establish a baseline to provide a means of measuring any tritium that might be inadvertently added from operations at Mound.

General Description

The Mound Laboratory total system designed by MRC, Dayton Laboratory, consists of a tritium field sampler and a separate laboratory sample recovery system.

Collection System A picture of the collection system is shown in Fig. 1. The collection system with sample inlet, sample outlet, and hydrogen inlet connections, mounted controls, and indicators occupies a space 44 in. wide, 16 in. deep, and 38 in. high. The

*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

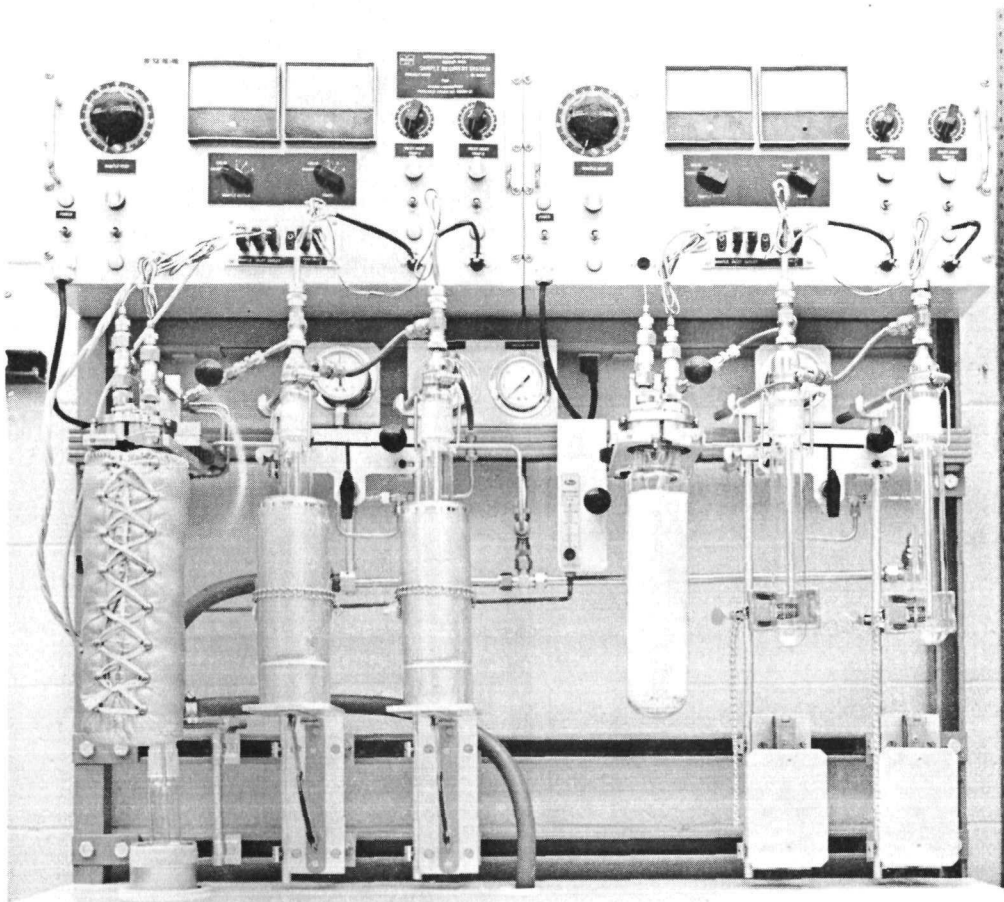


Fig. 1 - Tritium sample collection system.

sample collection/concentration bottles with protective wire-coated grid encasement are 3.88 in. in diameter and 17.55 in. overall length. The weight of the collection system, less bottles, is approximately 135 lb.

The collection system accommodates four sample collection bottles, the first two for the collection of tritium oxide and the other two for the collection of elemental tritium. The system valving permits a wide range of operating modes. Inadvertent introduction of hydrogen into the system (for the collection of elemental tritium) is averted by requiring air to flow past a sensor before a solenoid valve in the hydrogen line can be opened. Maintaining hydrogen supply within the limit of the flow meter provides assurance that the 4% lower explosive limit (L.E.L.), will not be exceeded. For normal air sample system flow rates of 30 to 75 liters/min with carrier hydrogen supplied at the rate of 100 cc/min, the mixture runs 0.2 to 0.4% of the L.E.L. The sensor and solenoid valve shut off the hydrogen inlet line in the event of low or no air flow during sampling.

Recovery System A two-station sample recovery system is shown in Fig. 2. This versatile, two-station system, was provided at Mound Laboratory to determine various operating parameters. The

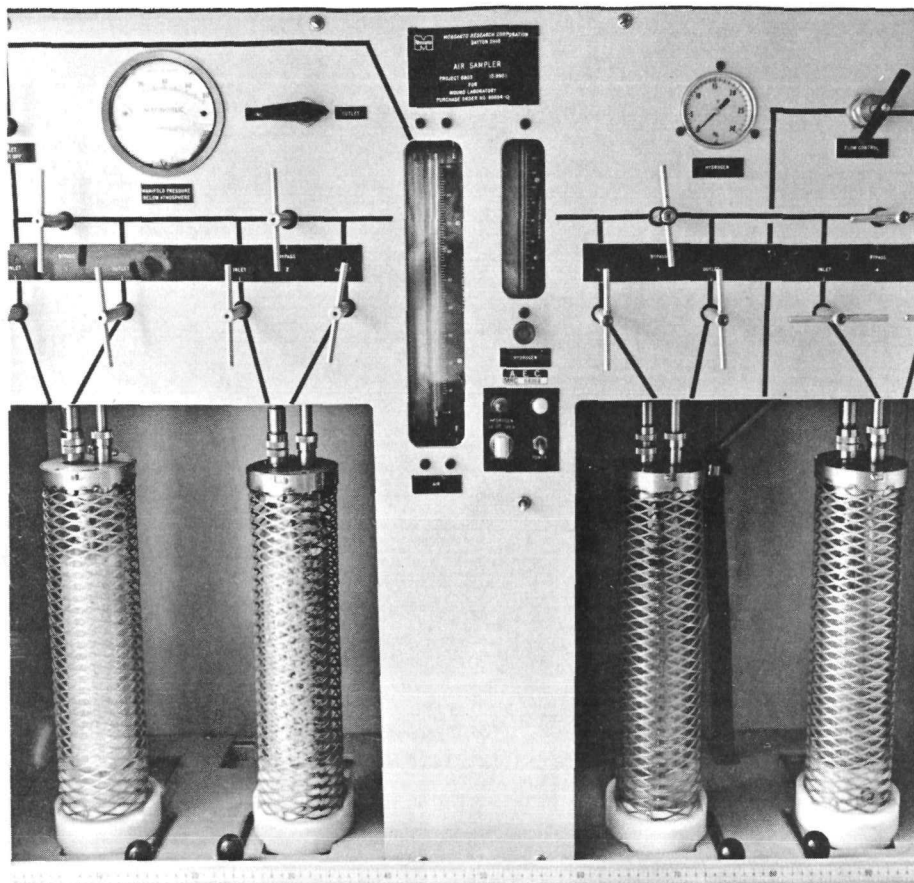


Fig. 2 - Two-station tritium sample recovery system.

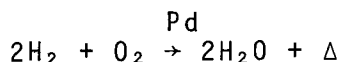
recovery system provides a laboratory facility for dry gas displacement or low pressure desorption of a collected sample at selected temperature and pressure into two cold traps in series.

Results and Discussion

Experience to date with the system at Mound Laboratory indicates that the silica gel used in the sample collection bottles must be thoroughly dehydrated prior to use. Typical air flow rate used in sampling has been 50 liters/min. At this flow rate, 750 g of silica gel in the collection bottles is adequate for a large volume grab sample in the range of 1500 to 3000 liters even under unfavorable ambient conditions of high relative humidity and temperature. Preliminary results in calibration of the system indicate 97-99% of the water vapor in the sampled air is recovered using one oxide collection bottle.

Seven hundred fifty grams of silica gel is also placed in the elemental tritium or gas collection bottles. The silica gel is coated with palladium black by adding 2 g of palladium powder to the 750-g charge of silica gel and agitating the mixture thoroughly until the palladium powder is completely coated on the surfaces of the silica gel particles. As the ambient air stream passes through this collection bottle, hydrogen gaseous isotopes are reacted with

the palladium to form oxides. The reaction is:



The resulting water is collected by the silica gel just as in the primary oxide collection bottles.

The cryogenic moisture recovery traps employed in the recovery system use liquid nitrogen. Essentially all of the water driven off the catalytic collector is captured in the first cold trap. No visible moisture has occurred in the second cold trap. Moisture is recovered in the system in a simple reverse of the method of collection. Recovery is completed in 30 min with the collection bottle at a temperature at 300°C. The heating mantle and its associated control variac are used to obtain the desired temperature. When the moisture has been driven off the sample, the exit temperature decreases. This temperature change can be monitored as the signal for desorption completion.

The use of palladium black is preferable to copper oxide since palladium reacts with hydrogen at ambient temperatures, whereas copper oxide must be heated before it will react. Since silica gel was found to be satisfactory, no other desiccant was evaluated.

Silica gel, as received from the manufacturer, contains residual moisture, so it must be thoroughly baked out before using at a temperature high enough to condition the silica gel for use in a reasonable period of time but not at a temperature high enough to destroy its sorption properties. Tests performed in the development of this monitoring system showed that a one-hour bakeout at 300°C adequately removed the moisture without affecting sorption properties. Normal residual moisture ranges from 0.03 to 0.07 g of water per gram of new silica gel.¹ To date, silica gel has been reused since there has been no indication that the bakeout reduced the collection efficiency in subsequent runs using known aqueous solutions of tritium oxide.²

The efficiency of the procedure for the determination of tritium oxide was tested using the set-up shown in Fig. 3. Known solutions of varying tritium oxide content, assayed by liquid scintillation counting and calibrated against a National Bureau of Standards (NBS) tritium oxide standard, were placed in the container marked "tritium oxide". Air was pulled through the drying trap at a flow rate of 50 liters/min, during which time the standardized tritium oxide was evaporated into the air stream and collected in the oxide collection bottle. Results indicate an overall recovery of 97-99%.

The efficiency of the procedure for the determination of elemental tritium was tested using the setup shown in Fig. 4. The flow rate of the calibration system was adjusted to 50 liters/min with valve #1 open and valves #2 and #3 closed. Hydrogen flow was adjusted to 150 cc/min. The system was operated in this configuration for 20 min after which time valves #2 and #3 were opened and valve #1 was closed. The tritium gas sample standard flask was flushed for 10 min. Gas samples used in calibration were standardized against an NBS gas standard prepared from the NBS tritium oxide standard.³ At the end of 30 min the set-up was shut down, the

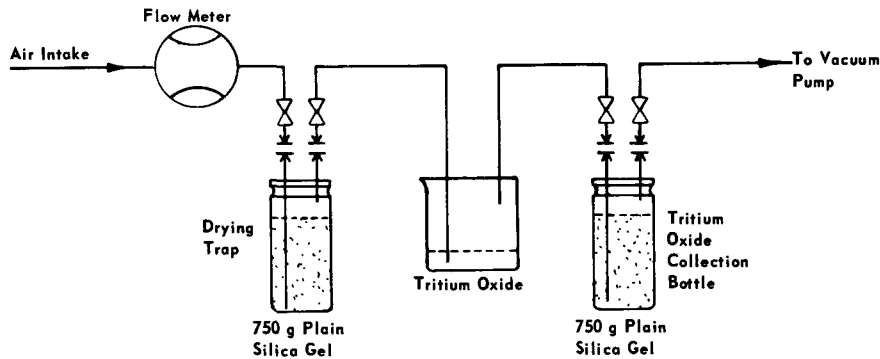


Fig. 3 - Setup used to test efficiency of tritium oxide collection.

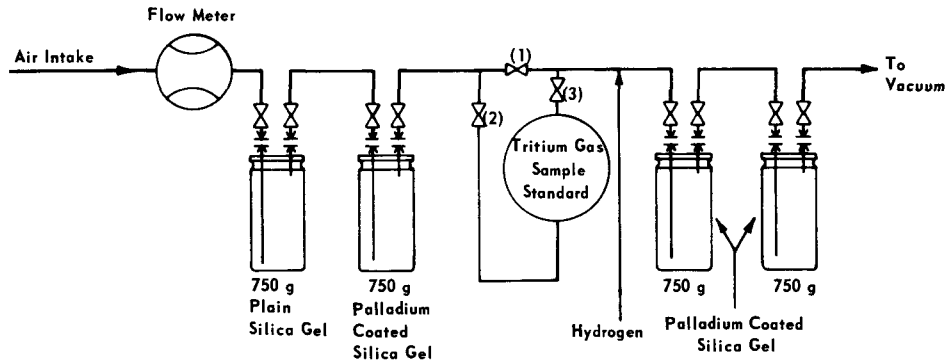


Fig. 4 - Setup used to test efficiency of elemental tritium collection.

collection flask containing the palladium-coated silica gel was removed, and the water was removed using the standard recovery procedure.

Two grams of palladium powder coated on the 750 g of silica gel are required to achieve recoveries in excess of 90% in the collection bottle. One gram of palladium powder achieved only 53 to 80.5% overall recovery in the collection bottle.

The theoretical sensitivity of this sampling system is 1.6×10^{-12} $\mu\text{Ci/cc}$ of air at the 95% confidence level for liquid scintillation counting with a 20 counts/min counter background, a 21% counter efficiency, and a 100 min sample counting time. A total of 1,223 liters of air sampled at a relative humidity of 50%, 68°F, and 760 mm barometric pressure would yield 10 ml of water which is the optimum amount that can be blended with a scintillation liquid for counting. A typical one-hour sample run at a flow rate of 50 liters/min would sample 3000 liters of air and yield some 25 ml of water. If all of the water could be counted as a single sample, the tritium oxide sensitivity would calculate to 0.6×10^{-12} $\mu\text{Ci/cc}$. The limitation in the system sensitivity is, therefore, not in the air sampling/collecting apparatus but in the liquid scintillation counting since only 10 ml or less of collected moisture can be analyzed. The lower detection limit could be achieved by concentrating the tritium in moisture collected to 10 ml by electrolysis. A sensitivity of 1.6×10^{-12} $\mu\text{Ci/cc}$ of air at the 95% confidence level, however, is in the range of tritium oxide background levels at Miami,

Florida⁴ and is adequate for an effective environmental oxide monitoring program.

A sensitivity of 0.6×10^{-12} $\mu\text{Ci/cc}$ of air is attainable for all samples containing elemental tritium since the total water in the collected gas sample can be controlled to a volume of 10 ml, all of which can be counted. The carrier hydrogen must be used to obtain sufficient water for analysis. Theoretically, 600 cc of hydrogen carrier gas produce 0.5 cc of water.

The use of silica gel, or other desiccants, for air sampling presents a "memory effect" that must be taken into consideration. The memory effect results from the fact that complete moisture desorption of silica gel is not attainable. The tritium content of the residual moisture will affect the results of each air sample collected. This factor has not been completely investigated with this sampling system, but based on limited data obtained to date, it appears that about 1% of the moisture from tritium oxide samples and from 3 to 10% of the moisture from tritium gas samples is not removed. In accurate sampling the memory effect can be corrected for by passing vapor of known tritium concentration through the sample collection bottle while it is being baked out under vacuum. Sufficient moisture is passed on to the silica gel to assure that all residual moisture is "pushed out" and replaced. When the sample collection bottle is used, the sample data are corrected to account for the controlled, known residual or background moisture and its tritium content.

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COMPARISON OF ALBEDO DOSIMETERS AND NUCLEAR
TRACK DETECTORS FOR NEUTRON MONITORING

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1. Abstract

The albedo neutron dosimeters so far used in personal dosimetry provided oversensitive indications of intermediate and thermal neutrons. Therefore, correction factors dependent on the location had to be used to assess the dose equivalent (Harvey¹, Hoy²). An albedo dosimeter tested at the Karlsruhe Nuclear Research Center showed promising results in measuring the dose equivalent of fast neutrons^{3 4}. Intercomparison measurements with nuclear track detectors were performed to indicate the dosimeter response, influence of the energy dependence and of the direction of the radiation incidence on the dosimeter reading.

2. Dosimeter and Radiation Sources

In a LiF-albedo dosimeter the thermal neutrons moderated and backscattered in the body of the wearer are detected by the ⁶Li (n,α)-reaction. If a pair of dosimeters is used, the neutron fraction of the reading is obtained from the difference in readings between a TLD-600 dosimeter (neutron + γ-dose reading) and a TLD-700 dosimeter (γ-dose reading).

Albedo dosimeters so far have been used preferably for the detection of intermediate and thermal neutrons. The detection response to fast neutrons is approximately 5 % of the response to thermal neutrons, but still 50 % of the γ-response. In this way, a LiF-albedo dosimeter is capable of detecting the dose equivalent of neutrons and of gamma rays over a dose range between 20 mrem and more than 1000 rem.

The following dosimeters were used for intercomparison measurements (cf. Table 1):

- Albedo neutron dosimeter according to Harvey: Absorption of incident thermal neutrons by means of a boron capsule¹. A pair of Harshaw ribbons of 3 x 3 x 1 mm³ size were used.

- Single albedo dosimeter:
Separate measurement of incident neutrons (D₂) outside the boron capsule and neutrons back-scattered from the body (D₁) within the boron capsule by means of one pair of dosimeters each⁴.

- Albedo dosimeter system:
Reduction of the influence of the body by wearing a dosimeter belt with one single albedo dosimeter each on the front and back of the body^{3 4}.
A functional relationship found

Detector	Energy	response
Kodak NTA Film	> 0.7 MeV	2·10 ⁴ tracks/cm ² ·rem [†])
²³⁷ Np+Makrofol E (40 μg/cm ²)	> 0.75 MeV	4 tracks/cm ² ·rem [†])
²³² Th+Makrofol E (0.05 mm foil)	> 1.2 MeV	37 tracks/cm ² ·rem [†])
Albedo Dosimeter Harvey	n _{th} , n _i	< 10 R/rem
Single Albedo Dosimeter	and > 100 keV	0.5 R/rem
Albedo Dosimeter System	-14 MeV	0.54 R/rem

[†]) Fluence-Dose conversion factor for track detectors
2.86 · 10⁷ n/cm² · rem

Tab.1: Neutron detector characteristic

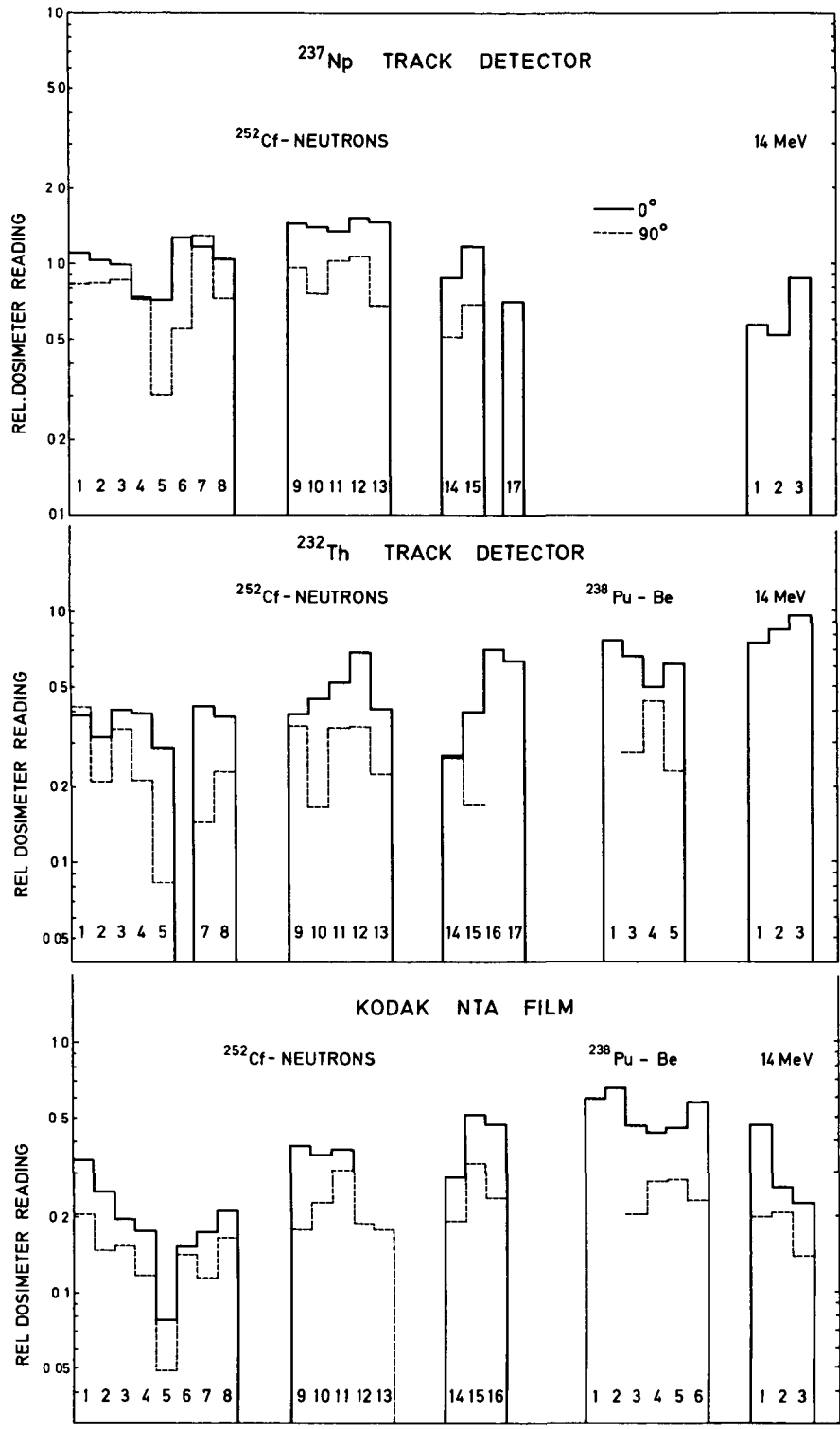


Fig.1: Relative dosimeter reading of nuclear track detectors after phantom irradiations with ^{252}Pu -Be- and 14 MeV neutrons

Position No.	Source	Distance source-detector
1	^{252}Cf in air + 5 cm Pb + 5 cm Al + 5 cm Fe + 16 cm Fe + 5 cm PVC + 11 cm PVC + 5 cm Concrete	2 m
2		
3		
4		
5		
6		
7		
8		
9	^{252}Cf in air 8 cm wall distance	1 m
10		
11		
12		
13	4 m	
14	^{252}Cf behind water layers of 4 cm 12.5 cm 43 cm 51 cm	2 m
15		
16		
17		
1	$^{238}\text{Pu-Be}$ in air in a small room:	1 m
2		2 m
3		1 m
4		2.3 m
5		2.2 m
6		3.6 m
1	14 MeV in air behind concrete (40 cm)	20 cm
2		30 cm
3		54 cm
4		1 m
5		1.5 m
6		2 m
7		3 m

Tab.2: Sources and exposure positions

Table 2 contains further details about the exposure positions. Irradiation with a ^{252}Cf -source of 1 mg was performed in free air and behind shieldings of PVC, concrete, aluminium, iron and lead. The source was set up at distances between 8 cm and 4 m from a concrete wall (wall effect), the distance of the detector remaining constant, or was suspended into a water tank of 60 cm diameter directly opposite the detector to generate water layers of different thicknesses ranging between 4 and 51 cm.

Irradiations with $^{238}\text{Pu-Be}$ neutrons were performed in free air and in a small chamber of $2 \times 3 \text{ m}^2$, the source being located in one corner of the room. 14 MeV neutrons were used for irradiation in free air and behind a wall, respectively. Exposures in a heavy water moderated power reactor (Obrigheim Nuclear Power Station) were performed directly on top of the reactor core outside the biological shield. Exposures in the FR 2 research reactor were performed in accessible places between the concrete and paraffin shields for in-pile experiments in horizontal beamholes.

3. Measured Results for Fast Neutrons

3.1 Nuclear Track Detectors

Figure 1 is a diagram of the reading of nuclear track detectors referred to the reading of the rem-counter for various exposure conditions. Due to fading within two weeks, the results obtained with the nuclear track emulsion are below of 60 %. An iron shield of 16 cm reduces the average neutron energy of the fission spectrum from 1.9 MeV to 0.88 MeV⁵. Here the NTA-film had higher fading and lower response, thus indicating only 25 % of the free air exposure. Because of the fading, the energy threshold of the NTA film is higher than in the ^{232}Th -detector. The reading of the ^{232}Th -detector for fission neutrons is lower by a factor of 2, while for 14 MeV neutrons it is only slightly higher than for Pu-Be

experimentally between D_1 and D_2/D_1 is used for correction:

$$D = k \times D_1$$

- Kodak NTA Type A nuclear emulsion welded in aluminium-plastic foil for the detection of fast neutrons > 0.7 MeV.
- ^{237}Np + Makrofol for the detection of fast neutrons > 0.75 MeV and
- ^{232}Th + Makrofol for the detection of fast neutrons > 1.2 MeV by counting of fission fragment tracks.

The exposures were performed with a human phantom (water bottles of 20 cm diameter, 40 cm height) at 1.4 m above ground with radiation incidence from the front, the sides and the back (0° , 90° , 180° , 270°). The respective dose equivalent were measured with an Anderson-Braun type rem-counter at the point of exposure of the phantom. In the case of 14 MeV neutrons also activation and threshold detectors were used. The rem-counter and the nuclear track detectors were calibrated in free air with an Am-Be-source of known source intensity.

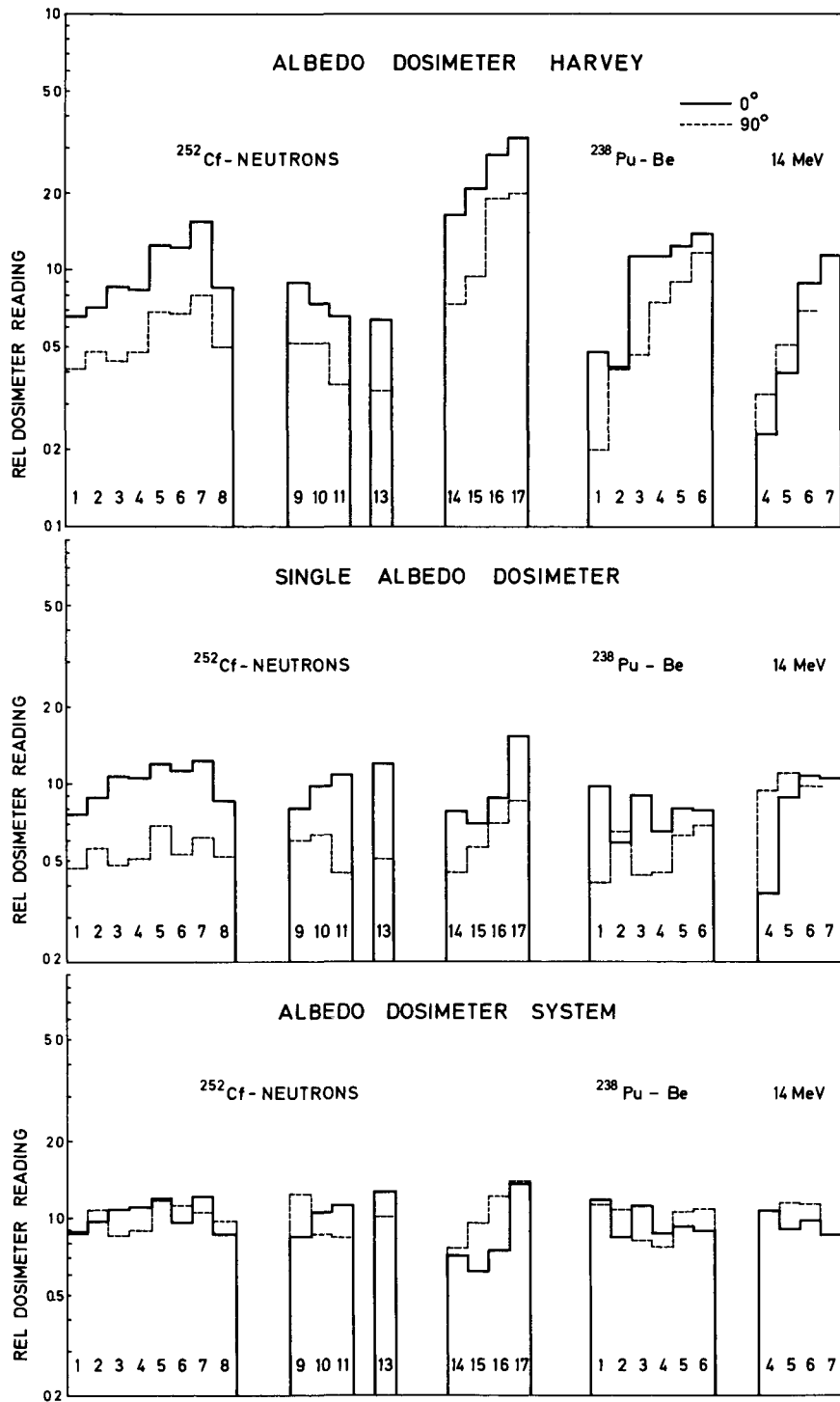


Fig.2: Relative dosimeter reading of albedo neutron dosimeters after phantom irradiations with ^{252}Cf -, ^{238}Pu -Be and 14 MeV neutrons

neutrons despite the high (n,f)-cross section, due to the fluence-dose conversion factor.

The deviation of the ^{237}Np reading from the reading of the rem-counter is between +50 % and -30 %. For 14 MeV neutrons, the deviations are larger because of the fluence-dose conversion factor. For radiation incidence under 90° nuclear track detectors in most cases indicate between 75 % and 45 % of the reading obtained by incidence upon the front.

3.2 Albedo Neutron Dosimeter (see Figure 2)

Because of its oversensitivity to thermal and intermediate neutrons, the albedo dosimeter according to Harvey indicates up to a factor of 6 more than the rem-counter. However, a modification of the fission neutron spectrum by 5 cm of shielding results in deviations only within $\pm 30\%$. In single albedo dosimeters the energy dependence and the influence of scattered thermal neutrons from the environment are reduced. For front incidence a deviation in readings for fission neutrons is found to be within $\pm 40\%$. For 14 MeV neutrons different correction factors were used⁴. As in nuclear track detectors, dosimeter readings between 75 % and 45 % of the reading obtained in free air were found for radiation incidence under 90° . The albedo dosimeter system reduces the influence of the direction dependence to approximately $\pm 15\%$ for radiation incidences under 0° , 90° , 270° and 180° . The total influence of the error due to energy and direction dependence of the dosimeter reading accordingly is $\pm 30\%$ for a dosimeter belt with two albedo dosimeters.

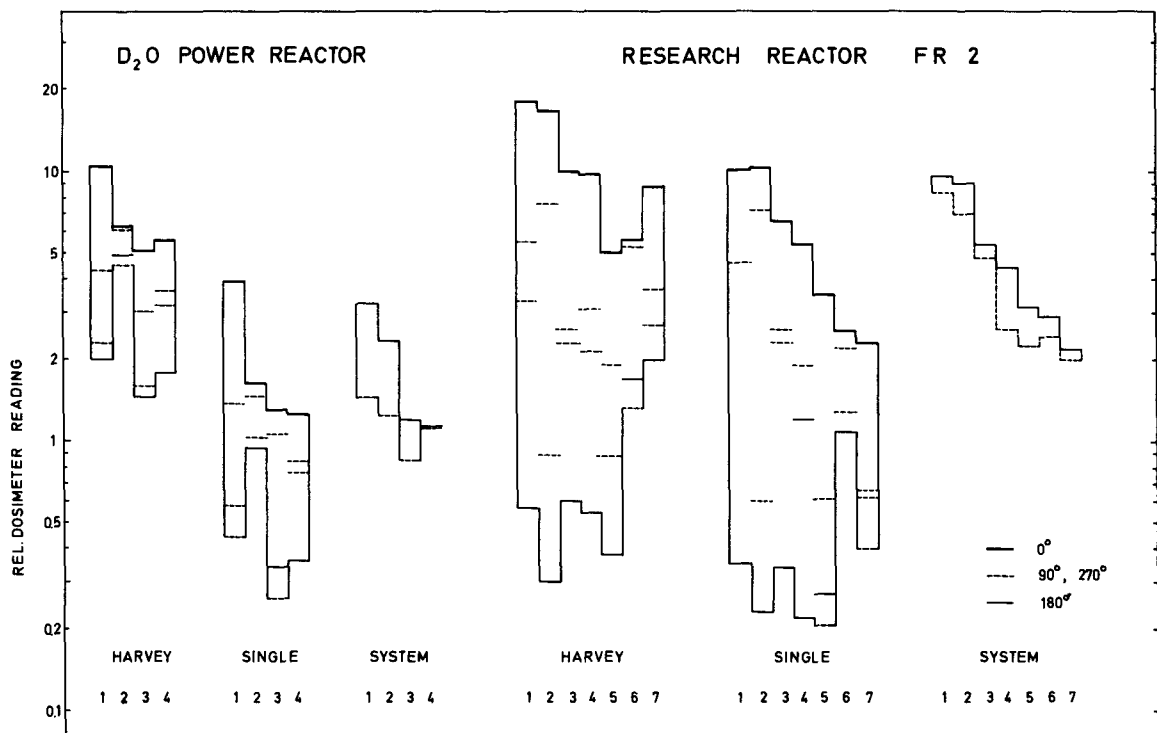


Fig.3: Relative dosimeter reading of albedo neutron dosimeters at reactor sites after phantom irradiations for different directions of radiation incidence

4. Results Measured at Reactor Sites (see Figure 3)

Chiefly thermal and intermediate neutrons are encountered near reactors, which requires correction factors to be applied in the evaluation of albedo dosimeters which depend on the location^{1 2}. The dosimeter reading varies by up to a factor of 2 in the case of incidence on the front near a power reactor and by a factor of up to 4 when measured near a research reactor. Harvey dosimeters have maximum sensitivities of 5 and 9 R/rem, respectively. In single albedo dosimeters this value is reduced to 2 and 5 R/rem, respectively. In albedo dosimeter systems the dosimeter reading shows a maximum variation of a factor of 2.5 except for the first two irradiation positions in the FR 2 research reactor. The maximum sensitivity is 2.5 R/rem. The albedo dosimeter system has the advantage of a non-directional dose reading for all radiation incidences between 0° and 180°. However the more unfavorable conditions existing near beamholes of research reactors are in no way representative for a personnel monitoring at reactors.

5. Summary

Because of the sometimes relatively high dose fraction of intermediate neutrons, location dependent correction factors must be taken into account in personnel monitoring at reactors by means of albedo dosimeters (corrections by up to a factor of 5). Because of the energy threshold, nuclear track detectors cannot be used for this purpose. In extreme cases, corrections by up to a factor of 10 are required near beamholes. This must be anticipated also in the energy range of intermediate neutrons from the calculated response of the dosimeter reading (see⁴, including results from⁶).

Albedo neutron dosimeters can be applied preferably to personnel monitoring near neutron sources in the range of energy between some 100 keV and 14 MeV. The readings are comparable to those obtained from a ²³⁷Np detector with respect to energy and direction dependence, while ²³⁷Th and the NTA-film show more unfavorable results because of the energy dependence and the energy dependence plus fading, respectively. For 14 MeV neutrons, ²³⁸Pu-Be neutrons and ²⁵²Cf-neutrons, a maximum deviation of ± 30 % was ascertained for the albedo dosimeter system, which is due to influences of scattered radiation, the neutron spectrum, and differences in the direction of radiation incidence between 0° and 180°. Compared with nuclear track detectors, albedo dosimeters have the advantage of a broader range of measurement (between 20 mrem and in excess of 1,000 rem), higher measuring accuracy (± 3 %), no energy threshold (with a detection of the correct dose > 100 keV), simple evaluation and simultaneous indication of the gamma dose.

Acknowledgments

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НЕКОТОРЫЕ ПЕРСПЕКТИВЫ ПРИМЕНЕНИЯ КРЕМНИЕВЫХ
ДЕТЕКТОРОВ ИОНИЗИРУЮЩИХ ИЗЛУЧЕНИЙ В ИССЛЕДОВАНИЯХ
ПО РАДИАЦИОННОЙ ЗАЩИТЕ

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The possibility of application of pulse rate measurements for x- and gamma-ray dosimetry with silicon radiation detectors was investigated. It was shown that this mode of operation ensures the sensitivity of 10^{-6} r.min⁻¹ for the detectors used in this work, and it is possible to decrease the energy dependence of sensitivity, for use in many radiation protection investigations by increasing the discrimination threshold. The general expression which can be used for rough estimates of sensitivity for these detectors was determined.

The background count rate of surface barrier detectors with different sensitive area from 0.2 to 60 cm² in alpha particles energy interval 3-9 MeV was determined. It was suggested for health physics and radiation protection monitoring of alpha-particles and simultaneous spectrometry to use the mosaic probe with large sensitive area (~ 60 cm²) which consists of many parallel united detectors one of which could be eliminated for spectrometric measurements.

Теперь уже хорошо известно, что неоднородные кремниевые детекторы ядерных излучений во многих случаях обеспечивают преимущества при использовании их для дозиметрии и радиометрии в исследованиях по радиационной защите. Тем не менее, ряд направлений возможного их применения изучен еще недостаточно, что и явилось основанием для настоящей работы.

ДОЗИМЕТРИЯ РЕНТГЕНОВСКОГО И ГАММА-ИЗЛУЧЕНИЯ
С ИСПОЛЬЗОВАНИЕМ ИМПУЛЬСНОГО РЕЖИМА РАБОТЫ
КРЕМНИЕВЫХ ДЕТЕКТОРОВ.

Служба радиационной безопасности для контроля радиационной обстановки нуждается в малогабаритном дозиметре рентгеновского и гамма излучения, обеспечивающем измерения в широком диапазоне мощностей экспозиционных доз. Однако, интегральный режим работы таких детекторов, подробно изученный рядом авторов (1,2,3 и др.), обладает низкой чувствительностью, ограничивающей использование их для оценки радиационной обстановки. С целью изучения возможности создания дозиметра с более высокой чувствительностью, выполнены исследования счетного режима работы золото-кремниевых

и кремний-литиевых детекторов. Измерялась их чувствительность (выраженная в скорости счета на единицу экспозиционной дозы) и зависимость ее от амплитудного порога регистрации импульсов, а также обсуждается способ оценки величины чувствительности расчетным путем.

Скорость счета на единицу экспозиционной дозы может быть выражена соотношением:

$$\frac{N_0}{PS} = \frac{1 - e^{-\mu_{Si} d}}{E_{\phi} \mu_{kt} \text{возд.}}; \quad (1)$$

где N_0 - скорость счета при уровне дискриминации, равном нулю; P - мощность экспозиционной дозы; E_{ϕ} - энергия фотонов; μ_{Si} - линейный коэффициент ослабления в кремнии; $\mu_{kt} \text{возд.}$ - массовый коэффициент передачи энергии в воздухе; d - ширина чувствительной области, S - площадь чувствительной поверхности детектора. Представляет интерес влияние величины порога регистрации (уровня амплитудной дискриминации) импульсов на чувствительность и "ход с жесткостью".

Обозначим через N_p скорость счета при пороге регистрации E_p . В интервале энергий фотонов $E_{\phi} = (0, 1+3) \text{ МэВ}$ в кремнии преобладает комптоновский эффект взаимодействия. Тогда, в случае однократного рассеяния и при толщине чувствительного слоя, превосходящей максимальный пробег вторичных электронов, интегральный спектр импульсов в первом приближении может быть представлен прямой линией, пересекающей энергетическую ось в точке максимальной энергии комптоновских электронов E_{max} , а ось скорости счета - в точке $N_0 \cdot \frac{E_p}{E_{\text{max}}}$. Аналитически такая линия представляется выражением: $N_p = N_0 \left(1 - \frac{E_p}{E_{\text{max}}}\right)$. Тогда зависимость чувствительности от уровня дискриминации можно выразить соотношением:

$$\frac{N_p}{PS} = \frac{1 - e^{-\mu_{Si} d}}{E_{\phi} \mu_{kt} \text{возд.}} \left(1 - \frac{E_p}{E_{\text{max}}}\right); \quad (2)$$

Соответствующие экспериментальные исследования выполнены с золото-кремниевыми и кремний-литиевыми детекторами, отличающимися толщиной чувствительного слоя.

Измерялись дифференциальные спектры вторичных электронов, по которым строились кривые зависимости чувствительности от уровня дискриминации для шести различных значений толщины чувствительной области в интервале (0,04+0,2) см.

На рис. 1 приведены экспериментальные результаты для толщины 0,08 и 0,2 см в сравнении с данными, рассчитанными по формуле (2).

Значения чувствительности, полученные в эксперименте для всех использованных толщин чувствительного слоя, отличаются от расчетных на фактор 2 и меньше, а само отклонение носит систематический характер. Такое расхождение определяется, по-видимому, смещением центра тяжести реального спектра импульсов по отношению к идеализированному в область низких энергий вторичных электронов. По этой же причине расхождение проявляет тенденцию к уменьшению при снижении порога регистрации, а значения чувствительности при пороге, равном нулю, полученные экстраполяцией экспериментальных кривых, отличаются от расчетных не более, чем на 30+40%, что находится в пределах экспериментальной ошибки измерения и погрешности в определении величины толщины чувствительного слоя.

Следовательно, формулу (2) можно использовать для грубой оценки чувствительности счетного режима работы кремниевых детекторов (с толщиной чувствительного слоя 0,04 см) в качестве дозиметров.

С целью исследования порога регистрации импульсов на "ход с жесткостью", по формуле (2) строились кривые зависимости чувствительности от энергии фотонов при различных уровнях дискриминации (рис. 2). Как показано на рисунке, "ход с жесткостью" существенно

зависит от уровня дискриминации, что дает возможность понижать его в известных пределах (для интервала энергий $0,3+3$ Мэв вплоть до $\pm 25\%$), правда, за счет некоторого уменьшения чувствительности, и применять кремниевые детекторы в дозиметрии полей с небольшим градиентом качества. На рис. 2. приведены экспериментальные значения дозовой чувствительности для энергий фотонов $0,661$ и $1,25$ Мэв. Из рисунка видно, что при переходе от энергии $0,661$ Мэв к $1,25$ Мэв с порогом регистрации $0,15$ Мэв чувствительность уменьшается в $(2,0 \pm 0,4)$ раза по сравнению с $1,9$, полученным в расчете, а при пороге $0,3$ Мэв - в $(1,4 \pm 0,3)$ раза против расчетного $1,3$.

Одним из критериев, определяющих область применения дозиметра, является интервал мощностей доз, доступных измерению. Максимальный уровень регистрируемых кремниевым детекторами в счетном режиме мощностей доз ограничивается разрешающим временем используемых импульсных электронных схем и составляет около $(0,1-0,01)$ р·мин⁻¹. Нижний предел чувствительности определяется наряду с геометрическими параметрами детектора (площадь и толщина чувствительной области), его фоновой скоростью счета. Минимальная мощность экспозиционной дозы, доступная измерению использованными в данной работе детекторами, составляет 10^{-6} р·мин⁻¹.

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Измерения малых активностей и идентификация их изотопного состава связаны с использованием аппаратуры, обладающей хорошим энергетическим разрешением и высокой чувствительностью к конкретному виду излучения. Как известно, чувствительность радиометрической аппаратуры определяется геометрической и физической эффективностью регистрации излучения и фоновой скоростью счета, которая у полупроводниковых детекторов, очевидно, в определенной степени зависит от условий их изготовления и радиационной частоты конструкционных материалов.

Проведено исследование фоновой скорости счета отечественных поверхностно-барьерных золото-кремниевых детекторов с площадью рабочей поверхности (S) от $0,2$ до 9 см² и мозаичных структур на их основе с площадью до 60 см², а также диффузионно-дрейфовых кремний-литиевых детекторов с $S=0,3+3$ см² и толщиной чувствительной области от $0,4$ до 2 мм. Энергетическое разрешение отдельных детекторов при комнатной температуре ($+20^\circ\text{C}$) составляло от 30 до 160 Кэв.

Обнаружено, что в области энергии $3+9$ Мэв, являющейся рабочей областью для альфа-спектрометрии радиоактивных изотопов, интегральная фоновая скорость счета как отдельных золото-кремниевых детекторов с площадью рабочей поверхности от 1 до 9 см², так и группы параллельно соединенных детекторов с общей площадью до 60 см², пропорциональна величине S и составляет $(0,22 \pm 0,02)$ час⁻¹см². Примерно $1/3$ этой величины обусловлена эманациями и аэрозолями,

и кремний-литиевых детекторов. Измерялась их чувствительность (выраженная в скорости счета на единицу экспозиционной дозы) и зависимость ее от амплитудного порога регистрации импульсов, а также обсуждается способ оценки величины чувствительности расчетным путем.

Скорость счета на единицу экспозиционной дозы может быть выражена соотношением:

$$\frac{n_0}{PS} = \frac{1 - e^{-\mu_{Si}d}}{E_{\phi} \mu_{кп} \text{возд.}}; \quad (1)$$

где n_0 - скорость счета при уровне дискриминации, равном нулю; P - мощность экспозиционной дозы; E_{ϕ} - энергия фотонов; μ_{Si} - линейный коэффициент ослабления в кремнии; $\mu_{кп} \text{возд.}$ - массовый коэффициент передачи энергии в воздухе; d - ширина чувствительной области, S - площадь чувствительной поверхности детектора. Представляет интерес влияние величины порога регистрации (уровня амплитудной дискриминации) импульсов на чувствительность и "ход с жесткостью".

Обозначим через n_{Γ} скорость счета при пороге регистрации E_{Γ} . В интервале энергий фотонов $E_{\phi} = (0, 1+3) \text{ МэВ}$ в кремнии преобладает комптоновский эффект взаимодействия. Тогда, в случае однократного рассеяния и при толщине чувствительного слоя, превосходящей максимальный пробег вторичных электронов, интегральный спектр импульсов в первом приближении может быть представлен прямой линией, пересекающей энергетическую ось в точке максимальной энергии комптоновских электронов E_{max} , а ось скорости счета - в точке $n_0 \cdot \frac{E_{\Gamma}}{E_{\text{max}}}$. Аналитически такая линия представляется выражением: $n_{\Gamma} = n_0 \left(1 - \frac{E_{\Gamma}}{E_{\text{max}}}\right)$. Тогда зависимость чувствительности от уровня дискриминации можно выразить соотношением:

$$\frac{n_{\Gamma}}{PS} = \frac{1 - e^{-\mu_{Si}d}}{E_{\phi} \mu_{кп} \text{возд.}} \left(1 - \frac{E_{\Gamma}}{E_{\text{max}}}\right); \quad (2)$$

Соответствующие экспериментальные исследования выполнены с золото-кремниевыми и кремний-литиевыми детекторами, отличающимися толщиной чувствительного слоя.

Измерялись дифференциальные спектры вторичных электронов, по которым строились кривые зависимости чувствительности от уровня дискриминации для шести различных значений толщины чувствительной области в интервале $(0,04+0,2)$ см.

На рис. 1 приведены экспериментальные результаты для толщины 0,08 и 0,2 см в сравнении с данными, рассчитанными по формуле (2).

Значения чувствительности, полученные в эксперименте для всех использованных толщин чувствительного слоя, отличаются от расчетных на фактор 2 и меньше, а само отклонение носит систематический характер. Такое расхождение определяется, по-видимому, смещением центра тяжести реального спектра импульсов по отношению к идеализированному в область низких энергий вторичных электронов. По этой же причине расхождение проявляет тенденцию к уменьшению при снижении порога регистрации, а значения чувствительности при пороге, равном нулю, полученные экстраполяцией экспериментальных кривых, отличаются от расчетных не более, чем на 30+40%, что находится в пределах экспериментальной ошибки измерения и погрешности в определении величины толщины чувствительного слоя.

Следовательно, формулу (2) можно использовать для грубой оценки чувствительности счетного режима работы кремниевых детекторов (с толщиной чувствительного слоя 0,04 см) в качестве дозиметров.

С целью исследования порога регистрации импульсов на "ход с жесткостью", по формуле (2) строились кривые зависимости чувствительности от энергии фотонов при различных уровнях дискриминации (рис. 2). Как показано на рисунке, "ход с жесткостью" существенно

зависит от уровня дискриминации, что дает возможность понижать его в известных пределах (для интервала энергий $0,3+3$ Мэв вплоть до $\pm 25\%$), правда, за счет некоторого уменьшения чувствительности, и применять кремниевые детекторы в дозиметрии полей с небольшим градиентом качества. На рис. 2. приведены экспериментальные значения дозовой чувствительности для энергий фотонов $0,661$ и $1,25$ Мэв. Из рисунка видно, что при переходе от энергии $0,661$ Мэв к $1,25$ Мэв с порогом регистрации $0,15$ Мэв чувствительность уменьшается в $(2,0 \pm 0,4)$ раза по сравнению с $1,9$, полученным в расчете, а при пороге $0,3$ Мэв - в $(1,4 \pm 0,3)$ раза против расчетного $1,3$.

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содержащимися в воздухе лабораторного помещения.

При увеличении площади рабочей поверхности золото-кремниевых детекторов до 8-10 см² их геометрическая эффективность возрастает до 30±35% для источников с площадью активного пятна около 6 см². Разрешающая способность детекторов таких размеров не хуже 2±3% на линии 5,15 Мэв, что обеспечивает возможность идентификации и измерения практически всех естественных альфа-радиоактивных изотопов, находящихся в смеси в малых количествах (порядка 10⁻¹⁴ кюри в пробе).

В оценке радиационной обстановки при измерении загрязненности рабочих поверхностей необходимы датчики с относительно большим S (~100 см²), пригодные для спектрометрического анализа изотопного состава загрязненности непосредственно на месте контроля. Для этой цели не пригодны ни отдельные золото-кремниевые детекторы с большой площадью, ни мозаичные структуры на основе детекторов серийного производства, поскольку разрешающая способность таких систем при комнатной температуре слишком мала, и не может быть повышена до удовлетворительной величины (2±3%) в силу ограничений принципиального свойства.

В данной работе предлагается использовать для измерения поверхностной загрязненности альфа-радиоактивными веществами и их идентификации мозаичный датчик с большой площадью, состоящий из нескольких параллельно соединенных полупроводниковых детекторов, один из которых отключается от остальных для выполнения спектрометрических измерений. Предложенный принцип сочетает достоинства мозаичной структуры, обеспечивающей возможность создания датчика с требуемой площадью чувствительной поверхности (до 100 см² и выше), с высокими спектрометрическими свойствами отдельного детектора, площадь которого (5±8 см²) не велика по сравнению с площадью мозаики.

Блок-схема прибора приведена на рис. 3. Мозаичный датчик, испытанный в настоящей работе, состоял из 12 поверхностно-барьерных золото-кремниевых детекторов, с площадью рабочей поверхности равной 5 см² и энергетическим разрешением близком к 70 кэв, производство которых освоено отечественной промышленностью.

Зарядочувствительный предусилитель с большой входной динамической емкостью смонтирован в блоке детектирования вместе с детекторами. Энергетическое разрешение прибора в спектрометрическом режиме работы составило 140 кэв при измерении спектра излучения источника плутония-239 в воздухе. Фоновая скорость счета всей мозаики составила 15 имп·час⁻¹.

Отбор детекторов проводился, в основном, по величине максимально допустимого напряжения смещения. Другой критерий - требование идентичности величины оптимального рабочего смещения для всех детекторов мозаики оказался менее критичным, поскольку зависимость энергетического разрешения от напряжения смещения, как правило, у таких детекторов не имеет острого минимума. Годными для использования в мозаике оказались 70% детекторов одной классификационной группы, что говорит о доступности использованного принципа и возможности его широкого применения.

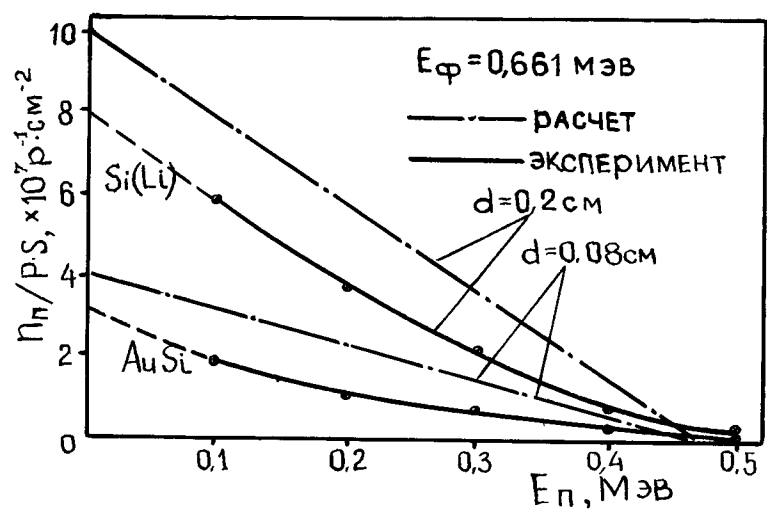


Рис. 1. Дозовая чувствительность в зависимости от порога регистрации импульсов для энергии излучения 0,661 Мэв.

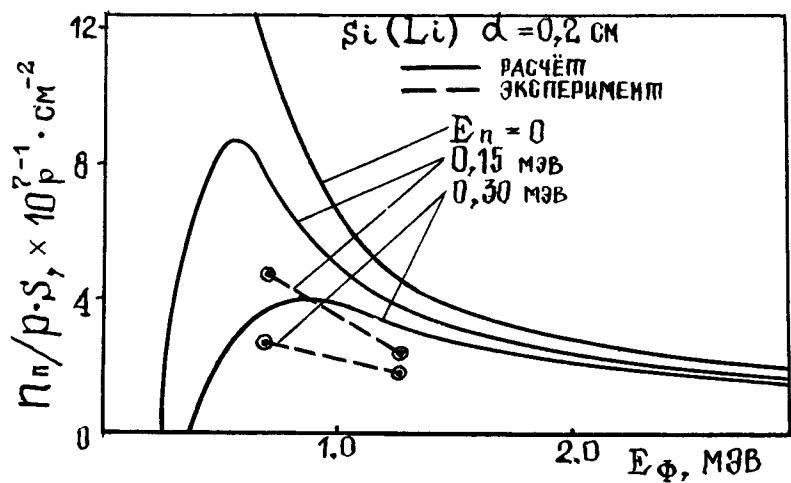


Рис. 2. Зависимость дозовой чувствительности от энергии излучения при различных порогах регистрации.

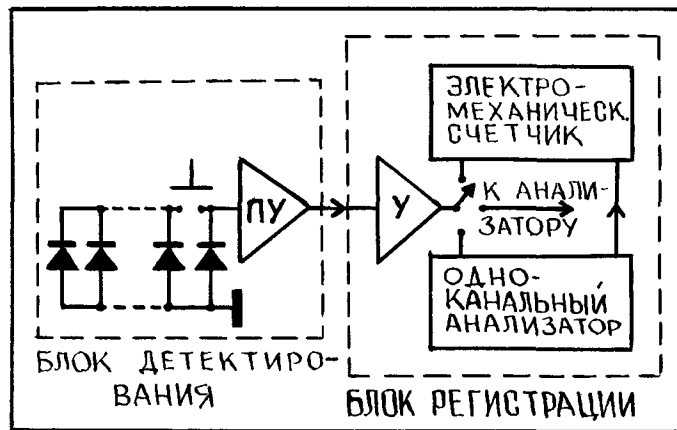


Рис. 3. Блок-схема прибора для измерения поверхностной загрязненности альфа-радиоактивными веществами и их идентификации.

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HIGH ENERGY PHOTON DETECTION
BY
FISSION TRACK REGISTRATION

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Abstract

An attempt is made in this report to detect high energy photons above 5 MeV by photo fission reaction. A solid state fission track detector, in which one face of a solid block was coated with fissionable material, was used as a high energy photon detector.

Typical results obtained with bremsstrahlung at maximum energy of 29 MV are as follows. Fission track density per 100 R was found to be about 1,000 tracks/cm². The number of high energy photons over 5 MeV per 100 R from the total bremsstrahlung was estimated to be 4×10^7 /cm², assuming that the effective cross section of natural uranium is approximately 0.05 barn for the bremsstrahlung.

The track detector presented in this report may provide a useful means to detect high energy photons even in the presence of other radiations except neutrons, and is expected to be utilized for the monitoring of high energy photons.

Introduction

For the measurement of high energy X- or gamma-rays above several MV or MeV, nuclear reactions of high energy photons have been applied in some instances in addition to usual measurement by ionizing process. The measurement by means of photo fission has an advantage in that the high energy photons can be distinguished from a mixed radiation field except neutrons.

In the present report, a method was examined for high energy photon detection by solid state fission track detector registering the tracks of fission fragments produced by photo fission reaction. A solid state fission track detector which had been previously developed by us for neutron detection was used to detect the high energy photons.¹

Experiment

The track detector consists of a solid for registration of fission fragment track and fissionable material. There are several ways of combination, e.g., U-doped glass or a glass block in contact with an uranium foil. The method adopted in the present study was to bring a solid in contact with a fissionable material. Silver activated phosphate glass of Toshiba FD-P8-3,

a commercially available dosimetric glass, was employed as the solid because of its easiness for etching and optically almost perfect surface. Uranylacetate, $\text{UO}_2(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$, an easily available reagent, was used as a fissionable material. A saturated solution of uranylacetate was made at $30 - 40^\circ\text{C}$, then $0.1 - 0.2$ ml of the solution was applied dropwise and dried on a glass block which was thoroughly pre-rinsed with distilled water, forming a layer of $15 - 30 \text{ mg/cm}^2$ thickness tightly attached on the surface of the glass block (Fig. 1). The layer is thicker than the maximum range of fission fragments, so that there is no effect of thickness on the track density.

The detectors were exposed in the range of 100 to 200 R to X-rays from the NIRS betatron (Toshiba) without any flattening filter at maximum bremsstrahlung energies of 15 to 29 MV. Exposure was measured by the Victoreen thimble ionization chamber, but the chamber could not be irradiated simultaneously with the detector owing to narrow X-ray beam.

To investigate the effect of exposure rate on track density, irradiation was made at variable exposure rate from 25 to 60 R/min at maximum energy of 29 MV.

After irradiation, detectors were cleaned thoroughly by use of an ultrasonic washer to remove the uranylacetate layer, and then radiophotoluminescence of the glass block was measured with a photoluminescence reader. Because of the high energy of X-rays in excess of the applicable range for the glass dosimeter, the reading does not correspond to the absolute exposure but rather its relative value. Finally, the glass blocks were etched by a 30 % solution of sodium hydroxide at 80°C for 10 min to make visible tracks which were counted under a microscope (Fig. 2). Track density was estimated by counting the number of tracks in 4 to 5 fields of 2.5 mm^2 on each glass block. Background was estimated from the opposite side of the glass block.

Experiments were carried out in the same way on thorium-chloride.

Result

Hereinafter, the value of radiophotoluminescence in roentgen obtained with the reader is expressed by (RPL).

The track density per 100 R of X-ray at maximum energies of 15, 20, 25 and 29 MV is shown in Fig. 3. The sensitivity of the detector for X-ray at maximum energy of 29 MV is about $1,000 \text{ tracks/cm}^2$ per 100 R, which is the average from several independent experiments. Effect of exposure rate on the track density is small as shown in Fig. 5.

In the case of thoriumchloride, significant result could not be drawn due to the insufficient number of fission tracks.

Discussion

Because of fluctuation in X-ray intensity during irradiation especially at low energy range and of the irradiation process of detector which was not made simultaneously with the ionization chamber, the relative exposure dosimetry with the glass block (RPL) is considered to be more reliable than that with the chamber. Fig. 4 shows the energy characteristic of the detector. The track density per (RPL) tends downward with decreasing energy.

Fig. 6 shows photo fission cross section of uranium and

thorium². Maximum cross section for photons was found to be about 15 MeV for both uranium and thorium. The higher the maximum energy of X-ray, the higher efficiency of detection was observed on the fission track detector as shown in Fig. 4. This seem to be due to the fact that the X-ray spectrum is continuous and that for the same exposure the photons of over 5 MeV is fewer at low maximum energy than at high maximum energy. In mono-energy photon flux such as gamma-rays, the highest efficiency of detection may be obtained at about 15 MeV. In either case, the detection of photons of below 5 MeV is impossible, since the cross section for them is practically zero.

The number of high energy photons contained in 100 R of X-rays at maximum energy of 29 MV are calculated as below. The relation between track density ρ and photon flux Φ is given by

$$\rho = \Phi \cdot \sigma \cdot N \cdot f$$

where σ is the photo fission cross section of uranium, N, the number of uranium atoms in 1 mg of uranylacetate, and f, the ratio of track density to the number of fissions occurred in 1 mg of uranylacetate layer which has a thickness larger than the range of fission fragments. From the experimental data on neutron irradiation, the value of f is estimated at $10/3(\text{track} \cdot \text{cm}^{-2}/\text{fission} \cdot \text{mg}^{-1})$. The number of high energy photons over 5 MeV per 100 R of X-ray was calculated to be approximately $4 \times 10^9/\text{cm}^2$, assuming that the effective fission cross section of uranium is 0.05 barn for the X-ray.

The tracks on a glass block indicate only the existence of high energy photons over 5 MeV in radiation field except neutrons. Therefore, the exposure of X-ray which has continuous energy spectrum of photons can not be determined from the track density. In such a case as to investigate the primary effect of high energy photons, the fission track detector may provide a useful means for the counting of high energy photons even in the presence of other type of radiations except neutron, and is expected to be utilized in the monitoring of high energy photons.

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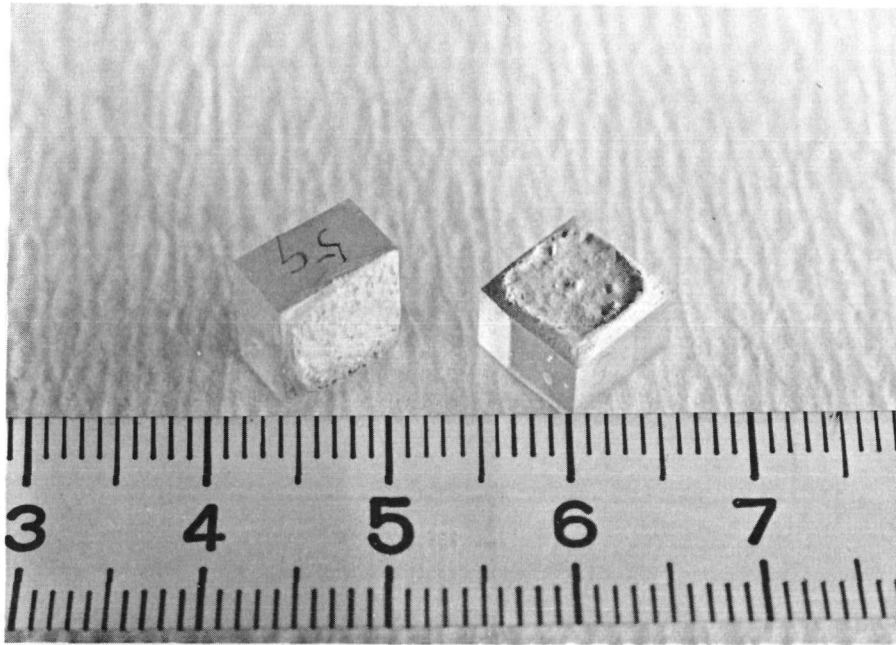


Fig. 1. Detectors, minor unit of scale in mm.

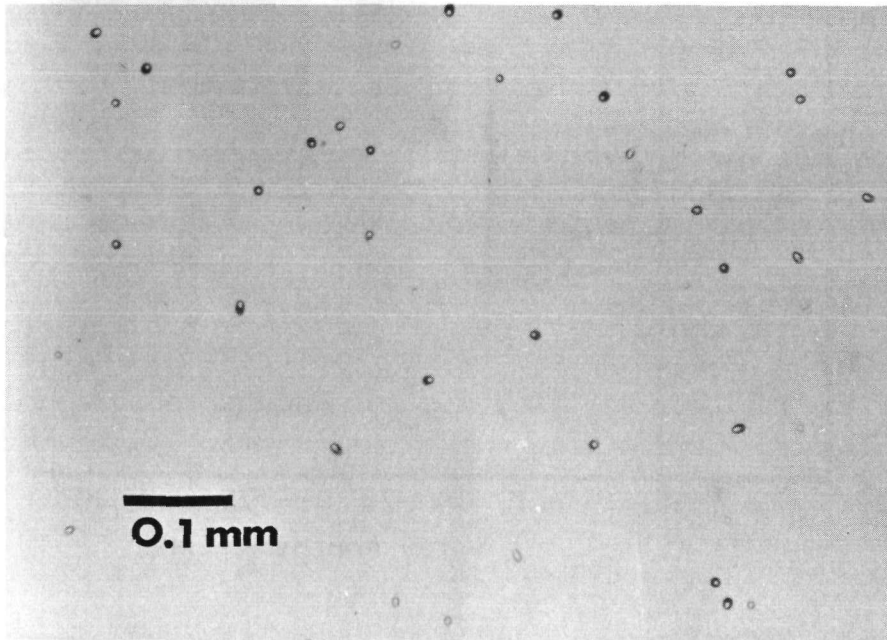


Fig. 2. Fission track etch pit in glass irradiated by X-ray at maximum energy of 29 MV from betatron.

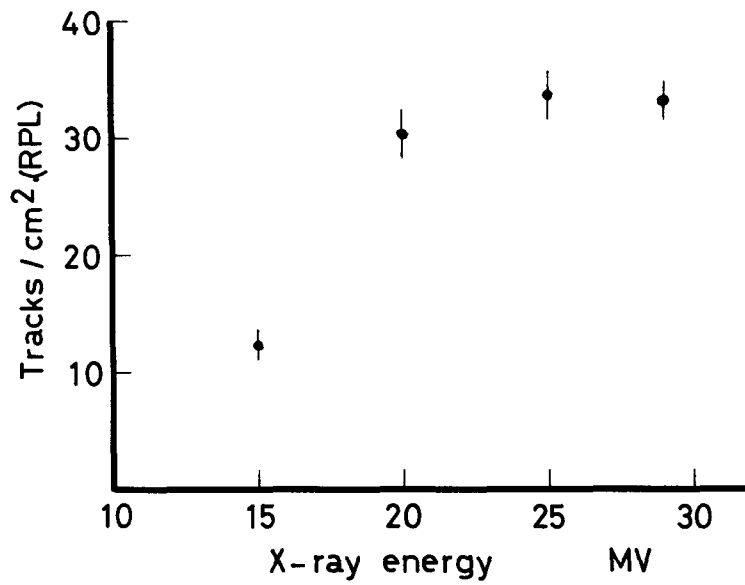


Fig. 3. Track density per 100 R of X-ray at maximum energies of 15 to 29 MV and (RPL).

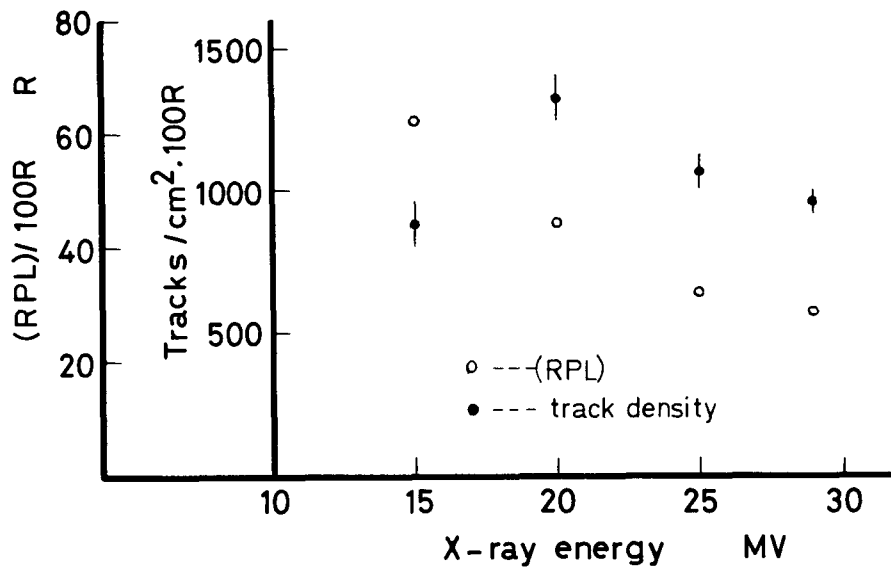


Fig. 4. Track density per (RPL) at maximum energies of 15 to 29 MV.

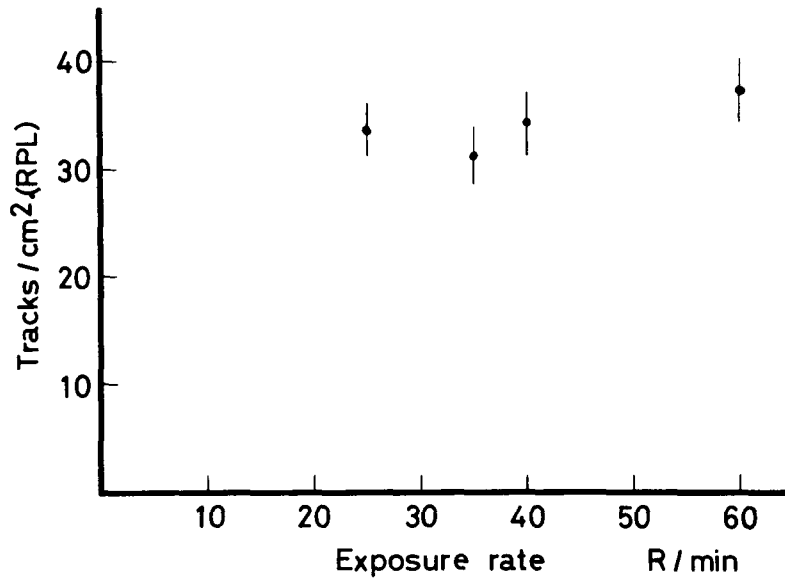


Fig. 5. Effect of exposure rate on track density at maximum energy of 29 MV.

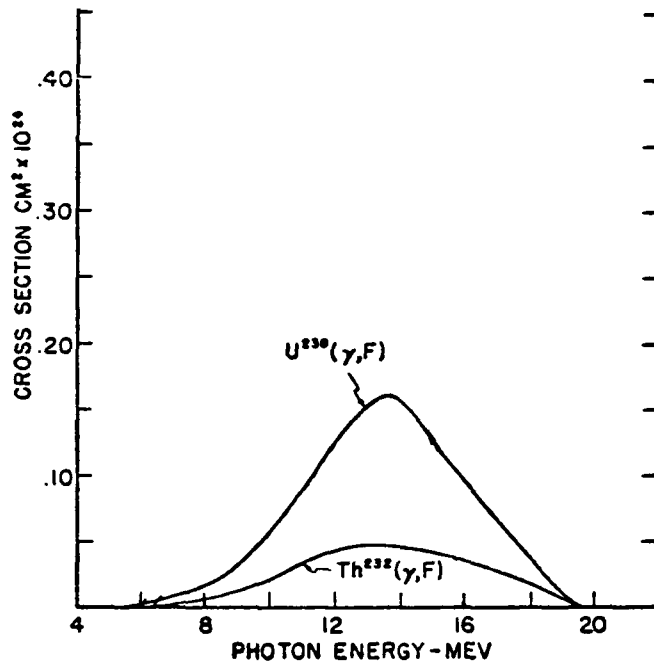


Fig. 6. Photo fission cross section of uranium and thorium.

AN INEXPENSIVE LIGHTWEIGHT ENVIRONMENTAL SURVEY INSTRUMENT

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Abstract

The operation of power producing nuclear reactors in the U.K. involves a statutory requirement to measure radiation levels in the area surrounding each nuclear site. The natural radiation background is measured at specified locations prior to operation of the reactor and afterwards routinely every three months. A survey instrument used for such measurements must therefore measure exposure levels from background (typically a few $\mu\text{R}\cdot\text{h}^{-1}$ in the U.K.) upwards. Other requirements for the instrument are that it should be very portable, be capable of being read accurately at low exposure rates, and should maintain its calibration.

The instrument described in this report weighs only 10 lbs, has a good energy response, $\pm 15\%$ over the range 50 keV to 6 MeV, and measures exposure levels from a few $\mu\text{R}\cdot\text{h}^{-1}$ to 100 $\text{mR}\cdot\text{h}^{-1}$. Ambiguities resulting from reading a fluctuating meter needle at low exposure rates are overcome by integrating the count over a preselected time, digital readout being provided. The report describes a comprehensive evaluation of the instrument and its comparison with other commercially used instruments. The instrument price is considerably cheaper than that of comparable instruments.

Introduction

Measurement of gamma radiation in the environment, natural prior to the operation of a power reactor and afterwards natural plus that due to the reactor, requires an instrument that records levels from a few $\mu\text{R}\cdot\text{h}^{-1}$ upwards. As the spectral content of this radiation may be unknown the exposure rate response of the instrument should be as flat as possible over a wide energy range, say 30 keV to 7 MeV.

Portable survey instruments used by the C.E.G.B. for this work indicate the measured exposure rate by means of a moving coil meter display.¹ The reading of such meters at low exposure rates involves problems of interpretation where a needle is fluctuating over a significant range of readings.

Recent advances in electronics have made it possible to produce small portable scalers with digital readout. In addition energy compensating filters are now commonly used to improve the poor energy characteristics of G.M. counters. To keep the total cost low, the instrument described in this report makes use of a commercially available portable scaler, G.M. circuitry and energy compensated G.M. counters.

Instrument Description

Four energy compensated G.M. counters, 20th Century type B6T's, are mounted radially on a tripod (Fig.1) to give the best directional response and to enable measurements to be made at 1 metre above ground. A short co-axial cable connects the detectors to the input of the electronics.

The electronics consist of the amplifier and E.H.T. unit of a Mini Monitor Mark V, and a Mini Instruments portable scaler type MS6.10. Both these units are housed in a small metal case, 20 cm x 14 cm x 12½ cm. Although the instrument is intended for outdoor use no special seals were fitted to the prototype to make it waterproof. During field trials a large polythene bag was slipped over the detectors and electronics and the measurements on Trawsfynydd Lake during a wet day and on rough water showed this to be adequate protection. Power supplies to the G.M. amplifier and E.H.T. unit are from two 9 volt batteries type PP6. The scaler unit is powered by four 6 ampere-hour Nickel-Cadmium rechargeable batteries and will run up to 10 hours continuously on a single charge.

Each G.M. counter was connected separately and its operating voltage plateau determined. The plateau was also measured with all four counters connected to verify that the same voltage range was obtained and the operating voltage for the counters was set at 680 volts.

Evaluation of Instrument

Laboratory Tests

Energy Response

The photon energy spectrum of a measured radiation field is frequently unknown and therefore an environmental survey instrument is required to have a 'flat' response over a wide energy range. Many reactors produce significant amounts of 6 MeV gamma radiation, and in gas cooled reactors this arises from the ^{16}O excited state which is formed by the fast neutron capture in ^{16}O of the CO_2 cooling gas and the subsequent beta decay of ^{16}N to $^{16}\text{O}^*$.

Tests were made at 29, 47, 59, 85, 107, 147, 183 and 210 keV using an improved low exposure-rate, filtered, X-ray series whose spectra have resolutions of 20%.

Radionuclide sources were used to measure the response above 200 keV up to 1.33 MeV.

The 335 keV resonance of the $^{19}\text{F}(p,\alpha)^{16}\text{O}$ reaction was used to determine the 6 MeV response, the radiation field being standardised by associated particle counting of the alpha particles with cross-checks by ionization measurements.

Fig.2 shows the energy response obtained. The response expressed as instrument reading divided by standardised exposure-rate is plotted against photon energy and has been normalised at 0.8 MeV.

Linearity of Response

Linearity was tested with standard ^{226}Ra sources and ^{60}Co sources. Results are given in Fig.3 and show that the instrument has a linear response up to 10 mR.h^{-1} . Although the response is non-linear above this exposure rate no fall-back effects are observed until about 4 R.h^{-1} and at approximately 100 R.h^{-1} the reading has reduced by 20% compared to that at 4 R.h^{-1} . The

meter reading, however, still remains greater than full scale for exposure rates up to 100 R.h^{-1} . The apparent non-linearity below $20 \text{ } \mu\text{R.h}^{-1}$ is discussed later on.

Temperature Tests

As the instrument will be used outdoors throughout the year it is important that its response should not change significantly with expected variations in temperature.

The instrument was placed in an environmental cabinet and irradiated in a constant field from a ^{60}Co source, the temperature was varied over the range -20°C to $+50^{\circ}\text{C}$. Before commencing these tests new 9 volt batteries were fitted and the Ni-Cd batteries were re-charged. The readings remained constant within $109.0 \text{ cps} \pm 7 \text{ cps}$ over this temperature range except at $+50^{\circ}\text{C}$ where a reading of 136 cps was obtained.

Variations in Instrument Readings with Supply Voltage

Tests were made to measure the variation in instrument reading with change in supply voltage for both battery supplies, the instrument being irradiated in a constant field. The lower limit markings for both battery tests were perfectly adequate.

Field Tests

Measurements were made at normal 'district survey' locations at two nuclear power stations.² Exposure rates were measured with the instrument and with other commonly used low exposure-rate instruments. The other instruments used in the comparison were the BNL 1 which has a plastic phosphor detector and a bottom range of $0-30 \text{ } \mu\text{R.h}^{-1}$, and the A.E.R.E. type 1368A which has 4 G.M. counters, 3 used in parallel for the lower ranges, and a bottom range of $0-50 \text{ } \mu\text{R.h}^{-1}$. For two of the locations measurements were also made with the Nuclear Enterprises N2601, which has an energy compensated G.M. counter and a single log range of $0 - 10 \text{ mR.h}^{-1}$, and with the General Radiological 1597A which uses a NaI detector and has a bottom range of $0 - 30 \text{ } \mu\text{R.h}^{-1}$. All the instruments had been previously calibrated against ^{226}Ra sources and the readings taken were all corrected for any non-linearities.

The first measurements were made at Trawsfynydd Lake, close to the Nuclear Power Station which was not operating at the time. Four sets of measurements were made above water depths between 20 to 30 feet and the following averaged results were obtained. Prototype background monitor $4 \text{ } \mu\text{R.h}^{-1}$, BNL 1 instrument $5 \text{ } \mu\text{R.h}^{-1}$ and 1368A instrument $7.5 \text{ } \mu\text{R.h}^{-1}$.

A second series of measurements were made at another station, with the reactors on load. Two different one mile locations were selected at which the results in Table 1 were obtained. Each reading for the meter display instruments was obtained by observing the needle for approximately one minute and taking the average reading, the figures in brackets show the range of fluctuations of the instantaneous reading.

Table 1

Instrument Type	Corrected Instrument Reading for 1 mile locations in $\mu\text{R.h}^{-1}$	
	Location 1	Location 2
Prototype background monitor	11	10.2
BNL 1	9(7 to 11.3)	8(6.5 to 10.2)
1368A	11.5(10 to 12)	11(10 to 11.7)
1597A	13(10.7 to 15.3)	10(8.5 to 11.5)
2601	12(9.5 to 17.3)	11.5(6.5 to 19.5)

The following conclusions have been made from these comparisons. Accurate assessment of very low exposure rate levels is difficult and differences of only $1 \mu\text{R.h}^{-1}$ between different instruments must be considered good. Measurements on Trawsfynydd Lake were made at lower exposure rate levels than the background radiation level of the room used for instrument calibration and corrections for non-linearity have therefore been obtained from extrapolation. No standard instrument will measure levels in the $\mu\text{R.h}^{-1}$ region and hence no accurate measurement can be made of the calibration room background level. The assumed value of $6.5 \mu\text{R.h}^{-1}$ is an averaged extrapolated result of measurements made in this room with a large number of different instruments of several types. Uncertainty in the absolute value of the room background may contribute to non-linearity of the prototype below $20 \mu\text{R.h}^{-1}$ (Fig. 3).

For the Lake measurements the radiation field is principally due to cosmic radiation with a small contribution from any radioactive content in the Lake. Levels of about $3.5 \mu\text{R.h}^{-1}$ are normally reported for cosmic radiation and so the readings of $4 \mu\text{R.h}^{-1}$ for the prototype and $5 \mu\text{R.h}^{-1}$ for the BNL 1 instrument appear realistic values.³ The reading of the 1368A is just over double the cosmic radiation level and may be due in part to the built-in radioactive content of the detectors. This built-in activity would be allowed for in setting the instrument up at $6.5 \mu\text{R.h}^{-1}$ but would cause it to read high if it were placed in a lower level radiation field.

Looking at the results for the other station it is interesting to see the large variations in readings obtained. For the one mile locations the BNL 1 instrument gave lower results than for all the other types. The 2601 instrument readings were approximately $1 \mu\text{R.h}^{-1}$ higher than the prototype results, this good agreement is not too surprising since the 2601 uses a single G.M. tube which is identical to the 4 detectors used in the prototype. The large fluctuations in the 2601 readings are due to the poor statistics obtained by using a single small detector. At these 1 mile locations the prototype and the 1368A are in good agreement and apart from location 1, so is the 1597A.

Conclusions

The tests on the prototype environmental survey monitor have shown that it compares favourably with other commercial low-level instruments whilst not having the interpretation problems associated with a meter display. The instrument has a good energy response ($\pm 15\%$ for 34 keV to 6 MeV), is simple and easy to use, and will cost approximately £280 (\sim \$670).

The environmental monitor described is only a prototype and a number of changes will be introduced in the operational instrument, for example the

controls will be simplified and the meter markings altered to $\mu\text{R.h}^{-1}$.

Many of the differences in readings observed when using different types of instruments arise from the problems of calibrating instruments at a few $\mu\text{R.h}^{-1}$ and further investigations are required on the calibration techniques and standardisation at these radiation levels.

Acknowledgements

It is a pleasure to acknowledge the helpful assistance of Dr. P. W. Roberts of Mini Instruments Limited who constructed the initial instrumentation. This paper is published by permission of the Central Electricity Generating Board.

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Fig.1 Lightweight Environmental Survey Meter

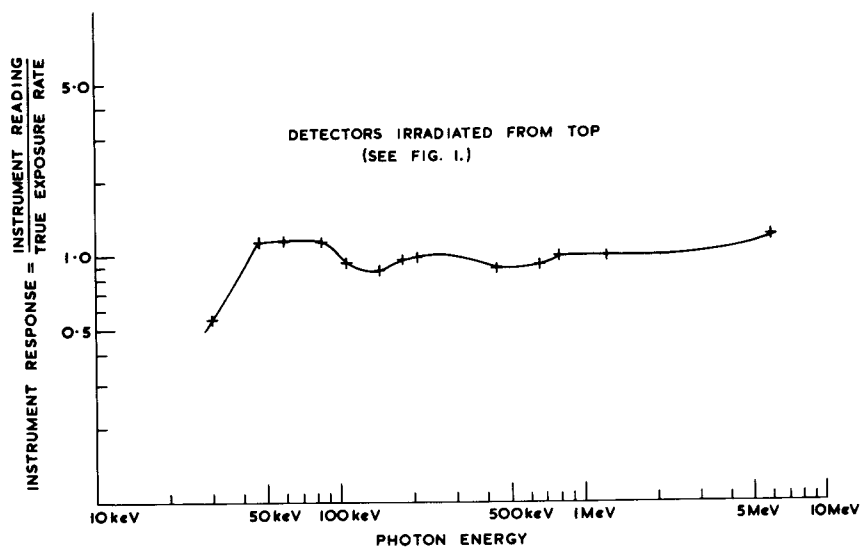


FIG. 2. PHOTON ENERGY RESPONSE.

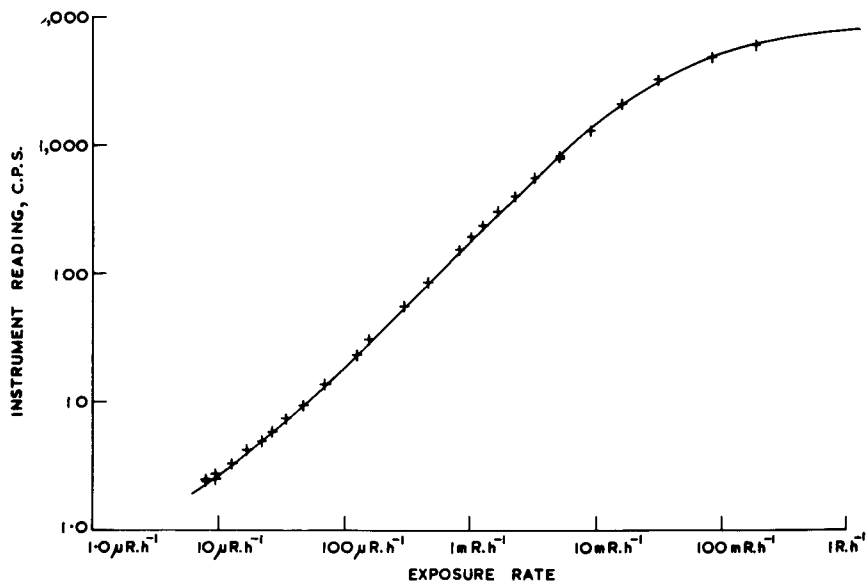


FIG. 3. LINEARITY OF BACKGROUND MONITOR.

A NEW TECHNIQUE IN
ENVIRONMENTAL NEUTRON SPECTROSCOPY

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Abstract

A new phoswich¹ has been developed that permits the measurement of the environmental neutron spectrum using Bonner spheres. The new phoswich consists of an 8 mm diameter, 8 mm long ⁶LiI(Eu) crystal surrounded by plastic scintillator. The use of the fast signals from the plastic scintillator in anticoincidence with the slow signals from the ⁶LiI(Eu) gives a nearly background free ⁶Li(n,α) ³He peak. Thus, the signal to noise ratio in the vicinity of the peak is very high (20-35 to 1). An adequate number of counts is obtainable in one day for most of the spheres.

Typical counting rates for polyethylene spheres of the diameters given below are:

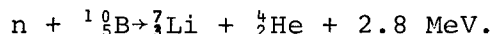
2 inch	2.69 c/hr
3 inch	13.9 c/hr
5 inch	14.0 c/hr
6.87 inch	12.4 c/hr
8 inch	9.07 c/hr
10 inch	6.91 c/hr
12 inch	5.96 c/hr
18 inch	3.77 c/hr

Details of circuitry, phoswich geometry, pulse height analyzer spectra and synthesized spectra will be given.

This paper is an elaboration of the preliminary results presented at an earlier time.²

The measurement of the neutron energy spectrum of environmental neutrons is besieged by difficulties caused by the gamma rays, muons, protons and other charged particles which are simultaneously present.

The commonly used neutron detection technique which is also applicable to environmental neutron studies is that of a ¹⁰B_{F₃} counter moderated with a hydrogenous material such as polyethylene or paraffin. The use of various thicknesses of moderator produces a family of different neutron detection efficiencies which are functions of the incident neutron energies. These responses have been studied by some authors,³ mostly in a direction perpendicular to the axis of the BF₃ counter. Responses versus energy for other polar angles have not been studied in detail to the best of our knowledge. As it is to be expected, there is a significant polar angle dependence in these counters. The popularity of the BF₃ counter is due to the ease with which charged particle events (either gamma ejected electrons or cosmic ray charged particles) are separated from neutron capture events by the exoergic reaction



The separation is made electronically with a simple integral discriminator. The other environmental events deposit typically a few KeV.

Another type of neutron detector is the Bonner Sphere.⁴ This is a spherical hydrogenous (polyethylene) moderator with a point like thermal neutron detector at its center. Hence, to a first approximation it has isotropically uniform energy responses. The energy dependence of its detection efficiency has been rather well studied.⁵ The most common point like detector is a ⁶Li(Eu) crystal. Here in

diameter spheres, 10 inches or greater, but it is questionable if it is meaningful in smaller ones. A second way to reject the charged particle background is to use a phoswich.

The phoswich used in the experiment described in this paper is shown in Figure 3. The pulses from the ${}^6\text{LiI}(\text{Eu})$ have a long decay time (1.4 μsec). The pulses from the plastic scintillator have a very short decay time (2-3 nsec). Due to the monochromaticity of the slow pulses, a very simple passive network was tried. This circuit permitted the separation of the slow and fast components after suitable passive shaping. Figure 4 shows a simplified diagram of the electronics used.

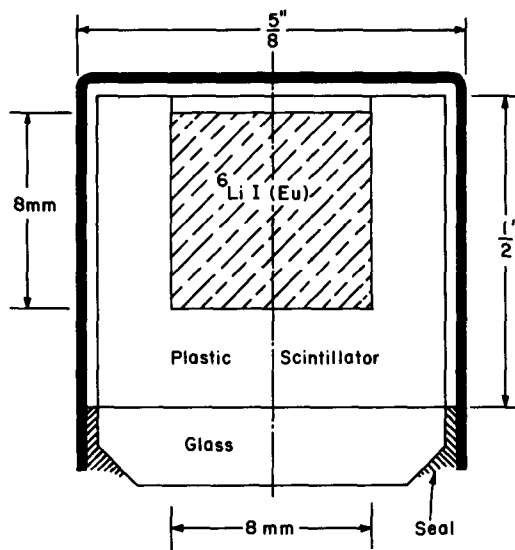


Figure 3. Cross-section of an 8 mm x 8 mm phoswich.

The values first tried and not necessarily optimum are

$$L = 102 \mu\text{H}, C = 33\text{pF}.$$

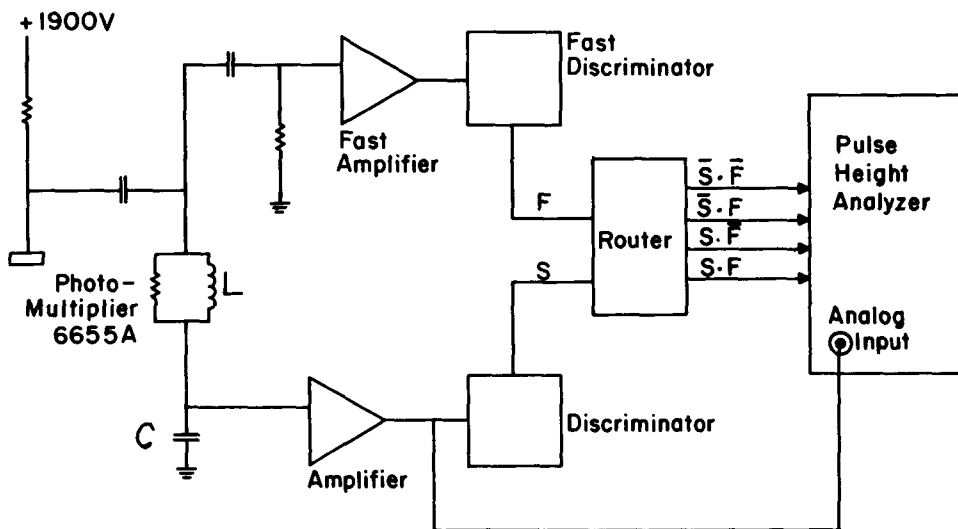
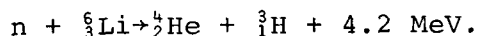


Figure 4. Electronics block diagram.

principle at least, the background charged particles are separated from the neutron capture events by choosing the size of the crystal such that most charged particles traversing it lose less energy than the 4.2 MeV of the reaction



Then again, the two types of events should be simply separable by an integral discriminator. In practice when measuring the extremely low environmental neutron flux the separation between neutron captures and other events is not good.

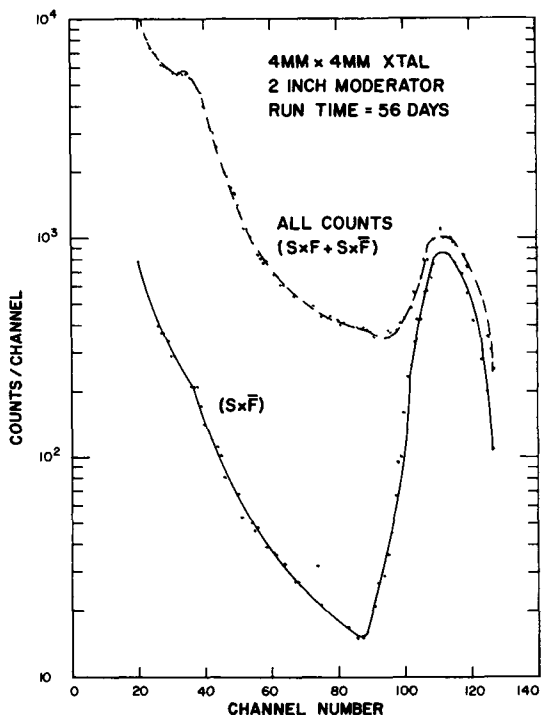


Figure 1. Upper curve is spectrum of all slow pulses. Lower curve is spectrum of slow pulses not accompanied by fast pulses, e.g., "neutrons".

Figure 1, upper curve, is a spectrum of pulses from a 4 mm x 4 mm ${}^6\text{LiI}(\text{Eu})$ crystal in the center of a 2 inch polyethylene spherical moderator while recording environmental neutrons for 56 days. The signal to noise ratio (peak to valley) is not as good as that obtainable while calibrating the system with a PuBe neutron source as shown in Figure 2, for two sizes of crystals.

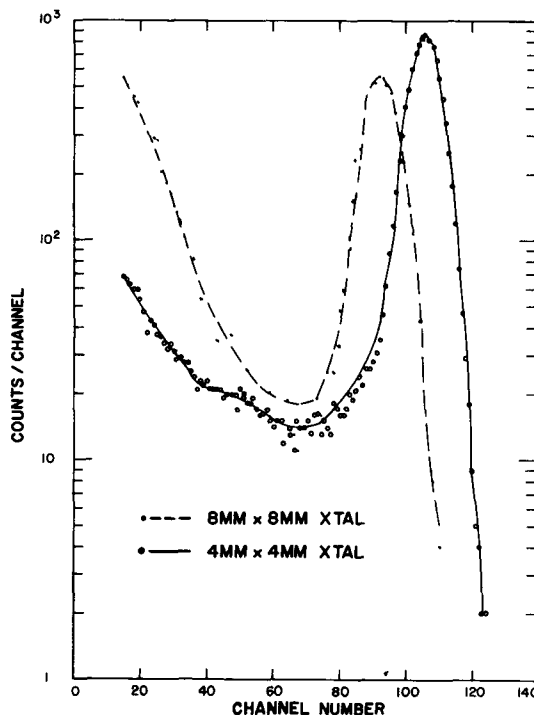


Figure 2. Spectra due to PuBe neutrons from two size ${}^6\text{LiI}(\text{Eu})$ crystals.

There are two ways to improve the rejection of unwanted counts. One is to use small proportional counters.⁶ This method almost works for larger

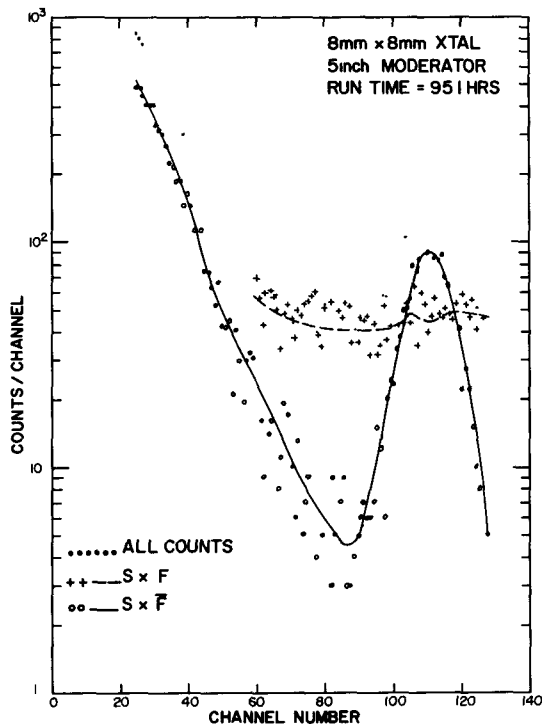


Figure 5. Pulse height spectra from an 8 mm x 8 mm phoswich in a 5 inch polyethylene spherical moderator exposed to environmental radiation for 95.1 hours.

Figure 5 shows the results of an environmental neutron measurement using an 8 mm x 8 mm ⁶LiI crystal in a 5 inch moderator. This run is 95.1 hours long. The solid curve shows the spectrum of slow pulses not accompanied by fast pulses ($S \times \bar{F}$). These event signatures are likely to be due to neutrons. The dashed curve shows the spectrum of slow pulses accompanied by fast pulses ($S \times F$).

These events are very likely "noise" or charged particles from the environment. The dotted curve shows all slow pulses ($S \times \bar{F} + S \times F$). The peak-to-valley ratio for the ($S \times \bar{F} + S \times F$) spectrum is about 2.8, while for the "neutron" pulse only spectrum ($S \times \bar{F}$) it is about 22. Therefore, the phoswich has improved the peak-to-valley ratios for neutron detection in environmental measurements by a factor of about five to six.

Figure 1, lower curve, shows the result of using the 4 mm x 4 mm crystal in a phoswich arrangement with background rejection. Another example of the improvement that is obtainable using this technique may be seen from the curve in Figure 5. Consider the neutron data as being in channels 90 to 125. Then we could derive two figures for the number of "neutron events". Case A, use the curve for "all" events. This gives (Signal + Noise) - Noise $\sim 3157 - 1440 = 1717$ counts + 68. Case B, use the curve for ($\bar{S} \times F$) type events. The corresponding difference is $1480 - 52 = 1438 + 39$. The fact that two standard deviations do not produce an overlap of the data is a measure of the errors committed in background subtraction in case A.

The dashed line also shows some feed through of neutron events. Hence, improvement in the rejection of the ($S \times F$) events is still possible.

The signal-to-noise ratio is now good enough that a set of Bonner spheres may be used for environmental neutron measure-

ments using a simple integral discriminator for noise rejection. Counting periods of about two days per sphere would be adequate.

Environmental Neutron Flux Measurement

A series of measurements was made inside a light frame house at the NAL site which is at an elevation of 740 feet above mean sea level. The corresponding count rates per hour are given below in Table I.

Table I

Detector Size	Counts Day	Detector Size	Counts Day
2 inch	64	8 inch	218
3 inch	334	10 inch	165
5 inch	336	12 inch	143
6.87 inch	297	18 inch	90

To interpret these results, the spectrum was unfolded using an iterative process.⁷

The absolute detection efficiency of the actual ⁶LiI(Eu) crystal used was calculated by measuring the neutron flux from a ²³⁸PuBe which had been calibrated by the National Bureau of Standards. The correction factor to the data in HASL-267, thus found, was 1.8₈.

Using this value, the cosmic ray neutron flux was calculated as well as the dose and dose equivalent per neutron/cm². These quantities are

cosmic ray
neutron flux = 63 n cm⁻² hr⁻¹

flux-to-dose
conversion
factor = 4.9 x 10⁻⁹ rad n⁻¹ cm²

flux-to-D. E. conversion
factor = 2.7 x 10⁻⁸ rem n⁻¹ cm²

Conclusions

A new technique has been developed which permits rapid low resolution environmental neutron spectroscopy using simple electronics.

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CONDUCTIMETRIC PROPERTIES OF SOME SOLID TRACK DETECTORS
AND HYPOTHESIS ON THE TRACK FORMATION MECHANISM
IN THESE IRRADIATED AND ETCHED DETECTORS

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Abstrat

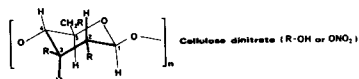
The purpose of this work is to study the conductivity variations of solid track detectors with ester function after etching and irradiation. The different conductimetric behaviour of these detectors allows to assume the chemical etching mechanism. This hypothesis seems to be confirmed by IR and RMN studies.

All the papers about latent track formation in solid track detectors point out the following condition : detectors must have electrical resistivity above 2 000 Ω /cm to record any track (1). So it is interesting to study the conductimetric properties of these insulators and especially the variation of their resistivity as a function of irradiation dose, etching time and concentration of etching reagent. For this purpose, we have studied cellulosic esters and polycarbonate films.

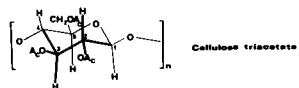
CONDUCTIMETRIC MEASUREMENTS

Experimental procedure

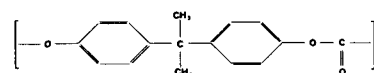
First of all, we studied a 40 μ m thick film of cellulose dinitrate. Analysis gave a nitrogen quantity of 12 % and a camphre quantity (plasticizer) of about .0 %. This material had been provided by Kodak Pathé Society (France).



Then, we used a 40 μ m thick film of cellulose triacetate without any plasticizer, purchased from Bayer chimie (France) (Triafol TN).



At last, Makrofol film (44' dioxidi-phenyl 22 propane polycarbonate) was chosen.



4,4' dioxyl diphenyl 2,2 propane polycarbonate

These samples were irradiated with the fission fragments from ²⁵²Cf (10⁶ fissions/cm²/hours). α particles from ²⁴¹Am were also used after being reduced to an energy in the range 0 to 2.27 Mev for a better recording.

Etching reagents were aqueous sodium hydroxyde solutions of 5N, 6,5N and 8N in a constant temperature bath (60° C).

Then etched samples were carefully washed by distilled water. At last they were kept in vacuum. No detectable variation in conductivity measurement of a particular sample was observed after a sufficient drying time (about 10 days). The electrical conductivity was measured with Wayne Kerr impedance bridge.

The measurement cell (fig. n°1) had been made in our laboratory. The electrodes were held by a insulating material in coaxial position inside two brass tubes, one of which was free of movement. This device was mounted in a rigid frame. The movable electrode was connected with the other by the mean of a screw. A torque wrench enabled a constant pression (45 Kg/cm²) on the electrodes.

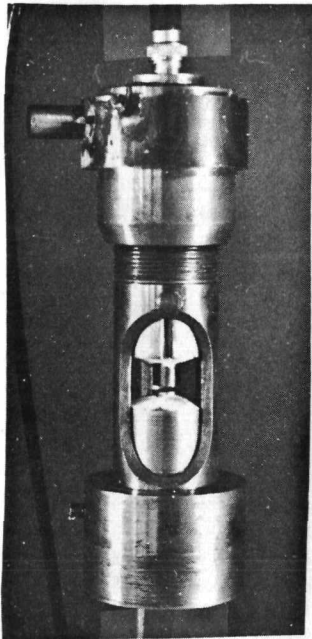


fig. 1

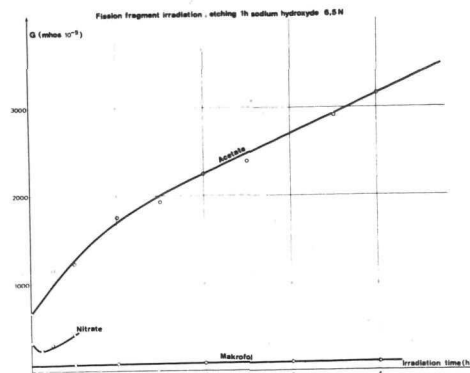


fig. 2

The following values were found :
 - cellulose acetate : 700 mhos 10^{-9}
 - cellulose nitrate : 310 mhos "
 - Polycarbonate : 60 mhos 10^{-9}

RESULTS AND INTERPRETATION

It is well known that ionizing particles produce breaking points in the macromolecular chain of organic polymer detectors.

The experiments were initiated in an effort to display clearly the existence of these defects by a conductimetric method. No conductimetric difference appeared when samples were irradiated but not etched even for large irradiation doses.

The etching process of tracks, making channels more or less wide, more or less deep and so substituting bulk material by air should have decreased the conductivity of the film (because air is a better insulator than cellulosic esters).

Besides, samples measured in vacuum showed a conductivity lower than in atmospheric conditions.

In fact, we observed that conductivity was increasing more or less with irradiating time, this leads to the idea that another phenomenon may be invoked, this phenomenon would increase the heterogeneity of material and consequently its conductivity. Fig. n° 2 summarises the results obtained with three detectors etched with sodium hydroxide 6,5 N up to one hour. First of all we notice the different conductivity of these three esters etched but non irradiated.

It also appears on fig. n°2 that cellulose acetate and Makrofol show a conductivity augmentation represented by a monotonous increasing function, but this variation is very different according to the detector. In the case of Makrofol indeed, it is nearly negligible.

For cellulose nitrate, the sample destruction etched one hour after 45 minutes irradiation constrained us to reduce the etching time to 35 minutes (fig. n° 3); so we obtained for this compound a linear increasing function of the irradiation time (for irradiation times longer than 30 minutes).

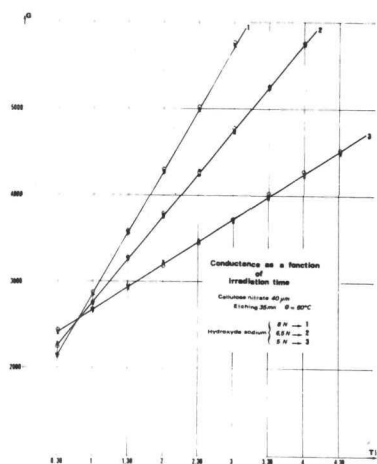


fig. 3

These conductivity augmentations can be naturally used to know the irradiation dose ; in a recent work, (2, 3) we perfected a method based on this observation in order to evaluate the track density. It is of interest to compare the behaviour of the three compounds (i.e. cellulose acetate, cellulose nitrate and polycarbonate) at short irradiation times (below 30 minutes) , the conductivity of cellulose nitrate (and only this polymer) is decreasing with irradiation dose. It is assumed that a different etching mechanism may be invoked for this detector ; this mechanism will be drawn later. Fig. n° 3 also shows the conductivity for a same irradiation time and a same etching time as a function of etching agent concentration. Of course, the augmentation of conductivity with concentration is greater, the irradiation dose is more important. In the case of irradiation by α particules, similar results have been obtained but the conductivity variations observed are not so important.

We tried to explain these conductivity variations.

It is very important to notice that conductivity increase is not due to sodium hydroxyde itself remaining in tracks because the samples were washed up as long as necessary to have no more change in conductivity measurement. Of course, the thickness of the film fell off after etching but the chemical attack was lasting the same time for a line of samples so that the thickness could be modified in the same manner at the etching step.

However, if track density is very important it is possible the channels become edge to edge ; so, the decrease of thickness would be a function of the irradiation dose. It seems likely this effect would take a part in the conductimetric decrease. But that is not enough to explain the results ; computation show indeed that the observed variations would be correlated with a thickness decrease greater than the undamaged film thickness itself. So, in all probability, another phenomenon would take place.

To be precise, we used a chemical reagent and we assume the conductimetric variation is the result of a chemical damage in polymer. It is therefore possible to invoke a saponifica-

tion reaction. In the case of non irradiated etched samples, the above reaction is limited to a thin layer at the surface of material. When irradiated, the chemical attack would occur deeply along the latent tracks that allow sodium hydroxyde diffusion. Then chemical change, especially formation of new hydroxyl groups should induce a conductivity enhancement.

It would seem this saponification reaction would be slightly efficient in the case of Makrofol. For cellulose nitrate, (4) sodium hydroxyde reacts in two ways : on one hand saponification (with a low rate), on the other hand oxydation degradation (with a fast rate) which would enlarge the tracks. So in case of short irradiation times, oxydation degradation would play a leading part. Perhaps that would explain the specific conductivity decreasing above mentioned for this polymer, because the presence of air, a better insulator, in tracks. When track density was important, reagent diffusion in bulk material became easier, so that hydroxyl group number would be sufficient to increase conductivity.

Furthermore oxydation degradation would be liable for nitrate sample destruction by 45 minutes etching whereas acetate film can suffer that etching time without being broken up.

CHEMICAL TRACK FORMATION MECHANISM AFTER ETCHING

The purpose of the following section is to test the above mentioned hypothesis, drawn from observations of conductimetric properties of samples exposed to different irradiation doses and then etched.

First, in order to prove the existence of oxydative degradation reaction, we changed etching condition. For cellulose nitrate we used a sodium hydroxyde solution in reducing condition (saturated by sodium nitrate). As a result (fig. n° 4) a general conductivity decrease was observed for each irradiation dose. This fact could be explain because hydroxyde sodium diffusion was more difficult, oxydative degradation reaction would not be so important, so film thickness is not so

reduced. The inverse phenomenon that is to say general conductivity increase was observed for an nitrate sodium oxydative bath.

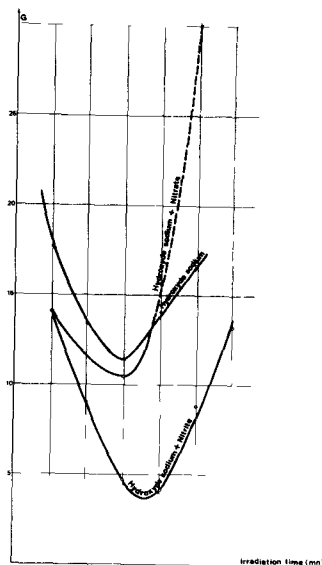


fig. 4

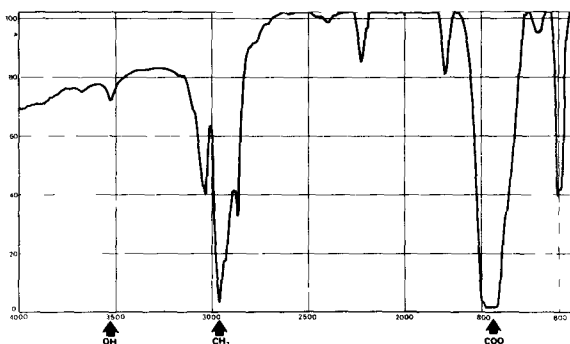


fig. 6

On the other hand, we tried to point out the important enhancement of saponification reaction with the irradiation dose and for a constant etching time. Two powerful tools, i.e. NMR and IR spectroscopies were used for such investigations.

First IR spectra of cellulose acetate are shown on fig. n° 5. It can be seen that the peak corresponding to OH groups (3500 cm^{-1}) is clearly increased.

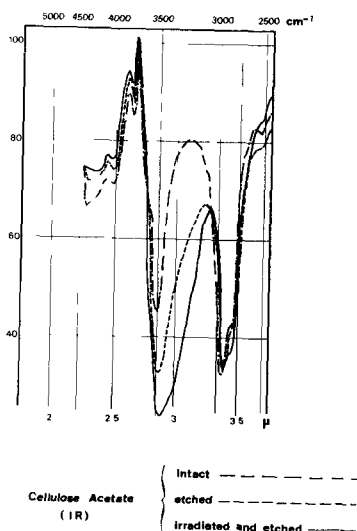


fig. 5

NMR studies confirmed the above observations : unfortunately, solubility problems appeared in the case of cellulose acetate samples. If intact films are soluble in chloroform and methylene chloride (NMR study of this polymer has been done by Gagnaire and Vincendon (5, 6) etched especially when irradiated films are only partly soluble in any solvent. This is an additional argument to support the hypothesis of the important chemical change due to irradiation and etching. One could suggest the following explanation : the result of the saponification is, in fact, to decrease the degree of acetylation. This reaction is only effective in irradiation damaged zones; consequently polymer solubility is affected.

Cellulose nitrate, probably due to the oxydation degradation process, is always soluble in acetone. NMR spectra of various cellulose nitrate films in deuterated acetone were recorded with a Varian HA 100 Spectrometer. On fig. 7, 8, 9, we can see spectra of intact film, of 35 minutes etched with 6,5N sodium hydroxyde film and 35 minutes etched film after irradiation. The

peak corresponding to oxydrile groups appears at $\delta = 3$ ppm (from TMS internal Standard) it must be noticed that any water trace would give a contribution to this peak. However it is possible to compare different film spectra because all the samples were dried in the same conditions. The area of this peak is slightly increased when the polymer film is etched but a considerable area enhancement is observed when irradiated and etched. At high field, it can be observed camphre peaks (plasticizer). Broad absorption lines in the range 3,5 to 6 p.p.m are connected with the seven cyclic protons.

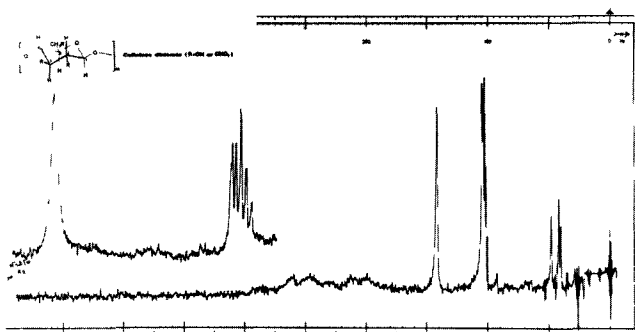


fig. 7

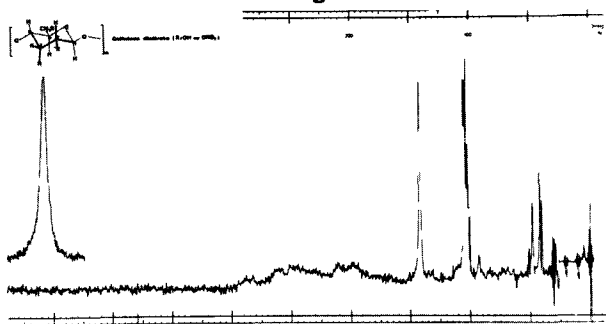


fig. 8

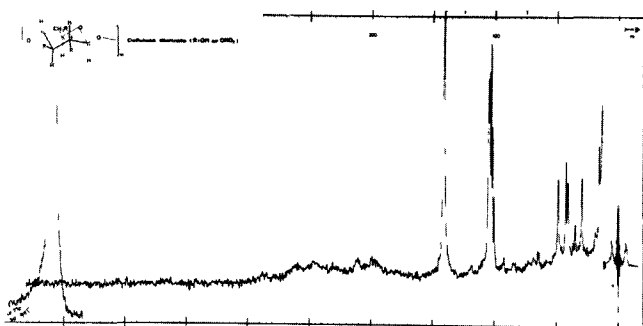


fig. 9

NMR Spectra of Makrofol films in deuterated chloroform were recorded with a 250 MHz Spectrometer from Cameca (Thomp-

son C.S.F.) This high field Spectrometer was needed in order to detect any slight variation that could have occurred. On fig. n° 10, a Makrofol film spectrum is shown. A typical para substituted aromatic signal is founded at about 7 p.p.m (8 phenyl protons). The methyl groups (6 protons) appear at $\delta = 1,35$ p.p.m. A small peak at $\delta = 1,25$ p.p.m is attributed to hydroxyl groups (probably the ends of macromolecular chains). This observation fit with the conclusions drawn from IR Spectra. The irradiation and etching effect is not clear because it is difficult to evaluate the hydroxyl peak importance at $\delta=1,25$ ppm which is closed by methyl group peak. Nevertheless, in order to obtain a better information we used TFA (trifluoroacetic acid) to shift out the OH group. We calculated the ratio of peak areas at 7 p.p.m and 1,3 p.p.m with and without TFA for each film (intact, etched, and irradiated etched). For instance, for the intact film the above ratio is 1,45 before TFA addition and 1,45 after.

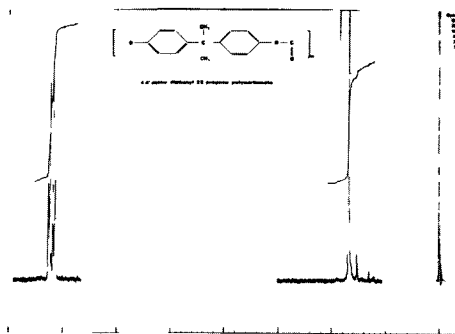


fig. 10

In the case of etched sample as irradiated etched samples the ratio is equal to 1,25 before TFA addition and 1,45 after TFA addition (fig. 11 and 12) (it can be observed on fig. 12 the shifted OH TFA peak at about $\delta = 10$ p.p.m) So it appears there is a weak augmentation of hydroxyl peak area for an etched sample compared with an intact film ; increase is no more important in the case of an irradiated etched sample. That is in good agreement with IR Spectra for Makrofol. In fact, it would seem likely that reagent diffusion is allowed even in non irradiated sample by flexibility of molecules, so that irradiation would not matter a good deal for saponification reaction.

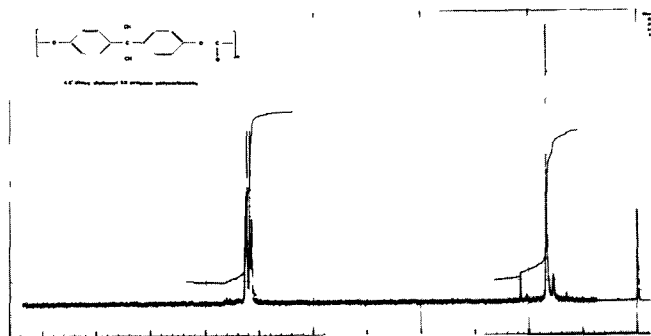


fig. 11

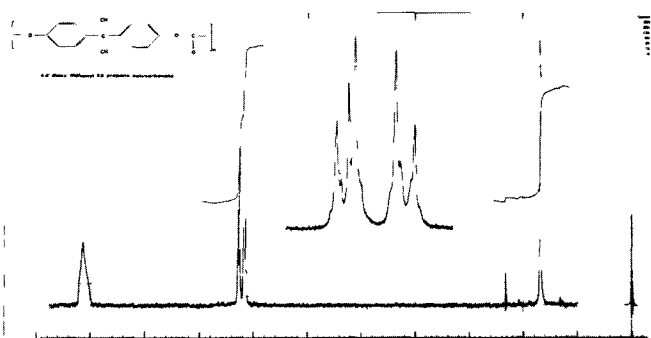


fig. 12

In conclusion, we have noticed correlation between conductimetric properties and saponification reaction for all studied detectors. That reaction is easier for cellulosic esters ; in the case of polycarbonate, it is more difficult.

At last it can be note that the sensitiveness of these detectors, characterized by the critical rate of energy loss $(\frac{dE}{dx})_c$, for the lightest recorded particle, rate which is 0,86 and 1,41 for nitrate and acetate (in the case of α particles) and which is 5,5 for polycarbonate (in the case of ^{16}O), was also correlated.

Thus we have been able to approach chemical track etching process by the mean of conductivity study i.e. by the mean of physical property study.

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We are grateful to Pr GAGNAIRE and his collaborators who gave us the opportunity to perform NMR experiments.

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