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Trans-Boundary Volcanic SO_2 Detected over Pakistan from Satellite Observations during the Time Period 2004–2012

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ABSTRACT

This study emphasizes on the amount of trans-boundary Sulfur dioxide (SO_2) present in the atmosphere of Pakistan as a consequence of various global volcanic eruptions, by using satellite data. The data products of SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY), Ozone Monitoring Instrument (OMI) and Global Ozone Monitoring Experiment-2 (GOME-2) were used for the time period of 2004–2012. SO_2 columns retrieved with Differential Optical Absorption Spectroscopy (DOAS) technique were used to perform both spatial and temporal analyses. The Nabro volcano eruption during 2011 had caused high SO_2 columns over East Africa, Middle East and South Asian regions. Daily satellite observations were used to study SO_2 plume pathway during this event. Other significant volcanic eruptions and their effects on atmospheric composition of Pakistan are also discussed. Back trajectory analysis is also performed to track the origin of air masses enriched with SO_2 column densities detected over Pakistan. Maximum SO_2 column densities of 9.4 Dobson Units (DU) were measured over Pakistan caused by Dalafilla volcanic eruption during November 2008.

Keywords: Trans-boundary; SO_2; Volcanic eruption; Satellite observations; Pakistan.

INTRODUCTION

Atmospheric sulfur dioxide (SO_2) results from both natural and anthropogenic sources (Kettle and Andreae, 2000; Halmer *et al.*, 2002; Vijay *et al.*, 2004; Dentener *et al.*, 2006; Lee *et al.*, 2008). Besides anthropogenic activities (fossil fuel combustion, metal smelting and biomass burning etc.), it is produced naturally from volcanic activity, oxidation in the soil, and the oxidation of hydrogen sulfide. Volcanic eruptions are sporadic and major contributor, emitting 7.5–13 Tg SO_2 per year (Andres and Kasgnoc, 1998; Halmer *et al.*, 2002). SO_2 life time varies from few days to several weeks in the troposphere due to its reaction with hydroxyl (OH) and other oxidizing agents (Finlayson-Pitts and Pitts, 2000; Platt and Stutz, 2004) by several homogeneous and heterogeneous processes. The final products are always sulfuric acid and sulfate particles, respectively. In the lower stratosphere lifetime of SO_2 exceeds from several weeks to 2 years (Eisinger and Burrows, 1998; Platt and Stutz, 2004). Tropospheric lifetime of SO_2 allows it to travel larger distances before it is washed-out or transformed into sulfuric acid and/or sulfate particles. Therefore, SO_2 can have impacts on air quality and regional climate.

Sulfur dioxide plays an important role in both tropospheric and stratospheric chemistry, and helps to determine the stratospheric aerosol loading (SPARC, 2006). In the stratosphere, sulfate aerosols provide sites for heterogeneous reactions which can convert chlorine from reservoir to radical forms (Solomon *et al.*, 1996). Sulfate aerosols can also affect air quality and climate by direct and indirect radiative effects (e.g., Graf *et al.*, 1997; Robock, 2000; Zhang *et al.*, 2007).

SO_2 is also one of the criteria pollutants (e.g., Aneja *et al.*, 2001) and a key precursor of acid rain; hazardous to forests and many fresh water ecosystems around the world (Ferrari and Salisbury, 1999).

SO_2 monitoring stations are sparse and monitoring on a global scale through these stations is not possible. Since last few decades, through satellite observations and the advancement of technology, now the atmospheric SO_2 monitoring on large scales has become possible (e.g., Krueger *et al.*, 1995; Eisinger and Burrows, 1998; Khokhar *et al.*, 2005; Carn *et al.*, 2008) with good temporal and spatial coverage. Space borne instruments, for example the Total Ozone Mapping Spectrometer (TOMS), since 1978 in space providing long-term SO_2 record (Krueger *et al.*, 1995), the Global Ozone Monitoring Experiment (GOME), 1996–2002 (Eisinger and Burrows, 1998; Khokhar *et al.*, 2005), SCanning Imaging Absorption spectroMeter for Atmospheric

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CHartographY (SCIAMACHY), 2002–2012 onboard ENVISAT-1 (Bovensmann *et al.*, 1999; Afe *et al.*, 2004; Lee *et al.*, 2008), Ozone Monitoring Instrument (OMI), since 2004-present (Krotkov *et al.*, 2006, 2008), GOME-2 since 2007-present (Callies *et al.*, 2000; Rix *et al*., 2012) in addition to hyperspectral Infrared Atmospheric Sounding Interferometer (IASI) launched in 2006 onboard MetOp-A (Clerbaux *et al.*, 2009) have opened new corridors by providing a continuous spatial and temporal analysis of various trace gas and green house gas monitoring around the globe and especially regions with limited or no alternate monitoring facilities.

This study has first time focused on the amount of volcanic SO_2 present in the atmosphere of Pakistan. It emphasizes mainly on spatial and temporal variation in SO_2 column densities over Pakistan during the time period of 2004–2012 as a consequence of trans-boundary SO_2 pollution by using satellite observations. For this purpose SO_2 column densities level-2 data from three satellite instruments: SCIAMACHY, OMI and GOME-2 was used. Differential Optical Absorption Spectroscopy (DOAS) (Platt and Perner, 1979; Van Roozendael, 1999; Khokhar *et al.*, 2005) technique is used to retrieve SO_2 column densities from back scattered UV radiances measured by satellite based nadir-viewing instruments. Both daily and monthly images are used to track the special events of high SO_2 columns detected over Pakistan as consequence of volcanic activity in the neighbouring and remote regions.

Pakistan has no continuous monitoring facility for the monitoring of ambient air quality. Pakistan Environmental Protection Agency (EPA) and Pakistan Council of Scientific and Industrial Research (PCSIR) have the facility of 7 fixed and 3 mobile (Wagon/Trucks with air quality monitoring instruments onboard) air quality monitoring stations with intermittent and rare measurements in few cities of Pakistan. According to report from economic survey of Pakistan 2012–2013, all of the air quality monitoring stations are non functional since year 2010 (ESoP, 2013).

A recent study (Khattak, 2013) has indicated temporal increase of 70–120 per cent (statistically significant) in the measured SO_2 column densities during the time period of 2004–2012 by using SCIAMACHY observations. Such studies are mandatory and play vital role in providing data/information about air quality of a region that has little or no air quality monitoring facilities.

METHODOLOGY AND DATA SOURCES

Data

Level-2 data from SCIAMACHY, GOME-2 and OMI instruments was used to evaluate high SO_2 columns during the time period of 2004–2012. Data was downloaded from the Website (http://temis.nl/) of tropospheric emission monitoring internet service (TEMIS) project. All of these satellite instruments have various similar features like they orbit the earth in Sun synchronised orbits and SO_2 column densities are retrieved from UV back scattered radiances measured in nadir view and by same DOAS method. They also differ in various aspects of different overpass time, spatial and spectral resolutions as listed in Table 1. The SCIAMACHY (Bovensmann *et al.*, 1999; Afe *et al.*, 2004) a multi-channel grating spectrometer onboard Environmental Satellite -1 (ENVISAT-1) by European Space Agency's (ESA), was launched on 1 March, 2002. It is functioning since August 2002, with a resolution of 60 × 30 km^2. The OMI instrument was launched on Earth Observing System (EOS) on 15 July, 2004. It has a spatial resolution of 13 × 24 km^2 at nadir (while 13 km × 128 km for the extreme viewing angles at the edges of the swath) and it provides daily coverage on global basis (Krotkov *et al.*, 2006; Levelt *et al.*, 2006). The GOME-2 instrument was launched on October 2006. It has been onboard the Meteorological Operational satellite-A (MetOp-A), with a resolution of 80 × 40 km^2 (Rix *et al.*, 2009, 2012). Details about the retrieval algorithms used to retrieve SO_2 column densities used in this study are not presented here just to avoid the repetition as this has been described in various studies (e.g., Afe *et al.*, 2004; Khokhar, 2006; Krotkov *et al.*, 2006; Levelt *et al.*, 2006; Lee *et al.*, 2009; Yang *et al.*, 2010; Rix *et al.*, 2012; Fioletov *et al.*, 2013, Theys *et al.*, 2013).

SO_2 Retrieval Analysis

The SO_2 slant column density are retrieved in the spectral window of 315–326 nm by using Differential Optical Absorption Spectroscopy (DOAS) technique (Platt and Perner, 1979; Platt *et al.*, 1994; Richter *et al.*, 1998a, b; Khokhar *et al.*, 2005; Wagner *et al.*, 2008).

The retrieval of the SO_2 slant column is affected largely by strong absorption by ozone (O_3), therefore, both SO_2 and O_3 should be taken into account in the retrieval process to avoid/minimize spectral interference. This "interference"

Table 1. Features of the three satellite instruments used in this study.

Features of the three satellite instruments used in this study			
Instrument	SCIAMACHY	OMI	GOME-2
Platform	ENVISAT-1	AURA	MetOp
Measurement Period	Oct. 2002–March 2012	2004–Present	2006–Present
Equator Crossing time*	10:00 am	01:45 pm	09:30 am
Nadir resolution (km^2)	60 × 30	24 × 13[1]	80 × 40
Spectral region	UV-Vis-NIR-SWIR	UV-Vis[2]	UV-Vis
Global Coverage	6 days	~1 day	1.5 days

* local time.

[1] at nadir and 13 km ×128 km for the extreme viewing angles at the edges of the swath.

[2] OMI covers only wavelengths < 500 nm.

when concentrations of SO_2 are low, may produce negative SO_2 slant column values, with an error of the same magnitude. This then represents the SO_2 background level, i.e., the apparent SO_2 absorption in the absence of emissions of SO_2. This is further dependent on solar zenith angle (SZA) and more prominent for higher SZA due to increased slant light path and larger horizontal ozone concentrations. For low SZA, i.e., at low and mid latitudes, the interference between SO_2 and O_3 absorption results in less negative SCD (low back ground level) and errors. Therefore, emissions of SO_2 (by pollution or volcano eruptions) can be detected against this background. This effect is corrected by applying background correction method used by various studies (e.g., Khokhar *et al.*, 2005; Lee *et al.*, 2009)

An air mass factor (AMF) is mandatory for the conversion of the retrieved slant column- integrated along the light path, into a vertical column densities (VCD- generally expressed in Dobson unit – 1 DU = 2.69×10^{16} molecules/cm^2). The SO_2 AMF depends strongly on surface albedo, clouds, aerosols optical properties, the vertical distribution of SO_2, and the ozone column (Khokhar *et al.*, 2005; Thomas *et al.*, 2005; Khokhar *et al.*, 2008; Krotkov *et al.*, 2008; Lee *et al.*, 2009).

In case of volcanic eruption, SO_2 AMF calculation is strongly dependent on the SO_2 plume height. In general, there is no information available on the altitude of the SO_2 cloud, so an assumption must be made. Therefore, VCD were computed for three different assumed plume heights 2 km for Low AMF, 6 km for Middle AMF and 14 km for High AMF, with an assumed plume thickness of 1 km (For further details see documents available at TEMIS website http://sacs.aeronomie.be/info/vcdamf.php). HYSPLIT trajectory model was used to have first guess of SO_2 cloud height and then VCD were computed by using the respective AMF (middle or High) during days of volcanic activity. Finally, monthly mean VCD were extracted over Pakistan. The retrieved SO_2 VCD over Pakistan from SCIAMACHY observations still experienced some negative values (see Figs. 4, 6 and 8) which are mainly due to remaining error caused by imperfect removal of ring effect and/or spectral interference with ozone (e.g., Khokhar *et al.*, 2005, 2006; Wagner *et al.*, 2008;). The SO_2 retrieval error from SCIAMACHY data is larger as compared to that from OMI and GOME-2 data sets.

Spatial Mapping

In order to investigate spatial distribution of SO_2 column densities, ArcGIS 10.1 software was used to extract data over Pakistan from global observations. For the statistical analysis of temporal trends and regression analysis, Partial Autocorrelation Function (PACF) statistical tools were used.

TRANS BOUNDARY VOLCANIC SO_2 POLLUTION OVER PAKISTAN

Temporal record of SO_2 column densities over Pakistan was prepared by plotting monthly average SO_2 variation during the time period of year 2004 to 2012. Analysis presented in Fig. 1 shows enhanced SO_2 column densities observed on various occasions during this time period.

To evaluate the strength of temporal increase in SO_2 columns over Pakistan, a trend analysis was performed by performing linear fits. The blue line represents the trend of SO_2 over Pakistan including the volcanic eruptions, whereas the red line represents the trend excluding the volcanic events. Analysis showed 120 percent increase in SO_2 with volcanic eruptions and 70 percent increase in SO_2 without volcanic eruptions over the time period of 2004 to 2012 (Khattak, 2013). Regression analysis indicated statistically significant data with *p-value* < *0.03*. Further investigations related these high SO_2 column densities detected over Pakistan to various volcanic activities (Red arrows in Fig.1 and indicated by triangles in Fig. 2) within Pakistan and its neighbouring regions/continents.

In Pakistan, only a historical fissure vent volcano named 'Tor Zawar' was active with minor seismic activity but no gaseous emissions were reported (GVP, 2013). It is located

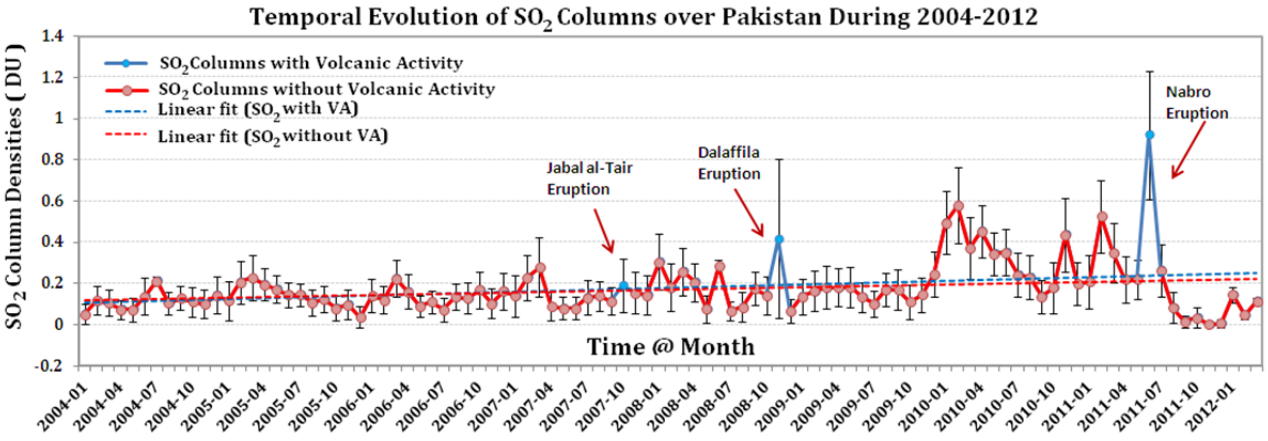
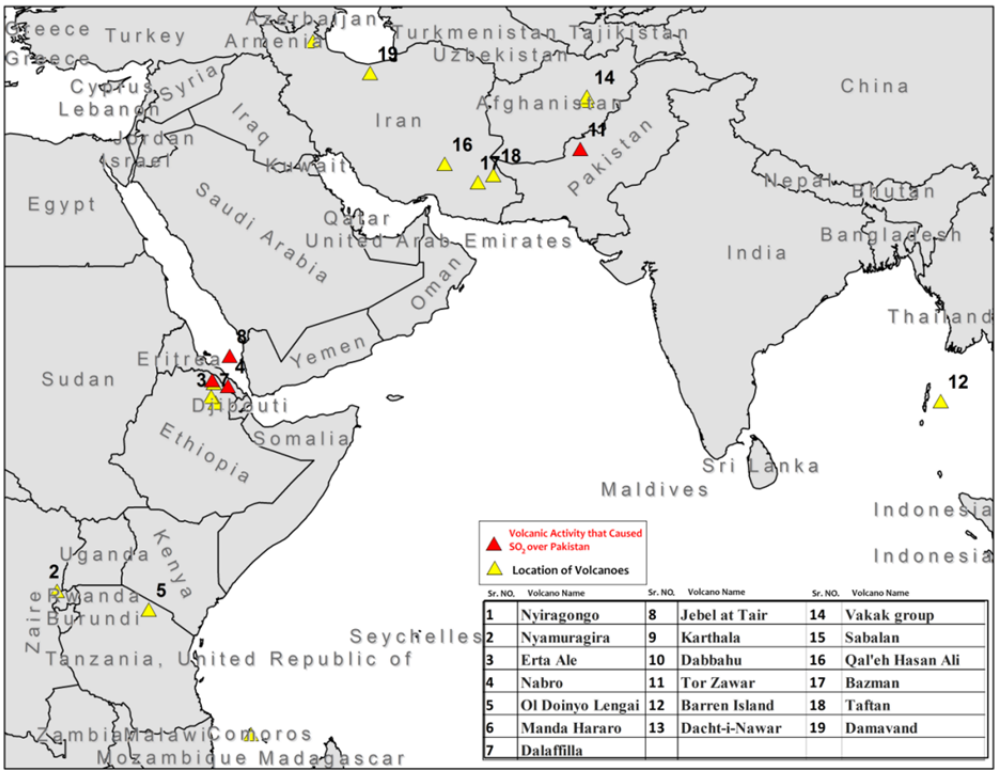


Fig. 1. Time series of SO_2 column densities (in Dobson units: 1 DU = 2.69×10^{16} molecules/cm^2) observed by SCIAMACHY instrument over Pakistan during the time period of 2004–2012. The arrows are indicating the month influenced by trans-boundary SO_2 pollution caused by respective volcanic eruptions. Dashed lines are showing the linear fits for respective datasets with and without volcanic activity. The vertical bars represent the calculated standard deviation for respective month (Figure adopted and modified from Khattak, 2013).



Sr. NO.	Volcano Name	Sr. NO.	Volcano Name	Sr. NO.	Volcano Name
1	Nyiragongo	8	Jebel at Tair	14	Vakak group
2	Nyamuragira	9	Karthala	15	Sabalan
3	Erta Ale	10	Dabbahu	16	Qal'eh Hasan Ali
4	Nabro	11	Tor Zawar	17	Bazman
5	Ol Doinyo Lengai	12	Barren Island	18	Taftan
6	Manda Hararo	13	Dacht-i-Nawar	19	Damavand
7	Dalaffilla				

Fig. 2. Location map of various volcanoes located (yellow triangle) within and in the neighbouring countries of Pakistan and in the African region as enlisted in the legend and Table 2. Red triangles indicate the volcanoes which contributed SO_2 column densities over Pakistan.

in Ziarat region of Pakistan at latitude 30.479°N and longitude 67.492°E. The most recent eruption took place in January 2010. None of three satellite instruments could detect SO_2 emissions from Tor Zawar and most probably the released SO_2 was below detection limit of satellite instruments. According to bulletins of global volcanism program (GVP), it was a minor volcanic outburst with no pre- and post-tertiary volcanic activity. There are few other volcanoes located in the neighbouring countries of Pakistan, as listed in Table 2 and presented in Fig. 2. It also describes the volcano type and status. According to volcano database of GVP, both in Iran and Afghanistan there are 11 Holocene volcanoes and one historical stratovolcano in India. None of these volcanoes experienced a significant volcanic activity during the past decade, therefore, did not contribute in the observed levels of SO_2 column densities over Pakistan.

Pakistan (33°40′N and 73°10′E) is situated in meteorological zone called subtropical high region (Ahrens, 2009). Around these latitudes, not all of the surface air moves equator ward. Some of air moves toward the poles and deflects toward the east due to Coriolis force, resulting in a more or less westerly air flow called the *prevailing westerlies* or simply, *westerlies* (Ahrens, 2009) depending on the location/movement of inter-tropical convergence zone (ITCZ). Therefore, this region is largely affected by the movement of both inter-continental and regional air masses coming from Middle East and African continent. Volcanic eruption occurring in the African region may contribute and affect the atmospheric composition of Pakistan through these westerlies. In the following sections the volcanic eruptions which have substantially contributed to the SO_2 column densities over Pakistan are discussed.

Jebel at Tair Volcano (15.55°N, 41.83°E) Red Sea, Yemen

Jebel at Tair, a stratovolcano is located in the Red sea. According to GVP's monthly report, the eruption started on the afternoon of 30 September, 2007 and continued until December, 2007. The eruption caused large SO_2 plumes, directed lava flows into the ocean and also caused the death of soldiers on the island (GVP, 2013).

Fig. 3 presents the daily maps of GOME-2 (Figs. 3(A), 3(C)) and OMI (Figs. 3(B), 3(D), 3(E) and 3(F)) observations for SO_2 plumes during the eruption of Jebel at Tair. The location of volcano is indicated by a black triangle symbol in each figure. On 1 October, 2007 SO_2 plume was first detected close to the vent and drifted northwest of the volcano. Beside the overpass time (stated in each figure) difference, both instruments OMI and GOME-2 detected the SO_2 plume efficiently. Gray color is indicating the region without satellite observations, as these instruments take more than one day for global coverage, especially, over the regions close to the equator. On the following days SO_2 plume has travelled a distance of 1430 km and 1210 km eastward on 2 and 3 October 2007, respectively. Finally, SO_2 plume reached over Pakistan on 4 October, 2007 affecting mainly the areas of Baluchistan and Punjab provinces as illustrated in Fig. 4; exhibiting monthly mean SO_2 column densities over Pakistan retrieved from SCIAMACHY observations.

Table 2. Volcanoes in the African region, Pakistan and its neighbouring countries*.

Volcanoes within in Pakistan, its neighbouring countries and the African Region with their last known Eruption					
Volcano Name	Region	Year	Location		SO_2 column densities detected over Pakistan
			Longitude (°E)	Latitude (°N)	
Nyiragongo	DR Congo, Central Africa	2012	29.25	–1.52	No
Nyamuragira	DR Congo, Central Africa	2012	29.20	–1.41	No
Erta Ale	Ethiopia, North-eastern Africa	2012	40.67	13.60	No
Nabro	Eritrea, Northeast Africa	2011	41.70	13.37	Yes
Ol Doinyo Lengai	Tanzania, Eastern Africa	2010	35.91	–2.76	No
Manda Hararo	Ethiopia, North-eastern Africa	2009	40.82	12.17	No
Dalaffilla	Ethiopia, North-eastern Africa	2008	40.55	13.79	Yes
Jebel at Tair	Yemen, Red Sea	2008	41.83	15.55	Yes
Karthala	Comoros, Western Indian Ocean	2007	43.38	–11.75	No
Dabbahu	Ethiopia, North-eastern Africa	2005	40.48	12.60	No
Tor Zawar	Pakistan	2010	67.492	30.479	Yes
Barren Island	Andaman Islands India	2011	93.858	12.278	No
Dacht-i-Nawar	Afghanistan	Unknown	67.92	33.95	No
Vakak group	Afghanistan	Unknown	67.97	34.25	No
Sabalan	Iran	Unknown	47.92	38.25	No
Qal'eh Hasan Ali	Iran	Unknown	57.57	29.4	No
Bazman	Iran	Unknown	60	28.07	No
Taftan	Iran	Unknown	61.13	28.6	No
Damavand	Turkmenistan	5350 BC	52.109	35.951	No

* Source: Bulletin of Global Volcanism Program.

Dalaffilla Volcano (13.79°N, 40.55°E) Ethiopia, North-Eastern Africa

Dalaffilla, a stratovolcano is located in Ethiopia. It has a twin volcano known as Alu. Both Dalaffilla and Alu volcanoes are part of the same volcanic system. In November 2008, major effusive eruption started in the summit of Dalaffilla volcano. According to GVP reports, the eruption started on 3 November, 2008 and continued till December, 2008.

Fig. 5 exhibits the maps of GOME-2 (Fig. 5(A)) and OMI (Figs. 5(B), 5(C) and 5(D)) observation from the day after eruption to the day SO_2 plume has reached Pakistan. A satellite overpass on 4 November, 2008 detected SO_2 plume from Dalaffilla volcano (black triangle in the Fig. 5). It has traveled approximately 1650 km towards North. On following days the plume propagated towards East. On 5 November, 2008 SO_2 plume travelled approximately a distance of 3300 km towards East and entered Pakistan on 5 November, 2008 (Fig. 5(C)). On the following days, SO_2 plumes spread over central Pakistan affecting the areas of Baluchistan and Punjab. Fig. 6 exhibits the monthly mean map of SO_2 from SCIAMACHY observations for the month of November, 2008. It clearly shows the elevated amount of SO_2 over the regions of Baluchistan and Punjab provinces. Maximum SO_2 columns observed were 9.4 DU on 6 November, 2008.

Nabro Volcano (13.37°N, 41.70°E) Eritrea, North-Eastern Africa

SO_2 datasets exhibited abnormally high column densities during June, 2011 over Pakistan. It was most significant over the northern and central parts of Pakistan as identified from Fig. 7. The reason behind such high SO_2 columns was investigated. It was found that the Nabro volcano eruption in Eritrea has caused these high SO_2 columns densities over Pakistan. Nabro is a stratovolcano in the north-eastern African nation of Eritrea. The eruption started explosively after a series of earthquakes on the evening of 12 June, 2011 (GPV, 2013) and continued till the mid of July, 2011. It produced the highest SO_2 column ever retrieved from space (Fee *et al.*, 2011; Carn *et al.*, 2012), for instance Clarisse *et al.* (2012) reported 3700 DU retrieved from daily IASI observations.

Nabro volcano eruption spread large amount of SO_2 and ash drifting over much of Eastern Africa, Middle East and South Asian regions. Fig. 7 shows GOME-2 (Figs. 7(A), 7(C) and 7(E) and OMI (Figs. 7(B), 7(D) and 7(F)) images of SO_2 plumes over East Africa and its transportation pattern towards Middle East and South Asia, especially over Pakistan. Both instruments tracked the SO_2 plumes pathway during 13 to 17 June, 2011 efficiently. On 13 June (day after volcanic eruption) plume is just confined to the north-eastern region of Africa. On 14 June, SO_2 plumes have covered almost 1650 km distance towards Northeast and it is drifted towards the Middle East region. On 15 June, plume has traveled around 5390 km towards East and reached closer to Pakistan. On 16 and 17 June, SO_2 plume split into two narrow plumes and spread over North and central parts of Pakistan. Maximum SO_2 columns measured were 8 DU on 17 June 2011 over central Pakistan. As mentioned earlier the eruption continued till the mid of July 2011 and significant SO_2 amount was detected for almost two weeks over Pakistan. It can be clearly identified from Fig. 8; presenting monthly mean map of SO_2 VCD from SCIAMACHY observations for June, 2011.

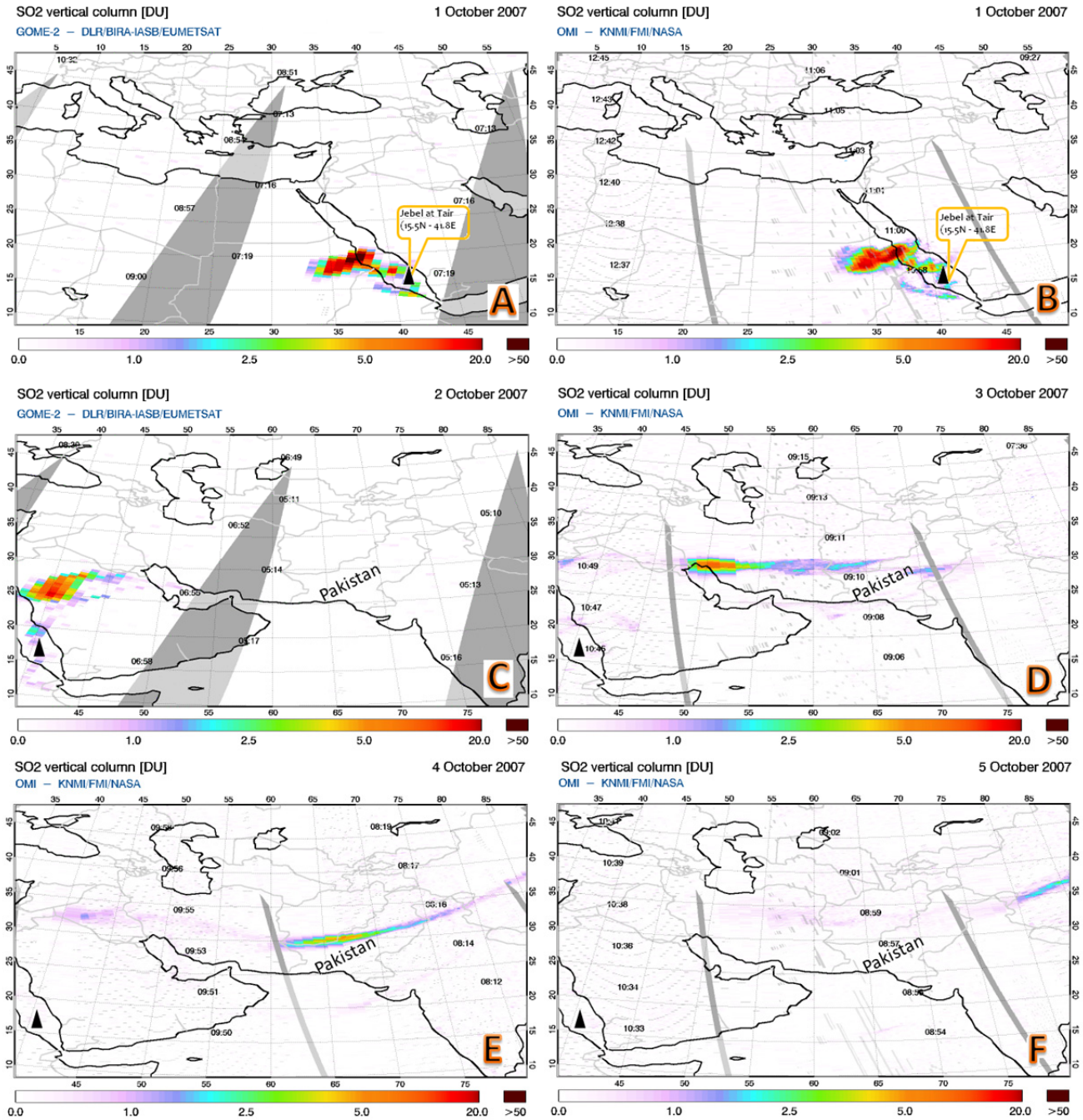


Fig. 3. Spatial maps of daily observations of GOME-2 (3(A) and 3(C)) and OMI (3(B), 3(D), 3(E) and 3(F)) instruments tracking the trans-boundary SO_2 column densities after the Jebel at Tair volcanic eruption in October 2007. It has caused some perturbations in the atmospheric SO_2 amount over Pakistan. Black triangles in each figure are indicating the location of Jebel at Tair (15.55°N, 41.83°E) volcano. Both instruments have efficiently monitored the SO_2 plume extent on respective days and compare well with each other. Overpass time in UTC for each orbit and instrument is given in each figure.

BACK TRAJECTORY ANALYSIS

In order to verify the trans-boundary SO_2 plume over Pakistan, backward trajectory analysis of air masses (Robinson *et al.*, 2011) was performed to track the origin of air masses enriched with SO_2 concentration detected over Pakistan. Maps consisting of backward trajectory were created for each volcanic event, for the identification of SO_2 source and its pathway through which it entered Pakistan's boundaries. Hybrid Single Particle Lagrangian Integrated Trajectory Model-4 (Draxler and Rolph, 2013; HYSPLIT4-Draxler *et al.*, 2013; Rolph, 2013) is an online available trajectory model. HYSPLIT model used for this purpose is developed by National Oceanic and Atmospheric Administration (NOAA)/Air Resources Laboratory (ARL). The meteorological data used for the analysis was from

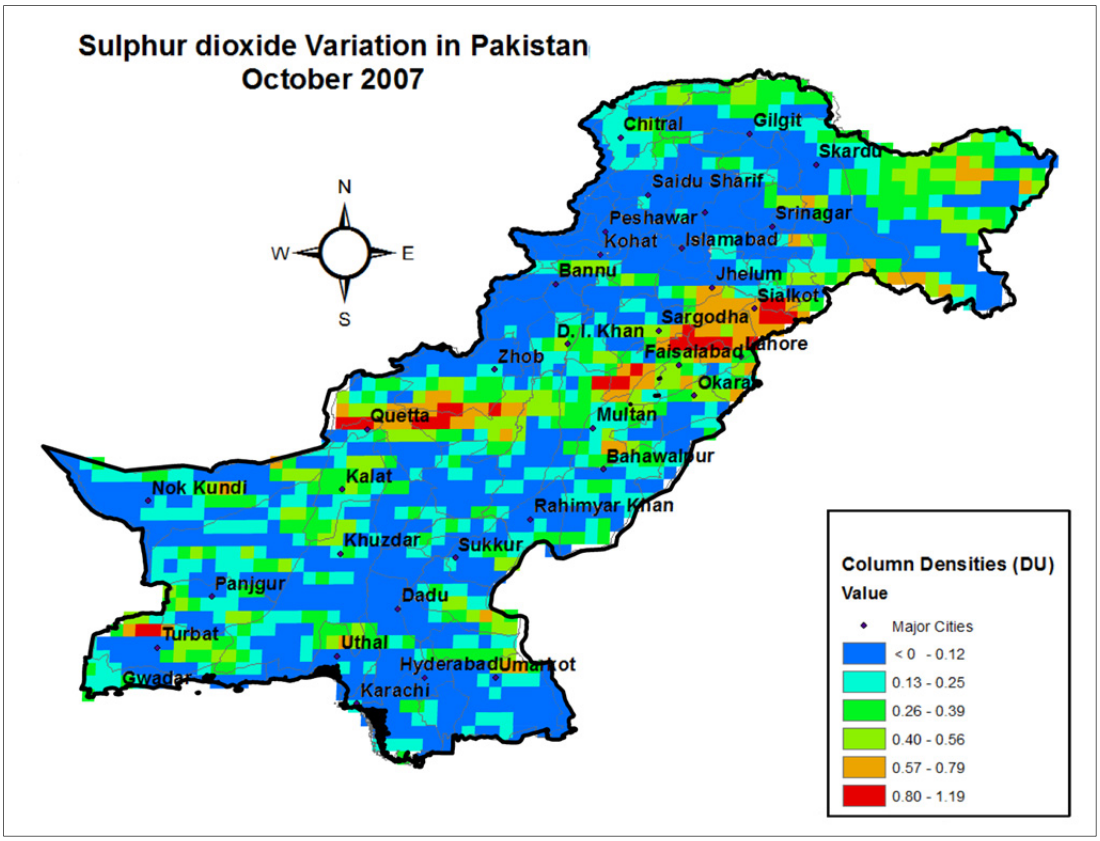


Fig. 4. Monthly mean map of SCIAMACHY data for SO_2 column densities observed over Pakistan during the month of October 2007. Enhanced SO_2 column amount can be clearly identified over Baluchistan and Punjab provinces caused by Jebel at Tair volcanic eruption. Negative SO_2 columns exhibited are mainly caused by remaining noise/error due to improper removal of ozone spectral interference and ring effect.

Global Data Assimilation System (GDAS1) of National Centers for Environmental Prediction (NCEP).

Metrological variables such as potential temperature, rain fall, mixed layer depth, and relative humidity were computed by HYSPLIT. There is a chance of 15 to 30 percent of error (of the travel distance) associated with the trajectories generated by HYSPLIT model (Stohl, 1998; Draxler and Hess, 2004) and it increases with the increasing distance (Sogacheva *et al.*, 2007). For this study, the back trajectory accuracy is sufficient enough to confirm the pathway of the polluted air masses and to determine the altitude of SO_2 plume reaching to Pakistan.

Jebel at Tair Volcano (15.55°N, 41.83°E) Red Sea, Yemen

Trajectory model was run for the backward trajectory of air masses on 4 October, 2007. The trajectory was calculated 120 hours backwards in time and at height 500, 5000 and 15000 meters above ground level (mAGL). Back trajectory showed that only the air masses originated from Ethiopia has caused SO_2 plume over Pakistan. It also indicated that SO_2 plume over Pakistan was present in the upper troposphere/tropopause region. It entered Pakistan from the west side. Air mass average altitude calculated with HYSPLIT is around 15000 mAGL. The trajectory confirmed the pathway of air masses with the pathway and altitude of SO_2 plume reaching Pakistan as shown in the Fig. 9.

Dalaffilla Volcano (13.79°N, 40.55°E) Ethiopia, North-Eastern Africa

Trajectory model simulations for 5 November, 2008 indicated that air masses with large SO_2 column amounts entered from west of Pakistan as shown in Fig. 10. The trajectory was calculated 72 hours backwards in time and at height 500, 4000 and 9000 mAGL. The backward trajectory analysis indicated that air masses have encountered with SO_2 plume in the vicinity of volcano and followed the SO_2 pathway accurately before reaching Pakistan. Other air masses at 9 km above ground level came across and intersected the air masses started from the Arabian Sea, and entered Pakistan on 5 November, 2008. The average altitude of the plume calculated with HYSPLIT is between 4000 and 9000 mAGL. The pathway and altitude of SO_2 plume reaching to Pakistan confirms the trans-boundary origin.

Nabro Volcano (13.37°N, 41.70°E) Eritrea, Northeast Africa

During 16 and 17 June, 2011 the HYPSLIT simulations indicated that SO_2 plumes were hitting Pakistan's boundaries over Northern areas and Baluchistan. The back trajectory of air masses and altitude of SO_2 plume reaching Pakistan is shown in the Fig. 11. According to GVP, initially the plume from volcanic eruption rose to the height of 9100–13700 m (30,000–45,000 ft) above sea level (ASL). Later

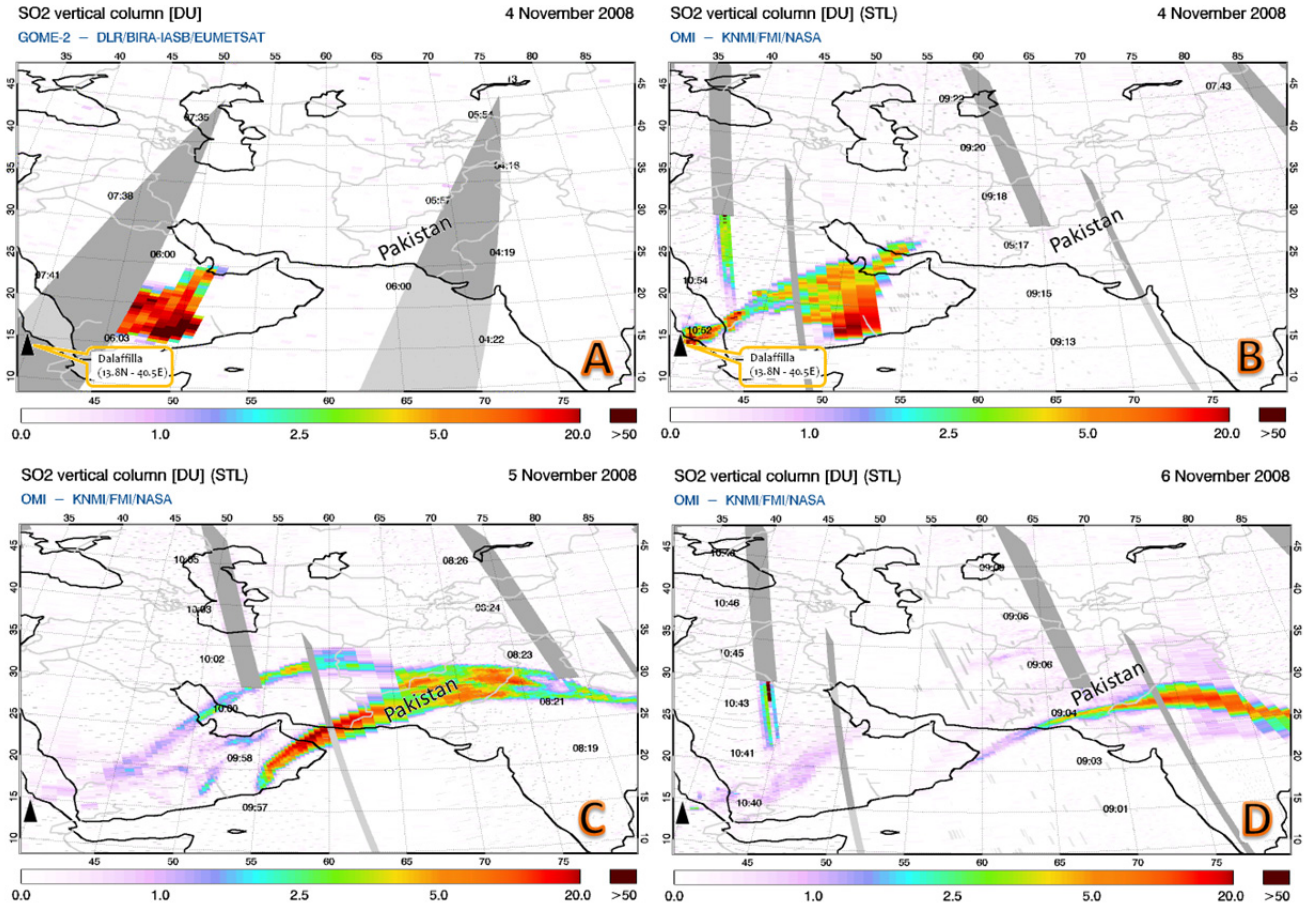


Fig. 5. Spatial maps of daily observations of GOME-2 (5(A)) and OMI (5(B), 5(C) and 5(D)) instruments tracking the trans-boundary SO_2 column densities after the Dalaffilla volcanic eruption in November 2008. It has caused significant perturbations in the atmospheric SO_2 amount over Pakistan. Black triangles in each figure are indicating the location of Dalaffilla Volcano (13.79°N, 40.55°E). Both instruments have efficiently monitored the SO_2 plume extent on respective days and compare well with each other. Overpass time in UTC for each orbit and instrument is given in each figure. Gray color is indicating the region without satellite observations, as these instruments take more than one day for global coverage, especially, over the regions close to the equator.

on 13 to 14 June the plume was detected at the altitude of 6100–10700 m ASL. The trajectory was calculated 120 hours backwards in time and at height of 400, 5000 and 11000 mAGL on 16 June, 2011. On 17 June, trajectory was calculated 144 hours backwards on 400, 7000 and 9000 mAGL. The back trajectory shows that the air mass originated over Bay of Bengal, crossed the volcanic eruption site and from there carried the SO_2 plume to Pakistan. Air masses carrying SO_2 pollution hit Pakistan over two different locations. The average altitude of these air masses calculated with HYSPLIT was around 9000 mAGL. Main objective of using HYSPLIT was to identify the SO_2 plume altitude and track the pathway of air masses reaching over Pakistan during volcanic activity in the African region.

DISCUSSION

Temporal record of SO_2 over Pakistan was analysed by plotting the monthly averaged SO_2 column from 2004 till 2012, as presented in Fig. 1. The bars show the spatial variation (standard deviation) for each month over Pakistan. Satellite database of SO_2 column amounts over Pakistan indicated high SO_2 column densities over cities and/or regions with high population, traffic density and industrial activities. Additionally, anomalous high SO_2 column densities were observed occasionally over northern, western and central parts of Pakistan. It was investigated and these enhanced levels of SO_2 over Pakistan were caused by some of global volcanic activities. For instance, in June, 2011 unexpected high SO_2 columns were observed over Pakistan caused by the Nabro volcanic eruption. It was very explosive and SO_2 rich volcanic eruption (Fee *et al.*, 2011; Carn *et al.*, 2012). For instance Clarisse *et al.* (2012) reported total SO_2 mass of 1.5 Tg retrieved from daily IASI observations and Carn *et al.* (2012) estimated 1 to 2 Tg SO_2 based on OMI and Atmospheric Infrared Sounder (AIRS) data. It has caused not only the stratospheric aerosol load (e.g., Bourassa *et al.*, 2012) but also large amounts of SO_2 pollution was detected over neighbouring regions. It has influenced Pakistan's atmosphere with maximum SO_2 of 7 DU detected on 17

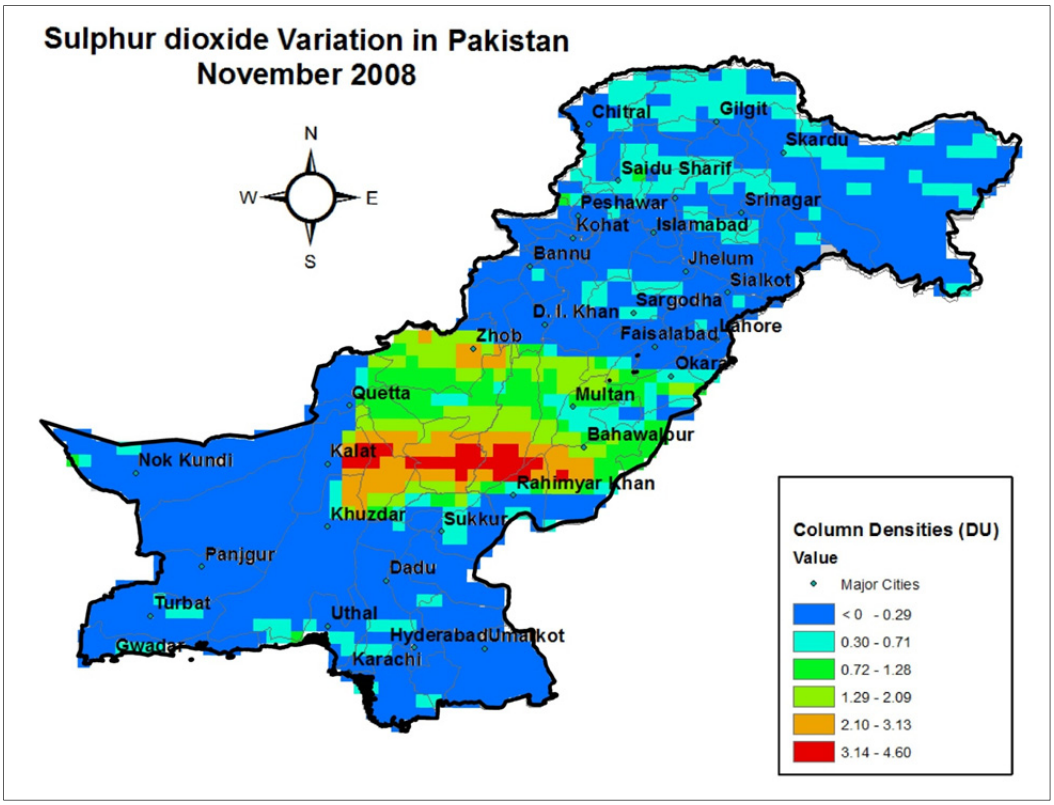


Fig. 6. Monthly mean map of SCIAMACHY data for SO_2 column densities observed over Pakistan during the month of November 2008. Enhanced SO_2 column amount can be clearly identified over Baluchistan and southern Punjab regions caused by Dalaffilla volcanic eruption.

June, 2011 from daily observations of SCIAMACHY instrument. Other SO_2 peaks in the time series presented in Fig. 1 were also related to Jabal al-Tair volcano from Red Sea during October, 2007 and Dalaffilla volcano from Ethiopia during November, 2008. The effect of volcanic eruption as enhanced SO_2 column densities over Pakistan can be clearly identified in Figs. 3–8. However, the effect is not significant in the case of Jebel at Tair volcano as compared to the Nabro and Dalaffilla volcanoes, but it is still identifiable. Another peak in SO_2 time series around January, 2007 is coincident with Tor Zawar volcano from Pakistan. It is difficult to relate this enhancement with Tor Zawar activity, because, none of three satellite instruments were able to detect the SO_2 emissions from this volcano. Although, volcanic activity has been reported by the GVP, but it cannot be speculated without any scientific and solid evidence. In principal whatever the amount of SO_2 was released that became the part of atmospheric composition of Pakistan. It might be due to the reason that amount of SO_2 released was well below the detection limit and/or background SO_2 levels that satellite instruments were not able to detect it over that region. Other enhanced SO_2 levels over Pakistan are observed during the months of March and April 2010. Similarly, 2010 eruption of Eyjafjallajökull volcano (Rix *et al.*, 2012) from Iceland is coincident with enhanced levels of SO_2 over Pakistan during the months of April–May 2010. It has caused a severe disruption to western and northern Europe air spaces. However, none of the satellite instrument could track volcanic gas plume over Pakistan during Eyjafjallajökull volcanic eruption.

In general, SO_2 column densities showed large seasonality due to its reaction with OH radical. Therefore, OH is the key parameter which determines the life time and seasonal cycle in the atmospheric SO_2 abundances. The SO_2 seasonal cycle may also be slightly dependent on both anthropogenic (coal, petroleum products and biomass burning) and natural (monsoon, snow) activities on regional basis. Fig. 1 exhibited a clear seasonal cycle in SO_2 amounts over Pakistan with maximum in winter and minimum in summer season. SO_2 released in the atmosphere is converted into sulfate particles and sulfuric acid (H_2SO_4) through its reaction with O_3, hydrogen peroxide (H_2O_2), hydroperoxyl (HO_2) and OH radicals. As this is the region of monsoon circulation and such major volcanic eruptions can inject SO_2 into stratosphere directly or through the monsoon circulation (Bourassa *et al.*, 2012). Such long-range transport as discussed in previous sections can be a significant source of stratospheric SO_2 and/or sulfate aerosols. According to (Bourassa *et al.*, 2012 and references therein), stratospheric aerosol can have significant impact on earth radiation budget and cause the cooling on the surface. A single Nabro volcano eruption during June 2011 caused approximately 1.3 terragrams of SO_2 in the upper troposphere (around altitude of 9–14 km) and also enhanced largely the stratospheric sulfate aerosols since Mount Pinatubo eruption (Bourassa *et al.*, 2012). Furthermore, stratospheric sulfate aerosols present at temperate latitudes

can play an important role in stratospheric ozone chemistry by providing sites for heterogeneous chemistry (e.g., Solomon *et al.*, 1996).

South Asian region is getting attention of world due to a decent growth in GDP of developing nations (India, Bangladesh and Pakistan) during last decade, and of scientific community due to consequent increase in atmospheric pollution. This region with worst air quality caused by rapid industrialization which has brought about more cars, factories, and more people to burn coal and wood for cooking purposes, is relatively less studied. According to (Ramanathan *et al.*, 2001), brown haze is a severe problem over the tropics. It is mainly due to increased emissions of aerosols and precursor gases like SO_2 that have increased over South Asia by a factor of 3 to 4 since 1970.

According to Pakistan Environmental protection Agency

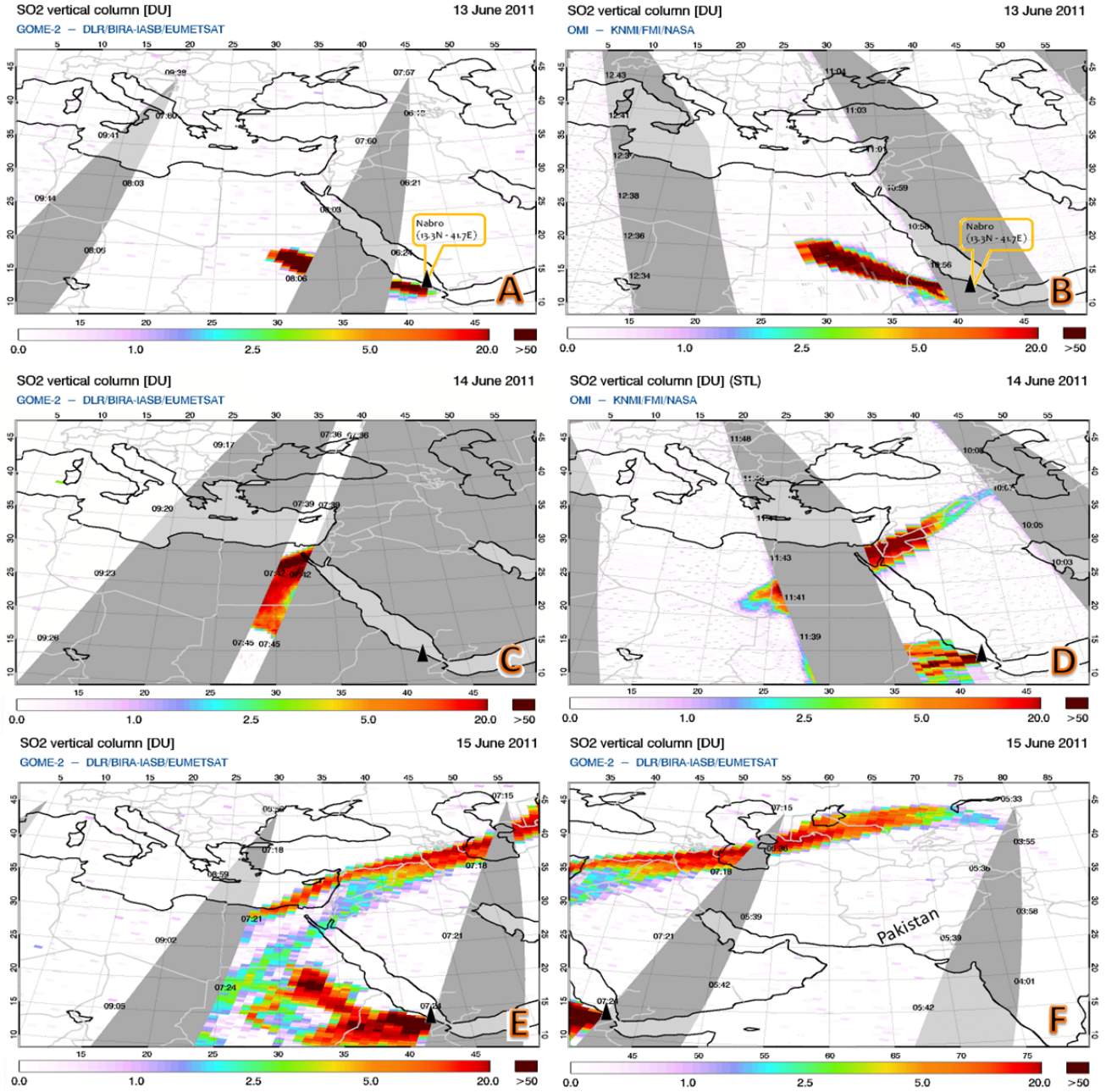


Fig. 7. Spatial maps of daily observations of GOME-2 (7(A), 7(C), 7(E) and 7(G)) and OMI (7(B), 7(D), 7(F) and 7(H)) instruments tracking the trans-boundary SO_2 column densities after the Nabro volcanic eruption in June 2011. It has caused significant perturbations in the atmospheric SO_2 amount over Pakistan. Black triangles in each figure are indicating the location of Nabro Volcano (13.3°N, 41.7°E). Both instruments have efficiently monitored the SO_2 plume extent on respective days and compare well with each other. Overpass time in UTC for each orbit and instrument is given in each figure. Gray color is indicating the region without satellite observations, as these instruments take more than one day for global coverage, especially, over the regions close to the equator.

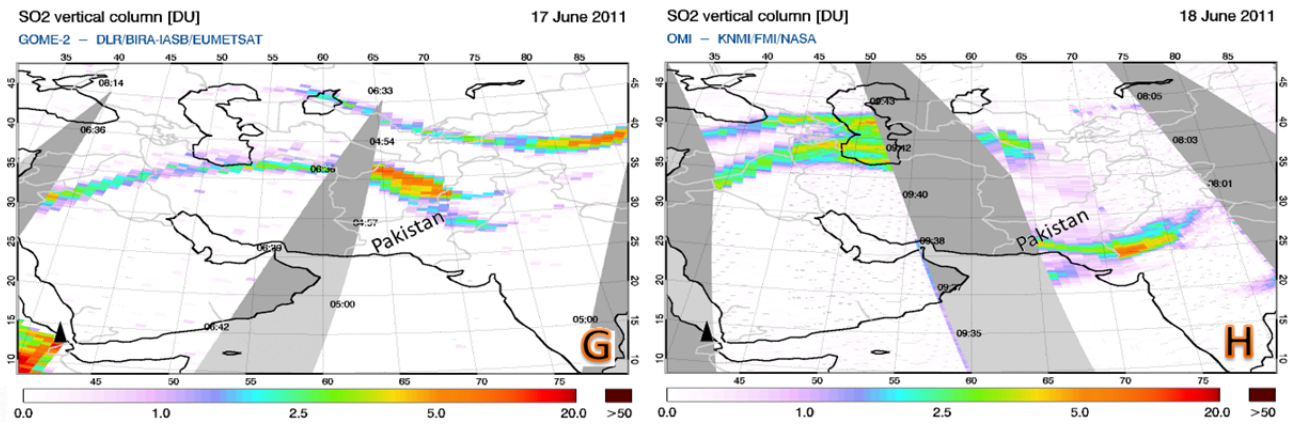


Fig. 7. *(continued).*

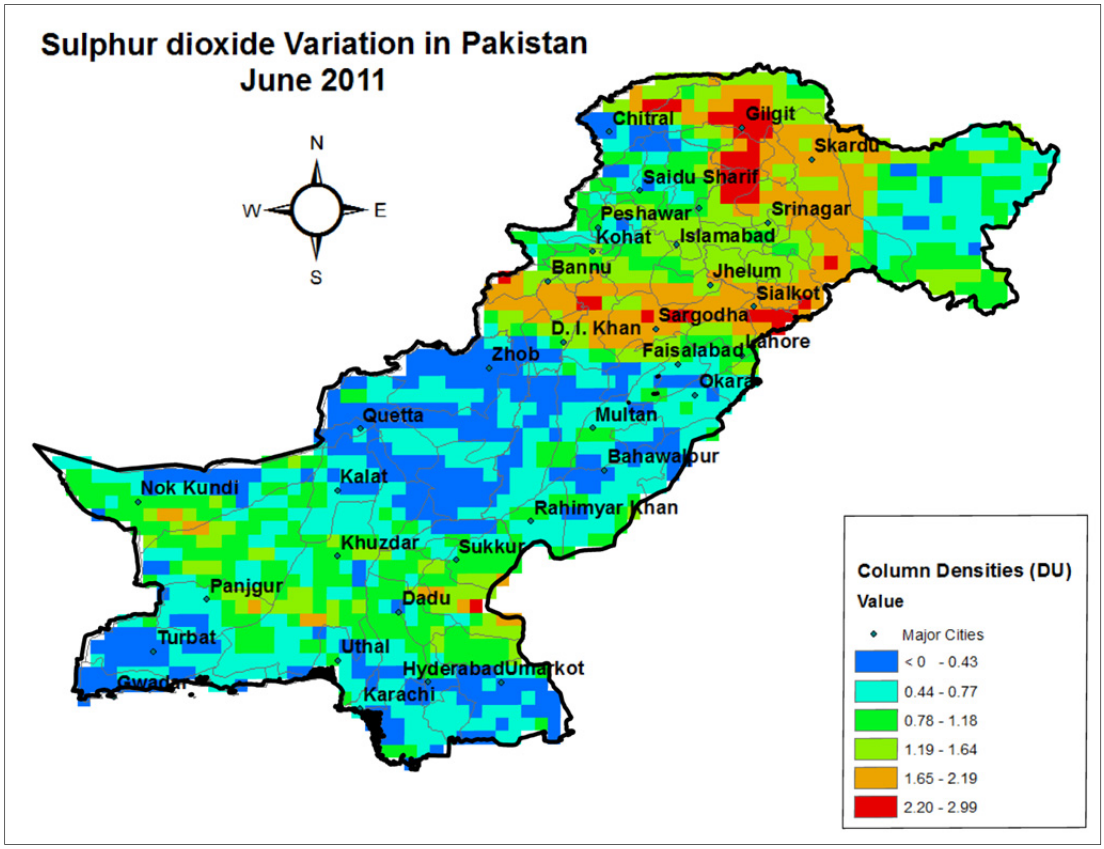


Fig. 8. Monthly mean map of SCIAMACHY data for SO_2 column densities observed over Pakistan during the month of June 2011. Enhanced SO_2 column amount can be clearly identified over northern regions of Pakistan caused by Nabro volcanic eruption.

(Pak-EPA), Pakistan has been suffering from trans-boundary air pollution, especially, from its immediate neighboring countries such as India and China. India is stated to be largest contributor to trans-boundary air pollution in Pakistan, especially, during winter seasons when dense smog is observed over northern India and northeastern parts of Pakistan. This study will help to identify that trans-boundary air pollution over Pakistan is not only caused by the anthropogenic sources but also due to global volcanic activities as well.

CONCLUSIONS

This study has first time emphasized on the amount of trans-boundary SO_2 pollution present in the atmosphere of Pakistan as a consequence of global and regional volcanic eruption by using satellite remote sensing data. SCIAMACHY, OMI and GOME-2 observations were used to study the spatial and temporal variation of SO_2 over Pakistan during the time period of year 2004 to 2012. An overall temporal increase of 120 percent with volcanic

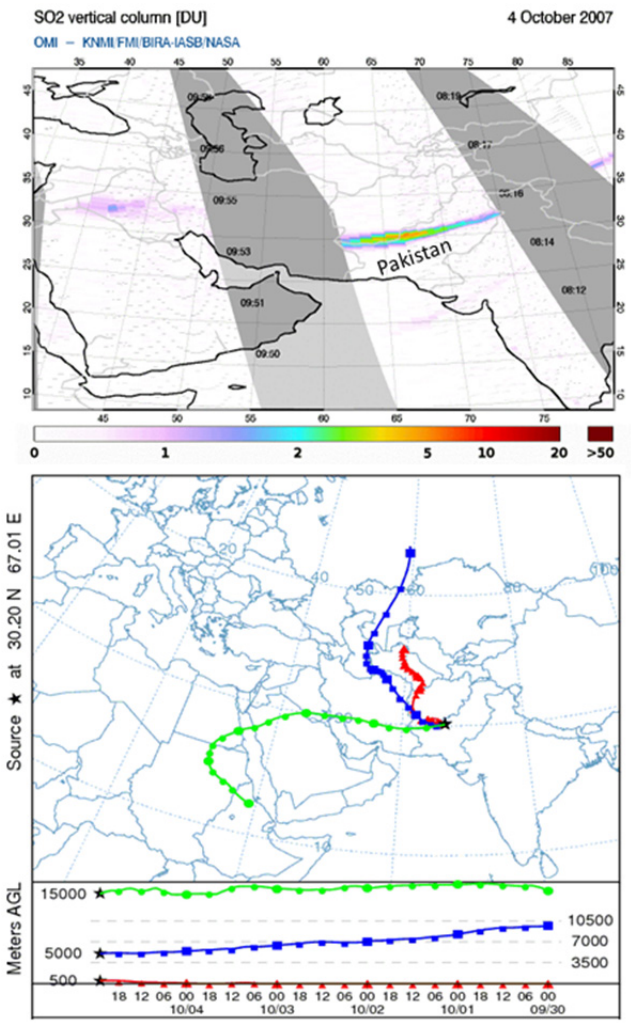


Fig. 9. HYSPLIT backward trajectory analysis performed at 500 (red), 5000 (blue), 15000 (green) mAGL for 4 October, 2007. Each gap represents a 6 hours interval. Trajectory following the SO_2 plume pathway (upper panel figure) is located at 15000 mAGL and passed over Middle East and Red Sea area.

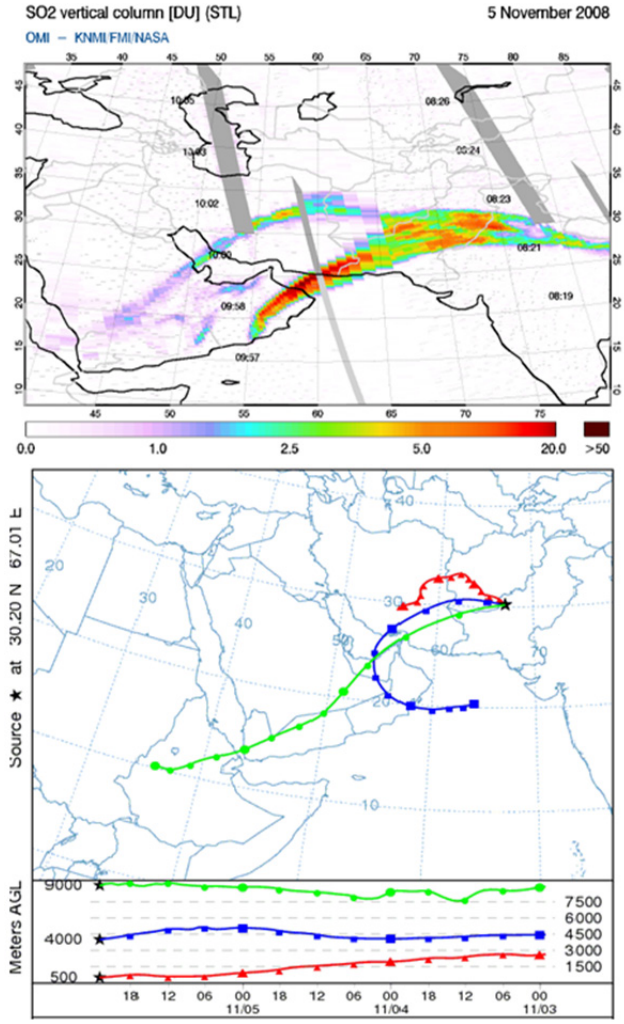


Fig. 10. HYSPLIT backward trajectory analysis performed at 500 (red), 4000 (blue), 9000 (green) mAGL for 5 November, 2008. Each gap represents a 6 hours interval. Trajectory following the SO_2 plume pathway (upper panel figure) is located at 9000 mAGL and passed over Yemen, Red Sea area and Eretria.

eruptions and 70 per cent without volcanic eruptions was calculated and statistical significance of data sets has been tested (p-value < 0.03, with confidence interval of 95%). Tor Zawar volcano located in Pakistan, erupted during January 2010 but no SO_2 emissions were reported and detected via satellite observations. Some major volcanic eruptions in the African region during last decade have affected atmosphere of Pakistan. Among them, Dalaffilla volcanic eruption in2008 and Nabro volcano event in 2011 caused the major effects by contributing high SO_2 column densities over the regions of northern areas, Baluchistan and Punjab province. Jebel at Tair volcanic activity during October 2007 has also affected few areas of Baluchistan and Punjab province. However, it contributed lesser amounts of SO_2 column densities as compared to the Nabro and Dalaffilla volcanic eruption events. The Back trajectory analyses performed by HYSPLIT model have confirmed the trans-boundary SO_2 plume pathways and also enabled to identify the plume altitude. Most of the time SO_2 plume was present at high altitude like in upper troposphere/tropopause region. Therefore, in order to have a better assessment of atmospheric composition there is a need to constrain SO_2 emissions from all sources, especially the trans-boundary fraction as a consequent of both intense anthropogenic and natural activities. Furthermore, this study will help different stake holders including policy makers to provide with concrete scientific record of atmospheric SO_2 column amounts over Pakistan in order to devise cost effective but efficient policies related to air quality issues in Pakistan.

ACKNOWLEDGEMENTS

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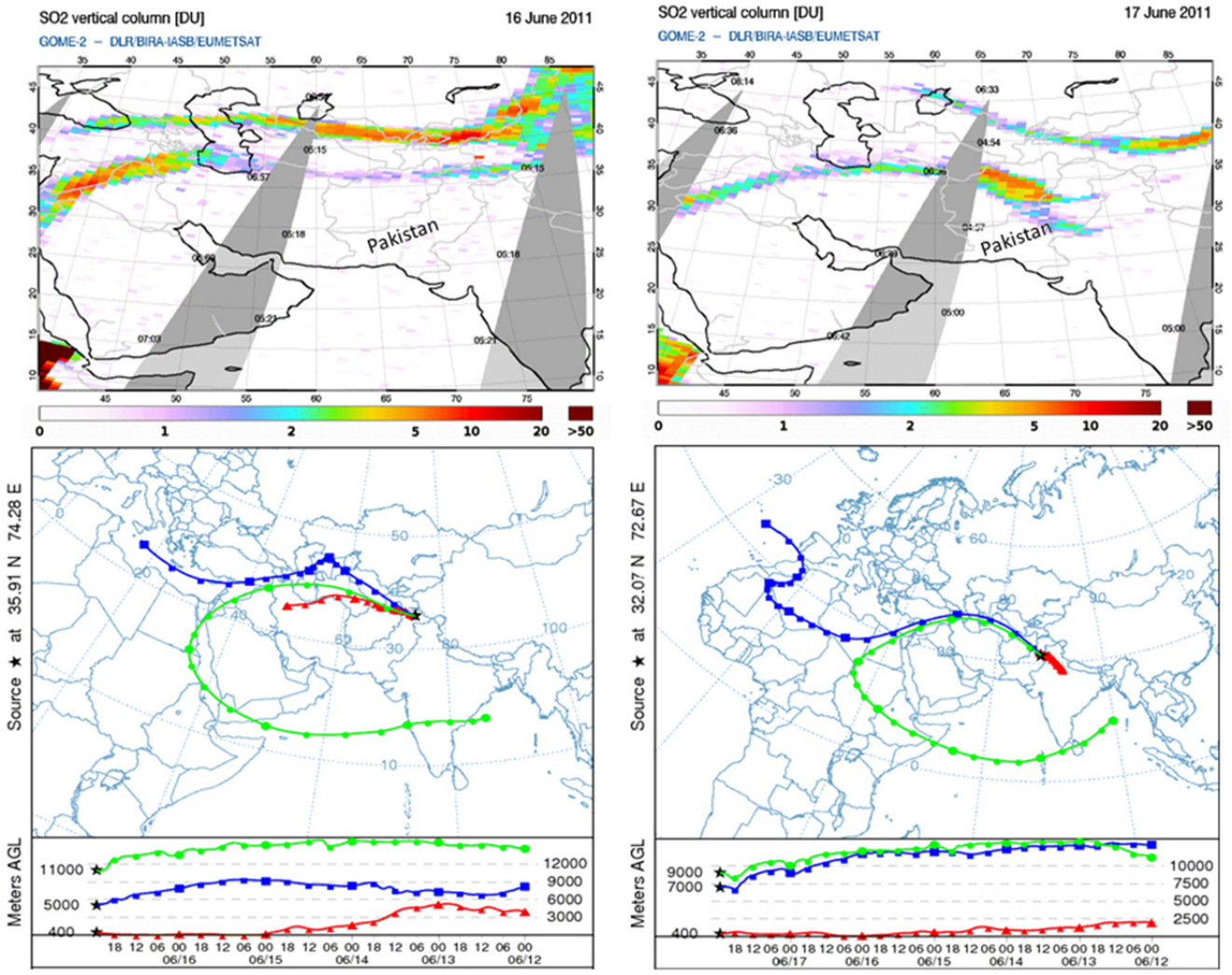

Fig. 11. HYSPLIT backward trajectory analyses performed at 400 (red), 5000 (blue), 11000 (green) mAGL for 16 June 2011 and at 400 (red), 7000 (blue), 9000 (green) mAGL for 17 June, 2011. Each gap represents a 6 hours interval. Trajectories following the SO_2 plume pathways (upper panel figure) are located at 7000, 9000 and 11000 mAGL. During both days, they originated from Bay of Bengal and passed over eastern Africa, headed North and then eastward over Middle Eastern region and finally reached to North of Pakistan.

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