SATELLITE REMOTE SENSING OF ATMOSPHERIC SO_2: VOLCANIC ERUPTIONS AND ANTHROPOGENIC EMISSIONS

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Abstract

In this article, we present satellite data analysis of atmospheric Sulfur Dioxide (SO_2) from volcanic eruptions and anthropogenic activities. Data from Global Ozone Monitoring Experiment (GOME) on board ERS-2 for the years 1996 to 2002 is analyzed using a DOAS based algorithm with the aim of retrieving SO_2 Slant Column Densities (SCDs). Difficulties in the retrieval of SO_2 SCDs due to instrumental effects are investigated in detail and significantly improved. The retrieved SCDs can be used to identify and monitor several volcanic eruptions. A brief introduction of different volcanic eruptions around the globe is presented Also informations about the anthropogenic SO_2 emissions can be easily achieved from the retrieved data set. A time series of anthropogenic SO_2 emissions over Eastern Europe is presented in this study. The time series showed high SO_2 SCDs over Eastern Europe during the winter months. The results demonstrate a high sensitivity of GOME instrument towards SO_2 emissions.

1. INTRODUCTION

Sulfur Dioxide (SO_2) is an important trace species in the atmosphere, under background conditions and in polluted areas. It is released to the troposphere as a result of both anthropogenic and natural phenomena.
Sulfur dioxide emissions are partly responsible for acid depositions on the surface and the occurrence of winter smog episodes. In addition, the oxidization of SO_2 in the troposphere leading to concentrations of sulfate aerosols in the atmosphere has also been found to contribute to visibility degradation.
Volcanoes are an important natural source of various atmospheric trace gases. Volcanic eruptions and their emissions are sporadic and intermittent and often occur in uninhabited regions. Therefore assessing the amount and size of the gaseous and particulate emission from volcanoes is difficult. Satellite remote sensing measurements provide a well-suited opportunity to overcome this difficulty.
The main anthropogenic sources of sulfur dioxide include: fossil fuel consumptions, the smelting of metal sulfide ores to obtain the pure metals and coal burning.

SO_2 when liberated to the atmosphere reacts rapidly with OH to form HSO_3 (Eq.1), which reacts with O_2 to form SO_3 (Eq.2). It is soluble in clouds and aerosols, where it reacts with H_2O. As a result of these processes, SO_2 is converted to H_2SO_4 (Eq.3), consequently causing acid rain and deforestation. In general the maximum concentration of SO_2 is close to its source and the amount of SO_2 decreases as the distance from the source increases, indicating a short tropospheric lifetime of typically a few days [1].

$$SO_2 + OH \xrightarrow{M} HSO_3 \quad (1)$$

$$HSO_3 + O_2 \rightarrow SO_3 + HO_2 \quad (2)$$

$$SO_3 + H_2O \xrightarrow{M} H_2SO_4 \quad (3)$$

In the dry stratosphere, particularly in the lower stratosphere where the concentration of OH is relatively small, the lifetime of SO_2 is longer than in the troposphere (of the order of several weeks) [1].
In this study, we give a brief overview of GOME instrument, data analysis with experienced difficulties probably due to some instrumental effects and their solutions. We further give an overview of GOME SO_2 observations during the period from the year 1996 to 2002, indicating different volcanic eruptions around the globe and anthropogenic SO_2 emissions.

2. The GOME INSTRUMENT AND DATA ANALYSIS

2.1. Global Ozone Monitoring Experiment

GOME is a nadir-scanning ultraviolet and visible spectrometer that measures the solar spectrum directly and Earthshine spectra, i.e.: the sunlight reflected and scattered back into space by the atmosphere and by the surface. The GOME instrument with spectral range from 240 to 790 nm [2] on-board ERS-2, was launched in April 1995 into a near-polar sun synchronous orbit at a mean altitude of 790 km with a local equator crossing time 10:30 am. Global coverage is obtained within 3 days at the equator by a 960 km across-track swath. For the measurements presented in this work, the ground pixel size is 40 (along track) * 320 (across track) km^2. A key feature of GOME is its ability to detect several chemically active atmospheric trace gases such as SO_2, NO_2, BrO, OClO and CH_2O etc.

2.2. Data Analysis

From the ratio of earthshine radiance and solar irradiance measurements, slant column densities (SCDs) of the respective absorbers can be derived by applying the technique of differential optical absorption spectroscopy (DOAS) [3, 4, 5, 6,]. SO_2 exhibits absorption structures in the UV spectral region, and a wavelength window between 312.5 and 327.6 nm (GOME channel 2b) is used for the retrieval. In this spectral region, the strong ozone Huggins band overlaps the weak SO_2 absorptions. Thus, a precise knowledge of the instrumental function, which describes the convolution of the incoming highly resolved signal to the instrumental resolution, is indispensable since small uncertainties in the spectral structure of strong absorbers can result in residuals that are larger than the weak SO_2 absorption itself. Narrow Fraunhofer lines are well suited to derive information on the instrumental function. According to the work of Van Roozendael et al., [8], we applied a nonlinear least squares fit to find an optimum slit function by fitting a highly resolved solar spectrum [9, 10] to the solar irradiance measurement of GOME. The slit function was found to be of an asymmetric Voigt line shape. Furthermore, the width and asymmetry are strongly wavelength dependent. We used this asymmetric and wavelength dependent slit function to convolve the highly resolved laboratory spectra of SO_2, two Ozone spectra at different temperatures and a Ring spectrum, thereby creating an optimal set of reference spectra at instrumental resolution. The retrieval of SO_2 column densities in the UV wavelength region is also strongly affected by the spectral interference due to the structures induced by the diffuser plate used for the solar irradiance measurements [11]. These structures vary with the position of the sun, thus exhibiting a seasonal dependency.

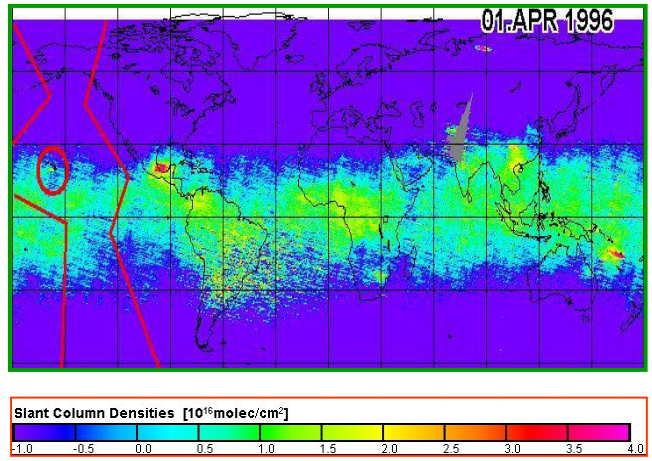

Fig. 1: Monthly mean of SO_2 SCDs for April 1996, the region indicated by the red line was selected for the calculation of offset values at each latitude.

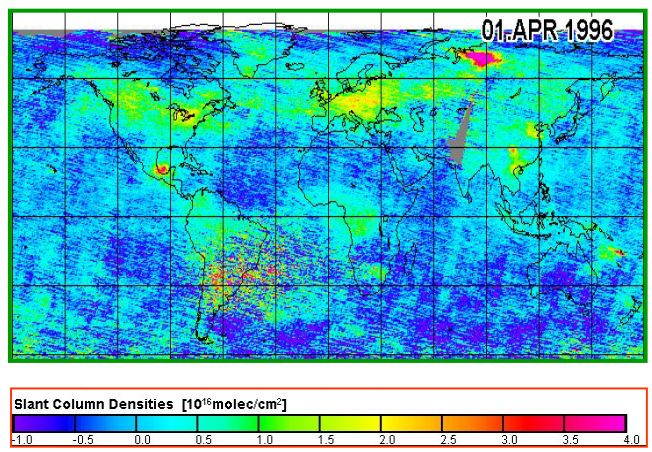

Fig. 2: Monthly mean of SO_2 SCDs (after correction by RSM) for April 1996, the regions of significant improvement, especially over Europe, Norilsk and USA can be clearly seen.

In addition to this seasonal pattern in the SO_2 SCDs, also a systematic latitudinal dependence was found, most probably caused by the interference with strong O_3

absorption, especially for larger solar zenith angles (SZA).
To avoid the latitudinal offsets described before, we normalize our retrieved columns by the reference sector method (RSM). This method uses a presumably SO_2 free reference sector over a remote area [Red line on Fig. 1] in order to calculate offset values of SO_2 columns at each latitude. In brief, this method applies a correction for each latitude so that the reference sector exhibits a SCD of zero after the correction [Fig. 2].

Apart from these complications, which result in latitudinal offsets in the retrieved SO_2 SCDs, we found artificially enhanced columns over high and bright surfaces e.g. Himalaya. The reason for these intereferences are yet unclear and we apply a preliminary and simple method to avoid these artefacts: We include a ratio spectrum, which contains the spectral structures of high and bright surfaces, in the DOAS fit. Therefore, we used the ratio of a pixel directly over the Himalaya and a nearby pixel in northern India from GOME orbit 61204045 on 12th of December 1996. Finally these pragmatic solutions helped us to overcome the inconsistency in the retrieved SO_2 SCDs.

3. SO_2 FROM VOLCANIC ERUPTIONS

Volcanic eruptions inject gases and particles into the atmosphere, leading to stratospheric and tropospheric aerosol formation, consequently contributing to climate radiative forcing. Mainly sulfur containing volcanic gases are responsible for the climate effects of explosive volcanic eruptions [12]. Highly explosive volcanic events, like Pinatubo in 1991, affect the climate on time scales of months to years [13].
The fate of volcanic aerosols in the stratosphere and their influence on chemistry, microphysics and dynamics are still under discussion [14].
In this section, we present a brief overview of GOME observed SO_2 SCDs from different volcanic events during the time-period from the year 1996 to 2002.

1. Nyamuragira (1.4°S, 29.2°E), Zaire / Congo

The Nyamuragira is a shield volcano in the Virunga volcano field of Zaire / Democratic Republic of Congo. It is the most active volcano from the central African region over last decade. According to the reports from the Global Volcanism Network (GVN) and observed by GOME, the eruption started on 1 December 1996. On 5 December, the plume had reached 12 km altitude [15]. GOME observed the volcanic activities of Nyamuragira during December 1996, October 1998, January - February 2000, February 2001 and July 2002, see Fig. (3A, 3B).

2. Popocatépetl (9.02°N, 98.62°W), Mexico

A large stratovolcano 70 km southeast of Mexico City, is the second highest and one of the most active volcanoes in Mexican history. In assessing the total amount of SO_2 emitted by Popocatépetl, it is more difficult to establish a threshold than other volcanoes because the large population of Mexico City and surrounds leads to a significant anthropogenic contribution to the total SO_2 emissions [1]. However, GOME observed volcanic activities during the months of March, April, July to October and Dec. 1996, July 1997, November 1998, December 2000, June 2001 and March 2002, see Fig. (3A, 3B).

3. Etna (1.4°S, 29.2°E) Sicily, Italy

Etna, Europe's most active volcano, is probably one of the most studied volcanoes on Earth. After 1992 Etna erupted in August 2001 and October 2002. The GOME instrument observed a SO_2 plume streaming southward from the volcano and out over the Mediterranean Sea. See Figure (3D).

4. Cerro Azul (0.9°S, 91.42°W) Galapagos Islands, Ecuador

Cerro Azul is a shield volcano located on Isabella in the Galapagos Islands of Ecuador. Volcanic eruption occurred in September and October 1998, as observed by GOME. See Figure (3B).

5. El Reventador (0.07°S, 77.67°W) and Tungurahu (1.46° S, 78.44° W) , Ecuador

El Reventador is the neighboring volcano of Tungurahua. GOME observed SO_2 emissions from out gassing of El Reventador in October 2002 following the huge eruption in November 2002. Tungurahua is an active stratovolcano also known as the "The Black Giant.". GOME observed first SO_2 signal on September 1999 and since that time perhaps there was a month without volcanic activity of Tungurahu with some periodic eruptions or out gassing until December 2002. See Figure (3C, 3D).

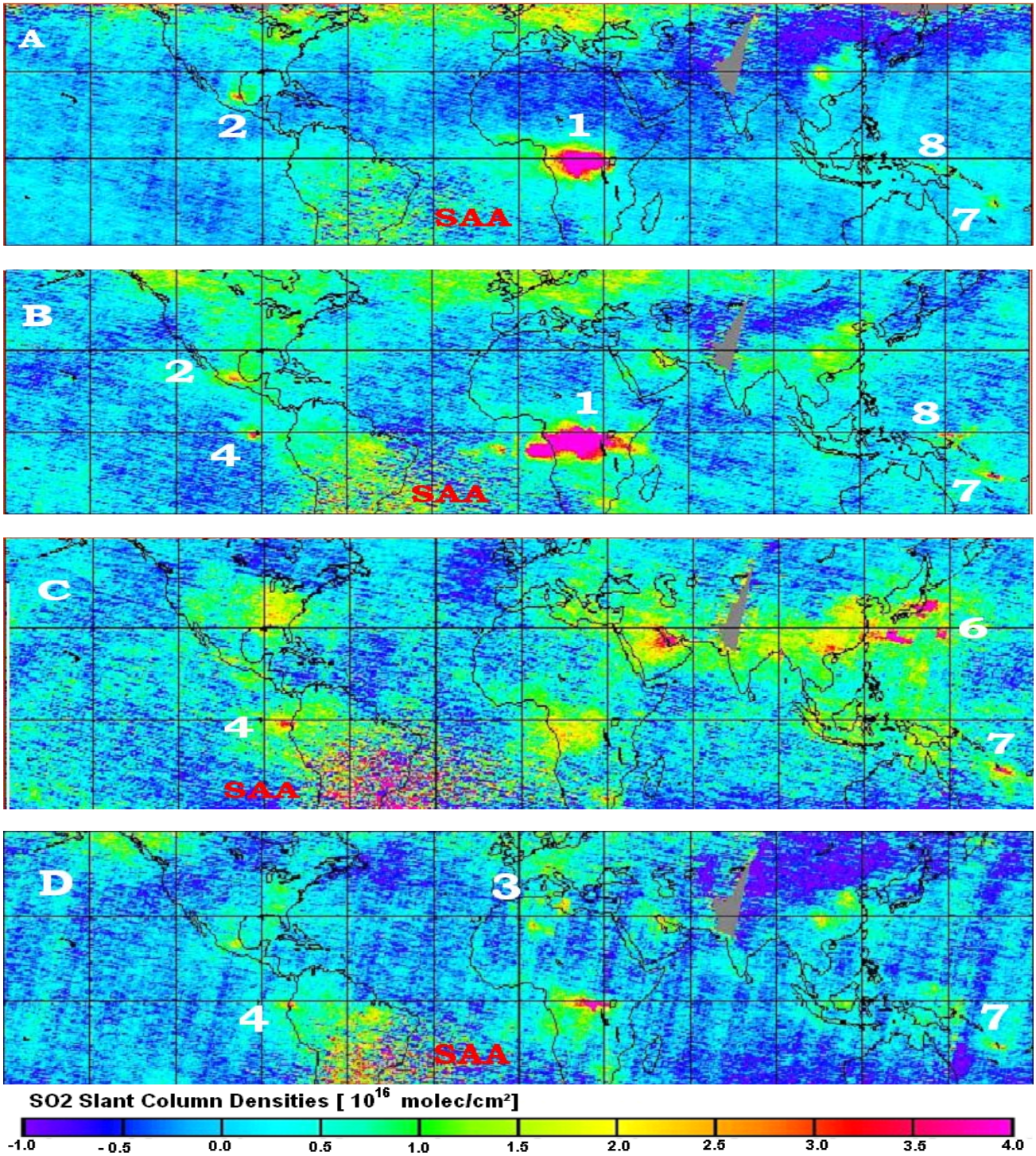

Fig. 3: Monthly mean of SO_2 SCDs for Dec. 1996(A), Oct. 1998(B), Aug. 2000(C) and for Oct. 2002(D) showing different volcanic vents as indicated by the name on maps. The region affected by South Atlantic Anomaly (SAA) is also mentioned. The indices on the figures represent the volcanic event described in the section 3.

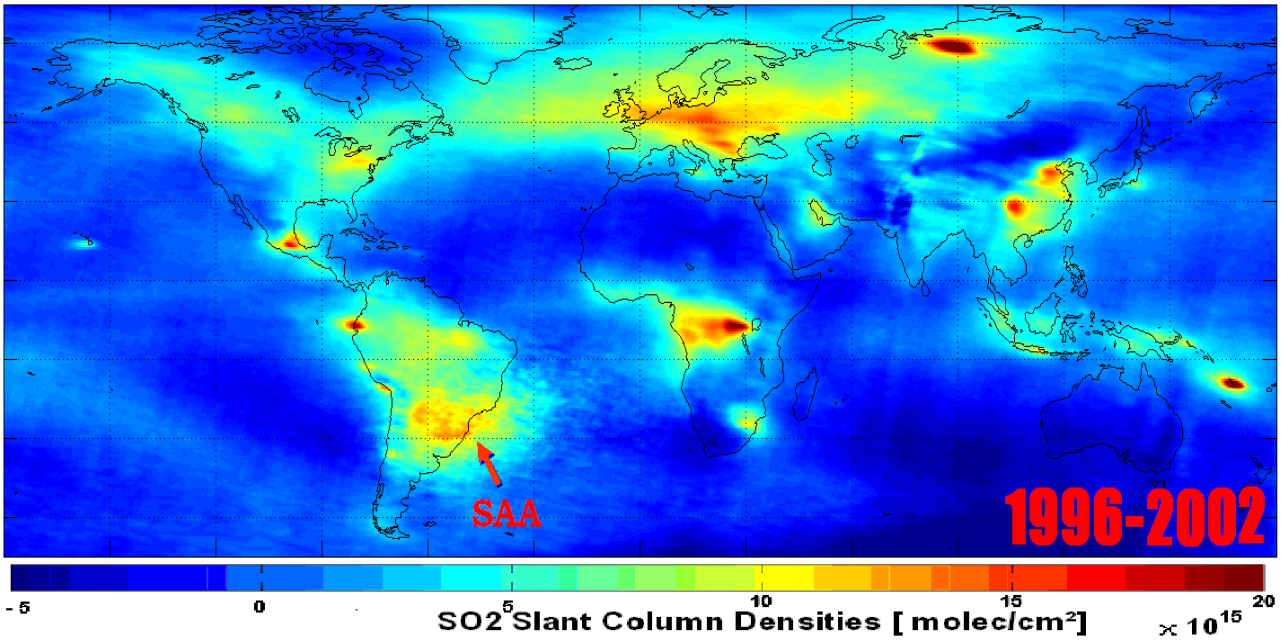

Fig. 4: 7 year mean map of SO_2 SCDs from 1996 to 2002; different volcanic activities like Nyamuragira from Congo-Zair, Etna (Sicily), Vanautu island and Popocatepetal from Mexico. Also regions of anthropogenic emissions like eastern coast of USA, South Africa, Norilsk (Russia) and Eastern Europe can be identified. Red arrow on figure indicates the region affected by South Atlantic Anomaly (SAA).

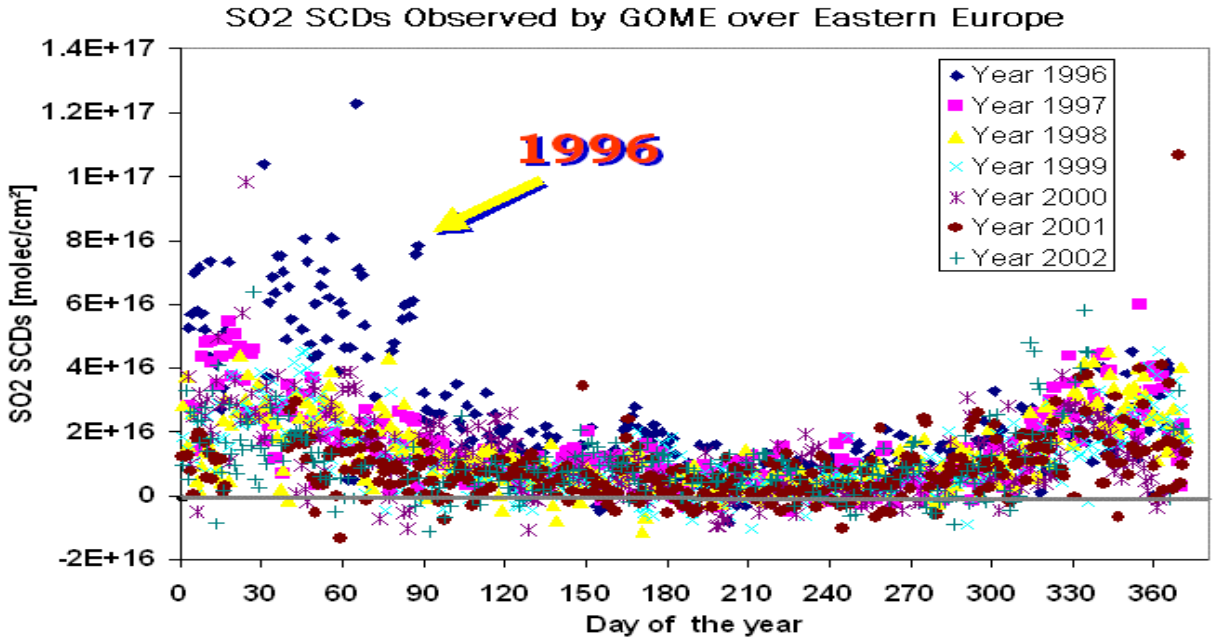

Fig. 5: Time series of SO_2 SCDs from Eastern Europe for the year 1996 to 2002
(Different colors represent the respective year)

6. Miyakejima (34.08N, 139.53E) and Bandai Honshu (37.6N, 140.1E), Japan

Bandai is a stratovolcano with a caldera in northern Honshu. GOME observed volcanic activity in August 2000. In August 2000, the Mount Oyama Volcano experienced its largest eruption in 17 years. GOME observed the SO_2 emissions during overpasses within the eruption period. See Figure (3C).

7. Central Islands (16.0° S, 168.5°E), Vanuatu

Lopevi, Yasur and Ambrym are the most active volcanoes from the Vanuatu region. Frequently SO_2 emissions are observed by GOME over this region. See Figure (3B, 3C).

8. Tavurvur Rabaul, new Britain island (4.3°S, 152.2°E) Papua New Guinea

The stratovolcano Tavurvur erupted sending thick clouds of ash over the town of Rabual in October 1998 and September 2000, GOME observed SO_2 emissions. See Figure (3B)

In addition to the above-mentioned volcanoes, GOME also observed SO_2 emissions from several other volcanic events like Mt. Cameroon, Montserrat, Stromboli, El Chichon, Mayon and Hawaiian volcanoes etc. All these observations demonstrate the capability of GOME instrument to monitor SO_2 emissions during volcanic eruptions and degassing scenarios over a longer period. GOME thereby contributes to the input data urgently needed for accurate modeling of the global sulfur budget [16].

4. SO_2 FROM ANTHROPOGENIC EMISSIONS

The industrial revolution increased the concentrations of greenhouse gases, aerosols and aerosol precursor gases. All of these have potential to alter the climate on global scale [17].

Anthropogenic SO_2 is much more difficult to detect than volcanic SO_2 because it is generally located at lower altitudes where the instrument's sensitivity is low [1].

In figure (4), examples for anthropogenic SO_2 emissions are shown, especially over China, Eastern USA, the Arabian Peninsula, Eastern Europe, South Africa and particularly Norilsk-Russia. In all these cases, burning of coal and smelting of metal ores are probably the source of the observed SO_2 SCDs. Also SO_2 emissions from biomass burning over central Africa, South East Asia and Amazon region can be clearly identified. GOME's ability to detect anthropogenic SO_2 emissions even at higher latitude particularly over Norilsk (Russia) makes it apart from the other conventional instruments carried by other spacecrafts.

4.1 Case Study: SO_2 observed Over Easter Europe

GOME enables us to monitor anthropogenic SO_2 emissions efficiently from various parts of the world. Here we present a case study of anthropogenic SO_2 emissions over Eastern Europe. We selected an area, 13°E to 30°E and 42°N to 52°N for the evaluation of time series from GOME measurements.

The time series showed high SO_2 SCDs during winter months, which are likely due to the higher rate of coal consumption for winter heating and also the good sensitivity of the instrument because of high surface albedo over snow covered regions.

In the beginning of the year 1996, SO_2 SCDs are relatively higher than the rest of years. As revealed from the NCEP data shown in Fig. 7, these winter months are relatively cooler as compared to rest of years with temperature anomaly of -3° to -4.5°C over the selected region.

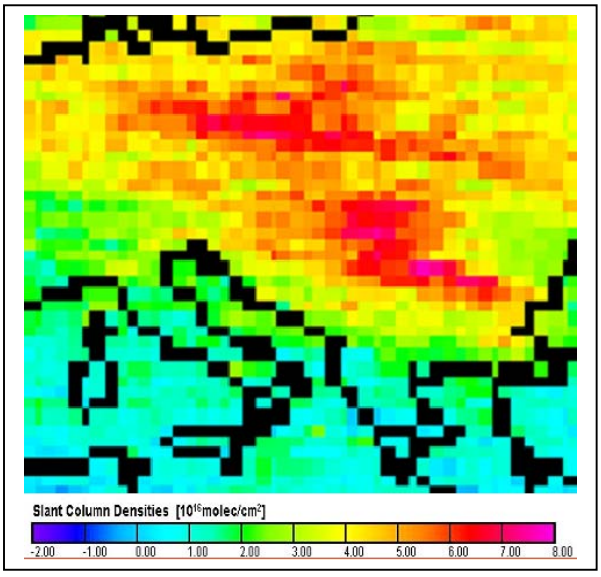

Fig. 6: Mean of SO_2 SCDs for the months January to March 1996 over Eastern Europe.

There is a good consistency in the mean of SO_2 SCDs for months January to March 1996 (Fig.6) and mean temperature anomaly for the same period (Fig.7) and same region. Therefore, the observed high SO_2 SCDs for the year 1996 are due to high rate of coal consumptions for winter heating and probably due to a longer period of enhanced surface albedo.

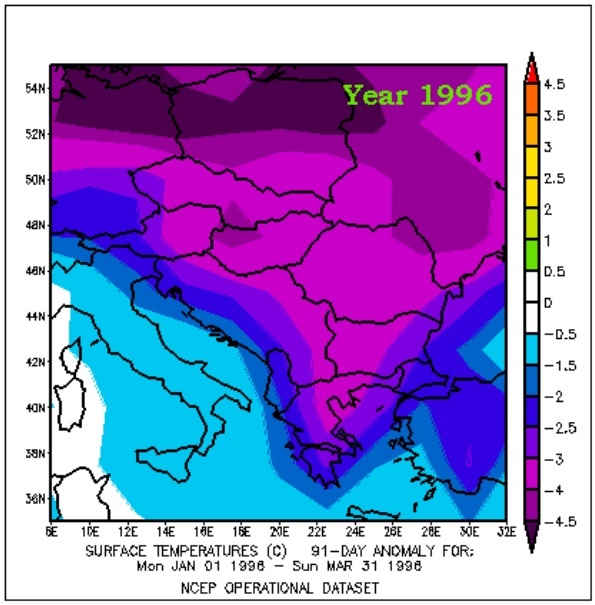

Fig. 7: Mean surface temperature anomaly for winter months (January- March) of the year 1996 over the selected region from Eastern Europe

5. CONCLUSIONS

Satellite remote sensing is a powerful tool to monitor SO_2 emissions from anthropogenic and volcanic eruptions especially. GOME on-board ERS-2 enabled us with the observations of atmospheric SO_2 for more than 7 years.
Selection of an appropriate instrumental slit function plays a key role in DOAS fitting analysis. By selecting asymmetric Voigt line shaped slit function for GOME Channel 2b we improved our analysis. Further improvements are achieved by using 'Ratio spectrum' and normalization of data set by Reference sector method (RSM).

Volcanic emissions are highly variable in space and time. To better quantify the impacts of volcanic eruptions on climate, there is a need to better constrain their sulfur emissions. GOME thereby contributes to the input data urgently needed for accurate modeling of the global sulfur budget [16].

The time series over Eastern Europe shows a seasonal cycle with higher SO_2 SCDs in winter months due to higher rate of coal consumptions and/or due to high surface albedo. Our results demonstrate the capability of GOME to monitor anthropogenic SO_2 emissions over a longer period.

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