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Tropospheric Sulfur Dioxide observed by the ERS-2 GOME Instrument

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Abstract. Since July 1995, the Global Ozone Monitoring Experiment (GOME) onboard ESA's 2nd European Remote Sensing Satellite (ERS-2) provides continuous spectral measurements of nadir backscattered earth radiances and solar irradiances in the UV/visible wavelength range. From these, column amounts of sulfur dioxide have been derived for the December 1996 eruption of Nyamuragira volcano, Zaire, and the ongoing activity of Popocatépetl volcano, Mexico. Maximal column densities of 33 DU for Nyamuragira and 6 DU for Popocatépetl have been found. Popocatépetl results compare well with the activity reported for this volcano. Furthermore, GOME observed enhanced SO_2 over Southeast Europe which is attributed to lignite combustion in local power plants. The results demonstrate a high sensitivity and specificity of this new sensor for sulfur dioxide. For cloud-free scenes at low solar zenith angles, the detection limit for SO_2 is estimated to be 0.4 DU.

Introduction

Sulfur dioxide enters the atmosphere as a result of both anthropogenic activity and natural phenomena. It is released directly into the troposphere in large amounts as a result of the combustion of fossil fuels, and to some extent by the oxidation of organic material in soils, the oxidation of dimethylsulfide (DMS) and H_2S over oceans, volcanic eruptions, and biomass burning.

SO_2 reacts rapidly with OH to form HSO_3, which reacts with O_2 to form SO_3. It is soluble in clouds and aerosols, where it reacts with H_2O_2. As a result of these processes, SO_2 is converted to H_2SO_4. In general the maximum concentration of SO_2 is close to its source and the amount of SO_2 decreases rapidly as the distance from the source increases, indicating a short tropospheric lifetime of typically a few days. Clean continental air contains less than 1 ppb SO_2, which corresponds to a total column density below 0.2 equivalent Dobson Units (1 DU = 1 matm cm) of SO_2 in a boundary layer of 2 km.

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 being of the order of several weeks. Injection of SO_2 by volcanic eruptions and its subsequent conversion to H_2SO_4 results in stratospheric aerosol formation.

Volcanoes are an important source of gases for the atmosphere. By their nature their emissions are sporadic and intermittent and often occur in uninhabited regions. Assessing the size of the gaseous and particulate emission from volcanoes by in-situ techniques is difficult if not impossible. Remote sensing measurements provide an opportunity to overcome this difficulty. As previously demonstrated using measurements by TOMS [*Krueger*, 1983; *Krueger et al.*, 1995, 1996] and SBUV/2 [*McPeters*, 1993], it is possible to measure SO_2 volcanic emissions using passive remote sensing in the UV.

This study describes the first SO_2 observations obtained from the new European satellite sensor GOME which combines the advantages of contiguous spectral sampling and contiguous ground coverage. The emissions from two volcanic case studies are presented: the African volcano Nyamuragira (1.41°S, 29.20°E, summit elevation 3058 m), and Popocatépetl volcano (19.02°N, 98.62°W, 5465 m) near Mexico City. The eruptions investigated did not have sufficient power to inject significant amounts of SO_2 into the stratosphere. In addition, first results on GOME observations of anthropogenic SO_2 will be presented.

The GOME Instrument on ERS-2

ERS-2 was launched in April 1995 into a near-polar sun-synchronous orbit at a mean altitude of 795 km. The descending node crosses the equator every 2800 km at 10:30 local time. GOME is a nadir-scanning double monochromator covering the 237 nm to 794 nm wavelength range at a spectral resolution of 0.17–0.33 nm. The spectrum is split into four spectral channels, each recorded quasi-simultaneously by a 1024-pixel Reticon photodiode array. Total ground coverage is obtained within 3 days at the equator by a 960 km across-track swath (4.5 s forward scan, 1.5 s back scan). For the measurements presented below, groundpixel size is 40×320 km^2 (40×80 km^2 before 11 March 1996). The solar irradiance is measured daily. GOME measurements are available since July 1995. Details of the overall scientific objectives of GOME, instrument concept, and some first scientific results are reported elsewhere [*Burrows et al.*, 1998, and references therein].

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Data analysis

The strong absorber ozone is GOME's primary target species. However, the column amounts of a number of important atmospheric trace gases which have comparably weak atmospheric absorptions, among them SO_2, can also be retrieved. SO_2 column densities are derived from the near-UV GOME radiance / irradiance measurements by a differential optical absorption spectroscopy (DOAS) algorithm [*Richter et al.*, 1998*a, b*] similar to that employed for ground-based zenith-sky measurements. The most important assumptions are that (i) Beer's law holds for the optical densities, (ii) Rayleigh and Mie scattering and albedo effects can be described by a broadband polynomial, and (iii) molecular absorption of ozone and SO_2, and Ring filling-in are the only relevant narrow-band effects. The temperature dependence of the SO_2 absorption cross sections is neglected. Empirical correction terms are not needed in the analysis.

The wavelength window 315.5–327.0 nm containing 100 detector pixels has been selected for SO_2 analysis. After the absorption signature of the ozone Huggins bands, which is typically one order of magnitude larger than the SO_2 absorption, has been subtracted from the measured optical density, a clear SO_2 signal is obtained in regions of elevated SO_2 (Figure 1). Slant column densities are then converted to vertical column densities by means of a simple geometric air mass factor, applicable for small solar zenith angles where the geometric light-path is an adequate approximation. The error induced by this assumption for tropospheric SO_2 is less than 20 %.

Case study 1: Nyamuragira volcano

GOME first observed SO_2 after an eruption of the Nyamuragira volcano, a shield volcano in the Virunga volcano field, Zaire / Democratic Republic of Kongo [*Eisinger et al.*, 1997]. According to the reports from the Global Volcanism Network (GVN), this eruption started on 1 December 1996. On 5 December the plume had reached 12 km altitude.

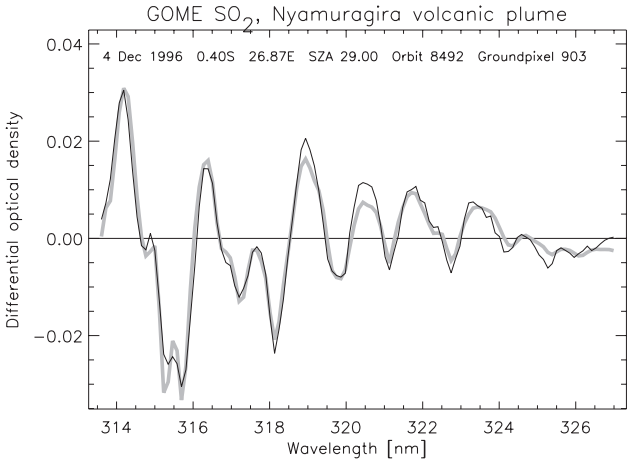

Figure 1. Differential laboratory reference spectrum of SO_2 measured by the GOME breadboard model (thick grey line) and GOME flight model measurement for a single groundpixel near Nyamuragira volcano (thin black line), after removing the contributions from scattering and ozone absorption.

The December 1996 Nyamuragira SO_2 plume could also be tracked by the two TOMS instruments onboard the Adeos and Earthprobe platforms [http://skye.gsfc.nasa.gov/].

Figure 2 displays vertical columns of SO_2, derived from the GOME radiances for the first 18 days of December 1996. The largest SO_2 column (33 DU) was observed on 1 December (immediately after the eruption started) for the groundpixel containing Nyamuragira. At the following GOME overpasses on 4 and 7 December the highest columns (around 18 DU) were still found near the volcano. It appears that SO_2 outgassing continued for at least two weeks, either continuously or in several large bursts. This is characteristic of an effusive eruption (in contrast to an explosive eruption), which is common for hot spot or rift volcanoes like Nyamuragira.

Throughout the observing period, the bulk of the SO_2 was transported to the west. The plume could be tracked as far as 2000 km to the west. Assuming tropospheric wind speeds between 5 and 10 m/s, this corresponds to a transport time of 2–5 days which is consistent with the lifetime of SO_2 in the troposphere.

SO_2 values well outside the Nyamuragira plume are typically between zero and 1 DU with low standard deviations (0.2 DU) indicating low background SO_2 where there are no strong local sources at this time of the year. By adding the SO_2 values, which exceed a threshold (e.g., 1.5 DU), set to delineate the plume, a lower limit of a few hundred kilotons (kt) can be derived for the December 1996 Nyamuragira eruption. In any case, the total SO_2 mass appears to be much lower than the 3 Mt observed by TOMS in the December 1981 eruption of the same volcano [*Krueger et al.*, 1996]. It is however comparable to its 1980 and 1986 eruptions.

Case study 2: Popocatépetl volcano

Popocatépetl, a large stratovolcano 70 km south-east of Mexico City, is the second highest and one of the most active volcanoes in Mexican history. After five comparably quiet decades, a new eruptive period began in December 1994. It is characterized by frequent exhalations of steam, ash, and gas with occasional minor explosions. On 30 June / 1 July 1997, the largest explosion since 1925 caused ash to fall on Mexico City.

The SO_2 measurements from GOME within 300 km of Popocatépetl are presented in Figure 3 from July 1995 to April 1998. These can be compared with the Popocatépetl activity reported by the GVN. Emissions of SO_2 are readily observable by GOME in the well reported period of March and April 1996. Maximum column densities, approximately 6 DU, were observed on 8 March 1996 and 1 July 1997, coinciding with the two largest eruptions reported by the GVN. GOME SO_2 columns are low from the beginning of the observations until February 1996. Throughout 1996 and sporadically in 1997, relatively large emissions from Popocatépetl were observed, in particular in the period from June to October 1997. We propose that the March 1996 event may have opened a vent which continued to outgas SO_2 through the summer and autumn of 1996.

In assessing the total amount of SO_2 emitted by Popocatépetl it is more difficult to establish a threshold because the large population of Mexico City and surrounds leads to a significant anthropogenic contribution to the total SO_2.

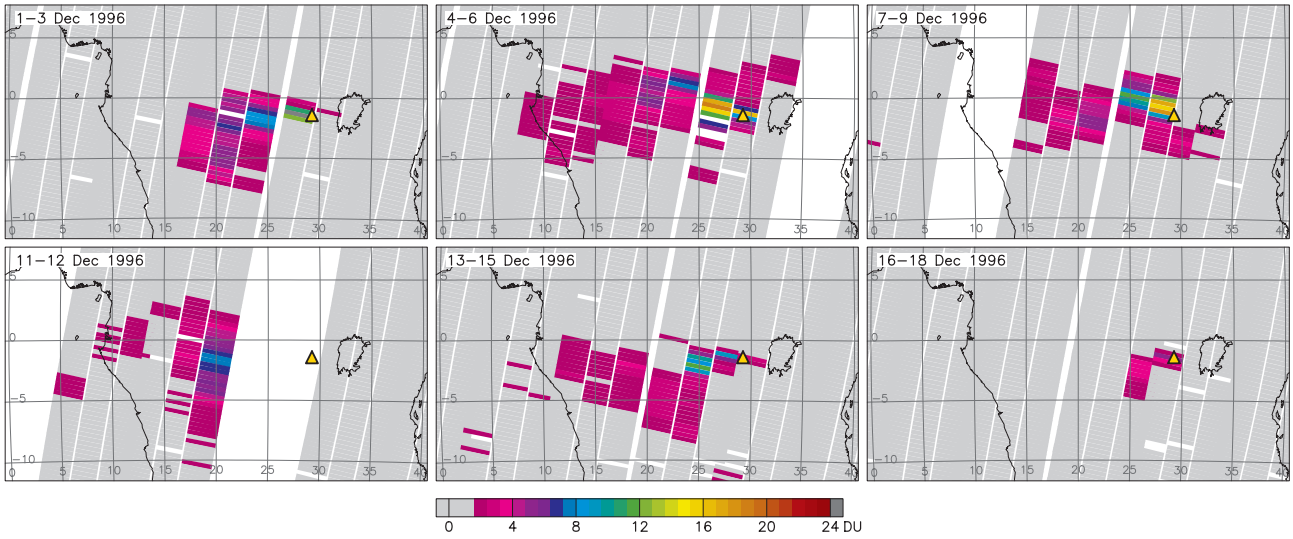

Figure 2. Time evolution of the SO_2 plume over Nyamuragira volcano (yellow triangle) during its December 1996 eruption. In order to visualize the extent of the volcanic SO_2 plume, three-day composites giving full ground coverage are shown. No background correction has been applied. GOME data are not available for 10 December. Before 1 Dec and after 18 Dec, no SO_2 above background levels could be observed.

Subtracting averaged background signals for each orbit and summing over all groundpixels within a 300 km distance from the volcano gives cumulated SO_2 masses of 230 kt for 1996, and 110 kt for 1997. As missing days, chemical losses, and SO_2 transported outside the 300 km radius are not accounted for, these values should be interpreted as lower limits for the Popocatépetl emissions.

Case study 3: Anthropogenic pollution event

Patches of enhanced SO_2 have been observed by GOME over the Balkans on 8 February 1998 (Figure 4). On this day a high pressure region was centred near Sofia [*Berliner Wetterkarte*, 1998] leading to favourable observing conditions.

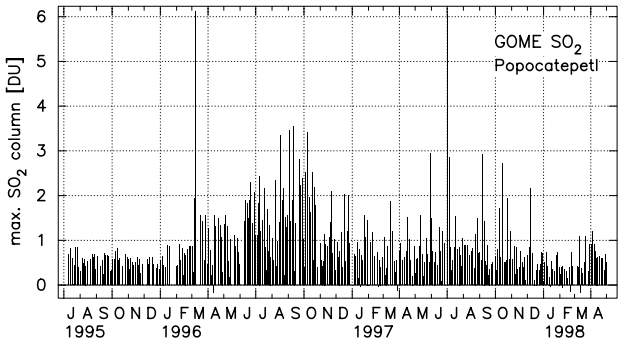

Figure 3. SO_2 vertical columns over Popocatépetl derived from the GOME radiances. For each overpass (approximately every third day), the maximum vertical SO_2 column in a 300 km region around Popocatépetl is shown. The average of the retrieved SO_2 columns between 300 and 2000 km distance of the volcano has been subtracted for clarity. This background correction amounts to (0.6±0.2) DU.

From the GOME broadband spectral measurements, the scene is classified as cloud-free, which is in agreement with the station reports from Belgrade and Bucharest. The meteorological situation favoured accumulation of pollutants.

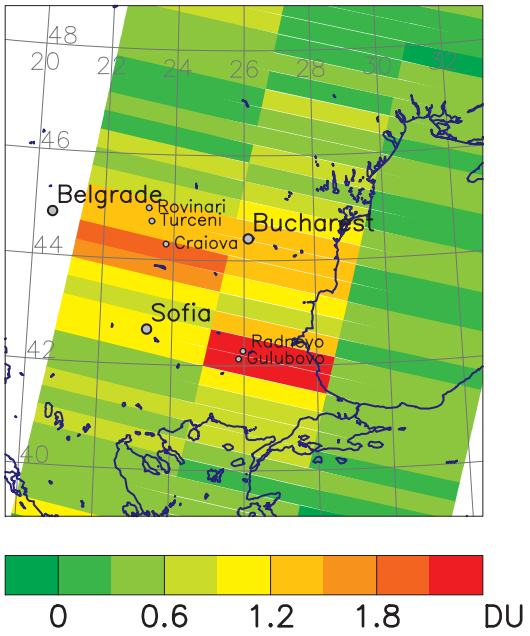

Figure 4. Enhanced SO_2 observed by GOME over the Balkans on 8 February 1998. The small dots indicate the locations of the largest thermal power plants burning lignite in Romania (Turceni, Rovinari, and Craiova, total 5065 MWe) and Bulgaria (Gulubovo and Radnevo, forming the Maritsa-Iztok complex, total 3485 MWe). Together, these plants represent 27 % of the electric power capacity installed in these countries [US Department of Energy].

Upward dispersal was prevented by a stable inversion, and SO_2 removal by wet deposition was effectively suppressed. Horizontal wind speeds were below 2.5 m/s, making other than local SO_2 sources improbable.

The largest SO_2 columns (up to 2.6 DU) have been found for the groundpixels containing Bulgaria's and Romania's major thermal power plants running on locally produced sulfur-rich lignite. We conclude that the enhanced SO_2 observed by GOME is mainly emitted by these power plants. This is consistent with the fact that public power generation is considered to contribute the major fraction to the SO_2 emissions in Bulgaria and Romania [European Environment Agency, CORINAIR database].

Discussion

The detection limit for SO_2 retrievals from GOME measurements depends on both the observing conditions and the retrieval algorithm. For cloud-free scenes at low solar zenith angles, a reasonable estimate is 0.4 DU in the vertical column, corresponding to 150 t of SO_2 in a $40 \times 320\,km^2$ GOME groundpixel. This comparably high sensitivity arises from the multi-wavelength advantage of GOME. Furthermore, interferences from varying ground albedo, aerosol and ozone content can be cleanly separated from the SO_2 signal, leading to highly specific measurements.

Sensitivity to tropospheric SO_2 is expected to be lower at high latitudes where less UV radiation penetrates to the surface. Similarly, larger effective absorptions of ozone are likely to result in a somewhat poorer detection limit. However, it has been demonstrated above that heavy and sustained anthropogenic SO_2 pollution events may be observed using GOME data. Further work is required to establish the detection limit at higher latitudes.

In addition to the two volcanoes presented in detail, GOME observed SO_2 also on several occasions above Etna (Sicily), Soufriere Hills (Montserrat), and Ambrym (Vanuatu).

The Popocatépetl timeseries demonstrates the capability of GOME to monitor volcanic degassing over a longer period. GOME thereby contributes to the input data urgently needed for accurate modelling of the global sulfur budget [*Graf et al.*, 1997]. For an accurate assessment of the SO_2 budget the short lifetime of SO_2 requires that the GOME measurements be coupled with models.

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