QUANTITATIVE ANALYSIS OF VOLCANIC SULFUR DIOXIDE EMISSIONS USING GOME BACKSCATTER MEASUREMENTS

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ABSTRACT

We analysed atmospheric UV backscatter spectra obtained by the Global Ozone Monitoring Experiment (GOME) spectrometer on board the ESA European Remote Sensing Satellite (ERS-2) after the last major eruptions of Mt. Etna on Sicily (Italy) in July/August 2001 and October/November 2002. Sulfur dioxide columnar amounts were retrieved from satellite measurements and ground-based measurements. The analysis revealed that information about the plume height of volcanic eruptions and aerosol parameters is necessary for a reliable quantitative retrieval of sulfur dioxide from space-borne sensor data at periods perturbed by volcanic eruptions.

1 INTRODUCTION

A major natural contribution to the global sulfur dioxide budget originates from volcanic activity (4). A relatively silent phase of Mt. Etna on Sicily (Italy) finished in November 1999 and since then, several major outbreaks of Europe's largest and most active volcano were registered. The eruptions in July/August 2001 and the more powerful outbreaks in October/November 2002 were also observed by the GOME spectrometer.

First SO_2 column measurements from space using the Total Ozone Mapping Spectrometer (TOMS) were presented by (6) and limitations of TOMS were discussed in a later paper by (7). First examples of GOME's capability to measure total atmospheric SO_2 columns after volcanic eruptions were shown by (3).

In general, the retrieval of volcanic sulfur dioxide emissions from space suffers from the sparse temporal and spatial coverage of actual satellite sensors but also from clouds in the troposphere. The short tropospheric lifetime of SO_2 of several days only and the separation of the background SO_2 content from volcanic emissions also hampers the estimation of the volcanic source strength. Other typically unknown but important parameters are the aerosol loading in and around the volcanic plume and the height above ground level of emissions (particles and gases). We show that the combination of trajectory analysis, ground-based measurements and GOME observations can be used to confirm the presence of large amounts of sulfur dioxide of volcanic origin away from the volcano. For the first time, GOME-derived SO_2 total columns were successfully validated against ground-based measurements from a Brewer spectrophotometer.

2 GOME INSTRUMENT AND DATA PROCESSING

The GOME spectrometer, an atmospheric chemistry instrument on board ESA's ERS-2, is able to measure the content of a number of minor atmospheric trace constituents including sulfur dioxide. GOME is a nadir-looking across-track scanning instrument with a typical footprint size of about 320×40 km^2. It measures the back-scattered radiation from the earth-atmosphere system between 240 nm and 790 nm with a moderately high spectral resolution of about 0.2 nm to 0.4 nm. In addition, a sun spectrum is recorded every day via a diffuser plate. The GOME instrument is described in detail e.g., in (2).

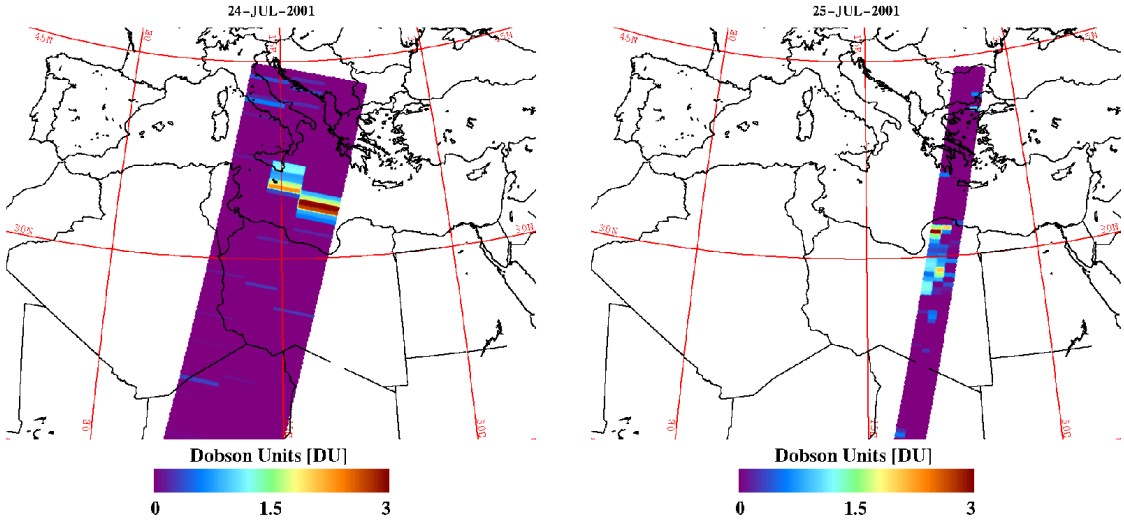

Fig. 1. Slant columns of sulfur dioxide over the Mediterranean Sea as retrieved from GOME measurements on July 24th 2001 (left panel) and July 25th 2001 (right panel). The smaller GOME footprint on July 25th 2001 is due to narrow scan operations where the ground pixel size is 80 $\times$ 40 km^2 only. The observed maximum slant column content on July 24th 2001 was around 3.7 DU (Dobson Units).

In our study we used an enhanced version of the GOME Data Processor (GDP) as described in detail in (9). The trace gas retrieval is based on a DOAS (Differential Optical Absorption Spectroscopy) fitting technique that provides trace gas slant column amounts along the viewing path of the instrument (2). The sulfur dioxide retrieval was performed between 315.8 nm and 327 nm (around 120 spectral points). Slant columns are finally converted to geometry-independent vertical column amounts through division by appropriate air mass factors (AMF) that originate from radiative transfer simulations (8). AMFs depend on the viewing geometry, on the reflection properties of the underlying surface (ground or clouds), the height above sea level (ground or cloud-top), the aerosol optical properties, and the atmospheric profiles of temperature, pressure and trace gas concentrations.

3 SLANT COLUMN RESULTS

3.1 July/August 2001

The meteorological conditions during the eruptions in 2001 were characterized by a stable high pressure system over the Southern Mediterranean and relatively constant westerly winds. Cloud coverage was below 10% throughout the entire period, providing optimum observation conditions. The most active phase of Mt. Etna began on 20th July 2001 and a direct overpass of GOME on 24th July 2001 clearly showed an SO_2 plume over the Mediterranean (Fig. 1, left panel) between Sicily and the Libyan coast. The maximum slant column content was around 3.6 DU $\pm$ 1.05 DU, which is around 10 times higher than the typical background content. On July 25th, enhanced levels of SO_2 were even found over the Saharan desert in Libya (Fig. 1 right panel). A second major eruption was observed on July 27th where SO_2 column densities of 5.25 DU $\pm$ 1.05 DU were detected. Emissions became lower on July 31st and fell back to pre-eruptive levels after August 6th.

3.2 October/November 2002

The meteorological conditions during the eruptions in fall 2002 were more variable. Also the overall cloudiness in the Mediterranean area was higher than during the year before. GOME data of 29th October 2002 was analyzed and maximum values of about 12.2 DU $\pm$ 1.16 DU were found close to the volcano. In the following, north-easterly winds transported the bulk of SO_2 to the Algerian coast (Fig. 2, left panel). From 31st October 2002 to 1st November 2002 the wind direction changed to West and enhanced SO_2 levels were found over parts of Greece and the Aegean sea on the following days (Fig. 2, right panel). On 2nd November 2002 enhanced levels of SO_2 were measured over Greece while another major eruption was observed on 4th November 2002 (Fig. 3).

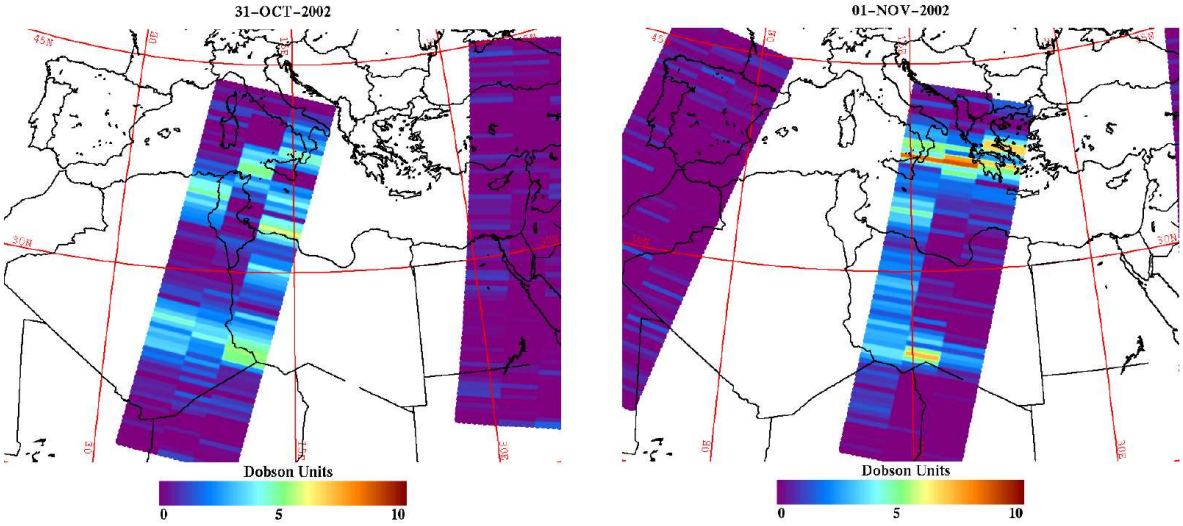

Fig. 2. Slant columns of sulfur dioxide over the Mediterranean Sea as retrieved from GOME measurements on October 31th 2002 (left panel) and November 1st 2002 (right panel). The observed maximum slant column content on November 1st 2002 was around 9.3 DU (Dobson Units).

4 AIR MASS FACTOR CALCULATION

Air Mass Factors for SO_2 vary by 15% (ground albedo 0.05, sun zenith angle $< 60°$) across the spectral fitting window between 315 nm and 327, that is dominated by strong ozone absorption in the Huggins bands. Results from a closed-loop test using pure synthetic data indicate that an AMF calculated around 320 nm is suitable to reproduce the input total content within 1%. The approximation breaks down for larger solar zenith angles because the retrieval then suffers from the large path length in stratospheric levels where ozone absorption is dominant, which considerably reduces the penetration depth of photons in the UV. As long as the ground albedo remains low (values in the UV are typically below 0.1), AMFs decrease rapidly to values even below 0.5 which indicates that GOME is no longer sensitive to sulfur dioxide in the lower troposphere.

The calculation of appropriate AMFs for species with major tropospheric loading (e.g., SO_2, NO_2) requires knowledge about its vertical distribution. Since the actual concentration profile is commonly unknown, climatological information is used instead and this may cause significant errors in the calculated column densities. External information about the trace gas profiles is therefore in favor. We studied the sensitivity of AMFs by modifying a standard SO_2 profile by adding sulfur dioxide plumes at 2 km, 3 km, 4 km, 5 km, and 6 km above sea level.

The aerosol loading (optical thickness, aerosol type) has an impact on calculated sulphur dioxide AMFs and column densities. These parameters are typically not independent from the height of the sulfur dioxide plume (7) and the optical properties of volcanic aerosol and mineral aerosol differ remarkably from that of e.g., maritime or urban components. The simulation of sulphur dioxide AMFs during and after volcanic outbreaks requires therefore knowledge about the plume height and the aerosol extinction properties. A sensitivity analysis on the aerosol loading and the plume height was performed which is described in detail in a corresponding paper by (11). Major results are that a moderate aerosol optical thickness of 0.3 causes changes of AMFS in the order of $\pm$5%, relative to an aerosol-free scenario and only slightly varying with the aerosol type. A higher aerosol loading (aerosol optical thickness 0.7) causes lower sulfur dioxide AMFs and changes up to -20% occur if a desert-like aerosol loading is present in the lower troposphere. Relative changes of AMFs with plume height remain small but an almost linear increase by a factor of 2 of absolute AMFs with varying plume height from 2 km to 6 km is observed. An improper knowledge of plume height will therefore cause large uncertainties in retrieved total sulfur dioxide column values.

5 TOTAL COLUMN RESULTS AND VALIDATION

5.1 Trajectory analysis

We investigated meteorological analysis data and data from other space-borne sensors that can provide information about the a priori unknown plume height of volcanic emissions. The APOLLO retrieval scheme (5) applied to AVHRR (NOAA-14, NOAA-16) data classified Etnas smoke and ash plumes as mid-level cloud between 700 hPa and 400 hPa above sea level. However, our sensitivity analysis gave evidence for a more precise knowledge of the plume height. We therefore analyzed forward and backward trajectories that were calculated using the FLEXTRA

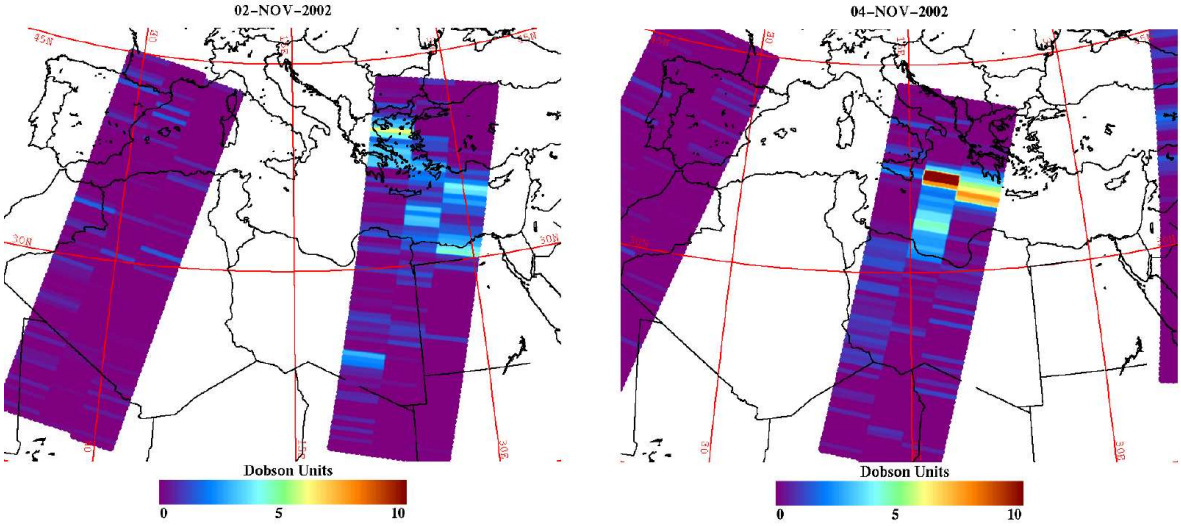

Fig. 3. Slant columns of sulfur dioxide over the Mediterranean Sea as retrieved from GOME measurements on November 2nd 2002 (left panel) and November 4th 2002 (right panel). The observed maximum slant column content on November 4th 2002 was around 13 DU (Dobson Units).

model (10). Ensembles of 3D forward trajectories were released at Mt.Etna at different pressure levels from 700 hPa to 200 hPa. By matching the parcel trajectories with the first guess GOME SO_2 retrievals the height of the SO_2 plume is determined. Forward runs started on 24th July 2001 at 1020 UTC at Mt. Etna. It could be shown that sulfur dioxide was transported within 24 hours to South-East directions between 500 hPa and 700 hPa. Enhanced levels of sulfur dioxide at the Libyan coast (see Fig. 1) can therefore be attributed to emissions from Mt. Etna on 23rd July 2001 and 24th July 2001. This is confirmed by consistent results from corresponding backward trajectories that were released at SO_2 spots. Similar results of the second event in Fall 2002 showed that strong westerly winds transported the emissions of Etna between 600 hPa and 400 hPa within 24 hours to regions easterly of Greece (Fig. 4). In general, volcanic emissions in fall 2002 were emitted into higher atmospheric levels and observed sulfur dioxide levels were higher, which underlines that these eruptions were more powerful than in summer 2001.

5.2 GOME measurements and error budget

Sulfur dioxide AMFs and vertical columns were calculated taking into account the information from the trajectory analysis (SO_2 plume height) and from lidar measurements (fall 2002 only). The summer scenarios were calculated using an average single SO_2 profile with a peak at 4 km above sea level while the fall scenario has a relative maximum of SO_2 at 5 km above sea level. Corresponding aerosol extinction profiles were set accordingly. The most plausible maritime aerosol model (aerosol optical thickness 0.3) was applied in the PBL for AMF calculations. On average, the impact of improperly known aerosol properties on calculated trace gas total columns is in the order of $< 10\%$, provided that the aerosol optical thickness remains below 0.3.

In the area of interest AMFs vary smoothly with the solar zenith angle. For the summer scenario SO_2 AMFs are between 1.4 an 1.6 while AMFs increase in fall 2002 to values around 1.7 to 2.0. AMFs my even be lower close to the volcano, due to the presence of large particles in the plume. Application of simple geometric AMFs (2.3 - 2.7) however will systematically underestimate total columns.

Application of trajectory analysis technique may considerably improve the knowledge of plume height but the remaining uncertainty will typically be in the order of $\pm$ 1 km. If the uncertainty of the SO_2 plume height is assumed to be in this range, the corresponding uncertainty of AMFs over water surfaces (maritime aerosol model with aer.opt. density 0.3 assumed) is around $\pm$ 15% (summer 2001) and $\pm$ 12% (fall 2002). Total error levels of SO_2 columns are not better than $\pm$ 30% for the first episode in Summer 2001 while errors are in the order of $\pm$ 20% for the second episode in fall 2002. A possible bias of at least 12% needs to be taken into account for insufficiently known profile data, mainly the plume height. A further uncertainty of about $\pm$ 30% must to be assumed for AMFs of pixels close to the volcano, due to the insufficiently known aerosol composition of the volcanic ash layer.

5.3 Comparison of space-borne data and ground-based results

Ground-based measurements of sulphur dioxide (Brewer spectrophotometer) and an aerosol extinction profile taken from Lidar measurements of a Raman Lidar at Thessaloniki (Greece) are partially available for the second active

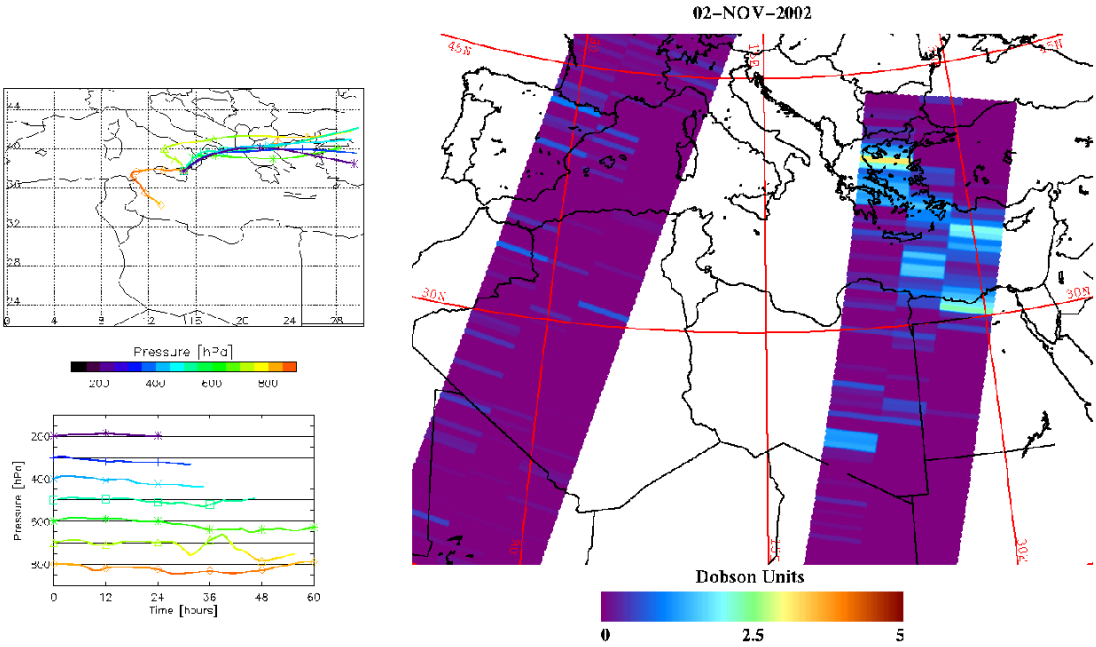

Fig. 4. Forward trajectory analysis started on 1st November 2002 (1020 UTC) at Mt. Etna (upper left panel) and corresponding height levels as a function of time (lower left panel). The forward trajectories released at 400 hPa and 500 hPa are crossing the region around Thessaloniki (Greece) indicated by the red symbol (see also Fig. 3). Trajectories stop in the lower panel if the given geographic area is left. Vertical columns of sulfur dioxide over the Mediterranean Sea as retrieved from GOME measurements on November 2nd 2002 are shown on the right panel. The observed maximum content was around 3 DU (Dobson Units).

phase of Mt. Etna in fall 2002 (1). The nighttime lidar measurements from 31st October 2002 to 1st November 2002 indicated the presence of volcanic aerosols by an increased aerosol extinction coefficient between 4500 m and 5500 m above sea level. GOME measurements show enhanced levels of SO_2 over Thessaloniki on 2nd November 2002 (Fig. 3), which happens, as already noticed by (12) in a recent paper about the important role of Mt. Etna for the SO_2 budget above Northern Greece. Forward trajectories were released from Mt. Etna between 300 hPa and 700 hPa on 1st November 2002 at 1020 UTC and the region of Thessaloniki was reached by the 400 hPa and 500 hPa trajectories 24 hours later (Fig. 4). The Brewer measurements indicate an SO_2 background content of about 2-4 DU before the arrival of the volcanic plume which increases to 5-7 DU on 02 November 2002 (see Fig. 5). The additional volcanic SO_2 content is therefore in the order of 3 DU which is similar to corresponding GOME results (Fig. 3). Note also that GOME pixels cover a much larger area (here: large parts over the Aegean Sea).

6 CONCLUSION

We retrieved the atmospheric sulfur dioxide content from GOME backscatter measurements during two eruptive periods of Mt. Etna. We could show that the large uncertainty of SO_2 total column measurements can be improved if information about the SO_2 profile and especially the plume height of volcanic emissions is available from other sources. A knowledge of the plume height not better than 1 km causes an uncertainty of derived SO_2 values in the order of 15%. Also the aerosol loading is of some importance if the aerosol optical thickness becomes larger than 0.3. The large GOME footprints however make it unlikely that such values occur frequently. The knowledge of the aerosol loading in the PBL is more important if desert aerosol components are likely to be present.

GOME results from the eruptive period of Mt. Etna in fall 2002 were compared against ground-based data from the Brewer station at Thessaloniki (Greece). Forward and backward trajectory analysis data confirmed the presence of volcanic emissions from Mt. Etna in the region of Thessaloniki. As expected, the SO_2 total columns measured from ground were higher than corresponding GOME measurements but the relative increase of SO_2 levels was in the same order as GOME total column results. Measurements of the global volcanic SO_2 budget with space-borne instruments like GOME are therefore limited, but re-analysis of historical data can be performed by using information from external sources. Instruments like the upcoming GOME-2/METOP will improve the situation through a better spatial and temporal coverage, but the need for auxiliary data remains until more advanced retrieval methods for e.g., the SO_2 profile and the aerosol loading from these sensors are available.

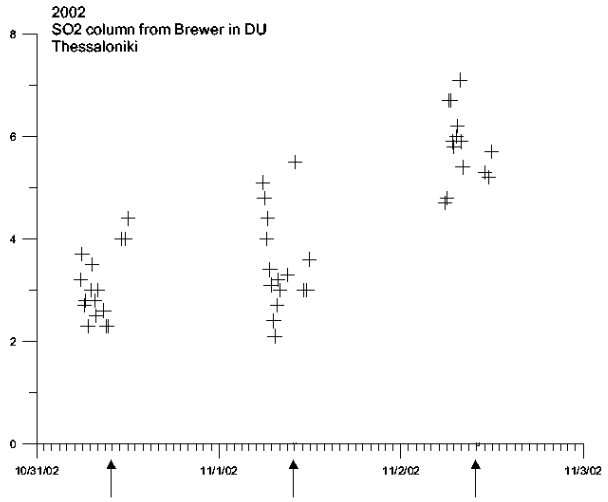

Fig. 5. Brewer measurements of sulfur dioxide from Thessaloniki station between 31 October 2002 and 02 November 2002. Vertical arrows indicate the GOME observation time. The increase of sulfur dioxide levels from 2-4 DU on 31 October 2002 to 5-7 DU on 02 November 2002 is consistent with GOME total column measurements of about 3 DU (see text).

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