Monitoring Changes in Tropospheric Constitution from Space

A contribution to subproject ACCENT-TROPOSAT-2 (AT2), Task Group 1

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Summary

Data from the two UV/visible instruments GOME and SCIAMACHY has been analysed for tropospheric SO_2 columns. A number of sensitivity studies was performed, and potential error sources were identified and partly quantified. The data have been compared to GOME measurements and good overall agreement was found with the exception of China, where a large increase is observed in the last years.

To improve the retrieval of SO_2 for large volcanic eruptions, sensitivity studies were performed on the dependence of airmass factors on altitude, wavelength, surface albedo, and SO_2 amount. It was found that for large SO_2 columns, the sensitivity depends strongly on wavelength, an effect that potentially can be used to derive SO_2 profile information. The results of the studies were confirmed in SCIAMACHY measurements during the massive Sierra Negra eruption.

Introduction

Sulphur dioxide is an important trace gas in the troposphere. It contributes to acid rain and is a key precursor for sulphuric acid aerosol formation. At high concentration, it also adversely affects human health, in particular in combination with fog (London smog). Both anthropogenic and volcanic sources contribute to the emissions. SO_2 is emitted mainly from coal burning, combustion of fossil fuels in general, but also from non-ferrous smeltering. Emission controls have greatly reduced anthropogenic emissions in some regions such as Europe and the US, but increased use of fossil fuels leads to increasing emissions in other areas e.g. Asia. Natural emissions are mainly from volcanic sources, from quite degassing as well as from explosive eruptions. The latter can pose a threat to both local population and to air travel. Therefore, global observations of SO_2 are relevant for environmental control, tropospheric chemistry, and climate change.

SO_2 can be measured by remote sensing of scattered sunlight both from the ground and from space. Continuous monitoring of volcanic SO_2 is performed using the TOMS satellite instrument (*Krueger et al, 1995)*, and for very large SO_2 concentrations, anthropogenic sources could also be observed. Owing to its higher spectral resolution and larger wavelength coverage, the GOME instrument provides improved sensitivity in particular to anthropogenic sources such as large power plants which emit SO_2 in the boundary layer (*Eisinger and Burrows*, 1998, *Khokar et al.*, 2005). By applying the SO_2-TOMS algorithm to OMI data, a more complete picture can be obtained owing to the higher spatial resolution and improved coverage (*Krotkov et al.*, 2006). In the infrared spectral region, SO_2 can also be measured (*Carn et al.*, 2005) but the sensitivity is highest in the middle troposphere and the measurements usually do not penetrate the boundary layer.

Scientific activities

Analysis of SCIAMACHY data

The SO_2 analysis for SCIAMACHY is based on the retrieval developed for the GOME instrument, and is described in *Afe et al.* (2004). Briefly, the fitting window for the DOAS fit is 315 - 327 nm, and absorption cross-sections of ozone at two temperatures, SO_2, the Ring effect, undersampling, and the polarisation dependency of the SCIAMACHY instrument are included. Daily solar irradiance measurements taken with the ASM diffuser are used as background spectrum. As SO_2 columns from both GOME and SCIAMACHY suffer from latitude dependent offsets, the data are normalized using a reference sector method similar to that employed for NO_2 by subtracting data taken on the same day at the same latitude in the 180° - 230° longitude region.
An important aspect of the retrieval is the conversion of the DOAS fit result, the slant column which provides the integrated absorber amount averaged over all contributing light paths through the atmosphere to the vertical column. To do this, a priori information is needed on surface albedo, aerosol loading and most importantly the vertical profile of SO_2 in the atmosphere. In particular the latter is usually not known, and has either to be taken from independent measurements as e.g. discussed in *Thomas et al.*(2005) or from climatologies or model results. Here, a simplified volcanic profile with a maximum between 10 and 15 km is used globally. This leads to systematic underestimation of SO_2 columns in regions with emissions close to the surface and to an overestimation over bright surfaces such as ice or clouds. This aspect of the retrieval is still very preliminary and will be improved in the future.

SO_2 Sensitivity Studies

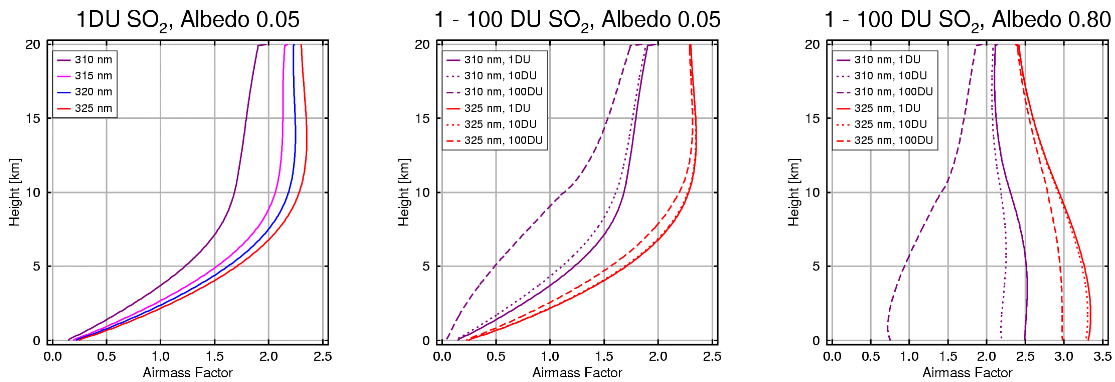

Fig. 1 Vertical sensitivity of nadir SO_2 measurements at 40° solar zenith angle in a Rayleigh atmosphere as function of surface albedo, wavelength and total SO_2 column.

The sensitivity of the measurements depends critically on surface albedo and the vertical profile of SO_2. In addition, the sensitivity also changes with wavelength as Rayleigh scattering increases to shorter wavelengths. A special situation occurs during large volcanic eruptions, when SO_2 itself becomes a strong absorber, and the sensitivity depends not only on where the SO_2 is located, but also on how much SO_2 is above the layer of interest.
This is illustrated in Fig. 1, where the results of a series of radiative transfer calculations performed with SCIATRAN are shown. Assuming a Rayleigh atmosphere without aerosols, the vertical sensitivity (airmass factor) is displayed for different surface albedo (0.05% and 0.80%) and at several wavelengths. Also, the effect of changing the total amount of SO_2 in the troposphere is investigated by using scenarios with 1, 10, and 100 DU of SO_2 well mixed in the lowest 10 km.
The main results of the sensitivity studies are as follows:

- For dark surfaces, sensitivity below 10 km decreases and is very low close to the ground. This is the result of strong Rayleigh scattering that reduces the number of photons penetrating low in the atmosphere, and accordingly is more pronounced at smaller wavelengths.
- If the amount of SO_2 is increased, the sensitivity is decreased, but much more so at the smallest wavelengths.
- Over bright surfaces, sensitivity to the lower layers is actually increased compared to higher latitudes at all wavelengths.
- The dependence on SO_2 column amount is similar over dark and bright surfaces.

Therefore, by evaluating the ratio of SO_2 columns retrieved at different wavelengths, some information on the vertical distribution can be obtained in the case of large SO_2 columns.

Scientific results and highlights

Comparison of GOME and SCIAMACHY measurements

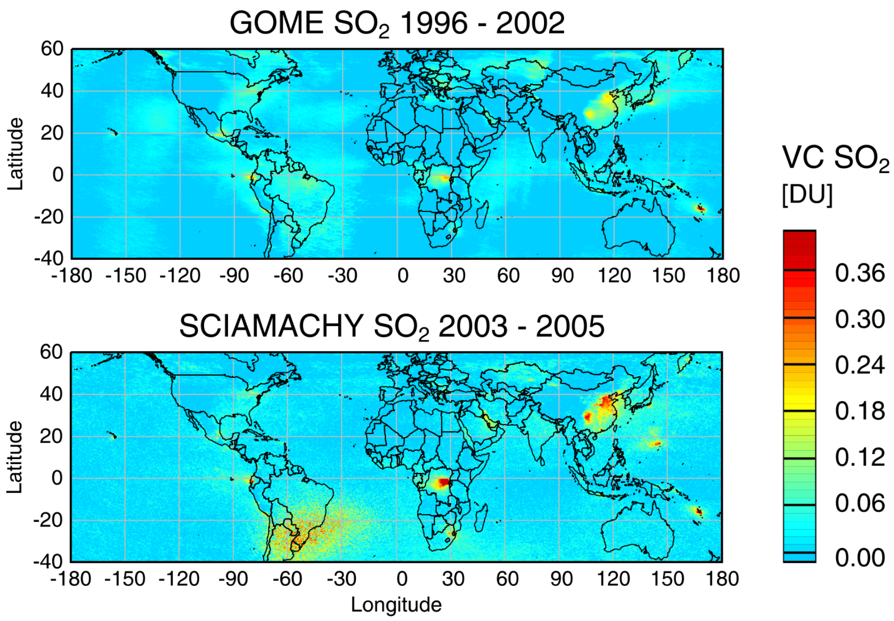

Fig. 2: Long-term averages of GOME (upper panel) and SCIAMACHY (lower panel) SO_2 measurements. As an airmass factor appropriate for volcanic eruptions was used, the absolute columns are too low over polluted areas by about a factor of two.

In Fig. 2, the long term averages for SO_2 from the GOME and SCIAMACHY data are compared. As can be seen, the overall agreement is good with a number of differences:

- SCIAMACHY measurements appear less “stripy”, a known problem in the GOME SO_2 data related to changes in the solar spectrum used.
- The large scatter in the Southern Atlantic Anomaly region in the SCIAMACHY data which is the result of a lower per pixel signal to noise ratio.
- Some differences in SO_2 column over volcanic regions which are related to variations in volcanic activity.
- Higher SO_2 columns over China.

On a daily or monthly average, the much higher spatial resolution of the SCIAMACHY measurements is apparent, but in the long-term average, this is effect smoothes out to a large degree. In particular the increase of SO_2 over China is intriguing and possibly related to the increases in NO_x emissions over that area which is also observed in GOME and SCIAMACHY data (*Richter et al.*, 2005), It therefore will be studied in more detail during the next year.

Analysis of SCIAMACHY SO2 columns during the Sierra Negra eruption

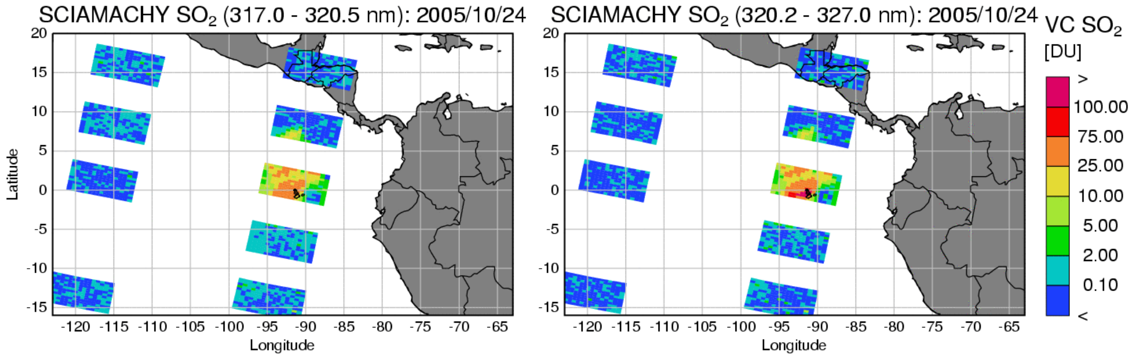

Fig. 3 SCIAMACHY measurements of SO_2 columns from the Sierra Negra Eruption using two different fitting windows. While the overall agreement is very good, the analysis in the 317 - 320 nm window (left) strongly underestimates the SO_2 columns in the vicinity of the volcano. Please note the non-linear colour scale.

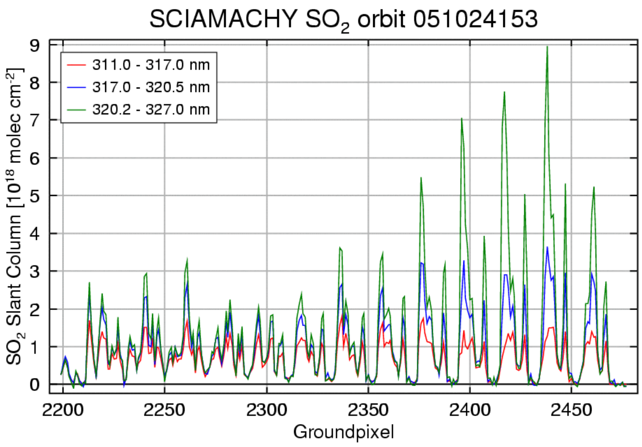

Fig. 4: SO_2 slant columns for the state covering the Sierra Negra eruption on October 24th (see Fig. 3). Here the values are displayed as function of the time of measurement. While for areas with low SO_2 the results from the three different fitting windows agree well, the increase over the areas with the largest columns is most pronounced at 320 - 327 nm and not seen at all at 310 - 317 nm.

On October 22nd, 2005, the Sierra Negra Volcano on the Galapagos Islands erupted and emitted large quantities of SO_2 into the troposphere. Emissions continued until the end of October and could be observed in SCIAMACHY data but also in TOMS and OMI measurements (*N. Krotkov and Arlin Krueger, personal communication*).

Using the standard SO_2 analysis described above results in very poor fits for the large SO_2 columns observed on the first days of the eruption. Closer inspection of the fit residuals showed, that the optical depth of the SO_2 bands changes by more than a factor of two over the fitting window. Therefore, smaller retrieval windows were used in a more detailed analysis. In Fig. 3, SCIAMACHY SO_2 columns retrieved in two windows are shown. While the overall agreement is good, the short wave window completely misses a plume with very large SO_2

columns extending over several hundreds of km from the volcano to the west. This is further illustrated in Fig. 4, where the data from the state covering the eruption are shown for three different fitting windows. Clearly, the agreement is very good for low and moderate SO_2 columns but for the highest values, a factor of more than 4 is found between the retrievals at 320 - 327 nm compared to the 311 - 317 nm fit.

Taking into account the results from the sensitivity study, the observations can be understood as the effect of the strong absorption in the areas with more than 50 DU of SO_2. A rough estimate is that if the additional SO_2 is located below 5 km, then the ratio seen in the measurements is similar to that in the simulations (compare Fig. 1). This would correspond to a situation, where two SO_2 plumes are present, a large (and older) one at somewhat higher altitudes and a smaller less diluted cloud close to the volcano at lower altitude. It is important to note that although some of the pixels were contaminated by clouds, the relative effect is similar as long as the SO_2 is located above the clouds (see Fig. 1, right panel). While a more detailed analysis is needed to come to quantitative results, this study demonstrates the feasibility to derive some information on the vertical SO_2 distribution under these conditions.

Future outlook

In the next year, data analysis of SCIAMACHY measurements will continue and a detailed comparison between GOME and SCIAMACHY measurements for the time period of overlapping measurements will be performed. It is planned to compare the results for the Sierra Negra eruption with the NASA/University of Maryland OMI retrievals to study the consistency between the two data sets and the effect of spatial resolution.

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