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 three UV/visible instruments GOME, SCIAMACHY, and GOME-2 has been analysed for tropospheric SO_2 columns. First results of SO_2 columns derived from GOME-2 show good agreement with OMI SO_2 columns for a volcanic eruption case and with SCIAMACHY data over China.

Extending the analysis presented in the previous report, GOME and SCIAMACHY SO_2 columns over the industrialised parts of China were evaluated, and a rapidly increasing trend in the SCIAMACHY data was identified. Comparison of GOME and SCIAMACHY columns in the time period of overlap shows good consistency in 2002/2003, and agreement with GOME-2 data in April 2007 confirms the recent SCIAMACHY results. Finally, possible origins of the observed changes are discussed.

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). Data from the OMI instrument have also been used to monitor the emissions of individual smelters in Peru (*Carn et al., 2007*). 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

Retrievals of GOME-2 SO_2 columns

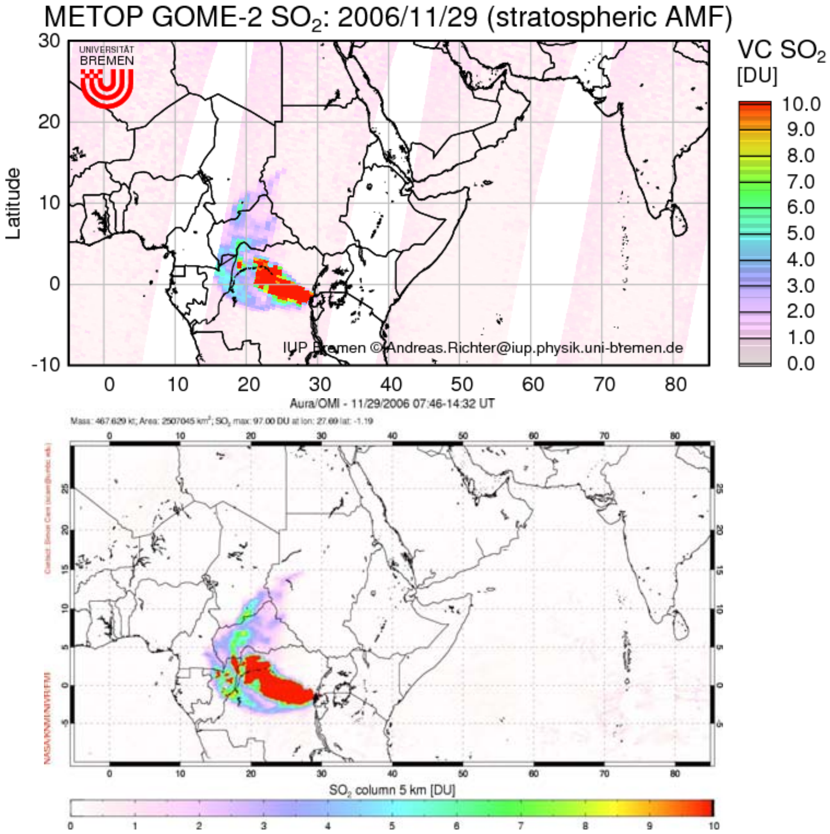

Fig. 1 GOME-2 measurements of SO_2 from the eruption of Mt Nyamuragira taken on November 29, 2006 (top) compared to the OMI measurements from that day. OMI data are courtesy of N. Krotkov and are available on http://SO2.umbc.edu/omi/pix/2006/nyam/imgloop.php?yr=06&mo=11&dy=29. The agreement is excellent and the differences can at least partly be understood by transport during the several hours time difference between the two measurements.

On October 19, 2006, the first of a series of three GOME-2 instruments was launched into orbit on MetOp-a. The GOME-2 instrument is very similar to GOME but provides better spatial coverage (global coverage every 1.5 days instead of every 3 days) and better spatial resolution (40 x 80 km^2 instead of 40 x 320 km^2). Compared to SCIAMACHY, the spatial resolution is somewhat poorer but the spatial coverage is much improved.

First calibrated spectra from GOME-2 became available in the current commissioning phase since early March 2007, but some orbits were already distributed earlier. One of the days released covers a large eruption of the Nyamuragyra volcano and the results of a SO_2 analysis of these orbits are shown in Fig. 1, together with the corresponding OMI map. The retrieval settings used are identical to those applied to GOME and SCIAMACHY measurements with the exception of the use of GOME-2 measured O_3 cross-sections (*Spietz et al., personal communication*) and convolution with the appropriate slit function for the other reference spectra. The agreement with OMI data is very nice with indication of a small scan angle dependence which could be related to the partial calibration of the data (the detectors were still not at nominal temperature on that day).

The change in plume position visible in the two images highlights the potential for using data from GOME-2, SCIAMACHY, and OMI to track the evolution of volcanic SO_2 plumes and better constrain the amount of sulphur injected into the atmosphere.

Scientific results and highlights

Comparison of GOME and SCIAMACHY measurements

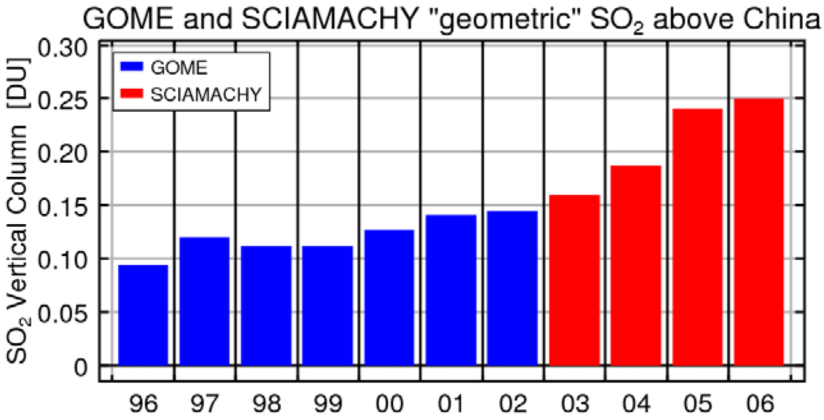

Fig. 2: Long-term time series of GOME (blue) and SCIAMACHY (red) SO_2-columns above the industrialised part of China (20°N, 100°E) – (40°N, 125°E). As an airmass factor appropriate for volcanic eruptions was used, the absolute columns are too low over polluted areas by about a factor of two.

In the last report, the increase of SO_2 over parts of China was noted and a link was made to the increases in NO_x emissions over that area which is observed in GOME and SCIAMACHY data (*Richter et al.*, 2005). In Fig. 2, the annual means of SO_2 over the region (20°N, 100°E) – (40°N, 125°E) are shown for the combined GOME and SCIAMACHY time series. A systematic increase is observed in the GOME data, followed by a marked upward trend in the SCIAMACHY measurements which appears to be less pronounced in 2006. Overall, the observed SO_2 column has increased by more than a factor of two over the measurement period.

These satellite based findings are in apparent contrast to results from air quality measurements performed in Beijing which report no increasing trend for SO_2 over the last years in spite of increasing energy use and traffic.

Therefore, it has first to be checked whether the SCIAMACHY time series is consistent with other measurements. This is discussed in the next section using GOME and GOME-2 data.

Verification of SCIAMACHY SO_2 columns with GOME and GOME-2 data

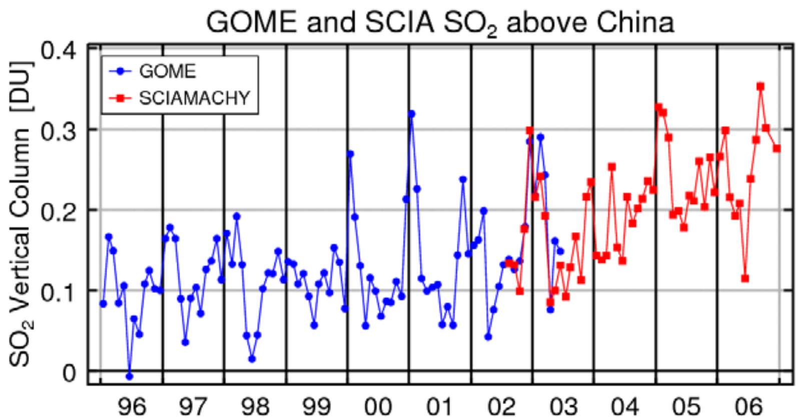

Fig. 3 As Fig. 2, but monthly values. Please note the good agreement between GOME and SCIAMACHY measurements in the overlapping time period.

In comparison to the signal of tropospheric NO_2, the sulphur dioxide signature is relatively weak for SO_2 in the boundary layer. This is mainly the result of the rapid increase in Rayleigh scattering towards the UV where the large SO_2 absorption bands are situated and the strong ozone absorption. Consequently, the SO_2 signal has a larger uncertainty than the NO_2 columns. In Fig. 3 the combined GOME and SCIAMACHY time series is shown as monthly averages. Again, the change over time can clearly be seen, first with enhanced winter values in the GOME data and then as a general increase in the SCIAMACHY columns.

In 2002 / 2003, the GOME and SCIAMACHY instruments were both fully operational for several months of overlap and as shown in Fig. 3, the agreement in the monthly averages is excellent over this time. Therefore, the SCIAMACHY and GOME time series are consistent at least during this time.

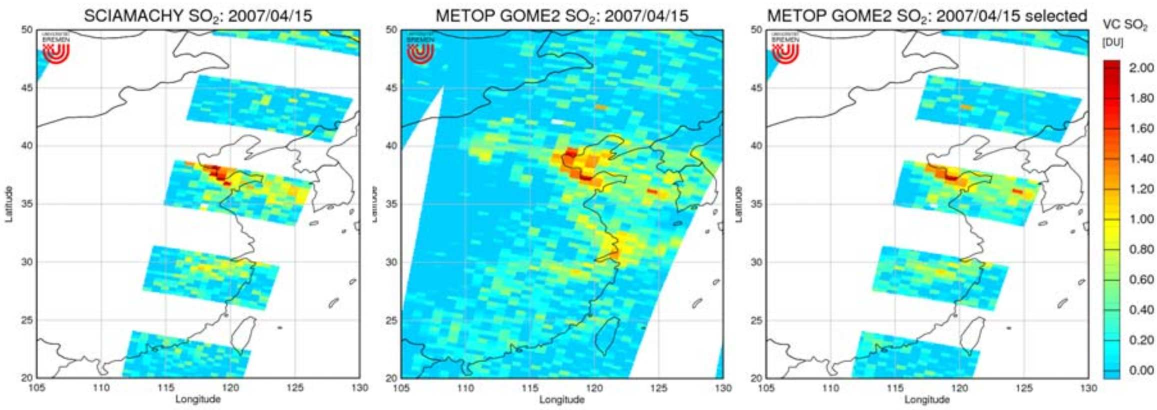

Fig. 4: SO_2 slant columns for April 15, 2007 as measured by SCIAMACHY (left) and GOME-2 (middle). The right panel shows the GOME-2 data limited to those areas where SCIAMACHY data are also available. Considering the time difference (30 minutes) and the different foot print (30 x 60 km^2 for SCIAMACHY; 40 x 80 km^2 for GOME-2), the agreement is excellent.

The other end of the SCIAMACHY time series can be compared to measurements from GOME-2. So far, not enough data from this new instrument are available for a statistical comparison, but as an example, one day is shown in Fig. 4. After removing those GOME-2 data for which no SCIAMACHY measurement is available, the very good agreement between the two data sets is apparent. Therefore, the high SO_2 columns at the end of the SCIAMACHY time series can be considered as tentatively verified by GOME-2 measurements.

As both the low end and the high end of the SCIAMACHY time series are confirmed by data from other instruments, it is concluded that the increase in SO_2 column is real and must be related to changes in China.

Discussion of SO_2 time series above China

Several different effects could potentially contribute to the increase in SO_2 column observed over China.

The most obvious explanation is an increase in SO_2 emissions, linked to the rapid economic and industrial growth witnessed in china over the last years. According to bottom-up emission estimates, SO_2 emissions have in fact increased over the last decade, but not to the extend suggested by the satellite data. This could point at deficiencies in the SO_2 emission inventories, but could also be related to the sensitivity of the satellite measurements.

One important aspect of the SO_2 column measurements is the vertical sensitivity profile. As discussed in the previous report, the sensitivity of the measurements to SO_2 in the boundary layer is relatively small and increases towards the free troposphere. Any change in vertical distribution of the SO_2 can therefore have an effect on the satellite measurement even if the total amount of SO_2 is not changing. One possible example is the switch from coal use for domestic heating to coal fired power plants. The latter inject SO_2 higher in the atmosphere

making it more accessible to satellite measurements. This could also explain the lack of trend in the air quality measurements in Beijing – part of the SO_2 would simply be transported downwind before reaching the surface.
In addition a change in vertical distribution, changes in aerosol loading could also contribute to a change in SO_2 signal. Depending on the relative altitude distribution of aerosols and SO_2, the signal seen from satellite could either increase (if the aerosols are reflective and within or below the SO_2 layer) or decrease (should the aerosol be above the SO_2). As SO_2 itself is involved in sulphate aerosol formation, the vertical profiles of both should in first approximation be similar which would lead to decreasing SO_2 columns for increasing aerosol. Still, an impact of changing aerosol on the observed SO_2 columns can not be ruled out.

Future outlook

In the next year, data analysis of SCIAMACHY and GOME-2 measurements will continue and a detailed comparison between SCIAMACHY and GOME-2 measurements will be performed. The study of SO_2 over China will be finalised and a comparison of GOME-2 SO_2 columns with SO_2 derived from AIRS measurements will be undertaken in collaboration with F. Prata from NILU.

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