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retrieval from
SCIAMACHY

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SO₂ Retrieval from SCIAMACHY using the Weighting Function DOAS (WFDOAS) Technique: comparison with Standard DOAS retrieval

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Abstract

Atmospheric SO₂ can be measured by remote sensing of scattered sunlight from space, using its unique absorption features in the ultraviolet region. However, the sensitivity of the SO₂ measurement depends critically on spectral interference, surface albedo and varies with wavelength as Rayleigh scattering increases at shorter wavelengths. The Weighting Function Differential Optical Absorption Spectroscopy (WFDOAS) method was used to solve these problems. The Ring spectra included in the WFDOAS fit were determined as a function of total ozone column density, solar zenith angle, surface albedo, and effective scene altitude. The WFDOAS SO₂ retrieval from SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) data onboard the ENVISAT satellite are presented here and compared to those of the Standard DOAS (SDOAS) method for cases of background conditions and volcanic eruption. The study demonstrates that the problems in the SO₂ retrieval with SDOAS, such as the positive offsets over remote (clean) regions and the negative offsets at high solar zenith angles and high ozone, can be solved by the WFDOAS approach.

1 Introduction

Sulfur dioxide (SO₂) plays an important role in the atmospheric sulfur cycle, in the formation of new aerosol, and the modification of existing aerosol (Chin and Jacob, 1996; Berglen et al., 2004). There is concern about the impact of SO₂ on global climate through the formation and modification of aerosols and their effects on the radiation balance of the atmosphere (Intergovernmental Panel on Climate Change (IPCC), 2001; Myhre et al., 2004), as well as on ecosystems and human health. The SO₂ is released into the atmosphere as a result of both anthropogenic activities and natural (e.g., volcanic) phenomena (Seinfeld and Pandis, 1998; Finlayson-Pitts and Pitts, 2000).

Anthropogenic emissions of SO₂ occur predominantly from fossil fuel combustion at

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the continental surface and chemical conversion and loss processes take place during transport (Tan et al., 2002; Koike et al., 2003; Guttikunda et al., 2005). Volcanoes are an important natural source of various atmospheric trace gases (in particular SO₂). Volcanic eruptions inject gases and particles into the atmosphere, leading to tropospheric and stratospheric aerosol formation. Highly explosive volcanic events affect the climate on time scales of months to years (McCormic et al., 1995; Robock, 2000). Neither the estimates of the anthropogenic SO₂ flux nor that from volcanic emissions is easy to determine precisely, on account of the dispersion of anthropogenic sources over large areas. Since volcanic eruptions and their emissions are sporadic and intermittent and often occur in uninhabited regions, it is difficult to assess the amount and size of the emissions from volcanoes. Satellite remote sensing measurements are well-suited to overcome these difficulties. The SO₂ observations from space was first performed using Total Ozone Mapping Spectrometer (TOMS) (Krueger, 1983), and then have been done from TOMS and other satellite instruments such as the Global Ozone Monitoring Experiment (GOME), Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) and Ozone Measurement Instrument (OMI) (e.g., Krueger et al., 1995; Eisinger and Burrows, 1998; Afe et al., 2004; Carn et al., 2004; Khokhar et al., 2005; Thomas et al., 2005; Krotkov et al., 2006; Richter et al., 2006; Carn et al., 2007; Yang et al., 2007; Krotkov et al., 2008). Modification and development of algorithms used to retrieve data from satellite observations have been of primary interest. The satellite remote sensing approach associated with the Differential Optical Absorption Spectroscopy (DOAS) technique (Platt, 1994) has been successfully employed for measurements of atmospheric trace gases on global and regional scales (e.g., Chance, 1998; Eisinger et al., 1998; Wagner et al., 1998; Martin et al., 2002; Palmer et al., 2003; Afe et al., 2004; Richter et al., 2005; Wittrock et al., 2006; Lee et al., 2008).

The Standard DOAS (called SDOAS here) retrieval of SO₂ from measurements of the GOME (Burrows et al., 1999) and SCIAMACHY (Bovensmann et al., 1999) works well over areas with strongly enhanced SO₂ after volcanic eruptions (Khokhar et al,

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2005; Richter et al., 2006). These plumes can be easily identified and tracked with high accuracy. Also, areas suffering from strong SO₂ pollution can be identified at least in monthly averages. However, there are a number of problems with the SO₂ data that need to be solved for a better exploitation of the satellite measurements:

1. There is an offset in the SO₂ slant columns that leads to positive values over remote (clean) regions. This offset varies over time and with latitude. Up to now, the approach used to account for this is to subtract the values over a clean reference sector which greatly reduces the problem at low and middle latitudes but often fails at higher latitudes. The origin of the offset is unknown but it depends on the exact choice of fitting window and cross-sections indicating a spectral interference problem.
2. At high latitudes/high ozone levels, the retrieved SO₂ columns are negative as a result of interference with ozone and its temperature dependence. The airmass factor of O₃ varies significantly with wavelength and its changes are correlated with the O₃ bands. In addition, the sensitivity of SO₂ retrievals also varies with wavelength as Rayleigh scattering increases at shorter wavelengths.
3. Over elevated areas, the SO₂ column is smaller and after offset correction often negative. This is the case over the Himalaya, the Andes, and the Alps but also over the Western part of the US (Khokhar et al., 2005). It is speculated that this is related to interference from Ring but no clear conclusion has been drawn so far (Van Roozendaal et al., 2002).
4. Over bright surfaces such as Greenland or the South Pole, often unrealistically large SO₂ columns are observed.
5. Overall the scatter in the SO₂ values is large, in particular for SCIAMACHY measurements. The Southern Atlantic Anomaly has a large impact over an area covering large parts of South America and the South Atlantic reaching the African

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coast. This can be improved by extending the SO_2 fitting window to the UV, but this reduces sensitivity to boundary layer SO_2 .

In an attempt to solve at least some of these problems, SO_2 retrievals using the Weighting Function Differential Optical Absorption Spectroscopy (WFDOAS) method were performed and results compared to those from Standard DOAS. The WFDOAS is distinguished from the Standard DOAS by the use of wavelength-dependent weighting functions of ozone and temperature, which describe the relative radiance change due to a vertical profile change. The WFDOAS method has been first demonstrated to be applicable to retrievals of trace gas columns in the near infrared region of SCIAMACHY (Buchwitz et al., 2000). It is also being used for direct retrieval of vertical O_3 amounts (Coldewey-Egbers et al., 2005; Weber et al., 2005, Bracher et al., 2005). It is distinct from the SDOAS by the use of wavelength-dependent weighting functions, which describe the relative radiance change due to a vertical profile change. The weighting function for the ozone column change is obtained by integrating vertically the altitude dependent weighting function. Such an approach is particularly suited to DOAS retrievals of strong absorbers like ozone in the UV spectral region. In addition, the effect of rotational Raman scattering (Ring effect) is accounted for by using a pre-calculated data base of Raman reference spectra which is tabulated as a function of ozone column density, solar zenith angle, surface albedo, and altitude (Coldewey-Egbers et al., 2005). Here, we limit the weighting functions to ozone and temperature, and SO_2 is only fitted as an additional absorber, which means that SO_2 cross-section is fitted to obtain the slant column amount. The WFDOAS SO_2 retrievals are still expected to be superior to those of SDOAS as the strong interference by ozone and Ring effect is optimally handled.

In the following, the SCIAMACHY SO_2 retrievals with WFDOAS and SDOAS as applied here will be described, the results from the two techniques be compared and the effect from different Ring parameterizations be evaluated. In this study we concentrated on selected scenarios with anthropogenic and volcanic emissions as well as background conditions in December 2006. The airmass factor (AMF) calculation to

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convert the slant columns (SC) to vertical columns (VC) is not covered here, but will be considered in future work.

2 Experimental

2.1 SCIAMACHY instrument

5 SCIAMACHY is one of ten instruments onboard ESA's Environmental Satellite (ENVISAT) which was launched into a sun-synchronous orbit in March 2002. The SCIAMACHY instrument is an 8 channel grating spectrometer measuring in nadir, limb, and occultation (both solar and lunar) viewing geometries (Bovensmann et al., 1999). SCIAMACHY covers the spectral region from 220 to 2400 nm with a spectral resolution
10 of 0.25 nm in the UV, 0.4 nm in the visible and less in the NIR. In these wavelength regions several trace gases were detected in the past (e.g., Afe et al., 2004; Buchwitz et al., 2005; Richter et al., 2005; Wittrock et al., 2006). The size of the nadir ground-pixels depends on wavelength range and solar elevation and can be as small as $30 \times 30 \text{ km}^2$. The SO_2 column retrieval is done using spectra from Channel 2 (310-405 nm) in nadir
15 viewing geometry.

2.2 Standard DOAS retrieval

The SDOAS method has been used for the SO_2 retrieval for GOME and SCIAMACHY in previous studies (Eisinger and Burrows, 1998; Afe et al., 2004; Khokhar et al., 2005; Richter et al., 2006; Lee et al., 2008). In this work, the wavelength range of 315–327 nm
20 is used for the SO_2 DOAS fit as the differential absorptions are large and interference by other species (apart from ozone) is small. In addition to the SO_2 cross-section (Vandaele et al., 1994), two ozone cross-sections (Bogumil et al., 2003), a synthetic Ring spectrum (Vountas et al., 1998), an undersampling correction (Chance, 1998), and the polarization dependency of the SCIAMACHY instrument are included in the

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fit. The specifications of the SDOAS SO₂ retrieval are summarized in Table 1. Daily solar irradiation measurements taken with the ASM diffuser are used as background spectrum. In the comparison with the WFDOAS retrieval, the reference sector method (RSM, for details see Martin et al., 2002; Richter and Burrows, 2002) is applied to remove a meridional offset in the SDOAS SO₂ SC by subtracting the columns of a presumably SO₂ free reference sector over a remote area, which were taken on the same day at the same latitude in the 180° E – 230° E longitudinal range.

2.3 Weighting function DOAS retrieval

Weighting Functions are the derivative of the radiation field with respect to the atmospheric parameters and they are used in the retrieval of strongly absorbing species (Rozanov et al., 1998). The WFDOAS algorithm was originally applied to retrieval of vertical O₃ amounts from GOME and SCIAMACHY data in the wavelength region of 326.6–335.0 nm (Coldewey-Egbers et al., 2005). Here we extend it for the SO₂ retrieval by changing the wavelength region and adding SO₂ as an additional absorber. Figure 1 shows the WFDOAS algorithm scheme used for the SO₂ retrieval in this work. The radiative transfer model, SCIATRAN 2.0 (Rozanov et al., 1997) was integrated into the iterative retrieval scheme. In earlier versions the radiative transfer quantities were read from look-up-tables. In the WFDOAS retrieval the measured atmospheric optical depth is approximated by a Taylor expansion around the reference intensity. A third order polynomial P_i is added to account for all broad band contributions from surface albedo and aerosol. The Ring spectrum, BrO absorption cross-section as well as SO₂ absorption cross-section for all of which slant column fitting is applied are included in the WFDOAS fitting. The optical depth equation can be written as follows (Coldewey-

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Egbers et al., 2005):

$$\begin{aligned} \ln I_i^{\text{mea}}(V^t, \vec{b}^t) &\approx \ln I_i^{\text{mod}}(\bar{V}, \vec{b}) + \frac{\partial \ln I_i^{\text{mod}}}{\partial V} \Big|_{\bar{V}} \times (\hat{V} - \bar{V}) + \frac{\partial \ln I_i^{\text{mod}}}{\partial T} \Big|_{\bar{T}} \times (\hat{T} - \bar{T}) \\ &+ SCD_{\text{SO}_2} \cdot \sigma_{i,\text{SO}_2} \\ &+ SCD_{\text{BrO}} \cdot \sigma_{i,\text{BrO}} \\ &+ SCD_{\text{Ring}} \cdot \sigma_{i,\text{Ring}} \\ &+ P_i \end{aligned} \quad (1)$$

where I^{mea} is the measured intensity and I^{mod} is the sun-normalized reference intensity as provided by SCIATRAN 2.0. Index t denotes the true atmospheric state. The entire right-hand side of the equation (excluding the reference intensity) has to be adjusted to the measured intensity (left-hand side) for all spectral points (index i) simultaneously. \bar{V} is the reference ozone column corresponding to the reference intensity, and \bar{T} is the reference surface temperature. \hat{V} and \hat{T} denote the corresponding fit parameters. The specifications for the WFDOAS SO₂ retrieval are shown in Table 1.

In the WFDOAS fitting, the atmospheric scenario which most closely matched the O₃ vertical column derived from the IUP climatology (Lamsal et al., 2004) was selected using linear interpolation between effective albedo, relative azimuth angle and geo-location data. A reference spectrum to account for the Ring effect, the “filling-in” of solar Fraunhofer lines in the scattered light, was calculated online as a function of total ozone column including ozone profile shape, solar zenith angle, surface albedo, altitude, and viewing geometries using SCIATRAN 2.0, but no interpolation was done (Weber et al., 2007). Ozone and temperature profiles are taken from IUP climatology (Lamsal et al., 2004) which contains different profile shapes for three latitude belts (tropics, middle and high latitudes in each hemisphere) and two seasons (winter/spring and summer/fall) as a function of the total ozone column. The Ring spectrum σ_{Ring} has

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been approximated as follows:

$$\sigma_{\text{Ring}} = \frac{I_0^{rrs}}{I_0} \exp \left(\frac{\tau_{O_3}^v}{\cos(\theta_0)} \left(1 - \frac{\sigma_{O_3}^{rrs}}{\sigma_{O_3}} \right) \right) \quad (2)$$

where the index *rrs* indicates the Raman smoothed quantity, I_0 the solar irradiance, σ_{O_3} ozone cross-section, θ_0 the solar zenith angle, and $\tau_{O_3}^v$ the vertical optical density of O_3 (Weber et al., 2007).

The ozone and temperature weighting functions and modeled radiances (reference intensity) are calculated as a function of solar zenith angle, line of sight, relative azimuth angle, surface height, and effective albedo using SCIATRAN 2.0. The DOAS fitting utilized a non-linear least square method, including wavelength shifts and squeeze for the nadir measurement spectrum to make the direct comparison between measured and modeled radiances (Coldewey-Egbers et al., 2005; Weber et al., 2005). After DOAS fitting the retrieved ozone column was compared with that of the reference scenario. The fit is repeated with an updated set of weighting functions until the change in retrieved value is below a convergence limit (see Fig. 1).

2.4 OMI SO_2 data

In addition, OMI SO_2 SC data converted from the publicly released Planetary Boundary Layer (PBL) OMI SO_2 Level 2 VC products (<http://daac.gsfc.nasa.gov/>) are added for further comparison with the WFD OAS SCIAMACHY SO_2 . The OMI SO_2 SC data are retrieved with the Band Residual Difference (BRD) algorithm (Krotkov et al., 2006, 2008). In this retrieval, the ‘sliding median’ empirical correction is used to reduce any cross-track and meridional biases, which subtracts a median residual for a sliding group of SO_2 free pixels covering $\pm 15^\circ$ latitude along the orbit track from each spectral band and cross-track position (Yang et al., 2007, Krotkov et al., 2008). There is cloud screening in these publicly released PBL SO_2 data using the removal of all measurements with surface reflectivity larger than 30 %. The OMI PBL SO_2 VCs were converted back to

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SC using the original constant AMF of 0.36.

3 Results

3.1 Ring effect in the WFDOAS SO₂ retrieval

Figure 2 shows the WFDOAS fitting results of two cases: one using the online Ring spectrum calculation (ORSC) as described in Sect. 2.3 and the other using just a single Ring spectrum (Vountas et al., 1998), which is based on one standard ozone profile and only depends on the solar zenith angle. The spectra representing background conditions as shown in Fig. 2 were taken in the 60° E and 100° E longitudinal region on 30 November 2005 (SCIAMACHY orbit no. 19611). As seen in the top panel of Fig. 2, the WFDOAS retrieval including a single Ring spectrum has positive offsets over the tropics and mid latitudes. These offsets decrease slightly at higher latitudes, while they are decreasing and even reaching negative values at high latitudes in the SDOAS retrievals. These offsets over the tropics and mid latitudes are not present when using the ORSC. Even the slightly decreasing offsets over higher latitudes are removed by using the ORSC. O₃ columns at higher latitudes are slightly lower compared to those obtained by using a single Ring spectrum. Also important, the fit residual decreased in the WFDOAS retrieval using the ORSC. It seems that depending on how the Ring effect is calculated, it may be responsible for positive offsets in the SO₂ SCs.

3.2 Comparison of WFDOAS with SDOAS results

Figure 3 shows the SO₂ slant columns retrieved with WFDOAS and SDOAS for one orbit with background conditions (a), where SO₂ is expected to be below the detection limit and another orbit containing a volcanic eruption (b). The orbit used for the background condition is the same as that shown in Fig. 2. Volcanic eruption spectra were obtained in the 80° W–120° W longitudinal region on 2 December 2006 (SCIA-

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MACHY orbit no. 24868). The OMI SO₂ SC data corresponding to geo-locations of SCIAMACHY data measured on the same day are plotted in Fig. 3.

As shown in Fig. 3a, the SDOAS results have positive offsets over tropical and mid latitude areas, and are decreasing and even reaching negative values at higher latitudes under background conditions. SO₂ levels in the SDOAS retrieval of volcanic emissions are overestimated compared to that WFDOAS retrieval (see Fig. 3b). The differences between SDOAS and WFDOAS are not a result from different polynomial degrees and absorbers (e.g. BrO in WFDOAS only, ETA and undersampling only in SDOAS), which were summarized in Table 1. There is little difference in the difference polynomial degrees in the WFDOAS retrieval, and BrO is a minor absorber in the retrieval wavelength so that it rarely contributes to the differences between SDOAS and WFDOAS retrieval. The constant and latitudinal offsets in the retrieved SO₂ SC have also been reported in previous SDOAS SO₂ retrieval studies (e.g., Khokhar et al., 2005; Richter et al., 2006). SO₂ shows clear absorption features in the retrieval wavelength range of 315 – 327 nm. In this wavelength range ozone absorption is, however, strong and interferes with SO₂. Negative SO₂ SC values may be obtained, when the varying ozone dependence of the Ring effect is not properly accounted for, particularly, with increasing slant O₃ absorption at higher SZAs (see Fig. 3a). The levels of OMI SO₂ are likely to be in the agreement with the WFDOAS SCIAMACHY SO₂ and show less scatter compared to the SDOAS and WFDOAS SCIAMACHY SO₂. The OMI SO₂ SCs are stable over tropic and mid-latitudes but have relatively larger scatter at larger SZA. Note that the ‘sliding median’ empirical correction and cloud-screening are used in the OMI SO₂ retrieval and the number of OMI SO₂ data set is less than SCIAMACHY SC data.

Monthly mean SO₂ slant column maps from December 2006 are shown in Fig. 4 for the WFDOAS retrieval, the SDOAS retrieval, the SDOAS retrieval after applying the reference sector (RSM) correction, and the BRD OMI retrieval. The WFDOAS results show less meridional offsets, compared to those by the SDOAS retrieval, while it shows slightly higher offsets compared to the RSM-applied SDOAS results. The meridional

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offset over low and mid latitude areas are completely removed in the RSM-corrected SDOAS retrieval, but not in some areas at high latitudes where the instrument took measurements at large solar zenith angles. The seven day mean of BRD OMI SO₂ SC from 01 to 07 December 2006 is presented in Fig. 4. Its background level is in the agreement with those in the RSM-corrected SDOAS retrieval. Both SCIAMACHY and OMI instrument observed volcanic SO₂ signals around Nyamuragira volcano in Congo (erupted on 27 November 2006), as also seen in Fig. 3b. There are some breaks in these volcanic signals in the SCIAMACHY data because it takes turns about recording signals in nadir and limb modes. In monthly mean data the volcanic SO₂ signals observed from the OMI instrument for December 2006 are lower than those from SCIAMACHY. The difference in monthly average volcanic SO₂ amounts can be explained by sampling effect (the average OMI SC on volcanic plumes should be lower than the SCIAMACHY SC as the OMI samples each pixel every day and the SCIAMACHY needs 6 days to acquire a contiguous global map).

4 Conclusions

The crucial role of SO₂ in the oxidation capacity of the atmosphere and the assessment of natural radiative forcing of atmospheric sulfate aerosols are well known. The development of robust control strategies to reduce SO₂ levels in the atmosphere requires more precise evaluation of the amount of natural and anthropogenic SO₂ from space. Satellite measurements of SO₂ are now feasible, and help better to identify and quantify the amount of natural and anthropogenic emissions of SO₂ and their transport and conversion to sulfate aerosols in the atmosphere. However, the SO₂ slant column retrieval in the UV spectral region is quite challenging due to the strong interference with ozone, which can produce positive offsets over remote (clean) regions, negative offsets at high latitudes and in region with high ozone, and smaller values over elevated areas which after correction with the reference sector method often become negative.

Some of these problems in the SDOAS retrieval of SO₂ can be overcome by using

the WFDOAS approach which improves the fit because

1. the wavelength dependence of the ozone airmass factor is accounted for by using weighting functions, and
2. the treatment of the Ring effect is improved when obtained from a complex data base which depends on ozone column, reflectivity, and SZA, which appears to be the most crucial factor in the SO₂ retrieval using the WFDOAS technique.

The ability of WFDOAS retrieval as an alternative method for SDOAS methods was demonstrated here. Monitoring of volcanic emission and ambient air pollution is important in terms of global SO₂ budget and ambient air quality. The WFDOAS retrieval showed clearly volcanic emissions (e.g., near and around Nyamuragira volcano in Congo, erupted on 27 November 2006), and ambient air pollution of SO₂ (e.g., over northeastern China). The current version of the WFDOAS retrieval is computationally expensive (~4 h per one orbit data on our local machine compared to several minutes in the SDOAS retrieval). It may be considerably sped up by use of look-up-tables. Up to now, it is suitable mainly for case studies.

The retrieval of SO₂ emissions (in particular volcanic SO₂ emissions) from space suffers from sparse temporal and spatial coverage of current satellite sensors but also from clouds in the troposphere. However, satellite measurements of SO₂ are a crucial step forward for real time monitoring of air pollution change on a global scale, and can provide information for near real-time application and forecasting such as hazard warning and air-quality monitoring.

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- Afe, O. T., Richter, A., Sierk, B., Wittrock, F., and Burrows, J. P.: BrO emission from volcanoes: a survey using GOME and SCIAMACHY measurements, *Geophys. Res. Lett.*, 31, doi:10.1029/2004GL020994, 2004.
- 5 Berglen, T. F., Bernsten, T. K., Isaksen, I. S. A., and Sundet, J. K.: A global model of the coupled sulfur/oxidant chemistry in the troposphere: the sulfur cycle, *J. Geophys. Res.*, 109, doi: 10.1029/2003JD003948, 2004.
- Bogumil, K., Orphal, J., Homann, T., Voigt, S., Spietz, P., Fleischmann, O. C., Vogel, A., Hartmann, M., Bovensmann, H., Frerik, J., and Burrows, J. P.: Measurements of molecular absorption spectra with the SCIAMACHY Pre-Flight Model: Instrument characterization and reference data for atmospheric remote sensing in the 230–2380 nm region, *J. Photoch. Photo-*
- 10 *bio. A*, 157, 167–184, 2003.
- Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noel, S., Rozanov, V. V., Chance, K. V., and Goede, A. H. P.: SCIAMACHY-Mission objectives and measurement modes, *J.*
- 15 *Atmos. Sci.*, 56, 127–150, 1999.
- Bracher, A., Lamsal, L. N., Weber, M., Bramstedt, K., Coldewey-Egbers, M., and Burrows, J. P.: Global satellite validation of SCIAMACHY O₃ columns with GOME WFDOAS, *Atmos. Chem. Phys.* 5, 2357–2368, 2005.
- Buchwitz, M., de Beek, R., Burrows, J. P., Bovensmann, H., Warneke, T., Notholt, J., Meirink, J.
- 20 F., Goede, A. P. H., Bergamaschi, P., Körner, S., Heimann, M., and Schulz, A.: Atmospheric methane and carbon dioxide from SCIAMACHY satellite data: Initial comparison with chemistry and transport models, *Atmos. Chem. Phys.*, 5, 941–962, 2005,
<http://www.atmos-chem-phys.net/5/941/2005/>.
- Buchwitz, M., Rozanov, V. V., and Burrows, J. P.: A near-infrared optimized DOAS method for the fast global retrieval of atmospheric CH₄, CO, CO₂, H₂O, and N₂O total column amounts from SCIAMACHY Envisat-1 nadir radiances, *J. Geophys. Res.*, 105, 15 231–15 245, 2000.
- 25 Burrows, J. P., Weber, M., Buchwitz, M., Rozanov, V. V., Ladstädter-Weissenmayer, A., Richter, A., de Beek, R., Hoogen, R., Bramstedt, K., Eichmann, K.-U., Eisinger, M., and Perner, D.: The Global Ozone Monitoring Experiment (GOME): Mission concept and first scientific results, *J. Atmos. Sci.*, 56, 151–175, 1999.
- 30 Carn, S. A., Krotkov, N. A., Gray, M.A., Krueger, A. J.: Fire at Iraqi sulfur plant emits SO₂ clouds detected by Earth Probe TOMS, *Geophys. Res. Lett.*, 31, doi:10.1029/2004GL020719,

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8, 10817–10839, 2008

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2004.

Carn, S. A., Krueger, A. J., Krotkov, N. A., Yang, K., Levelt, P. F.: Sulfur dioxide emissions from Peruvian copper smelters detected by the Ozone Monitoring Instrument, *Geophys. Res. Lett.*, 34, doi:10.1029/2006GL029020, 2007.

5 Chin, M. and Jacob, D. J.: Anthropogenic and natural contributions to tropospheric sulfate: a global model analysis, *J. Geophys. Res.*, 101, 18 691–18 669, 1996.

Coldewey-Egbers, M., Weber, M., Lamsal, L., De Beek, R., Buchwitz, M., and Burrows, J. P.: Total ozone retrieval from GOME UV spectral data using the weighting function DOAS approach, *Atmos. Chem. Phys.*, 5, 1015–1025, 2005,

10 <http://www.atmos-chem-phys.net/5/1015/2005/>.

Eisinger, M. and Burrows, J. P.: Tropospheric sulfur dioxide observed by the ERS-2 GOME instrument, *Geophys. Res. Lett.*, 25, 4177–4180, 1998.

Finlayson-Pitts, B. J. and Pitts, J. N.: *Chemistry of the Upper and Lower Atmosphere*, Academic Press, San Diego, USA, 2000.

15 Guttikunda, S. K., Tang, Y., Carmichael, G. R., Kurata, G., Pan, L., Streets, D. G., Woo, J.-H., Thongboonchoo, N., and Fried A.: Impacts of Asian megacity emissions on regional air quality during spring 2001, *J. Geophys. Res.*, 110, doi: 10.1029/2004JD004921, 2005.

Khokhar, M. F., Frankenberg, C., Van Roozendaal, M., Beirle, S., Kühl, S., Richter, A., Platt, U., and Wagner, T.: Satellite observation of atmospheric SO₂ from volcanic eruptions during the time-period of 1996–2002, *Adv. Space Res.*, 36, 879–887, 2005.

20 Kokhanovsky, A. A., von Hoyningen-Huene, W., Rozanov, V. V., Noël, S. Gerilowski, K., Bovensmann, H., Bramstedt, M., Buchwitz, M., and Burrows, J. P.: The semianalytical cloud retrieval algorithm for SCIAMACHY II. The application to MERIS and SCIAMACHY data, *Atmos. Chem. Phys.*, 6, 4129–4136, 2006, <http://www.atmos-chem-phys.net/6/4129/2006/>.

25 Koike, M., Kondo, Y., Kita, K., Takegawa, N., Masui, Y., Miyazaki, Y., Ko, M. W., Weinheimer, A. J., Flocke, F., Weber, R. J., Thornton, D. C., Sachse, G. W., Vay, S. A., Blake, D. R., Streets, D. G., Eisele, F. L., Sandholm, S. T., Singh, H. B., and Talbot, R. W.: Export of anthropogenic reactive nitrogen and sulfur compounds from the East Asia region in spring, *J. Geophys. Res.*, 108, doi:10.1029/2002JD003284, 2003.

30 Krotkov, N. A., Carn, S. A., Krueger, A. J., Bhartia, P. K., and Yang, K.: Band residual difference algorithm for retrieval of SO₂ from the Aura Ozone Monitoring Instrument (OMI), *IEEE Transac. Geosci. Remote Sens.*, 44, 1259–1266, 2006.

Krotkov, N. A., McClure, B., Dickerson, R. R., Carn, S. A., Can, L., Bhartia, P. K., Yang, K.,

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8, 10817–10839, 2008

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Krueger, A. J., Li, Z., Levelt, P. F., Chen, H., Wang, P., and Daren, L.: Validation of SO₂ retrievals from the Ozone Monitoring Instrument (OMI) over NE China, *J. Geophys. Res.*, doi:10.1029/2007JD008818, 2008.

5 Krueger, A.: Sighting of El Chichon sulfur dioxide clouds with the Nimbus 7 total ozone mapping spectrometer, *Science*, 220, 1377–1379, 1983.

Krueger, A., Walter, L., Bhartia, P., Schnetzler, C., Krotkov, N., Sprod, I., and Bluth, G.: Volcanic sulfur dioxide measurements from the total ozone mapping spectrometer instruments, *J. Geophys. Res.*, 100, 14 057–14 076, 1995.

10 Lamsal, L. N., Weber, M., Labow, G., and Burrows, J. P.: Influence of ozone and temperature climatology on the accuracy of satellite total ozone retrieval, *J. Geophys. Res.*, 112, doi:10.1029/2005JD006865, 2007.

Lamsal, L. N., Weber, M., Tellmann, S., and Burrows, J. P.: Ozone column classified climatology of ozone and temperature profiles based on ozone sondes and satellite data, *J. Geophys. Res.*, 109, doi:10.1029/2004JD004680, 2004.

15 Lee, C., Richter, A., Burrows, J. P., Lee, H., Kim, Y. J., Lee, Y. G., and Choi, B. C.: Impact of transport of sulfur dioxide from the Asian continent on the air quality over Korea during May 2005, *Atmos. Environ.*, doi:10.1016/j.atmosenv.2007.11.006, 2008.

20 Martin, R. V., Chance, K., Jacob, D. J., Kurosu, T. P., Spurr, R. J. D., Bucsela, E., Gleason, J. F., Palmer, P. I., Bey, I., Fiore, A. M., Li, Q., Yantosca, R. M., and Koelemeijer, R. B. A.: An improved retrieval of tropospheric nitrogen dioxide from GOME, *J. Geophys. Res.*, 107, doi:10.1029/2001JD001027, 2002.

McCormick, M. P., Thomason, L. W., and Trepte, C. R.: Atmospheric effects of the Mt Pinatubo eruption, *Nature*, 373, 399–404, 1995.

25 Myhre, G., Berglen, T. F., Myhre, C. E. L., and Isaksen, I. S. A.: The radiative effect of the anthropogenic influence on the stratospheric sulfate aerosol layer, *Tellus B*, 56, doi:10.1111/j.1600-0889.2004.00106.x, 2004.

Palmer, P. I., Jacob, D. J., Fiore, A. M., Martin, R. V., Chance, K., and Kurosu, T. P.: Mapping isoprene emissions over North America using formaldehyde column observations from space, *J. Geophys. Res.*, 108, doi:10.1029/2002JD002153, 2003.

30 Platt, U.: Differential optical absorption spectroscopy (DOAS), in: *Monitoring by Spectroscopic Techniques*, Wiley & Sons, New York, USA, 27–84, 1994.

Richter, A., Burrows, J. P., Nüß, H., Granier, C., and Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, *Nature*, 437, 129–132, 2005.

- Richter, A., Wittrock, F., and Burrows, J. P.: SO₂ Measurements with SCIAMACHY, in Proc. Atmospheric Science Conference, ESRIN, Frascati, ESA publication SP-628, 8–12 May 2006.
- Robock, A.: Volcanic eruption and climate, *Review Geophys.*, **38**, 191–219, 2000.
- Seinfeld, J. H. and Pandis, S.: *Atmospheric Chemistry and Physics*, John Wiley & Sons, New York, USA, 1998.
- 5 Rozanov, V. V., Kurosu, T., and Burrows, J. P.: Retrieval of atmospheric constituents in the UV-visible: A new quasi-analytical approach for the calculation of weighting functions, *J. Quant. Spectrosc. Ra.*, **60**, 277–299, 1998.
- Rozanov, V. V., Diebel, D., Spurr, R. J. D., and Burrows, J. P.: GOMETRAN: A radiative transfer model for the satellite project GOME – The plane parallel version, *J. Geophys. Res.*, **102**, 16 683–16 695, 1997.
- 10 Tan, Q., Huang, Y., and Chameides, W. L.: Budget and export of anthropogenic SO_x from East Asia during continental outflow condition, *J. Geophys. Res.*, doi:10.1029/2001JD000769, 2002.
- 15 Thomas, W., Erbertseder, T., Ruppert, T., Van Roozendael, M., Verdebout, J., Balis, D., Meleti, C., and Zerefos, C.: On the retrieval of volcanic sulfur dioxide emissions from GOME backscatter measurements, *J. Atmos. Chem.*, **50**, 295–320, 2005.
- Vandaele, A. C., Simon, P. C., Guilmot, J. M., Carleer, M., and Colin, R.: SO₂ absorption cross section measurement in the UV using a Fourier transform spectrometer, *J. Geophys. Res.*, **99**, 25 599–25 605, 1994.
- 20 Van Roozendael, M., Soevijanta, V., Fayt, C., and Lamvert, J.-C.: Investigation of DOAS issues affecting the accuracy of the GDP version 3.0 total ozone product, in: ERS-2 GOME GDP 3.0 Implementation and Delta Validation, ERSE-DTEX-EOAD-TN-02-0006, ESA/ESRIN, Frascati, Italy, 97–129, 2002.
- 25 Vountas, M., Rozanov, V. V., and Burrows, J. P.: Ring effect: Impact of rotational Raman scattering on radiative transfer in earth's atmosphere, *J. Quant. Spectrosc. Ra.*, **60**, 943–961, 1998.
- Wagner, T. and Platt, U.: Satellite mapping of enhanced BrO concentrations in the troposphere, *Nature*, **395**, 486–490, 1998.
- 30 Wahner, A., Ravishankara, A. R., Sander, S. P., and Friedl, R. R.: Absorption cross section of BrO between 312 and 385 nm at 298 and 223 K, *Chem. Phys. Lett.*, **152**, 507–512, 1988.
- Weber, M., Lamsal, L. N., Coldewey-Egbers, M., Bramstedt, K., and Burrows, J. P.: Pole-to-pole validation of GOME WFDOAS total ozone with groundbased data, *Atmos. Chem. Phys.*, **5**,

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1341–1355, 2005, <http://www.atmos-chem-phys.net/5/1341/2005/>.

Weber, M., Lamsal, L. N., and Burrows, J. P.: Improved SCIAMACHY total ozone retrieval: Steps towards homogenizing long-term total ozone datasets from GOME, SCIAMACHY, and GOME2, The 2007 ESA ENVISAT Symposium, Montreux, Switzerland, 23–27 April 2007, 2007.

Wittrock, F., Richter, A., Oetjen, H., Burrows, J. P., Kanakidou, M., Myriokefalitakis, S., Volkamer, R., Beirle, S., Platt, U., and Wagner, T.: Simultaneous global observations of glyoxal and formaldehyde from space, *Geophys. Res. Lett.*, 33, L16804, doi:10.1029/2006GL026310, 2006.

Yang, K., Krotkov, N. A., Krueger, A. J., Carn, S. A., Bhartia, P. K., and Levelt, P. F.: Retrieval of large volcanic SO₂ columns from the Aura Ozone Monitoring Instrument: Comparison and limitations, *J. Geophys. Res.*, doi:10.1029/2007JD008825.

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Table 1. Specifications of SO₂ retrievals for SCIAMACHY data using Weighting Function DOAS (WFDOAS) and Standard DOAS (SDOAS) techniques.

	Wavelength range, nm	Polynomial order	Cross-sections included in the fit
WFDOAS	315–327	3	SO ₂ ^a , WF-O ₃ ^b , WF-T ^c , BrO ^d Ring-ORSC ^e
SDOAS	315–327	4	SO ₂ ^a , O ₃ ^f , Ring ^g , USamp ^h , ETA ⁱ

^a SO₂ cross-section at 295 K (Vandaele et al., 1994);

^b O₃ weighting function;

^c temperature weighting function;

^d BrO cross-section at 228 K (Wahner et al., 1988);

^e Online Ring spectrum calculation;

^f two O₃ cross sections obtained at 223 K and 243 K (Bogumil et al., 2003) were included in the fitting routine;

^g Ring spectrum from Vountas et al. (1998);

^h undersampling correction;

ⁱ polarization dependency of the SCIAMACHY instrument.

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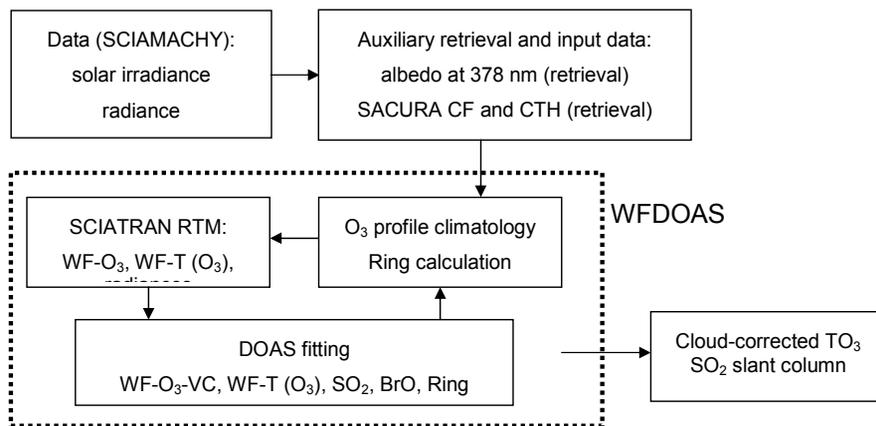


Fig. 1. Scheme of the WFDOAS algorithm for SO₂ retrieval from SCIAMACHY data. The radiative transfer model, SCIATRAN 2.0 was incorporated to calculate the modelled radiance and weighting functions. CF and CTH indicate cloud fraction and cloud-top-height from the SACURA algorithm (Kohkanovsky et al., 2006), respectively.

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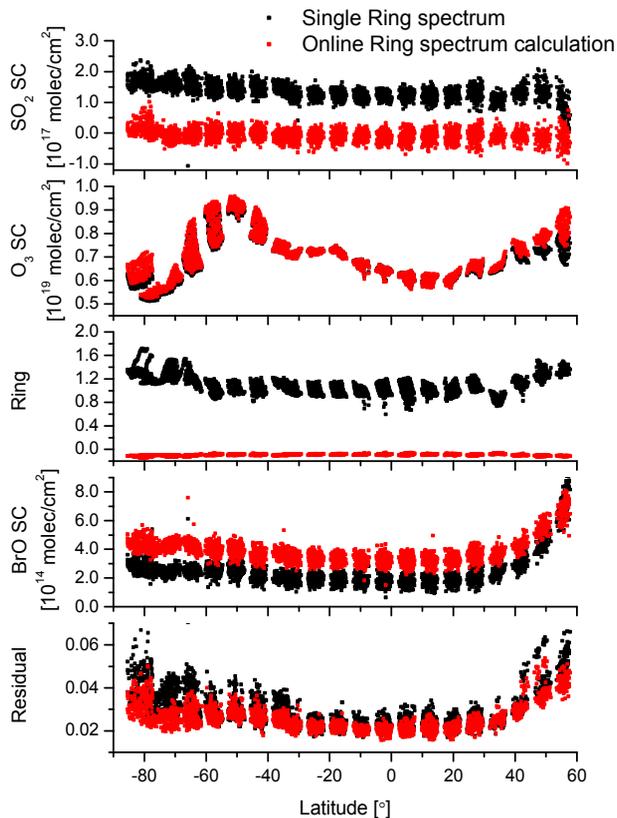


Fig. 2. Results of WFDOAS retrieval: (red dots) retrievals integrating the online Ring spectrum calculation (ORSC) and (black dots) incorporating a single Ring spectrum (Vountas et al., 1998). The spectra were taken in the 60° E and 100° E longitudinal region on 30 November 2005 (SCIAMACHY orbit no. 19611). Residuals are peak-to-peak values.

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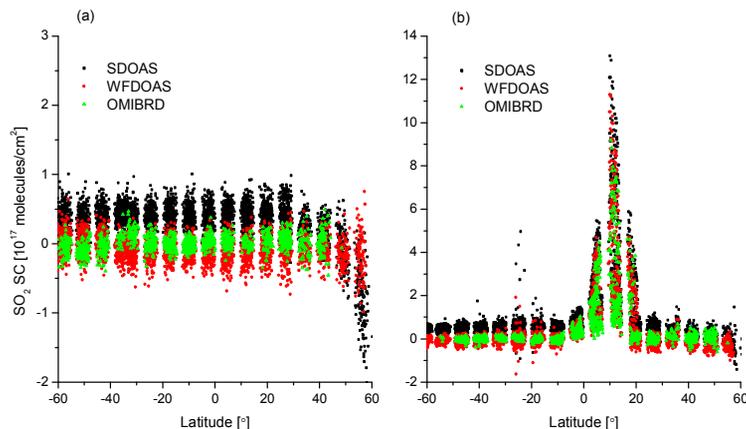


Fig. 3. SO₂ slant columns retrieved by the WFDOAS and SDOAS methods in the cases of remote background conditions **(a)** and volcanic eruptions **(b)**. The spectra for background condition were taken in the 60° E and 100° E longitudinal region on 30 November 2005 (SCIAMACHY orbit no. 19611, see also Fig. 2). Spectra of volcanic eruption (near and around Nyamuragira volcano in Congo, erupted on 27 November 2006) were obtained in the 80° W–120° W longitudinal region on 2 December 2006 (SCIAMACHY orbit no. 24868). For the comparison of SCIAMACHY SO₂ slant columns (SC) with OMI SO₂ SC are added, which are retrieved by the Band Residual Difference (BRD) algorithm (Krotkov et al., 2006, 2008).

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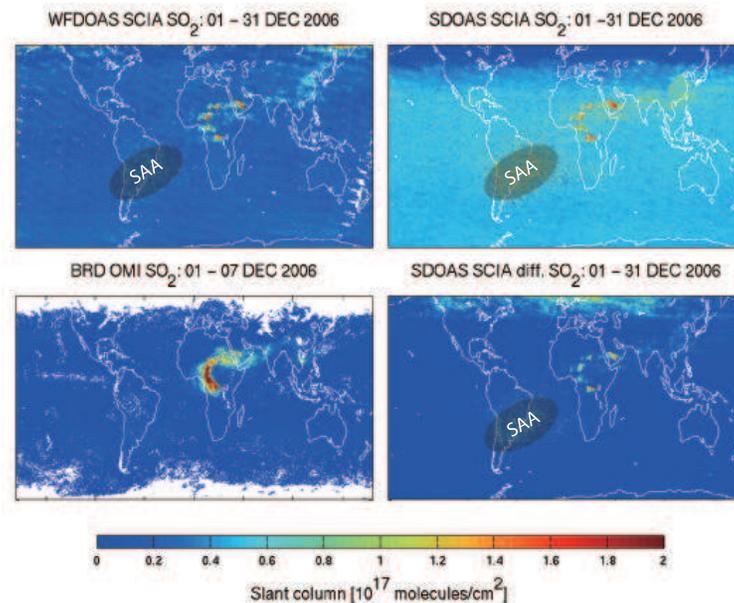


Fig. 4. Global maps of SO₂ slant columns averaged over December 2006. The panels show SO₂ slant columns from the WFDOAS retrieval, the SDOAS retrieval, the RSM-corrected SDOAS retrieval, and the OMI BRD algorithm. In the area of the Southern Atlantic Anomaly (SAA), large scatter in WFDOAS and SDOAS SCIAMACHY SO₂ results from exposure of the instrument to radiation and particles. RSM: Reference sector method (Martin et al., 2002; Richter and Burrows, 2002). There are the open areas in WFDOAS retrievals due to no SACURA data and in the BRD OMI retrievals due to the cloud-screening.

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