Belgian Institute
for Space Aeronomy

BIRA-IASB

Sulphur Dioxide Data Service

Algorithm description Document

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1 Introduction

1.1 Purpose of this document

A Sulphur Dioxide Data Service has been set up by BIRA-IASB, in collaboration with KNMI and DLR, in the framework of the projects TEMIS, PROMOTE and SACS. This Service has two branches:

- Archive Service
- NRT Service

The data are accessible via the TEMIS and SACS websites:
http://www.temis.nl/aviation/so2.php and http://sacs.aeronomie.be/

This document describes the algorithm used to retrieve the data products of the Sulphur Dioxide Data Service. The latest version of this document can be found on the SO2 Products Info Service website at
http://sacs.aeronomie.be/info/docs.php

The Data and Service version history is given in section 2.1 of the *Product Specification Document* [PSD].

1.2 Acronyms and abbreviations

AMF	Air-Mass Factor
BIRA-IASB	Belgian Institute for Space Aeronomy
DLR	German Centre for Air and Space Research
DOAS	Differential Optical Absorption Spectroscopy
DU	Dobson Units
ESA	European Space Agency
HDF	Hierarchical Data Format
KNMI	Royal Netherlands Meteorological Institute
NOVAC	Network for Observation of Volcanic and Atmospheric Change
NRT	Near-real-time
PROMOTE	Protocol Monitoring for the GMES Service Element: Atmosphere
SCD	Slant Column Density
SO2	Sulphur Dioxide
SZA	Solar Zenith Angle
TEMIS	Trophospheric Emission Monitoring Internet Service

VCD	Vertical Column Density

1.3 Applicable documents

[ADD]	Algorithm Description Document: BIRA_SO2_ADD_v11r1.doc
[PSD]	Product Specification Document: BIRA_SO2_PSD_v11r1.doc

1.4 Credits

The SO2 Data Service is set up and distributed as part of the following projects:

TEMIS -- Tropospheric Emission Monitoring Internet Service
http://www.temis.nl/

PROMOTE -- Protocol Monitoring for the GMES Service Element
http://www.gse-promote.org/

SACS -- Support to Aviation Control Service
http://sacs.aeronomie.be/

by the Belgian Institute for Space Aeronomy (BIRA-IASB, Brussels, Belgium) in collaboration with DLR (Oberpfaffenhofen, Germany) and KNMI (De Bilt, The Netherlands).

BIRA-IASB
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2 Background

Satellite UV-visible instruments, such as SCIAMACHY, OMI and GOME-2, measure the intensity of the sunlight in a nadir-viewing geometry, as function of the wavelength. The measured earthshine spectra accounts for all photons reaching the instruments that have been scattered by air molecules, aerosols and clouds or reflected by the surface of the Earth. This is depicted in Figure 1.

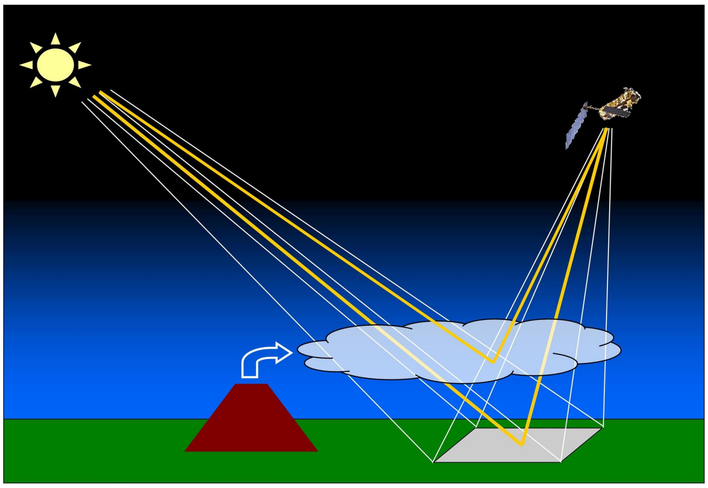

Figure 1: Sketch of a nadir-viewing satellite UV-visible measurement.

Along the different light paths, some of the photons are absorbed by the trace gases in the atmosphere (e.g., SO_2). Therefore the ratio between the measured earthshine spectra and the solar (absorption free) spectrum provides information on the amount of these absorbing trace gases in the atmosphere. However, it contains the signal from all species absorbing along the mean light path.

The SO_2 vertical columns are retrieved from the observations of the SCIAMACHY satellite instrument (see example in Figure 2) using the well-known DOAS (Differential Optical Absorption Spectroscopy) approach which consists in three main steps:

1. The atmospheric absorbers (in particular SO_2) are separated using the characteristic differential structures of their absorption cross-sections. The retrieved quantity can be interpreted as the integrated SO_2 concentration along the mean slant light path, it is the so-called slant column density (SCD). The spectral evaluation consists in a least-squares fit procedure where atmospheric cross-sections are adjusted to the log-ratio of a measured and a reference spectrum in a selected wavelength interval. The residual broadband features due to Rayleigh and Mie scattering are removed using a low-pass filtering method.
2. A background correction is applied to the retrieved SO_2 SCDs in order to avoid non-zero columns over regions known to have very low SO_2

and to insure a geophysical consistency of the results at high solar zenith angles.

3. The corrected SO_2 slant column is converted into a SO_2 vertical column (VCD). This implies the calculation of an air mass factor (AMF=SCD/VCD) which characterize the enhancement of the light path w.r.t. the vertical path. The AMF computation is done using simulations of the transfer of the radiation in the atmosphere. It requires accounting for parameters influencing the photons path (and hence the measurement sensitivity): solar zenith angle, instrument viewing angles, surface albedo, atmospheric absorption, scattering on molecules, clouds.

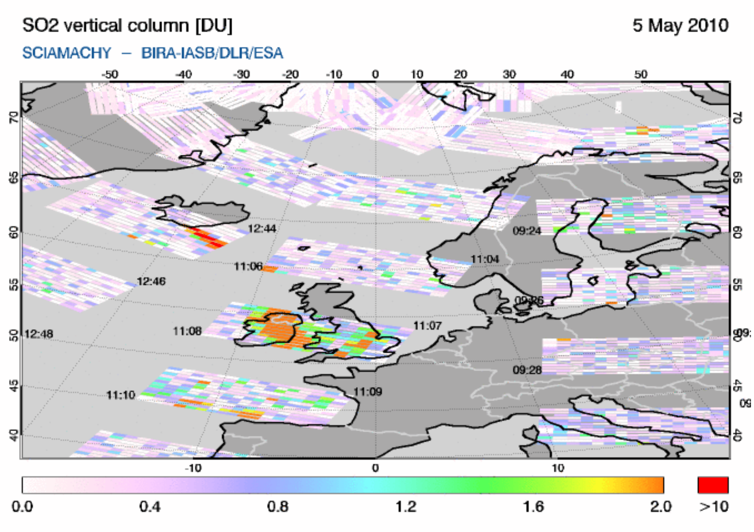

Figure 2: Example of SO_2 vertical columns from SCIAMACHY observations after the 2010 eruption of Eyjafjoll (Iceland).

3 The SO2 slant column density

Sulphur dioxide (SO2) slant column densities are the basis of the SO2 Data Service. This Service has two branches:

- Archive Service
- NRT Service

The Service provides global data files and maps (daily, and monthly average maps). Higher resolution maps over pre-defined regions known for SO2 emission from anthropogenic pollution or from volcanic activity are provided as well.

For a description of the data product, as well as of the data and service version history, see the *Product Specification Document* [PSD].

3.1 SO2 slant column retrieval

The SO2 slant column density, usually given in Dobson Units (DU), are retrieved from UV measurements made by SCIAMACHY using the Differential Optical Absorption Spectroscopy (DOAS) technique.

The retrieval of the SO2 slant column is done from the spectrum in the wavelength range 315-326 nm, which appears as a good compromise for accurate SO_2 slant column determination using DOAS. The detection limit however depends on the observing conditions (time and place) and the solar zenith angle. In particular, the strong ozone absorption in the UV can interfere with SO_2 retrieval (see Figure 3) so that larger effective ozone absorption is a source of bias and generally results in larger background noise.

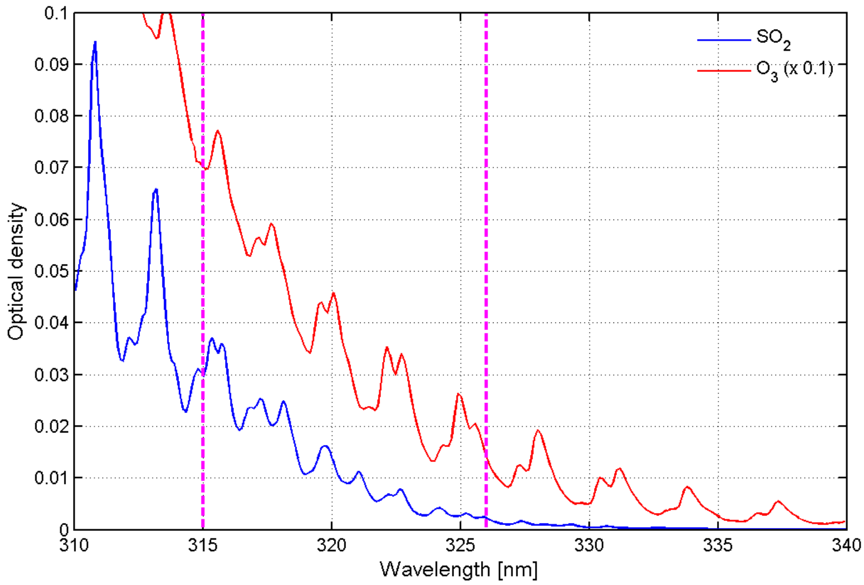

Figure 3: Typical optical densities of SO_2 and O_3 in the wavelength region from 310 to 340 nm. Note that the O_3 optical density has been divided by a factor of 10 for clarity reason.

The detailed settings for SCIAMACHY SO_2 DOAS fits are summarized in Table 1. In addition to SO_2, ozone cross-sections at two temperatures (218 and 243K) are included in the fit. Furthermore two Ring spectra are fitted. The latter spectra are being generated using SCIATRAN radiative transfer modeling.

Table 1: DOAS settings used for SCIAMACHY SO_2 slant column retrieval.

Fitting interval	315 - 326 nm
Sun reference	Equatorial earthshine spectrum
Wavelength calibration	Wavelength calibration of sun reference optimized by NLLS adjustment on convolved Chance and Spurr solar lines atlas
Absorption cross-sections	
- SO_2	SCIA Flight Model [Bogumil et al., 1999], 203 K (15 km), 243 K (6 km), 273 K (2.5 km)
- Ozone	Malicet et al. [1995], 218 K and 243 K
- Ring effect	2 Ring eigenvectors generated using SCIATRAN
Polynomial	3rd order (4 parameters)
Intensity offset correction	Constant offset

The result of the interference with O3 is that when concentrations of SO2 are low, the retrieval may give negative SO2 slant column values, with an error of the same magnitude. This then represents the SO2 background level, i.e. the apparent SO2 absorption in the absence of emissions of SO2. The negative values are, of course, not physical.

For low Solar Zenith Angles (SZAs), i.e. at low and mid latitudes, the SO2 background level is of the order of 1 Dobson Unit (DU) and emissions of SO2 (by pollution or volcano eruptions) will be visible against this background.

For higher SZAs, however, there is a problem. The higher the SZA, the longer the slant path is along which the retrieval takes place. Because of the vertical and horizontal distribution of ozone in the atmosphere (with a strong concentration in the lower stratosphere, the "ozone layer") a longer slant path means a larger relative absorption by ozone. For SO2 this is much less the case, as SO2 emissions generally show a rather limited distribution horizontally and vertically.

Effectively this means that at higher SZAs the interference between the SO2 and O3 absorption results in more negative SO2 slant column values, with large errors. This effect is corrected for by the background correction which is applied to the slant column data.

3.2 Background correction of the SO2 slant column

A background correction is applied to the SO2 slant columns retrieved with DOAS. There are two reasons to apply such a correction:

- The DOAS retrieval requires the use of a reference spectrum. For this a measurement is used from a location thought to be free of SO2, but it can lead to an offset in the SO2 slant column.
- At higher Solar Zenith Angels the "interference" between the SO2 and ozone absorption results in more negative SO2 slant column values, with large errors.

To find the offset 1, all SO2 slant column values for SZA < 50 degrees are averaged, with the SO2 slant column error as weights in the averaging. There may be large SO2 peak values that are real signals and these should therefore not be taken into account when computing the background that is assumed to be free of SO2. For that reason, once the above mentioned averaging gives the offset, another round of averaging is done of all the data that does not differ more than 5 DU from the average of the first round. The average of the second round then gives the offset value to be used in the background correction.

To find the offset 2 for the interference in the absorption signals at high SZA, a function is sought which relates the SO2 slant column against the ozone slant column value that is also given by the DOAS fit.
Figure 4 shows an example of the measured SO2 slant columns as function of the measured O3 slant columns for 01 March 2007. It is clear that the observed decrease of the SO2 slant column is due to the increase in ozone absorption along the slant path with increasing SZA.

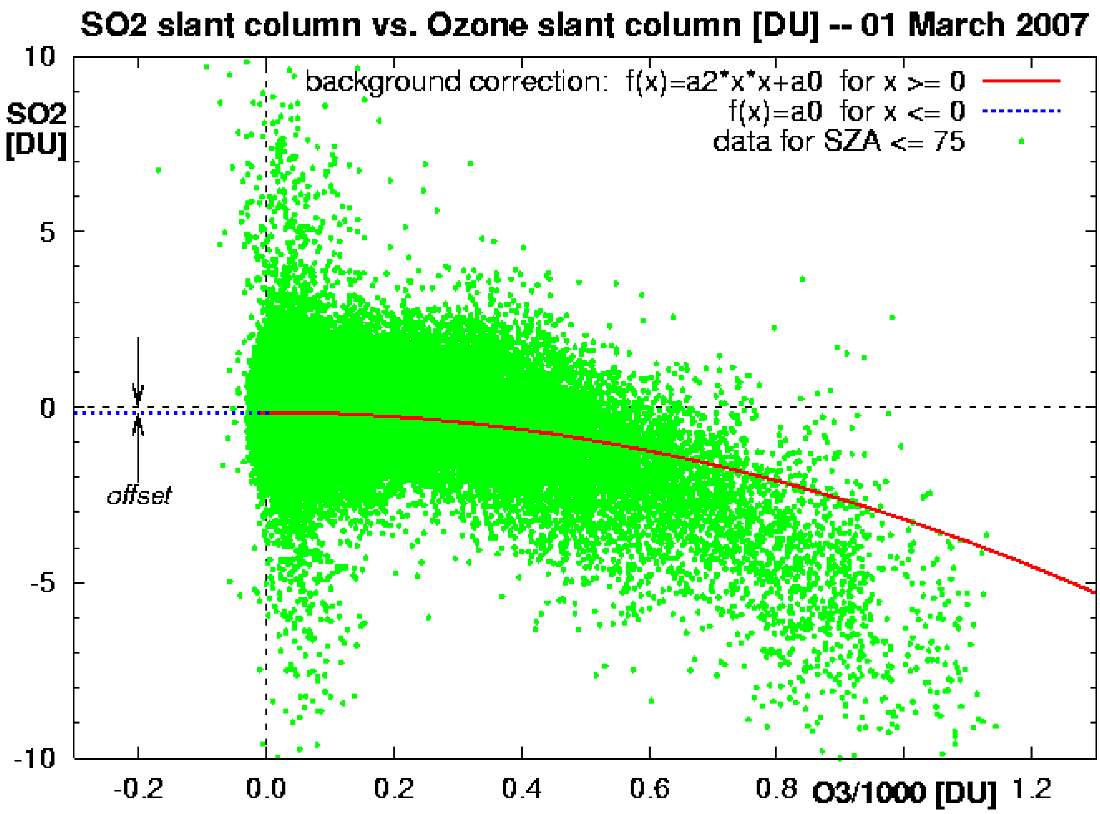

Figure 4 ***The green dots in this graph are all data points of 1 March 2007 with SZA < 75 degrees, with the SO2 slant column in DU as function of the ozone (O3) slant column in units of 1000 DU. The arrows and the blue dashed line on the left indicate the offset ("a0") due to the use of a reference spectrum determined from an average of all data points from March with SZA < 50 degrees. The solid red curve is the parabolic function of the background correction that follows from all data points of two years (2005-2006).***

Looking at the date points for SZA < 75 degrees for a given period of time indicates that this function is parabolic.

Hence the SO2 slant column correction is given by:

$$SO_2SCDcor[DU] = K.(\frac{O_3SCD[DU]}{1000})^2 \qquad (1)$$

The SO2 values at high ozone concentrations vary a lot from day to day and from month to month. Performing a parabolic fit through the data would then give different coefficients for different months, and these coefficients can vary really a lot (from -8 to close to zero). The reason for this large variation in the parabolic coefficient is that the number of data points for SZA > 50 degrees varies from month to month and is much smaller than for lower SZA.

The parabolic coefficient for the background correction, shown by the red line in the above figure, is based on a fit through all data points with SZA < 75 degrees for all days from 2004 to 2008.

Again to avoid real SO2 peak values from influencing the fit, the fit is done in two rounds: first with all points, then again but omitting those points that have an SO2 slant column of more than 5 DU away from the parabolic curve.

3.3 Reference spectra

The slant column retrieval algorithm requires a reference spectrum without the presence of absorption features of the trace gas to be retrieved, since the DOAS method is based on the difference in absorption in two spectra. For the SO2 slant column retrieval a reference spectrum without any SO2 absorption must therefore be selected. A good geographic region to do this is around the equator above the Pacific or Indian ocean, as there are no sources of SO2 located there. As instrument characteristics may vary over time, it is necessary to regularly update the reference spectrum with time.

For the retrieval for the Volcanic & Air Quality Services, a new reference spectrum is selected in principle once every month at around the middle of the month, depending on availability of the data, from a measurement south-west of the southern tip of India, at about 65 degrees East and just below the equator.

Since the reference spectrum changes every month, the correction for the SO2 background levels (section 3.2) is determined for each month separately:

$$K = a + b.Mn + c.Mn^2 \qquad (2)$$

Where Mn is the month number since January 2004 (01/2004=1) and a,b,c are the fit coefficients:

a=-3.92703
b=-0.00111463
c=0.000620777

4 The SO2 vertical column density

This chapter describes the meaning of a slant column and a vertical column density and how the first is converted to the second.

4.1 Slant Column and Vertical Column Densities

Nadir-viewing satellite based instrument, measure the sunlight scattered in the atmosphere and reflected by the surface of the Earth, as function of the wavelength of the light. In other words: the instruments measure earthshine spectra. Comparing such a spectrum with the spectrum of the sunlight itself provides information on the distribution and concentration of trace gases, such as ozone and SO2, because these gases absorb part of the incoming sunlight. (Instead of using the solar spectrum for this comparison, one can also use an earthshine spectrum from a part of the atmosphere free of the trace gases under study – this is the approach described in section 3.3 and applied in the SO2 Data Service.)

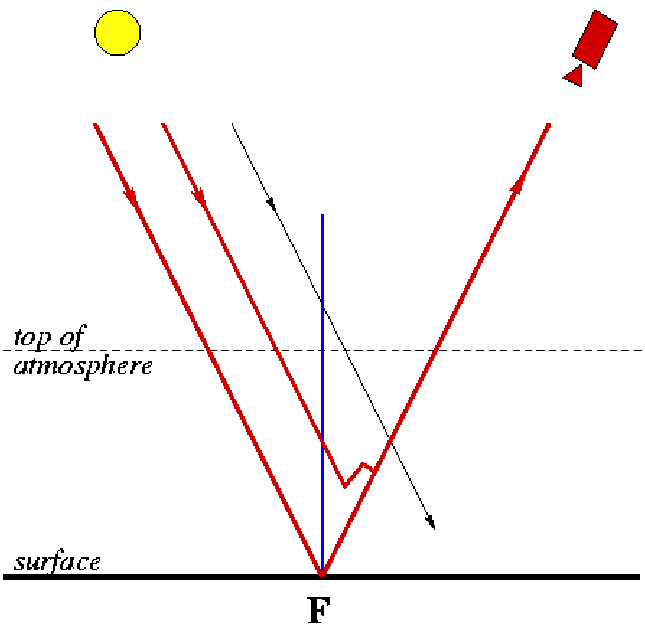

Figure 5

Figure 5 shows schematically different paths (red lines) of sunlight reaching the satellite through the atmosphere, reflected by the earth's surface and scattered in the atmosphere. **F** is the footpoint, the point on the earth's surface the satellite is looking at. A part of the light reaching the satellite is reflected by the earth's surface, another part is scattered higher in the atmosphere. The thick blue line represents the "vertical column" at footpoint **F**.

As the light follows these paths, part of the photons are absorbed by the trace gases in the atmosphere. The spectrum of the light measured by the satellite (the sum, as it were, of the thick red lines in the graph) thus provides information on the trace gases averaged *along all the light paths*. In other words, the total density of a given gas, such as SO2, is the concentration of this gas along the mean optical light path. This is usually called the *slant column density* (SCD) associated with footpoint F, the point on the earth's surface the satellite is looking at.

The SCD clearly does not provide the total concentration *right above* footpoint **F**, i.e. along the blue line in the graph. The total concentration along this line is called the *vertical column density* (VCD). It is this VCD that provides the most useful and directly interpretable information on the distribution and concentration of trace gases. It is therefore desirable to convert the SCD in the VCD.

As can be seen from the graph above, the VCD (along the blue line) is usually smaller than the SCD (along the red lines). The ratio between these two column densities:

$$AMF = SCD / VCD \tag{3}$$

is called the *Air-Mass Factor*. The value of the AMF depends on the length of the light path, the vertical distribution of absorbing trace gases in the atmosphere, the reflectivity (albedo) of the earth's surface, etc. The length of the light path depends on the position of the Sun (expressed in the Solar Zenith Angle, SZA) and the angle under which the satellite is looking at the atmosphere. The AMF is pre-calculated for a variety of these quantities and applied to the SCD to find the VCD at footpoint **F**.

Figure 6 shows a schematic representation of the slant path (thick red lines) of incoming sunlight through the earth's atmosphere to the satellite, associated with the footpoint **F**, the point on the earth's surface the satellite is looking at, in the presence of clouds. These clouds partly shield the satellite's view of the atmosphere above footpoint **F**.

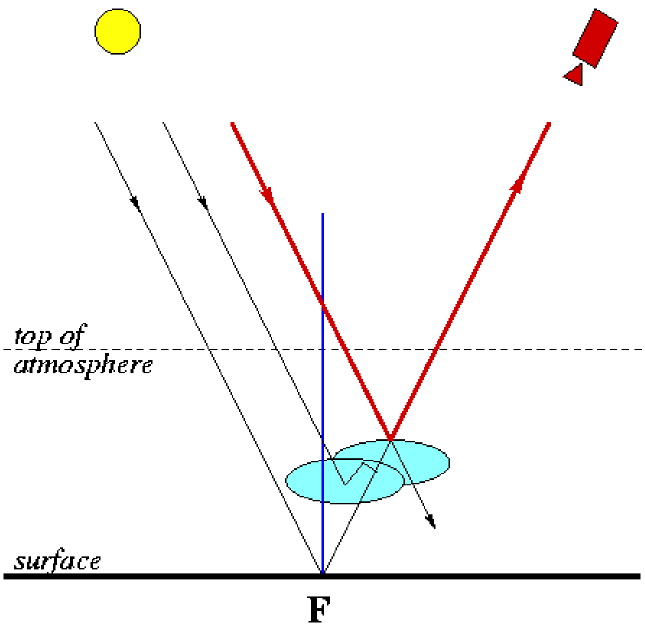

Figure 6

If there are clouds in the atmosphere, things become more complicated. Clouds reflect (and scatter) incoming sunlight and thus effectively shield all that is going on below the clouds from the satellite's view. Clearly, the satellite measurements provide an SCD which contains only information on the atmosphere above the clouds. To find the real VCD at footpoint **F**, for example of SO2, in such situations, an "effective" AMF is computed, taking the cloud fraction (which gives the percentage of the cloud cover) into account. In the presence of clouds the VCD is clearly less accurate than the VCD derived under clear-sky conditions.

4.2 Air Mass Factor using a Radiative Transfer Model

The air mass factors are determined using pre-calculate look-up tables, made with an off-line radiative transfer model (LIDORT). The look-up tables have a set of entries: the viewing geometry, the solar zenith angle, the surface albedo, the cloud fraction and cloud top pressure.

The treatment of partially cloudy pixels (characterized by an effective cloud fraction area *f*) is based on the hypothesis of the 'independent pixel approximation'. It is assumed that the intensity measured by the instrument is the sum of the intensities of a completely clear and a completely cloudy pixel weighted by (1-*f*) and *f* respectively. Under such circumstances, the AMF for a partially cloudy scene is given by:

$$AMF = \Phi \cdot AMF_{cloud} + (1 - \Phi) \cdot AMF_{clear} \quad (4)$$

where AMF_{cloud} is the AMF for a completely cloudy scene, and AMF_{clear} is the AMF for a corresponding cloud-free pixel. Φ is the intensity-weighted cloud fraction:

$$\Phi = \frac{f \; I_{cloud}}{f \; I_{cloud} + (1-f) \; I_{clear}} \tag{5}$$

I_{cloud} and I_{clear} being the backscattered intensities for fully cloudy and clear scenes, respectively. The variables in Eq. (4) are estimated using the cloud fraction retrieved by the FRESCO algorithm and by considering two Lambertian reflectors consistent with the FRESCO settings: one reflector representing the cloud at the retrieved cloud top pressure (albedo: 0.8) and the other one representative of the surface.

For the AMFs calculations, an a priori volcanic SO2 profile is assumed with a predefined central plume height. As the correct plume height is rarely available at the time of the measurement, the SCIAMACHY SO_2 product provides three different vertical column densities based on the assumption about the vertical profile of the SO_2 in the atmosphere.
The SO_2 is assumed to be in a 1 km thick layer centred around:

- 1 km above ground level
- 6 km above sea level, or 1 km above ground level if that is higher
- 14 km above sea level

The lowest level for each of these cases in general represents SO_2 in the planetary boundary layer (PBL) from anthropogenic activities or passive degassing of low volcanoes. The middle tropospheric level in general represents SO_2 from effusive volcanic eruptions or passive degassing of high volcanoes. The lower stratospheric level represents SO_2 from explosive volcanic eruptions.

As an illustration, the following image shows typical SO2 AMF for different plumes heights varying as a function of the solar zenith angle.

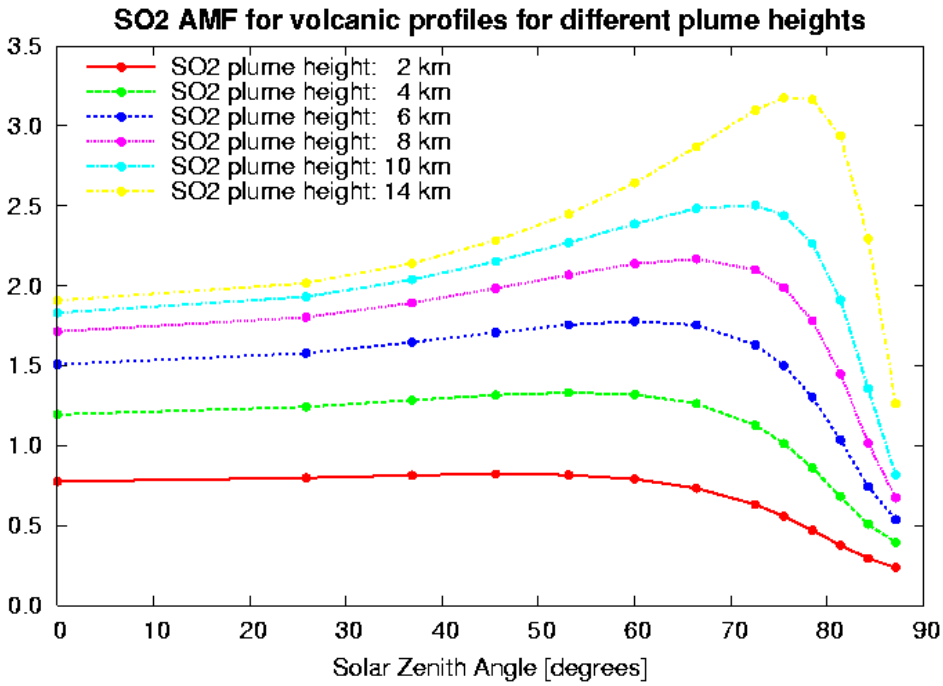

Figure 7. Air-mass factors as function of Solar Zenith Angle for an SO_2 plume of 1 km thickness and with low to moderate SO_2 concentrations (up to 10 DU). The viewing geometry is of nadir; the surface is at sea-level and with an albedo of 0.05.

All three VCDs s are available in the data files, whereas the lower stratospheric column is used for the plots presented on these web pages. To find an estimate of the VCD for another plume height, one could linearly interpolate between the three given values. The SCD is also plotted and can be accessed under "other plots".

If the VCD is available, images of it are shown on the website and the values are given in the data files. The data files also give an error estimate, which simply is the SCD error estimate divided by the AMF; possible errors in the AMF calculation are therefore not taken into account.

If no cloud cover information is available for a given ground pixel, the AMF and VCD of that ground pixel cannot be computed. In that case, the entries in the data files will get the "no data" value. Only the AMF for the clear-sky part will be given a value, as that can in principle be computed always.

5 References

The following is a list of some references regarding SO2 emissions associated with volcanic eruptions, and the DOAS technique used for the retrieval and related publications.

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- [More to come]