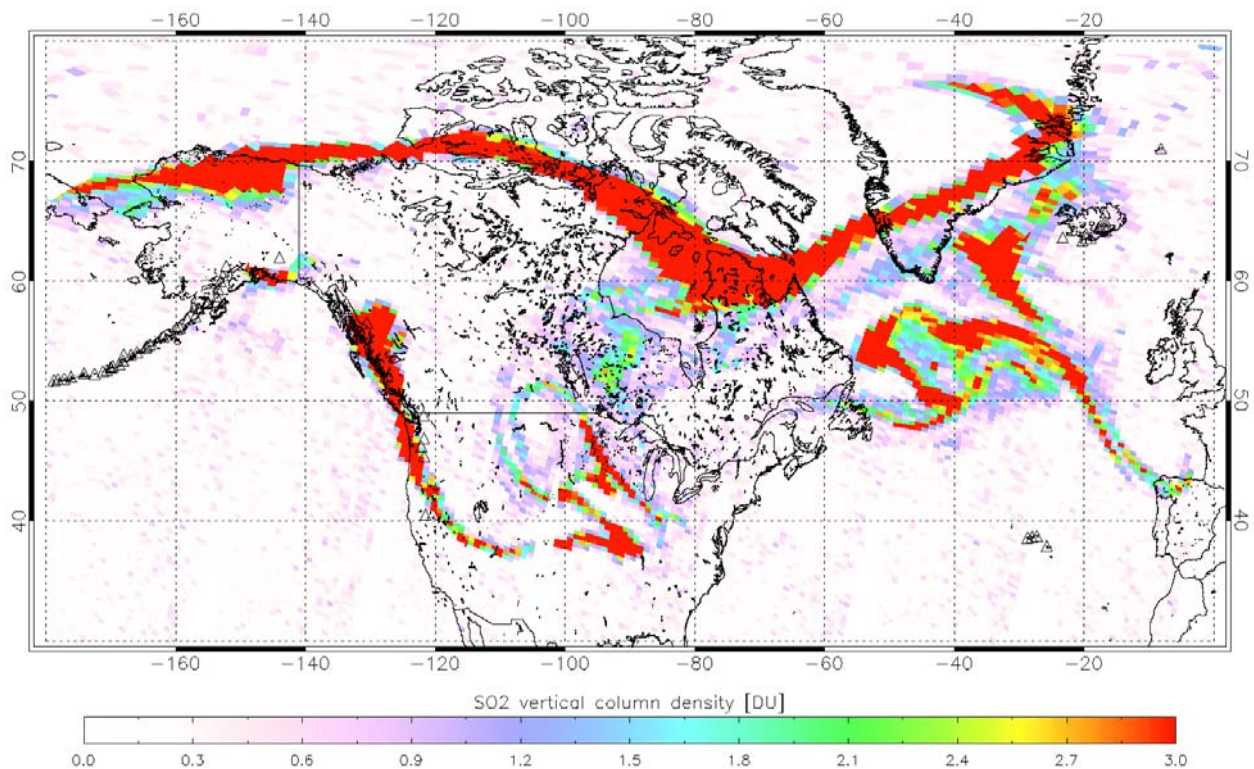


**O3MSAF – SATELLITE APPLICATION FACILITY ON
OZONE AND ATMOSPHERIC CHEMISTRY MONITORING**

Initial validation of GOME-2 GDP 4.2 SO₂ total columns (OTO/SO2) – ORR B



Distribution of SO₂ on 14 August 2008 as seen by GOME-2. The SO₂ originates from the eruption on 8 August of the Kasatochi volcano, which is located at 52.17°N, 175.51°W.

Validation reporting period March 2007 – August 2008
Input GOME-2 level-1B data v3.x and v4.0

reference / référence TN-IASB-GOME2-O3MSAF-SO2-01.1
issue / édition 1
revision / révision 1
date of issue / date d'édition 4 November 2008



auteurs / authors J. van Geffen, M. Van Roozendael, M. Rix, P. Valks
contributeurs / contributors --
reference / référence TN-IASB-GOME2-O3MSAF-SO2-01.1
issue / édition 1
revision / révision 1
status / état Final
date of issue / date d'édition 4 November 2008
document type / type de document Technical Note / Validation Report

distribution / distribution

Function	Organisation
O3M-SAF	EUMETSAT, BIRA-IASB, DLR, DMI, DWD, FMI, KNMI, RMI
GOME Team	DLR, ESA/ESRIN, BIRA-IASB, RTS, AUTH, various
UPAS Team	DLR-IMF, DLR-DFD

contributions externes / external contributors

--

historique du document / document change record

Issue	Rev.	Date	Section	Description of Change
0		15.09.08	All	Creation of document
1		29.09.08	All	Provisional validation report for ORR-B
1	1	04.11.08	A.2, D.2, E.4, E.3, G	Updated after ORR-B board report dd 30 Oct. 2008



Initial validation of GOME-2 GDP 4.2 SO₂ total columns (OTO/SO₂) – ORR B

CONTENTS

A. INTRODUCTION	- 4 -
A.1. SCOPE OF THIS DOCUMENT	- 4 -
A.2. PRELIMINARY NOTES	- 4 -
A.3. PLAN OF THIS DOCUMENT	- 4 -
B. SETTINGS FOR SO₂ COLUMN RETRIEVAL FROM GOME-2	- 6 -
B.1. EXPERIENCE FROM ERS-2/GOME AND SCIAMACHY	- 6 -
B.2. CHOICE OF SO ₂ SLANT COLUMN SETTINGS FOR GOME-2	- 7 -
C. VERIFICATION OF SO₂ SLANT COLUMNS AND SENSITIVITY TESTS	- 8 -
D. COMPARISON AGAINST SATELLITE DATA	- 11 -
D.1. ASSUMED ELEVATION OF THE SO ₂ IN THE ATMOSPHERE	- 11 -
D.2. COMPARISON OF VOLCANIC SO ₂ IN THE STRATOSPHERE	- 11 -
D.3. COMPARISON OF VOLCANIC SO ₂ IN THE MID-TROPOSPHERE	- 16 -
D.4. COMPARISON OF ANTHROPOGENIC SO ₂ IN THE LOWER TROPOSPHERE	- 19 -
E. COMPARISON AGAINST GROUND-BASED MEASUREMENTS	- 22 -
E.1. GROUND-BASED INSTRUMENTS USED FOR THE COMPARISONS	- 22 -
E.2. SATELLITE DATA USED FOR THE COMPARISONS	- 22 -
E.3. ANTHROPOGENIC SO ₂ TOTAL COLUMNS MEASURED IN BEIJING	- 22 -
E.4. THE PASSING OF VOLCANIC SO ₂ OVER GROUND STATIONS IN UCCLE AND MANCHESTER	- 24 -
F. CONCLUSION	- 27 -
G. REFERENCES	- 28 -



A. INTRODUCTION

A.1. Scope of this document

The present document reports on the verification and initial validation of MetOp-A GOME-2 SO₂ total column data over the March 2007 - August 2008 time period, produced by the GOME Data Processor (GDP) version 4.2 operated at DLR on behalf of EUMETSAT. This report includes verification work performed using the BIRA-IASB scientific retrieval tool synchronized on the GDP settings, as well as preliminary comparisons with SCIAMACHY, OMI and ground-based measurements.

A.2. Preliminary notes

SO₂ total columns as generated from GDP version 4.2 represent a new GOME-2 product, generated with the GDP operational environment system at DLR.

The aim of the present document is first to report on the status of the verification of the GOME-2 SO₂ column against a synchronised scientific algorithm available at BIRA. For this exercise, SO₂ retrieval settings selected by DLR scientists for GDP version 4.2 are being used. The consistency of this SO₂ product is then explored by performing various comparisons with SO₂ column data determined from measurements of satellite-based instruments SCIAMACHY and OMI, as well as selected ground-based instruments. Ground-based SO₂ column measurements available for volcanic eruptions (Kasatochi) and anthropogenic pollution (Beijing) are also used in an attempt to further document the geophysical consistency of the GOME-2 SO₂ product.

It should be noted here that validation of satellite-based SO₂ data products has not been done in any systematic way so far: the validation carried out so far is limited, because of the difficulties involved in comparisons between ground-based and satellite-based data, as well as difficulties in the comparison of the data from different satellites. These difficulties are, for example, related to measuring different air masses at different moment of the day. There have been some preliminary comparisons of OMI SO₂ data against in-situ aircraft measurements over NE China [Krotkov et al., 2008], and in-situ ground-based measurements at some stations in China has been compared against SCIAMACHY and OMI data [Zhang et al., 2008]. Additional validation exercises – both in-site vs. satellite comparisons and satellite inter-comparisons – are part of several projects related to monitoring of volcanic or anthropogenic SO₂ emissions; results of these comparisons are not published yet. The validation results presented here must be considered as preliminary and therefore subject to possible revisions.

Reported validation studies were carried out at the Belgian Institute for Space Aeronomy (BIRA-IASB, Brussels, Belgium) and at DLR Remote Sensing Technology Institute (DLR-IMF, Oberpfaffenhofen, Germany) in the framework of EUMETSAT Satellite Application Facility on Ozone and Atmospheric Chemistry Monitoring (O3M-SAF).

A.3. Plan of this document

This document is divided in 4 main parts, addressing respectively the description of the retrieval settings applied for the demonstration product, the verification of this product, comparisons against satellite data and comparisons against ground-based measurements. This is followed by concluding remarks and perspectives for future work.

A.4. Applicable O3MSAF Documents

- [ATBD] Algorithm Theoretical Basis Document for GOME-2 Total Column Products of Ozone, Minor Trace Gases, and Cloud Properties, DLR/GOME-2/ATBD/01, Rev. 2/A, Valks, P. and Loyola, 2008.
- [PUM] Product User Manual for GOME Total Column Products of Ozone, Minor Trace Gases, and Cloud Properties, DLR/GOME/PUM/01, Rev. 2/A, Loyola D., W. Zimmer, S. Kiemle, P. Valks, T. Ruppert, 2008.
- [PRD] Product Requirements Document, SAF/O3M/FMI/RQ/PRD/001/Rev. 06, D. Hovila, J., S. Kiemle, O. Tuinder, H. Joench-Soerensen, F. Karcher, 2008.

A.5. Technical information

GOME-2 product name SO2 total column (OTO/SO2)

Validation reporting period March 2007 - August 2008

Level-2 processor version GDP 4.2

Input GOME-2 Level-1B data version table

Start Date	Start Orbit	Level 1B Version
Mar. 08, 2007	1980	3.2.x
Apr. 03, 2007	2355	3.3.x
Sep. 03, 2007	4529	3.4.x
Oct. 04, 2007	4969	3.5.x
Nov. 14, 2007	5552	3.7.x
Jan. 31, 2008	6662	3.8.5
Mar. 10, 2008	7213	3.9.1
Jun. 26, 2008	8749	4.0.0

B. SETTINGS FOR SO₂ COLUMN RETRIEVAL FROM GOME-2

B.1. Experience from ERS-2/GOME and SCIAMACHY

Total columns of SO₂ emitted by volcanoes or from anthropogenic sources have been measured from UV-Visible space nadir sounders (GOME, SCIAMACHY, OMI) using differential absorption features around 320 nm [Eisinger and Burrows, 1998; Khokar et al., 2005; Thomas et al., 2005]. Experience from retrievals performed using these instruments have led to the selection of the 315-326 nm wavelength interval, which appears as a best compromise for accurate SO₂ 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 SO₂ retrieval (see Figure B.1.) so that larger effective ozone absorption is a source of bias and generally results in larger background noise. The detection limit for SO₂ therefore increases with ozone concentration. With the GOME and SCIAMACHY instruments, one generally admits that under most observational conditions (low and moderate SZAs) the detection limit for SO₂ columns lays around 1 DU. Due to the typically low surface albedo in the UV, combined with the effect of atmospheric scattering by air and particles as well as ozone absorption, the sensitivity to SO₂ is generally weak at the surface and increases quasi-linearly throughout the troposphere (see Figure B.2.). As a result, only major sources of emissions can be detected when these occur near the surface. The sensitivity to volcanic plumes emitted at higher altitude is typically significantly larger [Thomas et al., 2005]. In case of massive injection of volcanic SO₂ the penetration depth in the UV can be strongly reduced as a result of the SO₂ absorption, providing some profile information from the analysis at different wavelengths [Richter et al., 2006; Yang et al., 2008].

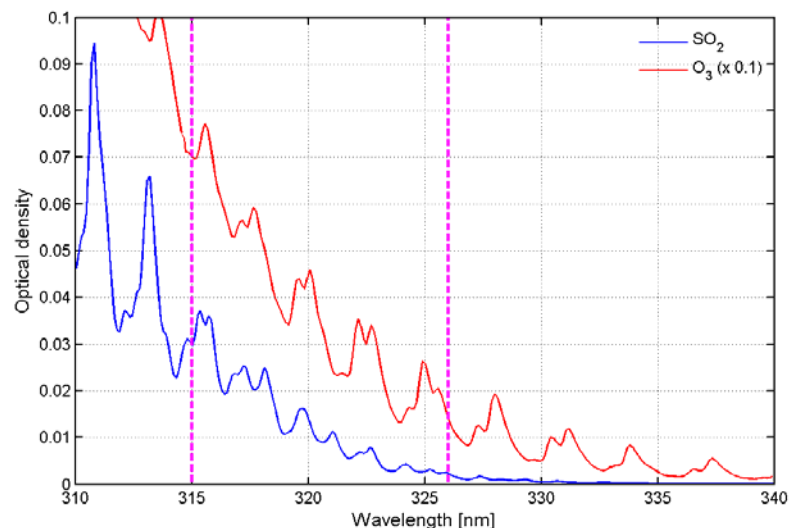


Figure B.1 – Typical optical densities of SO₂ and O₃ in the wavelength region from 310 to 340 nm. Note that the O₃ optical density has been divided by a factor of 10 for clarity reason. SO₂ columns have been retrieved from GOME and SCIAMACHY in the 315-326 nm interval.

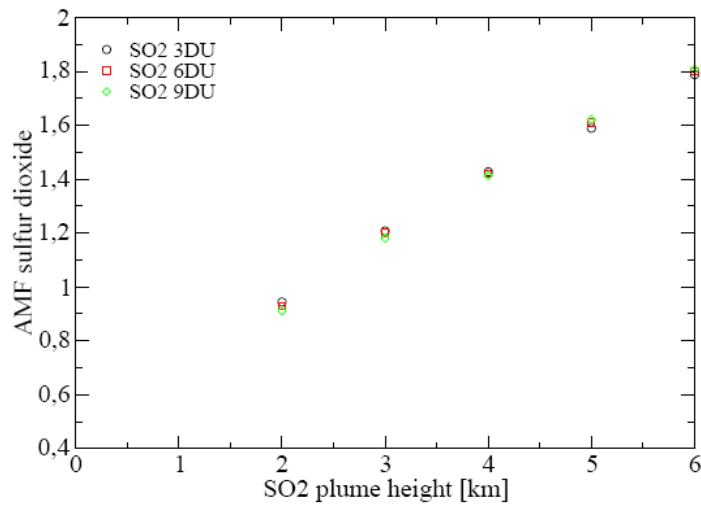


Figure B.2 – Sulphur dioxide AMFs as function of plume height and total SO₂ content. Aerosols were not taken into account. AMFs change from 0.91 to 1.81 or a factor of 2 if the plume height is varied from 2 km to 6 km. The variation with the total sulphur dioxide content remains in the order of 1% (adapted from Thomas et al., 2005).

B.2. Choice of SO₂ slant column settings for GOME-2

On the basis of past experience from GOME and SCIAMACHY, also supported by noise-driven considerations and test retrievals performed at DLR, the GOME/SCIAMACHY SO₂ DOAS fitting interval 315 – 326 nm was found to also represent an optimal choice for GOME-2 retrieval. This fitting interval was selected for the generation of a first data set, which we now consider for evaluation. In the following the consistency of the GDP 4.2 SO₂ column product is investigated from the point of view of (1) the verification (i.e. whether SO₂ retrievals performed with GDP 4.2 are consistent with scientific retrievals performed using same settings), (2) the comparison against other satellite instruments (OMI and SCIAMACHY), and (3) the comparison against independent ground-based observations.

C. VERIFICATION OF SO₂ SLANT COLUMNS AND SENSITIVITY TESTS

For verification purposes, the retrieval software of BIRA-IASB was synchronised with the GDP 4.2 processor, using a common set of slant column retrieval settings, as documented in Table C.1. Comparisons between the two processing systems were performed on a limited set of GOME-2 orbits. A high level of agreement was obtained, demonstrating the consistency between the two slant column fitting algorithms.

Table C.1 – *DOAS settings used for GOME-2 SO₂ slant column verification.*

Fitting interval	315 - 326 nm
Sun reference	Sun irradiance from GOME-2 L1 product
Wavelength calibration	Wavelength calibration of sun reference optimized by NLLS adjustment on convolved Chance and Spurr solar lines atlas
Absorption cross-sections	
- SO ₂	Reconvolved SCIA Flight Model [Bogumil et al., 1999], 203 K (15 km), 243 K (6 km), 273 K (2.5 km)
- NO ₂	GOME-2 Flight Model/CATGAS [Gür et al., 2005], 241 K
- Ozone	Malicet et al. [1995], 218 K and 243 K
- Ring effect	2 Ring eigenvectors generated using SCIATRAN
Polynomial	3 rd order (4 parameters)
Intensity offset correction	Constant offset

The level of agreement achieved when comparing SO₂ slant columns retrieved from the GDP 4.2 processor and from the BIRA-IASB scientific algorithm is illustrated in Figures C.1. to C.3. As can be seen, the elevated SO₂ slant columns (SCDs) observed respectively above Mt. Etna and Piton the La Fournaise, are captured in the same way by both algorithms. Also the level of the noise on the background SO₂ SCD retrieved in the remaining part of the orbits is very similar in both data sets, as well as the negative SCD bias typically observed at large solar zenith angle close to the terminator, mainly due to residual interference by the large stratospheric ozone absorption (note that for the operational retrieval of vertical SO₂ columns, a SO₂ background correction is applied to correct for this negative bias, see [Valks and Loyola, 2008]).

The correlation plot presented in Figure C.3. further demonstrates the good agreement obtained. One obtains a slope of 0.98 and a correlation coefficient larger than 0.99. Such results are considered as fully satisfying for the verification of the GDP 4.2 SO₂ slant column fit results. Given the close relationship between retrieval settings adopted for GOME-2 and for previous SCIAMACHY and GOME instruments, a high level of consistency is expected between these instruments for SO₂ total column retrieval. This is further investigated in the next sections. Also the consistency between GOME-2 SO₂ columns and those derived from OMI using a different retrieval approach (band-residual technique, see Krotkov et al., 2006) is explored.

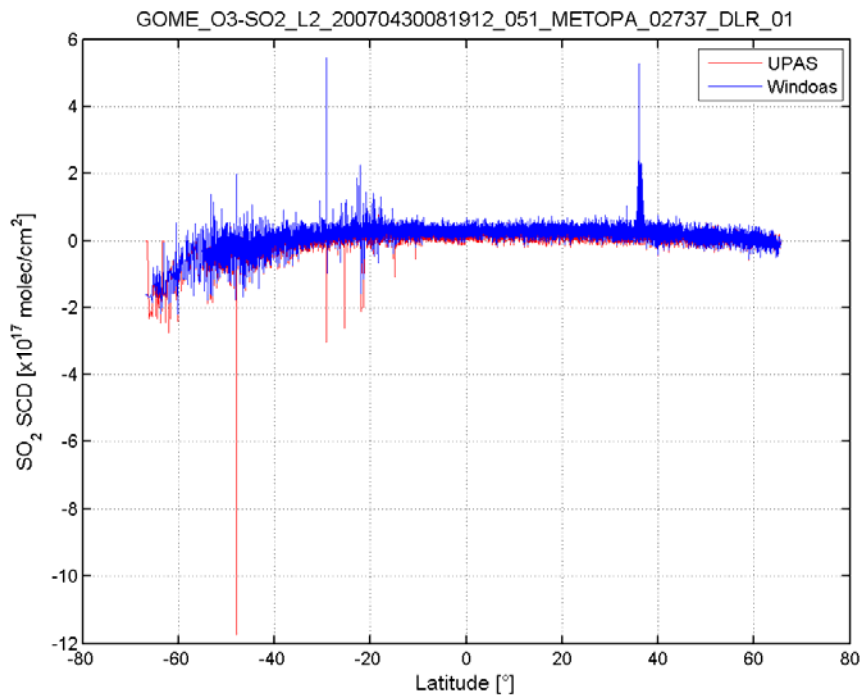


Figure C.1 – Comparison of SO_2 slant columns retrieved from GDP 4.2 and from the BIRA-IASB scientific algorithm for orbit 2737 on 30 April 2007, including an eruption of Mount Etna. DOAS settings were synchronised according to Table C.1.

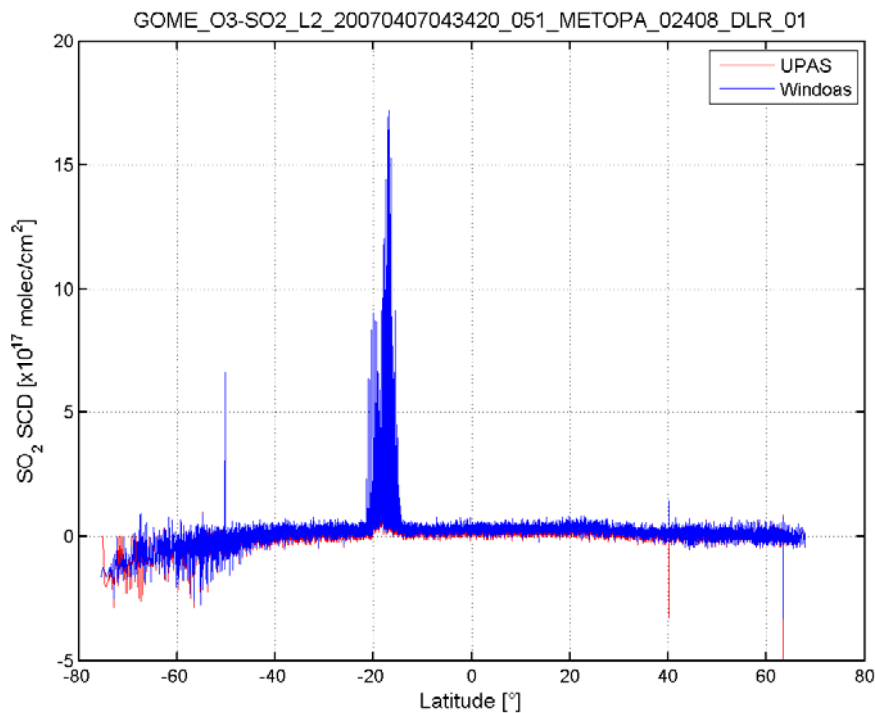


Figure C.2 – Comparison of SO_2 slant columns retrieved from GDP 4.2 and from the BIRA-IASB scientific algorithm for orbit 2408 on 7 April 2007, including an eruption of Piton de la Fournaise, Reunion. DOAS settings were synchronised according to Table C.1.

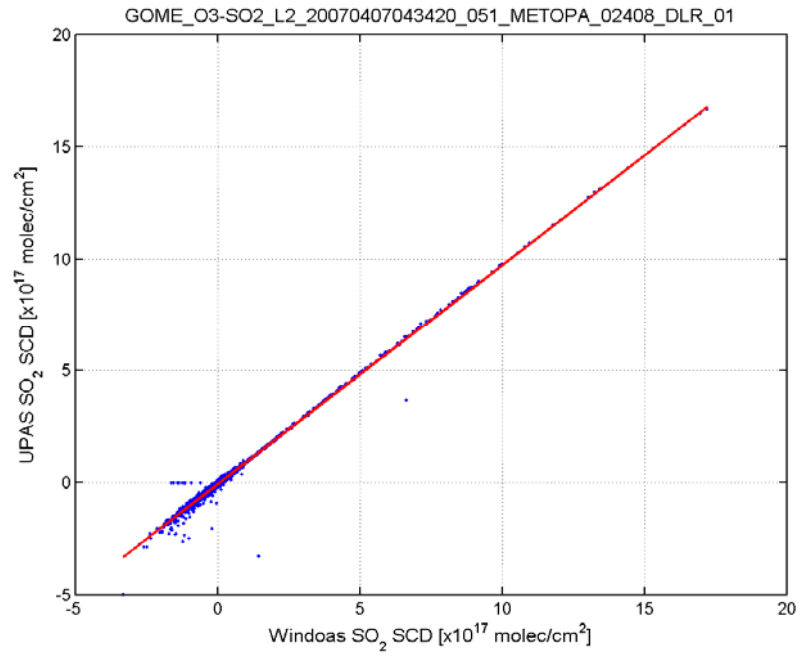


Figure C.3 – Correlation plot of SO₂ slant columns retrieved from the GDP 4.2 and the BIRA-IASB scientific algorithm.

D. COMPARISON AGAINST SATELLITE DATA

This chapter presents a comparison of SO₂ total columns derived from GOME-2 measurements against SO₂ total columns derived from SCIAMACHY and OMI measurements for a few typical cases.

D.1. Assumed elevation of the SO₂ in the atmosphere

The satellite SO₂ products provide 3 (GOME-2 & SCIAMACHY) or 4 (OMI) different vertical column densities based on the assumption about the vertical profile of the SO₂ in the atmosphere. For the three satellite instruments used here, these assumed heights are listed in Table D.1.

Table D.1 – Assumed vertical distribution of SO₂ for satellite data sets.

Instrument	SO₂ distribution
GOME-2	The SO ₂ is assumed to be in a 1 km thick layer centred around: <ul style="list-style-type: none"> • 2.5 km above sea level • 6 km above sea level • 15 km above sea level
SCIAMACHY	The SO ₂ is assumed to be in a 1 km thick layer centred around: <ul style="list-style-type: none"> • 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
OMI	The SO ₂ is assumed to be concentrated in the: <ul style="list-style-type: none"> • planetary boundary layer • lower troposphere: between 0 and 5 km • middle troposphere: between 5 and 10 km • lower stratosphere: between 15 and 20 km

The lowest level for each of these cases in general represents SO₂ in the planetary boundary layer (PBL) from anthropogenic activities or passive degassing of low volcanoes. The middle tropospheric level in general represents SO₂ from effusive volcanic eruptions or passive degassing of high volcanoes. The lower stratospheric level represents SO₂ from explosive volcanic eruptions.

The difference in time of the GOME-2 and SCIAMACHY measurements is about 40 minutes and the two are in approximately the same orbit. During that time any SO₂ will have moved a little and the cloud situation will be somewhat different, but a direct comparison of respective measurements is still possible. OMI measures 2-4 hours later (depending on the location on Earth) and a direct comparison with GOME-2 therefore has limited value.

D.2. Comparison of volcanic SO₂ in the stratosphere

On 7 August 2008 the Kasatochi volcano on one of the Aleutian Islands (52.17N, 175.51W; summit 314 m) erupted, a volcano that had not been active for more than 200 years. The eruption took place in at least three phases between about 20h UTC on 7 August and 04h30 UTC on 8 August and emitted large amounts of SO₂ into the atmosphere, reaching an altitude of about 12 km. Figure D.1 shows the SO₂ distribution as observed by GOME-2, SCIAMACHY and OMI on 8 August. For comparison, also the cloud cover fraction taken from the GOME-2 level-2 data product is shown.

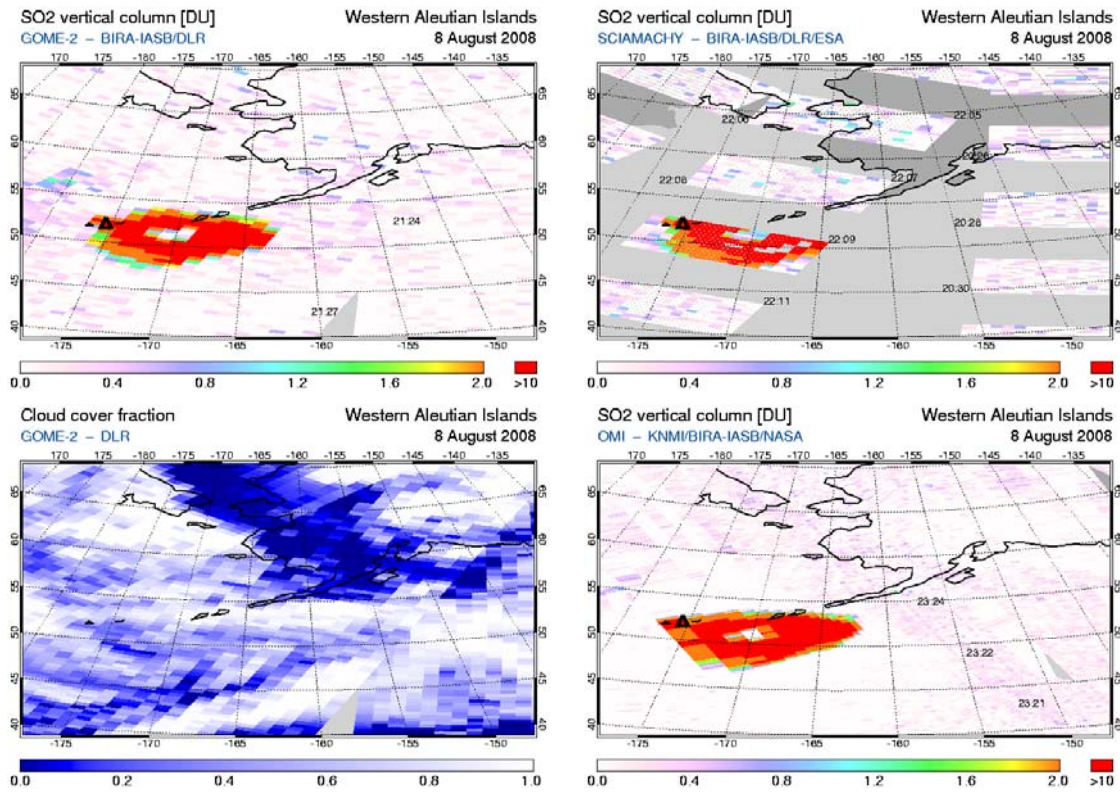


Figure D.1 – Total column SO_2 distribution on 8 August 2008 as measured by GOME-2 (top-left), SCIAMACHY (top-right) and OMI (bottom-right), as well as the cloud cover fraction from GOME-2 (bottom-left). The SO_2 was emitted during the eruption of the Kasatochi volcano, the position of which is marked by a triangle in the SO_2 graphs. The numbers in the SO_2 graphs show the measurement times in UTC – for GOME-2 at the begin and end of each PDU, for SCIAMACHY at begin and end of each nadir state, and for OMI at the centre of the orbit, one every 50 scans.

The SO_2 patch occurs in nicely overlapping GOME-2 and SCIAMACHY orbits and so a direct comparison on a pixel-to-pixel basis is possible. To that end an along-track and along-scan line through the GOME-2 orbit are defined and the corresponding track and scan of SCIAMACHY are determined – see Figure D.2. The match is not perfect, but near enough for a comparison.

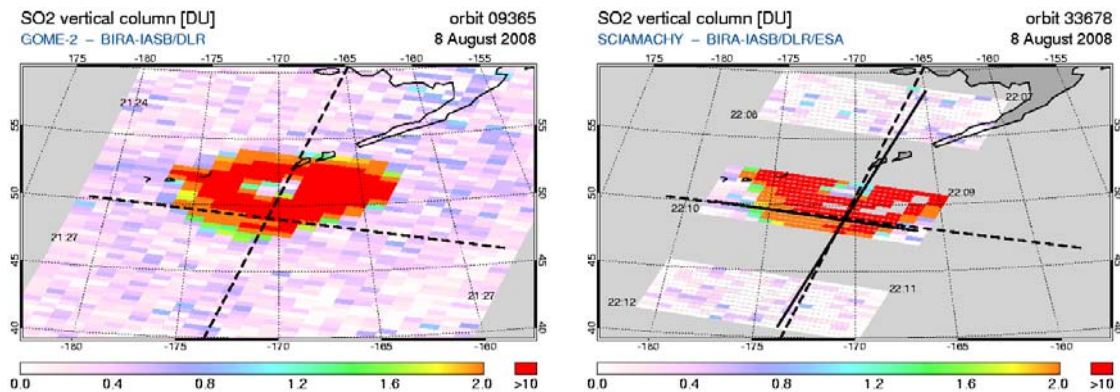


Figure D.2 – Along-track and along-scan lines in the GOME-2 (dashed) and SCIAMACHY (solid) orbit used for a direct pixel-to-pixel comparison of data on 8 August 2008.

Along-track and along-scan matches can only be used in cases where GOME-2 and SCIAMACHY orbits fully overlap, and the higher the latitude, the larger the angle between the comparison lines. And even then the (almost) match works only for about three SCIAMACHY nadir states. The focus lies here mainly on the central of these three nadir states, with the peak of the SO₂ cloud.

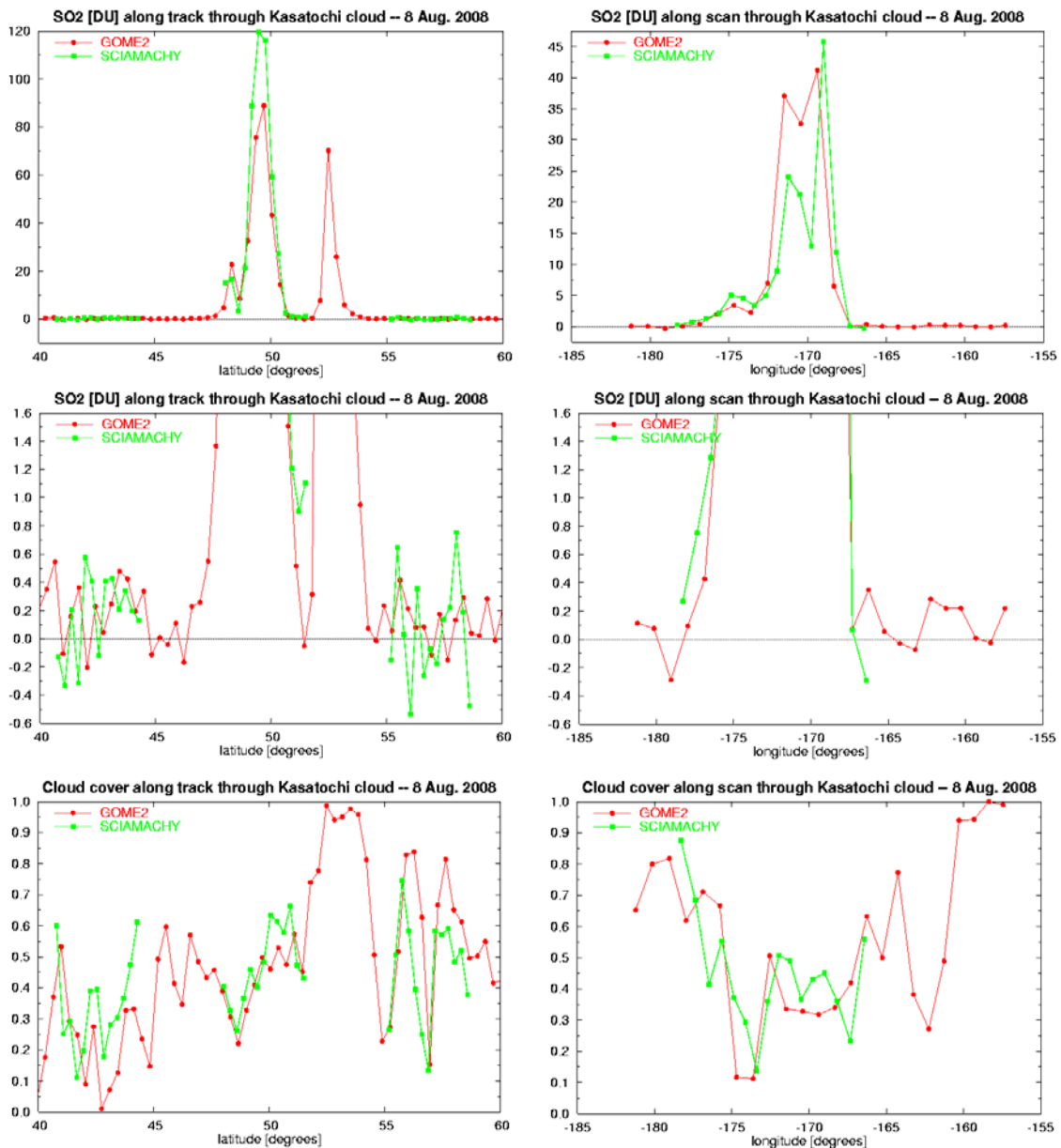


Figure D.3 – Comparison of the total SO₂ column (top row) and a zoom-in around zero SO₂ (middle row) from GOME-2 and SCIAMACHY, as well as of the cloud cover fraction (bottom row) for the along-track lines (left column) and along-scan lines (right column) drawn in Figure D.2.

The results of the pixel-to-pixel comparison along the lines in Figure D.2 are shown in Figure D.3. Since the SO₂ cloud is certainly in the lower stratosphere – most likely at around 12 km altitude – the comparison is done using the data set based on a stratospheric SO₂ plume.

The match between the location and magnitude of the SO₂ peak along and across the track (top row in Figure D.3) is very good, especially if one takes into account that there must have been quite some dynamical motion in the SO₂ cloud during the 40 minutes time difference. The maximum SO₂ values from SCIAMACHY are about 25 % higher compared to the GOME-2 columns. The resolution in the across-track direction of SCIAMACHY is higher than GOME-2's resolution, which means that the match along the scan (right column in Figure D.3) will always be a little less accurate than a match along a track.

The low-level SO₂ concentrations outside the SO₂ cloud itself (middle row in Figure D.3) match well too, as do the cloud cover fractions (bottom row), even for the SCIAMACHY nadir states north and south of the central one, where the along-track lines do not match completely. Note that the cloud data for SCIAMACHY are taken from the FRESCO+ data set (from the TEMIS website – www.temis.nl), whereas the GOME-2 cloud data come from the OCRA/ROCINN data available in the level-2 files.

Three days later, on 11 August, the SO₂ cloud lies off the coast of Alaska and another almost-match can be found, as Figure D.4 shows. Again the along-track match deviates about 1 SCIAMACHY ground pixel to the end of the depicted range, but the comparison can still be made.

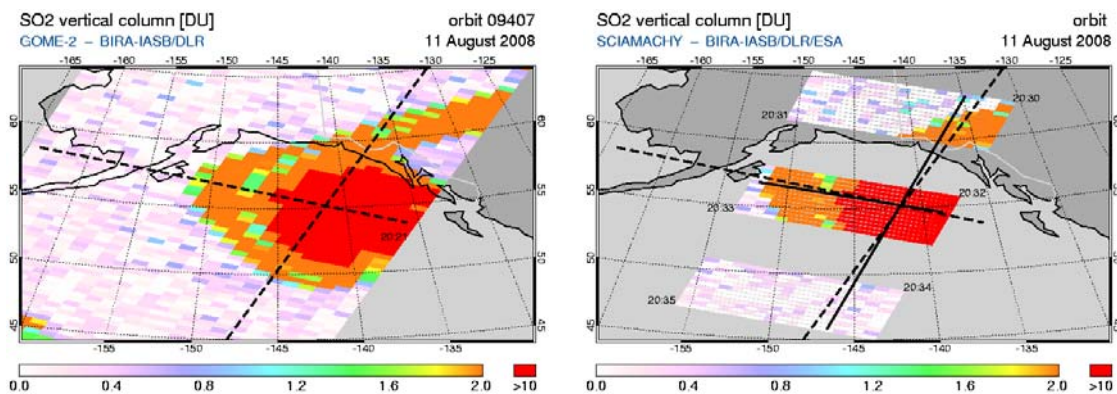


Figure D.4 – Along-track and along-scan lines in the GOME-2 (dashed) and SCIAMACHY (solid) orbit used for a direct pixel-to-pixel comparison of data on 11 August 2008.

The results of the comparisons along the lines in Figure D.4 are shown in Figure D.5. The comparison of the SO₂ total column (top row) shows that both instruments capture the structure of the SO₂ cloud very well, the locations of the peak SO₂ values and the dimensions of the SO₂ cloud match nicely for both instruments. Differences can be found in the total SO₂ columns, SCIAMACHY gives total columns that are of the order of 20 DU higher. A similar difference is seen in the comparison in Figure D.3, at least in the along-track comparison, though less clear because the SO₂ cloud is smaller in size. The agreement in the cloud cover fraction (bottom row) shows that the difference in SO₂ columns is not related to cloud issues.

Low level SO₂ concentrations (not shown) agree well, which means that the difference in total SO₂ column only appears clearly for very large SO₂ values. Though dynamics in the SO₂ cloud mean that the SO₂ is distributed somewhat differently for the moments the measurements are made, the difference must primarily be related to the retrieval of the SO₂ total column.

The total column of SO₂ is determined from the retrieved slant column, divided by an appropriate air-mass factor. The algorithm that is used to calculate the AMF look-up tables is the same for both instruments. Differences in the AMF between the two instruments are therefore mainly related to differences in the viewing geometry (which do not exactly match because the orbits are not exactly the same and due to the time difference). Clearly, the main reason for the observed differences lies in the retrieval of the SO₂ slant columns.

One important difference between the SO₂ slant column retrieval from SCIAMACHY and GOME-2 is the use of different cross-sections at different temperatures. GOME-2 uses reconvolved SCIA FM cross-sections at a temperature representative for the specific height (see Table C.1), in this case a stratospheric temperature for the 15 km retrieval, while for the SCIAMACHY retrieval a cross-section at a tropospheric temperature is used. This can result in up to 20% higher slant columns and could therefore explain part of the difference in total column SO₂. Another issue that could induce differences in the retrieved slant columns is the choice of the reference spectra, for the GOME-2 retrieval a daily solar spectrum is used as reference, whereas the SCIAMACHY retrieval uses an earthshine spectrum as reference (selected from an equatorial region without SO₂ sources).

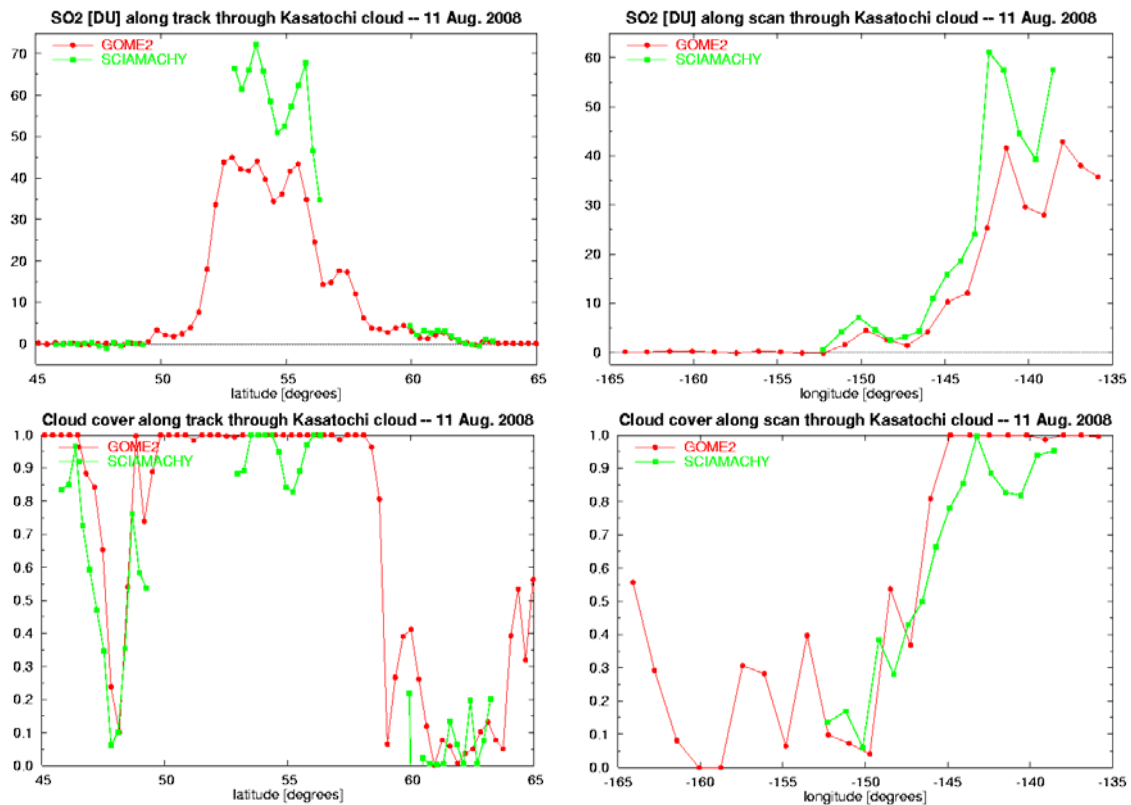


Figure D.5 – Comparison of the total SO₂ column (top row) from GOME-2 and SCIAMACHY, as well as the cloud cover fraction (bottom row) for the along-track lines (left column) and along-scan lines (right column) drawn in Figure D.4.

D.3. Comparison of volcanic SO₂ in the mid-troposphere

The Kilauea volcano on Hawaii (19.42N, 155.29W; summit 1222 m) started a new period of activity in March 2008, with a large number of low-level eruptions. The SO₂ emitted by the volcano seems to remain mostly in the neighbourhood of the volcano and remains visible for a day or two. This implies that the SO₂ is emitted at low altitudes, in the troposphere, where the lifetime of SO₂ is a few days. The total amounts of SO₂ emitted are not very large, with concentrations between 5 and 15 DU. The series of Kilauea eruptions can therefore be used for comparing low-level SO₂ concentrations in the middle troposphere. Since the SO₂ cloud of the volcano is limited in size, the situation can also be used to compare background SO₂ concentrations.

Consider as an example the SO₂ plume west of Hawaii on 17 May 2008. Figure D.6 shows the GOME-2 and SCIAMACHY orbits that passed over the plume that day. These orbits overlap completely and it is easy to find matching along-track and along-scan lines suitable for a comparison, because at this latitude the tracks of the instruments run nicely parallel. Due to the ground pixel widths of 60 km for SCIAMACHY and 80 km for GOME-2, a match is found every three GOME-2 tracks. Let us therefore look at a set of four tracks and three scans for the comparison.

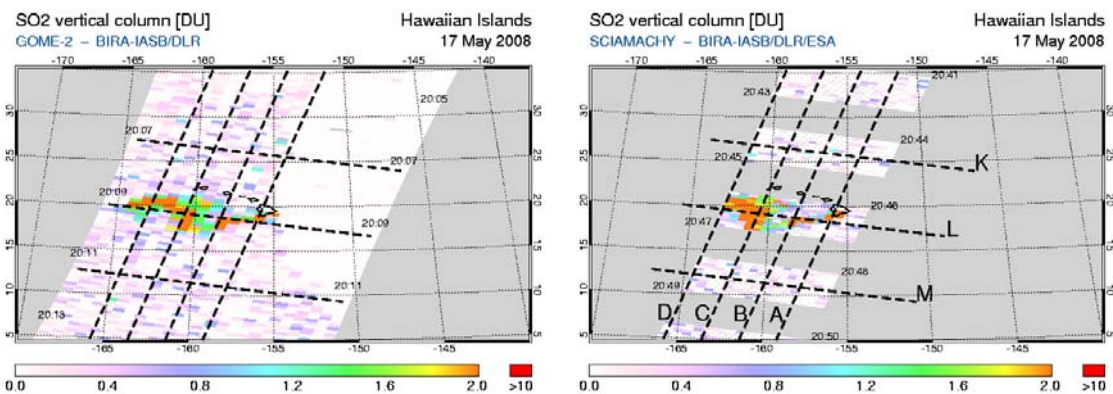


Figure D.6 – SO₂ distribution for GOME-2 orbit 8185 (left) and SCIAMACHY orbit 32490 (right), which passed over Hawaii on 17 May 2008. The dashed lines are used for along-track and along-scan comparisons, labelled with the letters in the right plot.

Figure D.7 shows the comparisons of the SO₂ total columns and cloud cover fractions along the four tracks A – D of Figure D.6, while the comparisons along the three scans K – M are shown in Figure D.8.

The correspondence between the SO₂ columns from GOME-2 and SCIAMACHY for these tracks and scans is very good, especially for those cases where the cloud cover fraction is not too different. In the 40 minutes between the GOME-2 and the SCIAMACHY observations the cloudiness has changed somewhat, which obviously affects the results of the SO₂ retrieval, as the cloud cover fraction is an important parameter for the calculation of the AMF. In particular on the eastern side of the most northern of the three SCIAMACHY states (i.e. the one of scan K) the cloud fraction for SCIAMACHY is considerably larger than the cloud fraction for GOME-2 (see top-right panel in Figure D.7).

The peak values of the SO₂ total columns correspond very well, keeping the different cloudiness in mind. This shows that the differences in SO₂ peak values found in Section D.2, when looking at very high SO₂ concentrations in the lower stratosphere, are limited mainly to that situation. The fact that the SO₂ cross-sections are used at different temperatures for the two instruments plays less of a role for tropospheric SO₂ as the temperature difference is smaller.

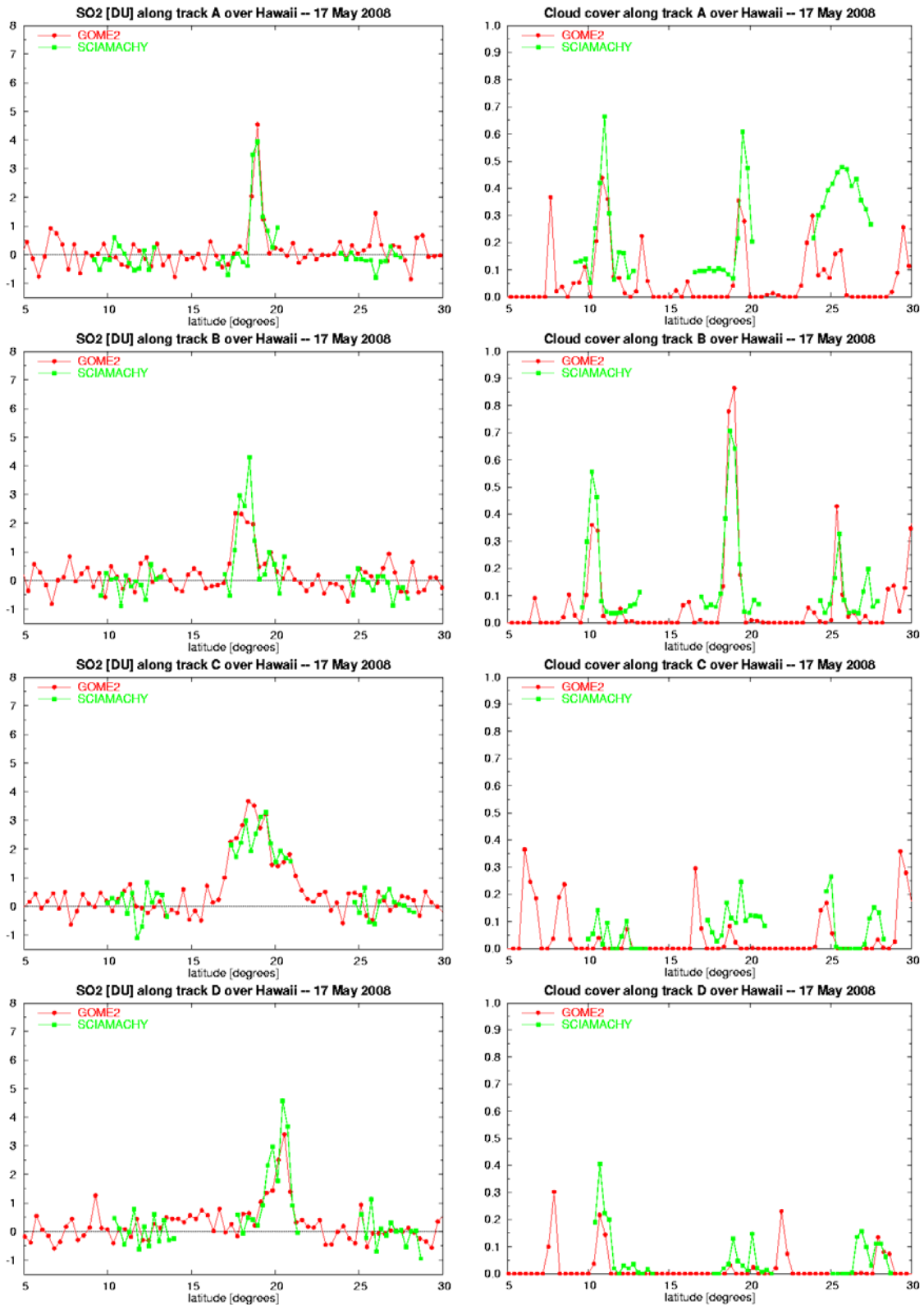


Figure D.7 – Comparison of the total SO₂ column (left column) and the cloud cover fraction (right column) from GOME-2 and SCIAMACHY for the along-track lines A – D (top to bottom) drawn in Figure D.6.

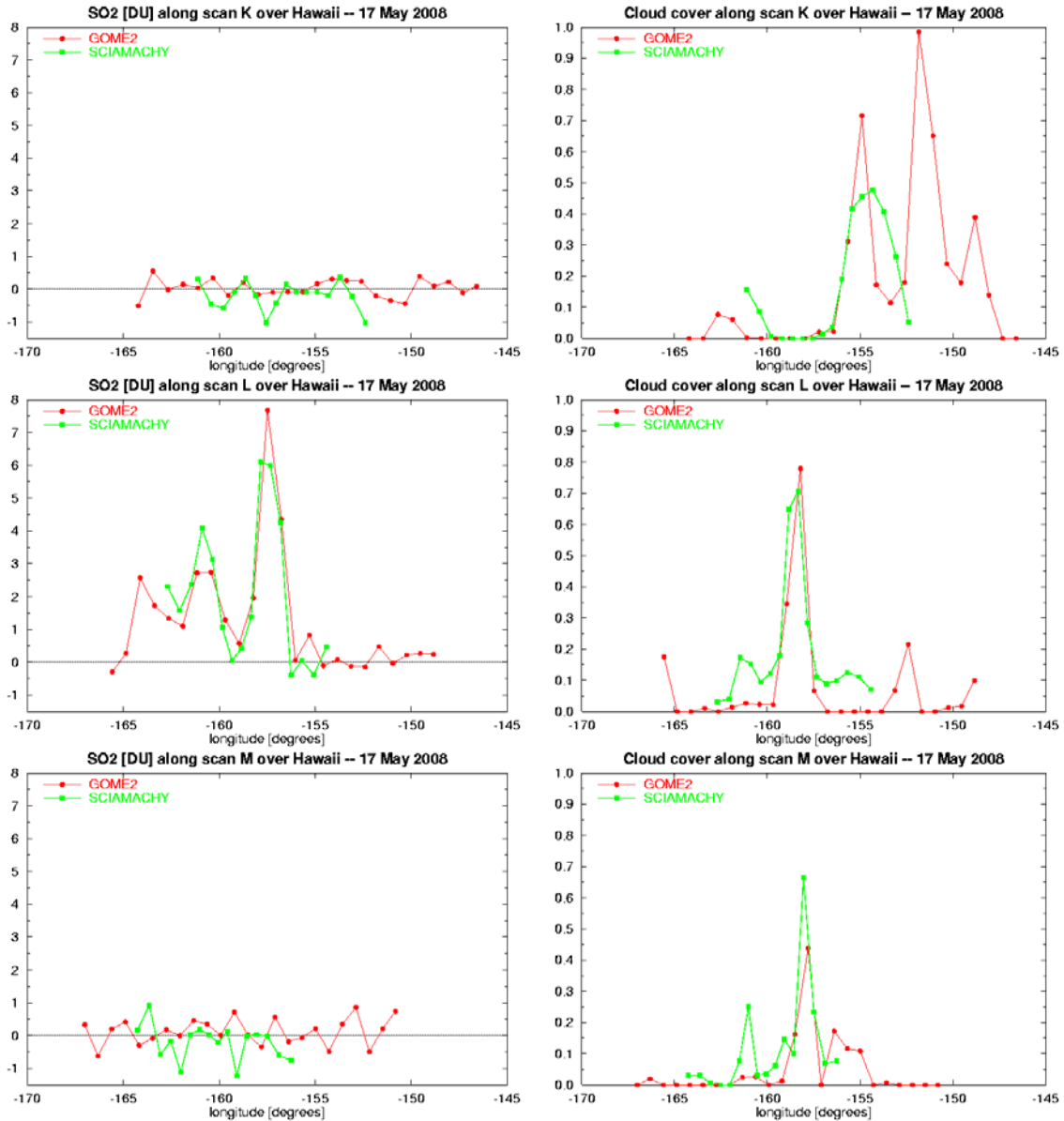


Figure D.8 – Comparison of the total SO_2 column (left column) and the cloud cover fraction (right column) from GOME-2 and SCIAMACHY for the along-scan lines K – L (top to bottom) drawn in Figure D.6.

As another comparison of a similar situation, consider the SO_2 cloud on 24 March 2008, the first day that SCIAMACHY detected SO_2 over Hawaii in the new period of activity of Kilauea. Figure D.9 shows the SO_2 distribution based on both GOME-2 and SCIAMACHY measurements and two lines used for an along-track and an along-scan comparison. The results of that comparison are presented in Figure D.10.

This comparison again shows good agreement between GOME-2 and SCIAMACHY for the SO_2 total column. That the SCIAMACHY data show a higher SO_2 peak than GOME-2 in the along-scan comparison can be attributed to a lower cloud fraction at the moment SCIAMACHY passed over the area.

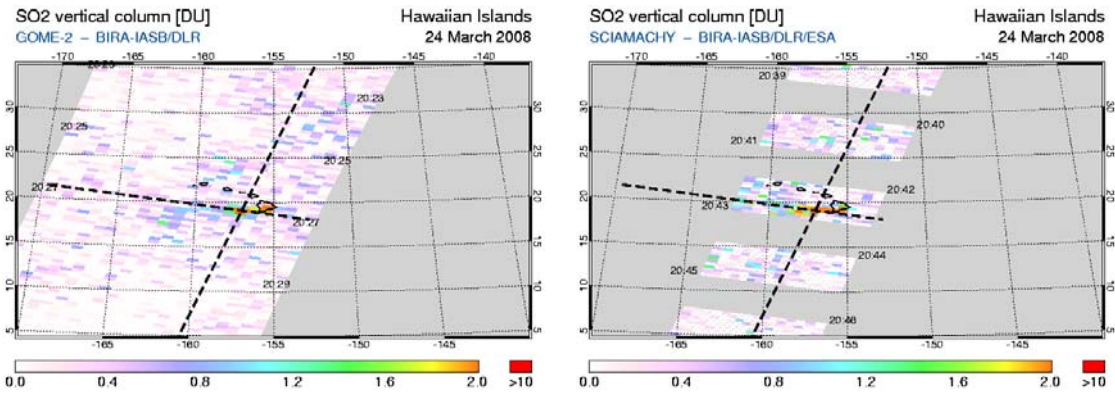


Figure D.9 – SO_2 distribution for GOME-2 orbit 7418 (left) and SCIAMACHY orbit 31717 (right), which passed over Hawaii on 24 March 2008. The dashed lines are used for along-track and along-scan comparisons.

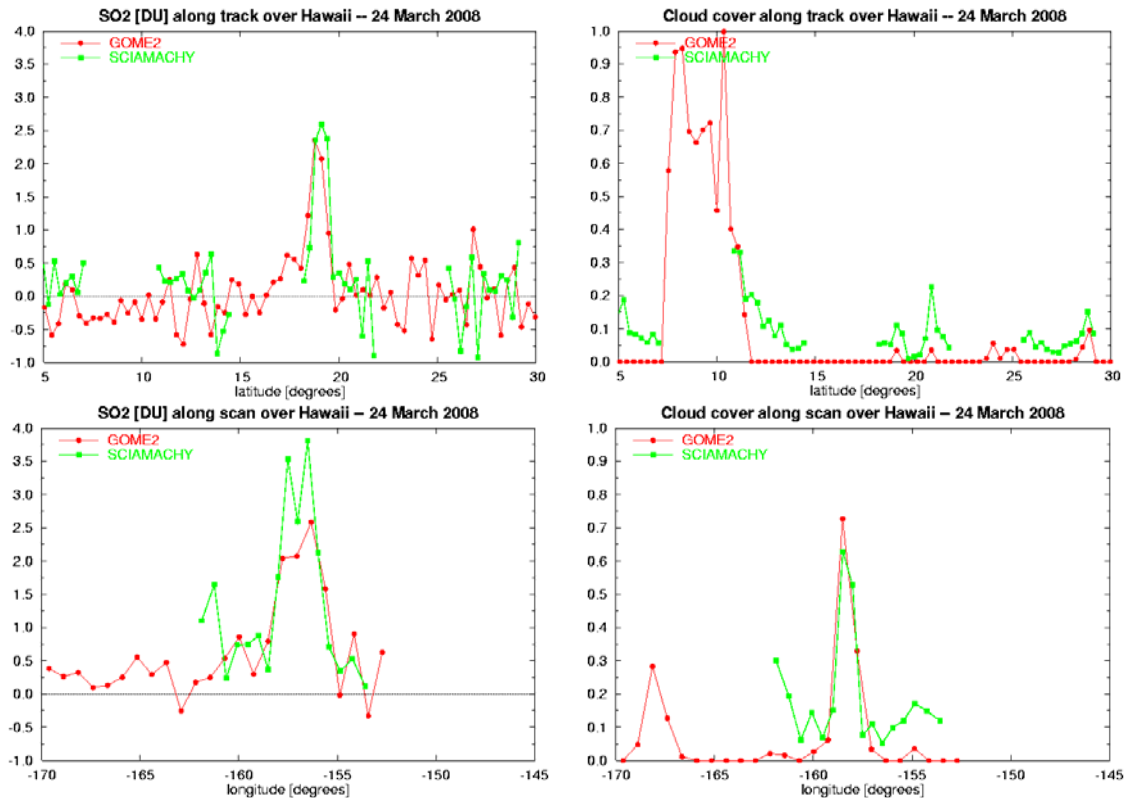


Figure D.10 – Comparison of the total SO_2 column (top row) and the cloud cover fraction (bottom row) from GOME-2 and SCIAMACHY for the along-track line (left column) and along-scan line (right column) drawn in Figure D.9.

D.4. Comparison of anthropogenic SO_2 in the lower troposphere

Some anthropogenic activities emit SO_2 into the atmosphere. This SO_2 usually remains in the planetary boundary layer (PBL), within the lower 2 km of the atmosphere. The lifetime of SO_2 in the PBL is a day or two: it combines with water, forming sulphuric acid, or aerosols which rain out. Retrieval of SO_2 close to the surface is rather difficult: it comes with large uncertainties, due to the reduced sensitivity of satellite instruments for SO_2 in the PBL (see section B.1) and it is hampered by the presence of clouds.

As an example consider the orbits that pass over Eastern China on 3 July 2008 – see Figure D.11 – and define two tracks and two scans for comparison. The results of the comparison of SO₂ are presented in Figure D.12. These graphs first of all show that there is a large variation in SO₂ column values, some of which are large negative. This indicates that the errors on the retrieved column amounts are quite large. And the SCIAMACHY data seems to have larger errors than the GOME-2 data, judging from the peak values along track A away from the SO₂ patch (top-left in Figure D.12), but it can be seen that both instruments capture the general structure of the anthropogenic SO₂ pollution, as they show elevated SO₂ concentration in the same areas.

The differences in the SO₂ column amounts that can be seen in this comparison are not related to cloud effects – the cloud cover fractions (not shown) do not differ significantly – and must be due mainly to the uncertainties in the retrieval of SO₂ close to the surface.

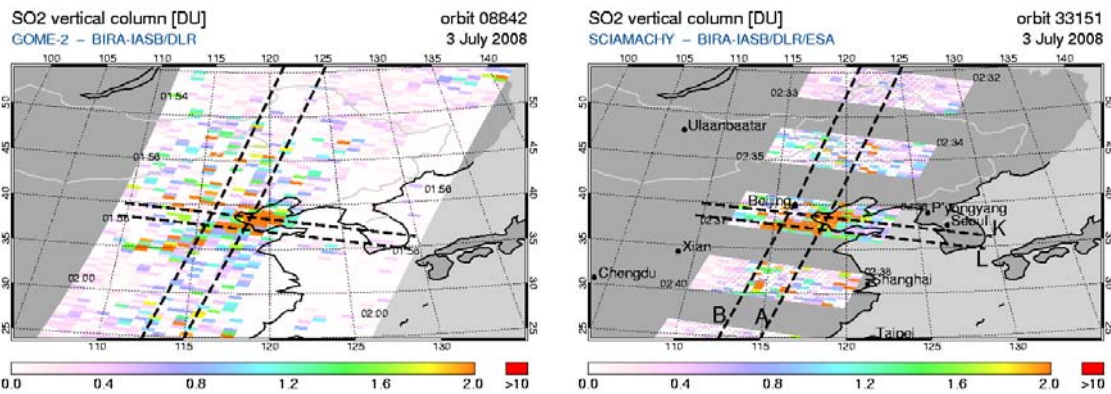


Figure D.11 – SO₂ distribution for the GOME-2 (left) and SCIAMACHY (right) orbits which passed over Eastern China on 3 July 2008. The dashed lines are used for along-track and along-scan comparisons, labelled with the letters in the SCIAMACHY plot.

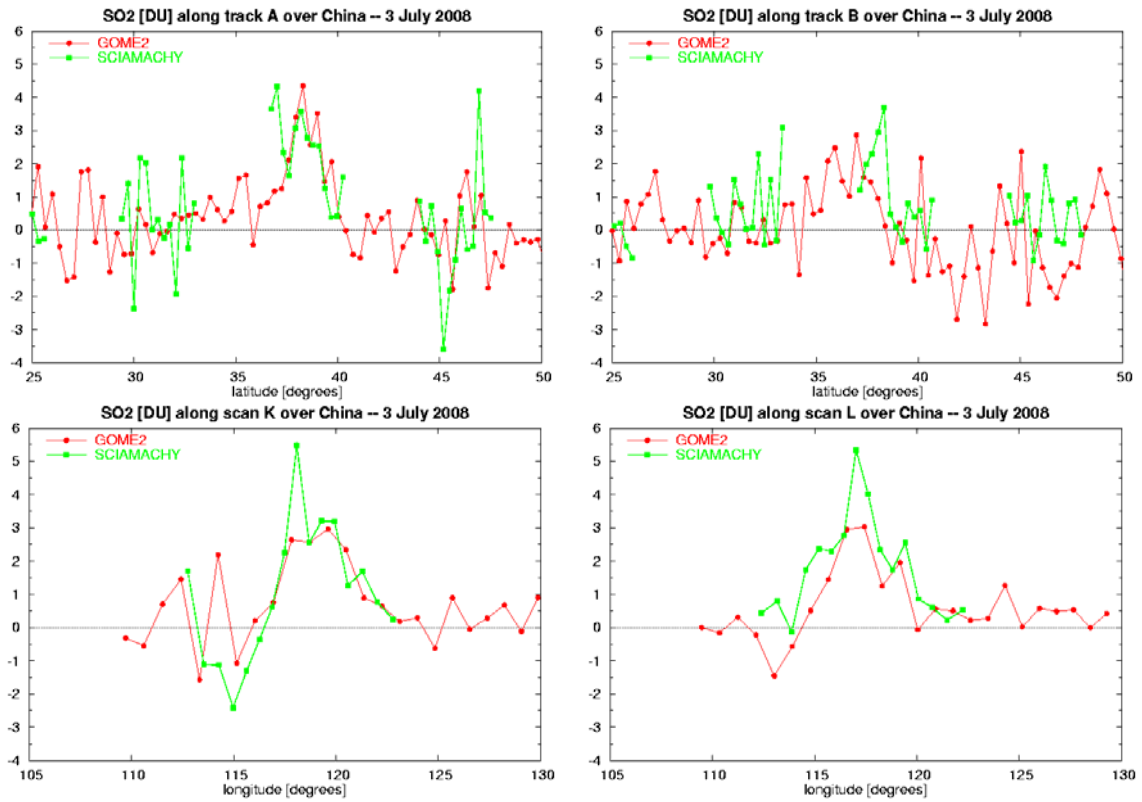


Figure D.12 – Comparison of the total SO₂ from GOME-2 and SCIAMACHY for the along-track lines A and B (top row) and the along-scan lines K and L (bottom row) drawn in Figure D.11.

E. COMPARISON AGAINST GROUND-BASED MEASUREMENTS

This chapter describes a preliminary comparison of SO₂ total columns derived from measurements by GOME-2, SCIAMACHY and OMI against a few selected data sets of ground-based instruments, focussing first on anthropogenic SO₂ in China, and next on an SO₂ cloud from volcanic origin passing over two ground stations in Europe.

E.1. Ground-based instruments used for the comparisons

Table E.1 lists the main characteristics of the ground-based instruments. From the Brewer data, only “direct sun” measurements are used for the comparison.

Table E.1 – Location and type of ground-based instruments used for the comparison.

<i>Ground station place</i>	<i>Longitude</i>	<i>Latitude</i>	<i>Instrument type</i>
Beijing	116.38 ° E	39.98 ° N	MaxDOAS – Multi-axis DOAS
Uccle	4.35 ° E	50.80 ° N	Brewer #178 – double spectrometer
Manchester	2.23 ° W	53.50 ° N	Brewer #172 – double spectrometer

The ground-based data from Uccle were kindly provided by Hugué De Backer from the Belgium Royal Meteorological Institute (KMI-IRM). The ground-based data from Manchester were kindly provided by John Rimmer of the University of Manchester (UK).

For the comparisons, the individual ground-based measurements of a given day are averaged and shown by vertical bars. In some cases there are several measurements per day, in other cases only one or two. This difference is not taken into account when computing the daily average. The absence of a vertical bar implies that there were no (valid) measurements that day.

E.2. Satellite data used for the comparisons

For the comparisons, data from GOME-2, SCIAMACHY and OMI is used. The satellite overpass data is determined by taking an average over all ground pixels with a centre coordinate within a radial distance of 50 km of the ground station. In case of data from GOME-2 and SCIAMACHY data, only the measurements of the forward scan ground pixels are taken into account. Retrieval errors are not considered in the averaging.

The OMI overpass data were kindly provided by Michael Van of NASA. OMI overpass data for a selected list of groundstations is provided via the AVDC website <http://avdc.gsfc.nasa.gov/>, including Uccle and Manchester; at the moment of writing, however, that website was unavailable due to technical problems. The overpass data for Beijing were extracted on request. GOME-2 and SCIAMACHY overpass data were extracted from the data products produced by DLR and BIRA-IASB, respectively.

E.3. Anthropogenic SO₂ total columns measured in Beijing

Close to the Olympic Stadium in Beijing, BIRA-IASB has placed a MaxDOAS instrument on the top of the roof of Institute of Atmospheric Physics (IAP), one of the Chinese partners in the project AMFIC (“Air

Quality Monitoring and Forecast In China). This instrument is measuring various trace gases since mid-July 2008, focussing on air quality related compounds. One of the species retrieved is SO₂.

The SO₂ total column density is determined from the measured slant column density – retrieved with a DOAS technique – with a geometrical approximation of the air-mass factor. The instrument measures at different viewing angles. The retrieval results of the measurements at 15° and 30° are compared and if these agree within 30%, the measurement is considered to be valid.

Figure E.1 shows a preliminary comparison of SO₂ total column data for July 2008. On several days characterized by persistent cloud coverage, the retrieval of SO₂ from the ground-based measurements appeared to be too unreliable to provide useful data. For the comparison, the SO₂ of the satellite data sets with the SO₂ assumed in the lowest layer (see Section D.1) are used.

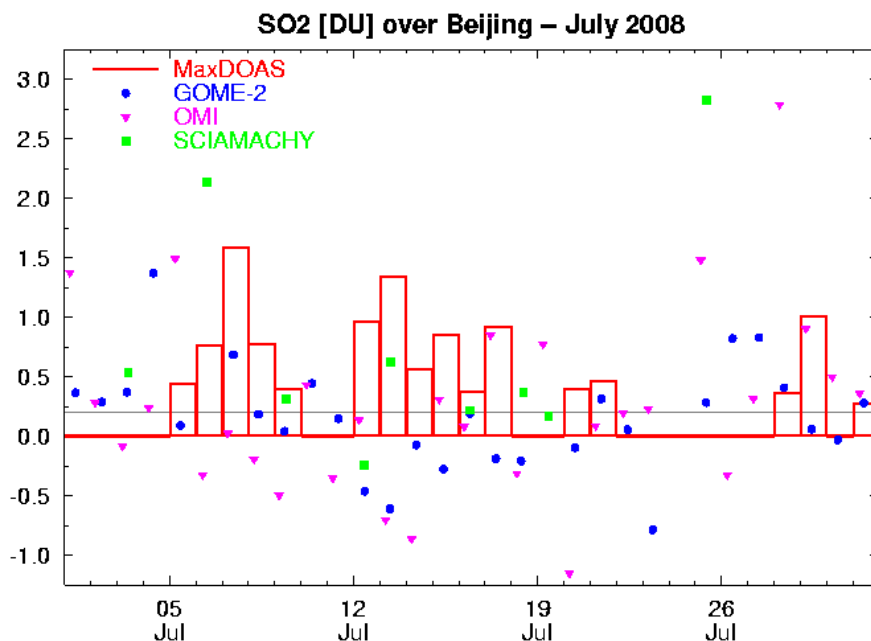


Figure E.1 – Comparison of SO₂ total column data [in DU] as measured in Beijing during July 2008 with satellite overpass data. The data is displayed in local time (UTC + 8h). The horizontal gray line symbolises the average background SO₂ level, away from any emission sources. Given the uncertainties in the satellite retrieval of SO₂ close to the ground, concentrations below 1 DU should be considered with care.

The SO₂ measured in this area in this period is of anthropogenic origin and does not show very high concentrations. The SO₂ can be assumed to be limited to the planetary boundary layer (PBL), i.e. the lower 2 km or so. The retrieval from satellite observations of SO₂ so close to the ground is difficult and shows rather large uncertainties. The overall background level of SO₂ is of the order of 0.2 DU, but given the uncertainties in the retrieval, which are around 0.5 – 1.5 DU for individual measurements for all three satellites, SO₂ concentrations below about 1 DU should be considered with great care.

The results presented here show first of all that there is a rather large scatter in the satellite data, with some outliers. On the whole, though, the concentrations found from satellite and ground-based data are in reasonable agreement. Note that for the days that ground-based measurements are not available, the scatter in the satellite data seems a little larger, possibly also due to a cloud contamination effect.

E.4. The passing of volcanic SO₂ over ground stations in Uccle and Manchester

The SO₂ released into the atmosphere by the eruption of the Kasatochi volcano, referred to in Chapter D, was transported across the Northern Hemisphere by stratospheric winds in a few “branches” during August 2008. The path these branches followed after the initial emission – which took place between about 20h UTC on 7 August and 04h30 UTC on 8 August – depended on the altitude reached by the SO₂. One of the branches passed over the ground stations in Uccle and Manchester on 17-18 August (see Figure E.2), where ground-stations with Brewer spectrometers measure ozone and SO₂ concentrations.

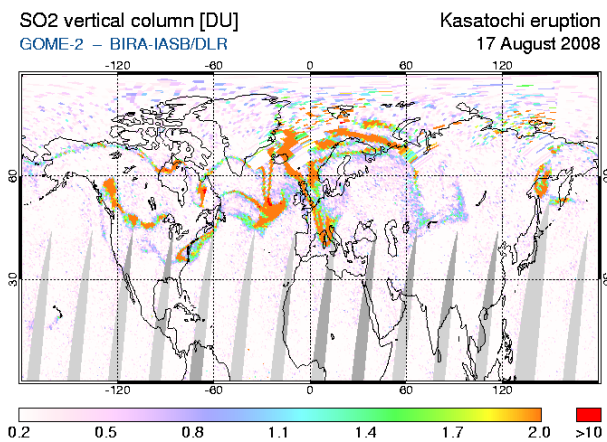


Figure E.2 – Distribution of SO₂ over the Northern Hemisphere on 17 August 2008, as seen by GOME-2. The SO₂ was released by the eruption of the Kasatochi volcano on 7-8 August 2008.

Figures E.3 and E.4 show a comparison of the daily averaged ground-based data with satellite overpass data. For the latter, the SO₂ data sets for the lower stratosphere (see Section D.1) were used. The Brewer data for Uccle showed an offset of +1.0 DU (the average over all data points from 1 to 15 August); this offset has been corrected for in the graphs. The Brewer data for Manchester showed no offset (the average over the data from 1 to 15 August is 0.02 DU).

The Brewer spectrometers are calibrated using reference instruments and this calibration is usually focussing on ozone. This means that SO₂ measurements and their calibration are secondary. For the calculation of the SO₂ concentration, a set of weighting coefficients is applied to the raw data when calculating the ratios used to derive the ozone concentration. These coefficients are designed to eliminate SO₂ and aerosols from the ozone calculation. The sum SO₂+O₃ is then calculated using a different set of weighting coefficients, designed to optimise for SO₂. The previously calculated ozone is then subtracted to leave the SO₂ concentration. These weighting coefficients are instrument specific, but in fact they are assumed to be the same for all Brewers and hard wired into the software, which means that the first calculation of O₃ is affected by some fraction of the SO₂ and aerosols. In the case of Manchester, for example, this leads to an underestimation of the O₃ by up to about 40% of the SO₂ present in the optical path (estimation at the moment; work is ongoing to quantify this). This means that subtracting the O₃ from the SO₂+O₃ measurement may lead to an overestimated SO₂ value. (John Rimmer, Manchester Univ.; priv.comm.)

Comparing the daily averaged data gives very good results in these two cases (Figures E.3 and E.4). Both the satellite and the Brewer measurements capture the enhanced SO₂ columns over Uccle and Manchester very well. The comparison uses daily averages of the Brewer data because it is often difficult to compare individual measurements: ground-based measurements are rarely at exactly the same moment as satellite observations. And, more importantly, satellite instruments – with their large foot print – observe a different air mass than the ground-based instruments, since the latter perform point measurements. Still, in this case the comparison of individual measurements seems to give quite good results, as the example of Figure E.5 shows.

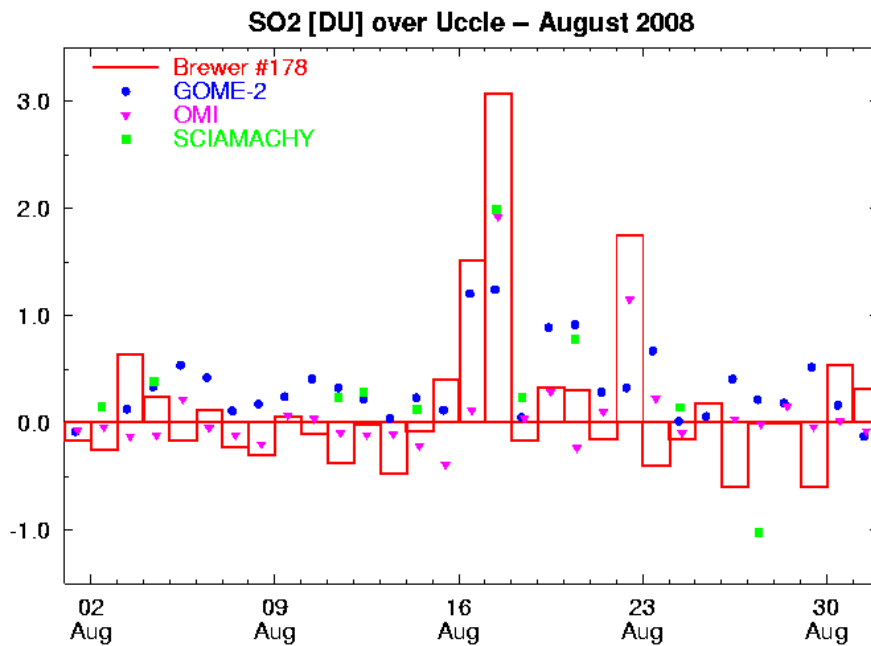


Figure E.3 – Comparison of SO₂ total column data [in DU] as measured by the double Brewer of the Belgian Royal Meteorological Institute (KMI-IRM) in Uccle during August 2008 with satellite overpass data. The data is displayed in UTC time.

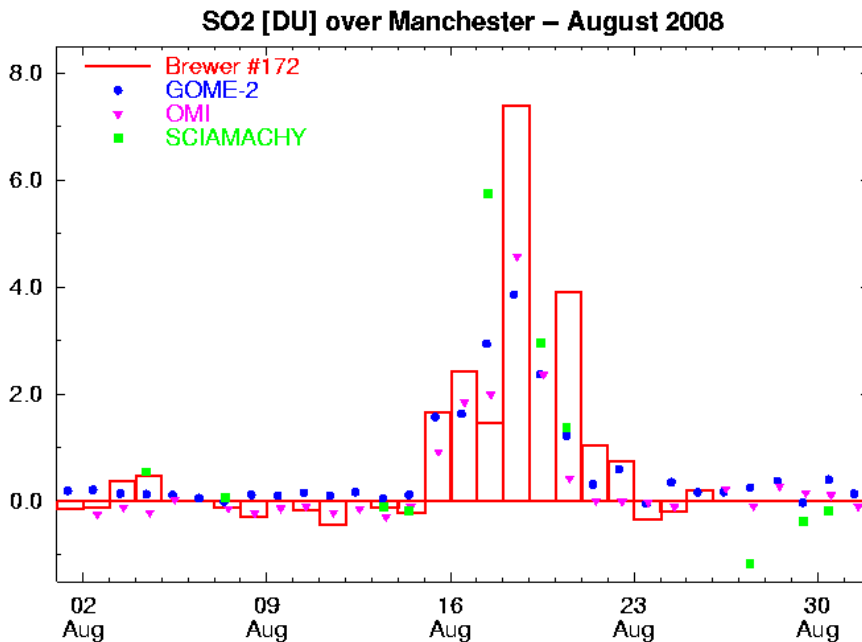


Figure E.4 – Comparison of SO₂ total column data [in DU] as measured by the double Brewer of the University of Manchester (UK) during August 2008 with satellite overpass data. Due to technical problems with the Brewer, the ground-based measurements end on 25 August. The data is displayed in UTC time.

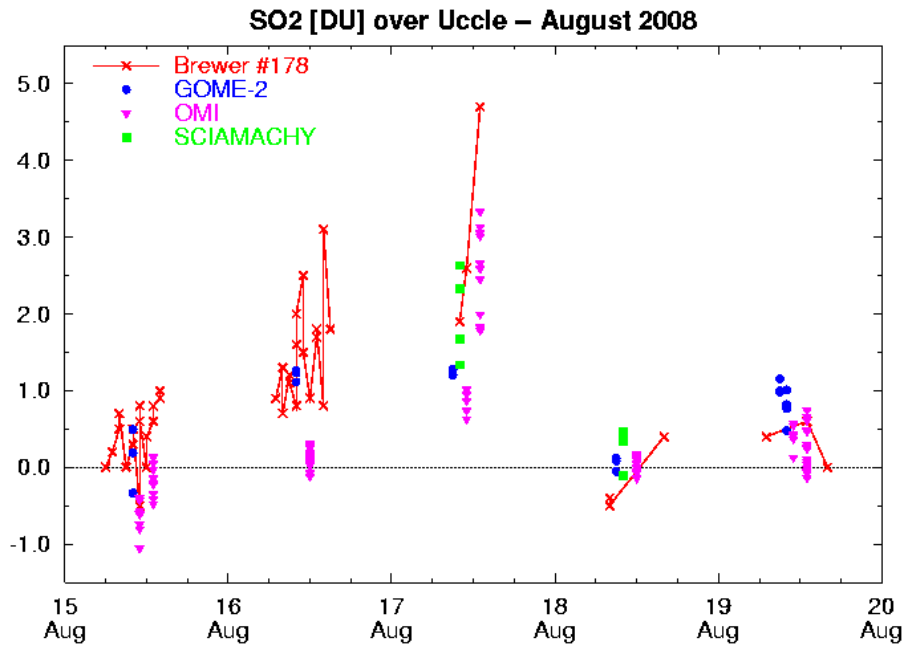


Figure E.5 – Comparison SO₂ total column data [in DU] of the individual ground-based and satellite measurement for the Uccle overpass of Fig. E.2 for 5 days. The lines between the symbols of the Brewer data are drawn to guide the eye; they do not represent actual SO₂ data.



F. CONCLUSION

The GOME-2 SO₂ total columns for the period March 2007 – August 2008 retrieved with version 4.2 of the GOME Data Processor (GDP) have been evaluated using (1) independent GOME-2 scientific retrievals performed at BIRA-IASB, (2) comparisons with correlative data sets from SCIAMACHY and OMI and (3) comparisons with independent ground-based SO₂ measurements in Beijing (MaxDOAS) and in Uccle/Brussels and Manchester (Brewer).

Our study first addressed the verification of the GOME-2 SO₂ slant columns, as retrieved with the GDP 4.2. For the DOAS fit, the 315-326 nm wavelength interval has been selected as an optimal choice for accurate SO₂ slant column determination. After synchronisation of the BIRA-IASB and GDP 4.2 fitting tools, a high level of agreement was found between the two data-sets for GOME-2, confirming the reliability of the GDP system for SO₂ slant column fitting.

In a second step, we focused on validation issues involving comparisons with currently available correlative SO₂ data sets from SCIAMACHY and OMI. Comparisons for the stratospheric SO₂ plume from the Kasatochi eruption show that the three satellite instruments capture the structure of the SO₂ cloud very well: the locations of the peak SO₂ values and the dimensions of the SO₂ cloud match nicely. The maximum GOME-2 total SO₂ columns in the volcanic plume are about 20-30% lower than the SO₂ columns from SCIAMACHY, but this difference can for a large part be explained by the use of different cross-sections at different temperatures in the slant column retrieval. Comparisons for the low-level SO₂ plume from the Kilauea eruptions on Hawaii show a very good agreement between the total SO₂ columns from GOME-2 and SCIAMACHY.

Finally, the validation of the GOME-2 SO₂ columns with ground-based Max-DOAS and Brewer measurements has been addressed. Although satellite measurements of anthropogenic SO₂ in the planetary boundary layer have relatively large uncertainties, the GOME-2 and the Max-DOAS SO₂ measurements in Beijing are in reasonable agreement. The passing of volcanic SO₂ from the Kasatochi eruption over Europe has been measured with Brewer spectrometers in Uccle and Manchester. Comparisons of the Brewer measurements with GOME-2 and other satellite measurements show a very good agreement. Both the GOME-2 and the Brewer measurements capture the enhanced SO₂ columns over Uccle and Manchester very well.

Based on the initial validation with ground-based measurements and the comparisons with correlative SCIAMACHY and OMI data, we concluded that the current GOME-2 GDP 4.2 SO₂ column product already fulfil the user requirements and can be declared as fully operational.

G. REFERENCES

- Bogumil, K., Orphal, J., Homann, T., Voigt, S., Spietz, P., Fleischmann, O.C., Vogel, A., Hartmann, M., Kromminga, H., Bovensmann, H., Frerick, J., Burrows, J.P.: 2003, "Measurements of molecular absorption spectra with the SCIAMACHY pre-flight model: instrument characterization and reference data for atmospheric remote-sensing in the 230 – 2380nm region", *J. of Photochemistry and Photobiology, A: Chemistry*, **157**, 167-184
- Carn, S.A., Krotkov, N.A., Yang, K., Hoff, R.M., Prata, A.J., Krueger, A.J., Loughlin, S.C. and Levelt P.F.: 2007, "Extended observations of volcanic SO₂ and sulfate aerosol in the stratosphere," *Atmos. Chem. Phys. Discuss.* **7**, 2857–2871.
- Eisinger, M. and Burrows, J.P.: 1998, "Tropospheric Sulphur Dioxide observed by the ERS-2 GOME Instrument", *Geophys. Res. Lett.* **25**, pp. 4177-4180.
- Gür, B., Spietz, P., Orphal, J., Burrows, J.: 2005, "Absorption Spectra Measurements with the GOME-2 FMs using the IUP/IFE-UB's Calibration Apparatus for Trace Gas Absorption Spectroscopy CATGAS", Final Report, IUP University of Bremen, Oct. 2005
- Khokhar, M.F., Frankenberg, C., Van Roozendaal, M., Beirle, S., Kühl, S., Richter, A., Platt U. and Wagner, T.: 2005, "Satellite observations of atmospheric SO₂ from volcanic eruptions during the time period of 1996 to 2002," *Adv. Space Res.* **36**, 879–887.
- Krotkov, N.A., Carn, S.A., Krueger, A.J., Bhartia, P.K. and Yang, K.: 2006, "Band Residual Difference Algorithm for Retrieval of SO₂ From the Aura Ozone Monitoring Instrument (OMI)," *IEEE Trans. Geosc. Remote Sens.* **44** (no. 5), May 2006, 1259–1266.
- Krotkov, N.A., McClure, B., Dickerson, R.R., Li, C., Carn, S.A., Bhartia, P.K., Yang, K., Krueger, A., Li, Z., Hains, J.C., Levelt, P.F., Chen, H., Yuan, J., Gong, F. and Bian, X.: 2008, "Ozone Monitoring Instrument (OMI) SO₂ validation over NE China," *J. Geophys Res.* **113**, doi:10.1029/2007JD008818.
- Loyola, D., Van Geffen, J., Valks, P., Erbertseder, T., Van Roozendaal, M., Thomas, W., Zimmer, W. and Wißkirchen, K.: 2008, "Satellite-based detection of volcanic sulphur dioxide from recent eruptions in Central and South America," *Advances in Geosciences* **14**, 35–40.
- Malicet, J., Daumont, D., Charbonnier, J., Parisse, C., Chakir, A., Brion, J.: 1995, "Ozone UV spectroscopy. II. Absorption cross-sections and temperature dependence", *J. Atmos. Chem.*, **21**, 263-273
- Platt, U.: 1994, "Differential Optical Absorption Spectroscopy (DOAS)," in *Air Monitoring by Spectroscopic Techniques*, M.W. Sigrist (ed.), *Chemical Analysis Series* **127**, John Wiley & Sons, New York.
- Richter, A., Wittrock, F. and Burrows, J.P.: 2006, "SO₂ measurements with SCIAMACHY," in *Proceedings of the First Convergence on Atmospheric Science*, 8-12 May 2006, Frascati, Italy, ESA publication SP-628.
- Thomas, W., Erbertseder, T., Ruppert, T., Van Roozendaal, M., Verdebout, J., Balis, D., Meleti, C. and Zerefos, C.: 2005, "On the retrieval of volcanic sulfur dioxide emissions from GOME backscatter measurements," *J. Atmos. Chem.* **50**, 295–320, doi:10.1007/s10874-005-5079-5.
- Valks, P. and Loyola, D.: 2008, "Algorithm Theoretical Basis Document for GOME-2 Total Column Products of Ozone, Minor Trace Gases, and Cloud Properties," DLR/GOME-2/ATBD/01.
- Van Geffen, J., Van Roozendaal, M., Di Nicolantonio, W., Tampellini, L., Valks, P., Erbertseder, T. and Van der A, R.: 2007, "Monitoring of volcanic activity from satellite as part of GSE PROMOTE," in *Proceedings of the 2007 ENVISAT Symposium*, 23-27 April 2007, Montreux, Switzerland, ESA publication SP-636.
- Yang, K. et al.: 2008, oral presentation at the OMI Science Team Meeting, Helsinki, Finland, June 2008.
- Zhang, X., Van Geffen, J., Zhang, P., Liao, H. and Lou, S.: 2008, "Satellite observations of tropospheric SO₂ over China during 2005-2007," *J. Geophys. Res.* , submitted.

