

VALIDATION OF OPERATIONAL SCIAMACHY LEVEL-2 PRODUCTS BY BALLOON-BORNE- DIFFERENTIAL OPTICAL ABSORPTION SPECTROSCOPY (DOAS)

Pfeilsticker¹ K., H. Bösch^{1,5}, A. Butz^{1,2}, C. Camy-Peyret², M. P. Chipperfield³, M. Dorf¹,
K. Grunow⁴, L. Kritten¹, A. Lindner¹, S. Payan², A. Rozanov⁷, C. von Savigny⁷, B. Simmes¹, C. Sioris⁸,
and F. Weidner¹

(1) Institut für Umweltp Physik, University of Heidelberg, Heidelberg, Germany

(2) Laboratoire de Physique Moléculaire pour l'Atmosphère et l'Astrophysique (LPMAA), Université Pierre et Marie Curie, Paris, France

(3) Institute for Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds, UK

(4) Meteorologisches Institut, Freie Universität Berlin, Berlin, Germany

(5) now with Jet Propulsion Laboratory (JPL), Pasadena, USA

(6) Max Planck Institut für Chemie, Mainz, Germany

(7) Institut für Umweltp Physik und Fernerkundung, University of Bremen, Bremen, Germany

(8) Harvard-Smithsonian Center for Astrophysics, Cambridge, USA

ABSTRACT

Level 2 products (profiles of O₃, NO₂, BrO) of the Envisat/SCIAMACHY instrument are validated by balloon-borne measurements of the same quantities using the same optical technique. The comparison studies with the level 2 retrievals of the operational processor OL 3.0 are encouraging. The satellite retrievals tend underestimate the balloon borne O₃ profiles in the 20 to 30 km altitude range by 10% to 30%. For NO₂, overestimation is observed below the concentration maximum around 28 km with deviations amounting up to 40%. For BrO, no operational product is available so far. Hence, we revisit the study of Dorf et al. (2006) which validates the scientific BrO product and finds a bias of -20% / +20% observed for altitudes above/below 25 km somewhat depending on the SCIAMACHY retrieval algorithm.

1. INTRODUCTION

The SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument onboard the European Envisat satellite is an UV/visible/near-IR spectrometer designed to measure direct and scattered sunlight in various viewing directions (Bovensmann et al. 1999). Here, we briefly report on the efforts made to validate SCIAMACHY level 1 and level 2 products by balloon-borne solar occultation UV/visible spectroscopy. So far, the LPMA/DOAS payload (Limb Profile Monitor of the Atmosphere / Differential Optical Absorption Spectroscopy) has been deployed on 4 validation campaigns conducted at high, mid and low-latitudes (Table 1). For all flights, the payload was equipped with a UV/visible DOAS spectrometer (Ferlemann et al. 2000), a Fourier transform spectrometer (Camy-Peyret, 1995) and a mini-DOAS instrument (Weidner et al. 2005). Correlative measurements with Envisat/SCIAMACHY were performed using an air mass trajectory matching technique. For photochemically sensitive gases a suitable

correction scheme was employed, in order to correct for illumination (daytime) mismatches in the individual measurements. For more details of the employed methods, techniques and scientific results see Gurlit et al. (2005), Weidner et al. (2005), Dufour et al. (2005), Butz et al. (2006), Dorf et al. (2006), Sioris et al. (2006), Feng et al. (2006), Frieler et al. (2006) and others.

2. METHODS

2.1 Instrumentation and trace gas retrieval

The French/German LPMA/DOAS balloon payload comprises three optical spectrometers, which analyze direct sunlight over virtually the entire wavelength range from the UV to the mid-IR and thus essentially cover the same wavelength range as SCIAMACHY. Details of the setup and operational performance of the instruments are described in Camy-Peyret (1995) and Ferlemann et al. (1998). Only a short description of the instrumental features important for SCIAMACHY validation is given here.

The LPMA/DOAS spectrometers are deployed on an azimuth-controlled gondola. It carries an automated sun-tracker (Hawat et al., 1998), which provides a beam of sunlight (beam diameter 10 cm) for the direct sun spectrometers. The inner core (beam diameter 5 cm) of the solar beam is directed into the LPMA Fourier Transform spectrometer (FT-IR) (effective field of view (FOV) = 0.2°). Two small telescopes (diameter = 1 cm, effective FOV = 0.53°) mounted into the beam's outer fringe, feed the collected sunlight into the two DOAS spectrometers. The optical setup guarantees that the UV/visible (DOAS) and IR (LPMA) spectrometers analyze direct light from either the whole solar disk (DOAS), or from its inner core (LPMA). Thus, the analyzed sunlight traverses almost the same atmospheric air masses (except for the slightly different effective FOV of both spectrometers). The measurements are performed during balloon ascent

Table 1: Compendium of LPMA / DOAS observations and coincident Envisat/SCIAMACHY overpasses. BA and SO indicate balloon ascent and solar occultation measurements, respectively. Adopted from Butz et al. (2006).

Balloon flight date, time/UT	Location	Geophysical condition	Available datasets	Satellite coincidence orbit, date, time/UT	Altitude range/km	Time delay/h	Spatial distance/km
04 Mar. 2003 13:20–16:17	Kiruna 67.9° N, 21.1° E	high lat. spring SZA: 71.1°–94.1°	SO: LPMA	5273, 4 Mar., 11:05	20–30	–5.1	369–496
				5285, 5 Mar., 7:17	23–24	+15.3	498–499
23 Mar. 2003 14.47–17:28	Kiruna 67.9° N, 21.1° E	high lat. spring SZA: 78.9°–94.7°	BA: LPMA, DOAS	5545, 23 Mar., 11:07	18–28	–5.2	268–496
				5558, 24 Mar., 9:01	19–29	+17.4	10–495
			SO: LPMA, DOAS	5545, 23 Mar., 11:07	20–30	–6.2	63–458
				5558, 24 Mar., 9:01	17–30	+16.0	256–453
9 Oct. 2003 15:39–17:09	Aire sur l'Adour 43.7° N, 0.3° W	mid-lat. fall SZA: 72.0°–87.8°	BA: DOAS	8407, 9 Oct., 9:51	17–31	–6.5	738–988
				8421, 10 Oct., 9:20	25–33	+17.2	547–977
24 Mar. 2004 14:04–17:31	Kiruna 67.9° N, 21.1° E	high lat. spring SZA: 74.5°–95.3°	BA: DOAS	10798, 24 Mar., 10:35	12–33	–5.4	371–499
				10812, 25 Mar., 10:04	6–16	+19.9	32–485
			SO: DOAS	10798, 24 Mar., 10:35	10–33	–6.9	191–436
				10812, 25 Mar., 10:04	10–20	+16.7	301–475
17 June 2005 18:32–21:13	Teresina 5.1° S, 42.9° W	tropical winter SZA: 60.6°–95.8°	BA: in preparation	17240, 17 June, 11:53	25–30	–8.1	382–491
				17255, 18 June, 13:02	5–33	+18.4	6–490
			SO: in preparation	17240, 17 June, 11:53	23–32	–9.1	519–971
				17255, 18 June, 13:02	8–33	+16.2	12–496

or descent and in solar occultation geometry, with moderate spectral resolution in the UV/visible (UV: FWHM = 0.5 nm, visible: FWHM = 1.5 nm) and high spectral resolution in the IR (unapodized resolution 0.02 cm⁻¹).

From the direct sunlight spectra, slant column amounts of the targeted atmospheric absorbers are inferred, using either the DOAS approach in the UV/visible (Platt and Stutz, 2006), or a forward modeling approach in dedicated IR micro-windows. The derived slant column amounts or absorptions of the measured species are inverted into trace gas profiles by applying the truncated Singular Value Decomposition (SVD) or the Maximum A Posteriori (MAP) inversion technique (Rodgers, 2000). For the profile inversion of reactive species (e.g. NO₂ and BrO), a correction based on photochemical modelling is included (e.g. Butz et al., 2006).

2.2 Trajectory modelling

Balloon-borne measurements are inherently restricted by different constraints, limiting their flexibility in satellite validation. Therefore, trajectory modelling was also included into the validation, in order to find best coincidences between air masses probed by the balloon-borne instruments and Envisat/SCIAMACHY observations. The trajectory model uses the operational analyses and forecasts of the European Centre for Medium Range Weather Forecasts (ECMWF) - or a combination of both - given every 6 h on a 2.5° x 2.5° latitude/longitude grid. The ECMWF data are interpolated to 25 user-defined isentropic levels extending from the surface up to 1600 K. The internal time step for integrating the path of the air masses is 10 min and the diabatic and climatological heating rates are based on

Newtonian cooling. The results (trajectory points) are stored for each hour (e.g. Langematz (1987)).

Backward and forward trajectories are started at the balloon measurement locations, which depend on the individual measurement technique. For the LPMA / DOAS remote-sensing payload, the start and end points are calculated from knowledge of the balloon flight trajectory and the known observation geometry given by the line-of-sight for each measurement. For post-flight analysis, forward and backward trajectories are calculated for up to 10 days, but for balloon flight planning purposes the time range is limited by the available ECMWF forecasts (analyses are available up to 12:00 UT of the day before, forecasts for every 6 h up to 72 h). The actual geolocations of SCIAMACHY observations are taken from the SCIAMACHY Operational Support Team (SOST) on their website (<http://atmos.af.op.dlr.de/projects/scops>). Here, the overpass time, the geolocation and detailed measurement specifications (e.g. swath, measurement duration, ground pixel size) are downloaded for the SCIAMACHY limb and for the SCIAMACHY nadir mode for each Envisat orbit. For the air mass trajectory-based matching technique only the area covered by tangent points (light blue areas in Fig. 1) of SCIAMACHY limb observation is considered in more detail. This information is used to find satellite measurement points along individual air mass trajectories, for which the spatial and temporal mismatch is as small as possible. The match criterion is chosen based on the experience of the ozone Match experiment e.g. Gathen et al. (1995): a time mismatch between the satellite observation and the air mass trajectory started at the balloon observation of < ± 1 h and an area mismatch of < ± 500 km. If SCIAMACHY

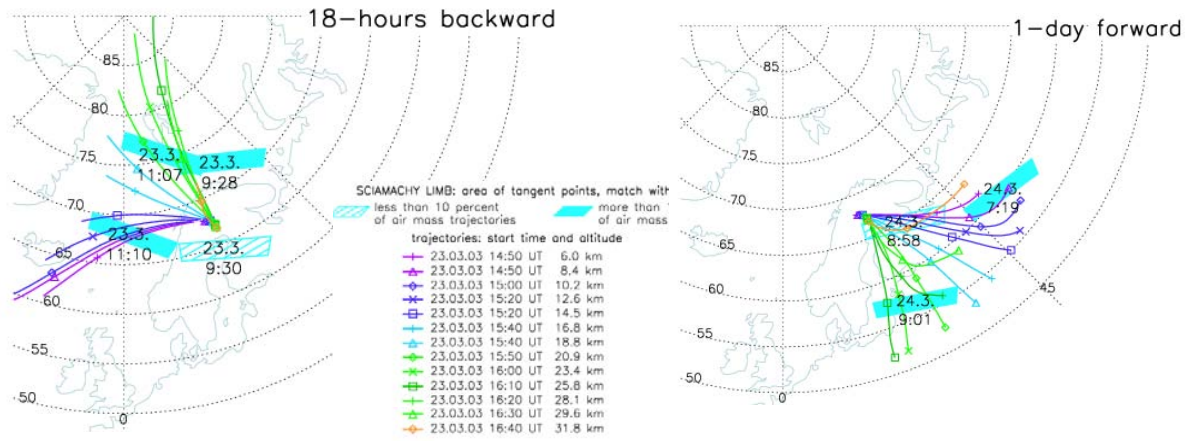


Fig. 1: Air mass trajectories modelled 18 hours backward (left panel) and 24 hours forward (right panel) in time for the balloon ascent measurements of the LPMA/DOAS payload at Kiruna on March 23, 2003. The trajectories are color-coded according to their starting altitude as indicated in the legend. A symbol is plotted every 12 hour interval. The areas covered by the tangent points of SCIAMACHY limb observations are projected onto the Earth's surface and illustrated as blue rectangles. Filled rectangles correspond to SCIAMACHY limb observations for which more than 10% of the calculated air masses are coincident with the LPMA/DOAS measurements. The shaded rectangles represent SCIAMACHY limb observations for which less than 10% of the calculated air masses are coincident with the LPMA/DOAS measurements. The time and date of the satellite measurements is given next to the rectangles (courtesy of FU-Berlin).

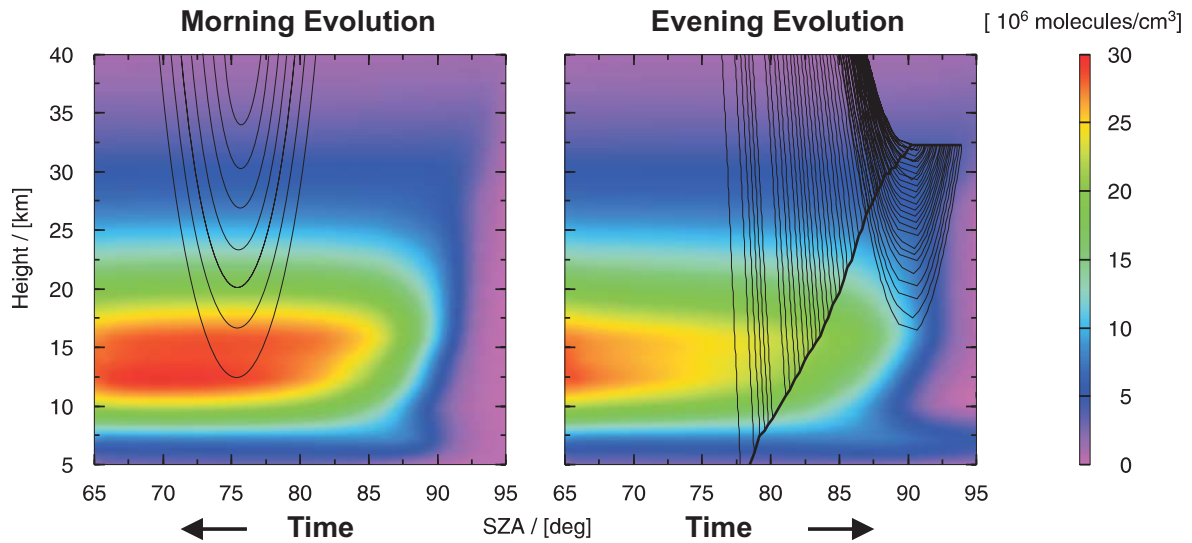


Fig. 2: Color-coded model concentration field of BrO as a function of height and SZA, for the DOAS balloon flight on 23 March 2003 at Kiruna (67.9°N, 22.1°E). Left and right panels show the morning and evening evolution of BrO, respectively. The black lines in the left panel represent the line-of-sight of a SCIAMACHY limb scan. In the right panel the observation geometry of the DOAS measurements is shown for every twentieth spectrum measured during ascent and every tenth spectrum during solar occultation. The thick black line represents the trajectory of the balloon and the thin black lines indicate the optical paths from the Sun to the balloon instrument for measurements during ascent and solar occultation. Note that in the real atmosphere the lines-of-sight are close to being straight lines, but the projection of the Earth's curvature on a straight x-axis causes the lines-of-sight to appear curved in the presentation. Figure adopted from Dorf et al. (2006).

observations do not satisfy these criteria, the distance criterion is extended up to 1000 km.

2.4 Photochemical modelling

As outlined above, the use of a validated photochemical model is necessary when different measurements of stratospheric radicals are compared and validated. Fig. 2 demonstrates why the model has to be used to compare SCIAMACHY BrO (or NO₂) limb measurements (left panel) with LPMA / DOAS balloon ascent and occultation observations (right panel). In both panels, the line-of-sights are indicated by thin black lines. In addition, the thick black line in the right panel represents the balloon trajectory. Here the observations are super-imposed by a photochemical simulation of stratospheric BrO from the SLIMCAT 3-D CTM Chipperfield (1999) for March 23, 2003.

SLIMCAT is a 3-D off-line CTM with detailed treatment of stratospheric photochemistry. The model temperatures and horizontal winds are specified from ECMWF analyses. The vertical transport in the stratosphere is diagnosed from radiative heating rates. In the stratosphere the model uses an isentropic coordinate extended down to the surface using hybrid sigma-theta levels (M.P. Chipperfield, private communication). The troposphere is assumed to be well-mixed.

The CTM is integrated with a horizontal resolution of 7.5° x 7.5° and 24 levels extending from the surface to about 55 km. The model is forced using ECMWF analyses and the simulation starts on 1 January 1977. The model halogen loading is specified from observed tropospheric CH₃Br and halon loadings WMO (2003). In addition, an extra 4 pptv bromine is modelled in a tracer to represent bromine-containing very short-lived substances (VSLS) and 1 pptv is assumed to be transported to the stratosphere as Br_y (Feng et al., 2006). Output is saved at 00:00 UT every 2 days, interpolated to the location of the balloon flights. A 1-D column model is then used to reconstruct the diurnal cycle for comparison with the observations.

The vertical 1-D column model simulates stratospheric photochemistry on forward and backward air mass trajectories (described above) with the aim to find best guess profiles for the satellite observations based on the different validation balloon measurements. The stratospheric photochemistry is modelled on 20 potential temperature (Θ) levels between Θ = 323 K and Θ = 1520 K. The 1-D column model is initialized, at each height level, at 00:00 UT with 3-D CTM SLIMCAT model results at an adjacent 48 hour model time step at the balloon launch site.

The model is constrained to follow the evolution of the SZA time-line, which is taken from the air mass trajectory calculations. In satellite validation, these measures guarantee that the photochemical evolution of the modeled air mass is a good approximation of the true evolution between

initialization of the model, the satellite measurement and balloon-borne observation. For simplicity a single representative SZA time-line is chosen for all Θ levels and the model is run with fixed pressure and temperature for each Θ level taken from the meteorological support data of the balloon flight.

Furthermore, each observation conducted by the remote sensing instruments SCIAMACHY and LPMA/DOAS is a composite of changing photochemical conditions (due to changing SZA) along the line-of-sight (Fig. 2).

Photochemical-weighting factors are calculated to scale balloon observations to the photochemical conditions of the satellite measurements. In the case of LPMA / DOAS measurements the scaling is implicitly considered by the profile inversion algorithm as described by Butz et al., (2006). A flow diagram of the overall validation procedure is given in Fig. 3.

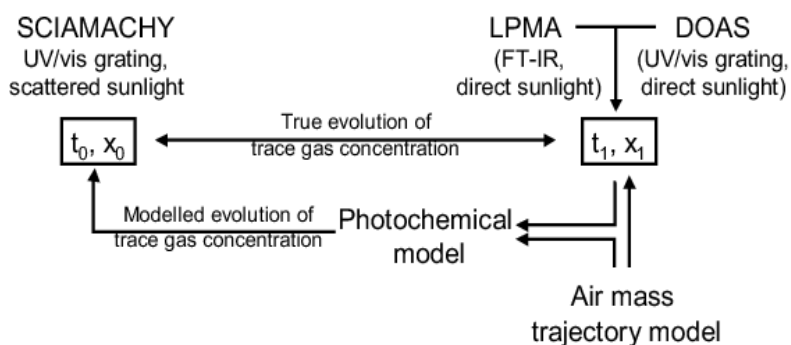


Fig. 3: Schematic drawing of the presented validation strategy. SCIAMACHY observations are conducted at time t_0 and location x_0 . Prior to the balloon flight dedicated to SCIAMACHY validation, an air mass trajectory model is used to optimize the time t_1 and location x_1 of the LPMA / DOAS balloon borne observations, e.g. by optimizing the launch time of the balloon. After the balloon flight, the trajectory model calculates the air mass history in order to identify satellite measurements, which actually sampled the same air masses as the balloon-borne instruments. For the validation of photochemically active trace species, the illumination history of the coincident air masses is fed into a photochemical model to reproduce the evolution of the considered trace gases between satellite and balloon-borne observations as realistically as possible and to infer appropriate scaling factors. Figure adopted from Butz et al. (2006).

3. PRODUCTS & RESULTS

We report on 5 LPMA / DOAS validation balloon flights performed since 2003. Three were conducted from ESRANGE, Kiruna, Sweden, one from Aire sur l'Adour in southern France (see Table 1) and one from Teresina, Brazil. For each balloon flight a satellite coincident measurement is identified in the morning before and after the balloon flight using the trajectory matching

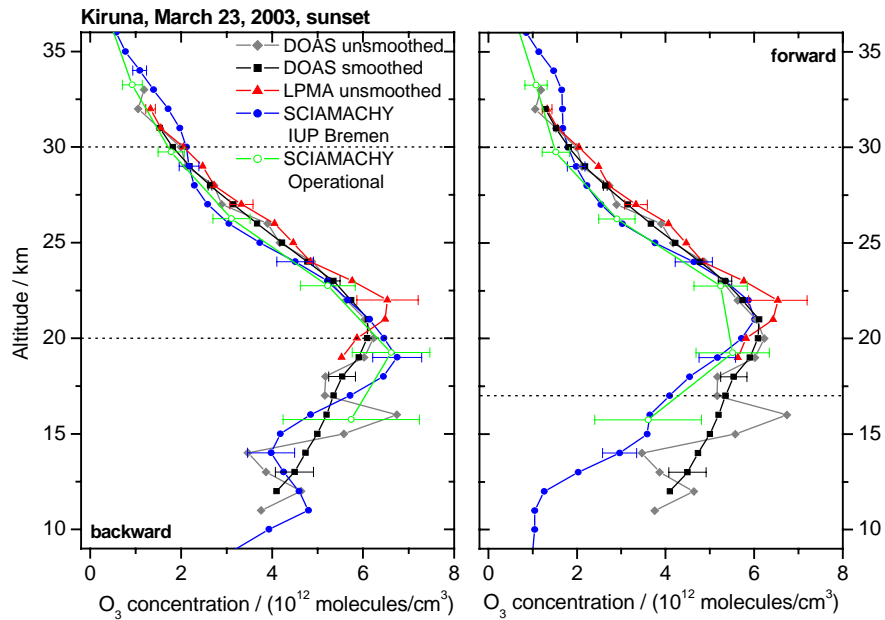


Fig. 4: Comparison of O_3 profiles inferred from SCIAMACHY limb observations with correlative balloon borne measurements. The observations are conducted during sunset at Kiruna on March 23, 2003. The left and right panel corresponds to a backward and a forward coincidence, respectively. Satellite data inferred by IUP-Bremen are shown as blue circles, inferred by the operational level 2 processor OL 3.0 by green open circles. Appropriately smoothed DOAS data are plotted as black boxes, LPMA data as red triangles. The grey diamonds represent DOAS profiles at full altitude resolution without smoothing. The altitude range between the horizontal dotted lines represents the range where coincident air masses are found. For better visibility, only selected error bars are shown. Figure adopted from Butz et al. (2006) with a recent update concerning the SCIAMACHY operational product.

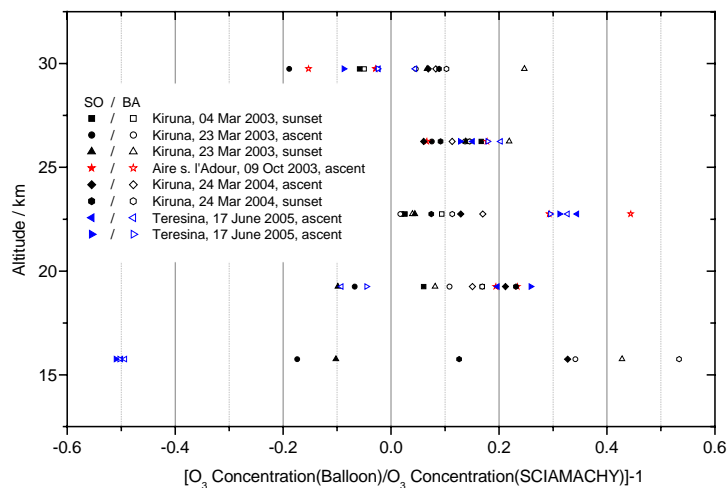


Fig. 5: Relative deviations between SCIAMACHY operational products and LPMA / DOAS measurements of O_3 . Filled and open symbols correspond to backward and forward coincidences, respectively. Observation sites and conditions are indicated in the legend. High latitudes are shown in black, mid-latitudes in red and low latitudes in blue.

technique described above (Fig.1). In the following we refer to these coincidences as backward and forward coincidences. If trace gas profiles inferred from balloon ascent and solar occultation are available, the satellite coincidences are identified separately. For each balloon flight table 1 provides information on the measurement site, the geophysical condition, the SZA range covered by the balloon-borne observations, the available data sets and some details on the selected SCIAMACHY limb scans.

3.1 O_3 validation

Figure 4 shows an illustrative example for the agreement between SCIAMACHY O_3 profiles

inferred by the operational level 2 processor OL 3.0 as well as the IUP-Bremen scientific retrieval and the coinciding LPMA / DOAS balloon-borne observations. Figure 5 summarizes all considered data by showing the relative differences between the balloon borne observations and the operational SCIAMACHY retrievals of O_3 stratospheric profiles. SCIAMACHY limb O_3 profiles underestimate the validation data in the 20 km to 30 km altitude range. Deviations can be as large as 40% but are commonly on the order of 10% to 20%. Below 20 km, there is no clear trend inferable from the limited data set considered here. Remaining small tangent height errors cannot be excluded from our study

3.2 NO₂ validation:

Figure 6 shows an illustrative comparison between SCIAMACHY NO₂ profiles inferred by the operational level 2 processor OL 3.0 as well as the IUP-Bremen scientific retrieval and the coinciding LPMA / DOAS balloon-borne observations. The relative deviations between the operational satellite retrievals and the balloon-borne observations are shown in Fig. 7. In the 20 km to 30 km altitude range, the operational SCIAMACHY retrievals overestimate the balloon-borne NO₂ profiles. Commonly, deviations are on the order of 10% to 30% but most often fall within the combined error large. Maximum deviations are found around 23 km altitude, in bars, in particular since the error bars of the operational retrievals are general below the

concentration maximum of NO₂. This finding could point to the fact that, for operational SCIAMACHY retrievals, low NO₂ concentrations in the lower stratosphere are masked by the concentration maximum higher up.

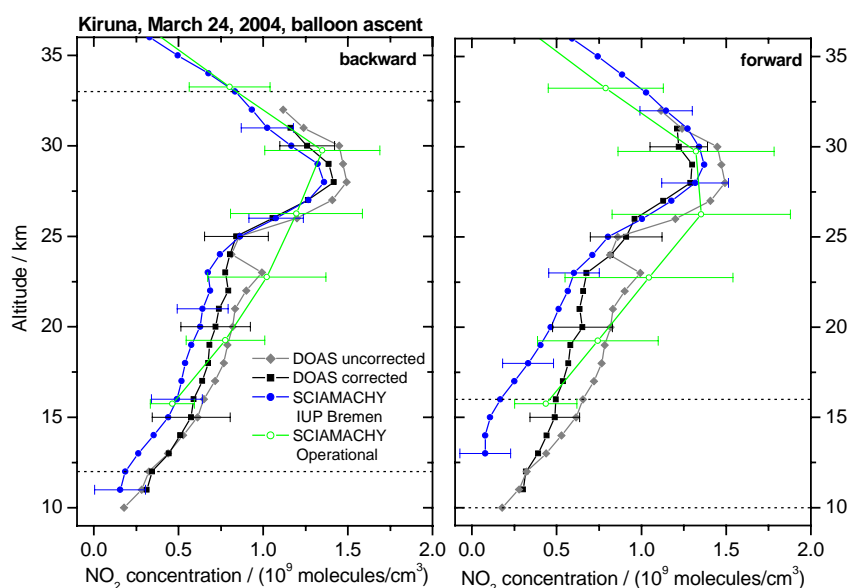


Fig. 6: Comparison of NO₂ profiles inferred from SCIAMACHY limb observations with correlative balloon-borne measurements. The observations were conducted at Kiruna on March 24, 2004, during sunset. The left and right panel corresponds to a backward and a forward coincidence, respectively. Satellite data inferred by IUP-Bremen are shown as blue circles, by the operational level 2 processor OL 3.0 as green open circles. Appropriately smoothed DOAS data are plotted as black boxes. The grey diamonds represent DOAS profiles at full altitude resolution without smoothing. The altitude range between the horizontal dotted lines represents the range where coincident air masses are found. For better visibility, only selected error bars are shown. Figure adopted from Butz et al. (2006) with a recent update concerning the SCIAMACHY operational product.

3.3 BrO validation:

So far, no operational products of BrO from SCIAMACHY limb are available. Therefore, we revisit the validation study of Dorf et al. (2006) that investigates the quality of the scientific BrO retrievals. We recommend our dataset to be used for validation of a future operational BrO product from SCIAMACHY limb observations. Figure 8 shows an example of a comparison of BrO profiles measured by the DOAS balloon instrument and SCIAMACHY. Stratospheric BrO abundances measured from 3 different balloon sensors (DOAS, SAOZ, Triple) are compared in Dorf et al. (2006) with first retrieval exercises of SCIAMACHY BrO limb profiling from the Harvard group. Total Br_y =

(20.1 ± 2.8) pptv, obtained from DOAS BrO observations at mid-latitudes in 2003, serves as an upper limit of the comparison. The good agreement of balloon trace gas measurements with the SLIMCAT model indicates that vertical transport is considered correctly and was not a major source of error. Within the given range of errors of the different measurement techniques, most of the balloon observations agree with model BrO. Initial BrO profiles available from the Harvard-Smithsonian SCIAMACHY retrieval agree on average to around 20 % with the photochemically corrected balloon observations (SAOZ and DOAS).

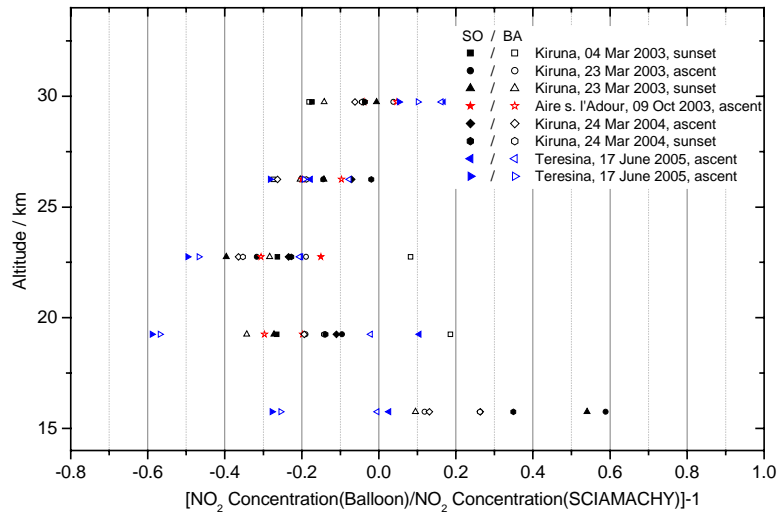


Fig. 7: Relative deviations between SCIAMACHY operational products and LPMA / DOAS measurements of NO_2 . Filled and open symbols correspond to backward and forward coincidences, respectively. Observation sites and conditions are indicated in the legend. Observation sites and conditions are indicated in the legend. High latitudes are shown in black, mid-latitudes in red and low latitudes in blue.

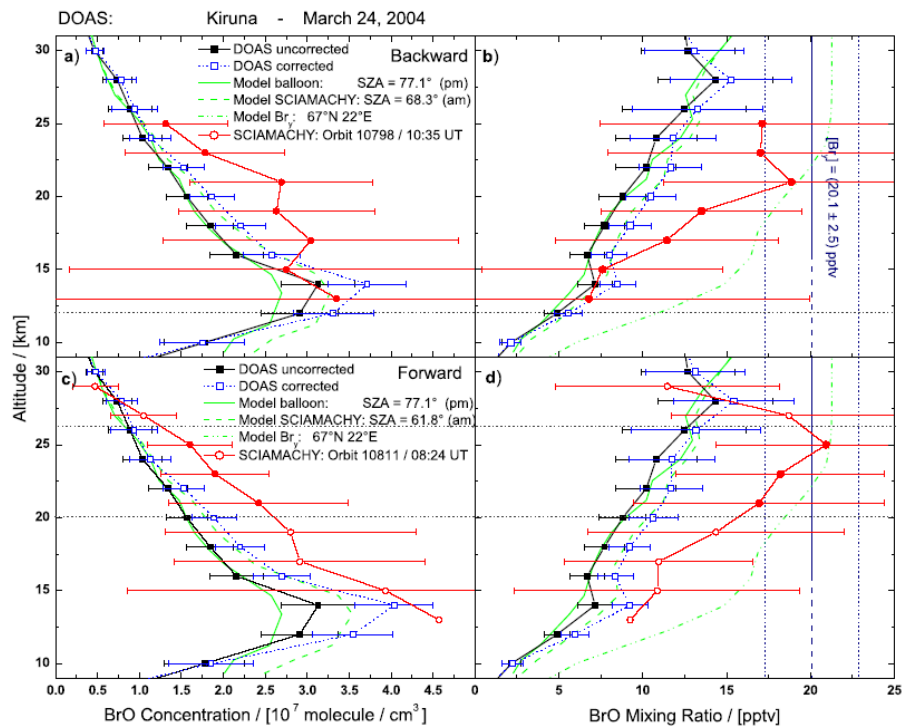


Fig. 8: Comparison of a BrO profile measured by DOAS during balloon ascent on March 24, 2004 at Kiruna with model calculations and SCIAMACHY limb retrievals. Black squares represent the photochemically uncorrected balloon measurement and blue squares the balloon profile photochemically corrected to the SZA of the SCIAMACHY measurement. Corresponding model profiles at the SZA of the balloon and satellite observations are shown as solid and dashed green lines respectively. Total inorganic Br_y volume mixing ratios as used in the model (green dash-dotted line) and as retrieved by DOAS measurements (vertical dark blue lines, see text for details) are also indicated. SCIAMACHY measurements are shown as solid red circles in the altitude range for the match, which is indicated as thin dotted horizontal lines, and as open red circles outside this range. Panels (a) and (b) show calculations for the ‘best’ backward match and Panels (c) and (d) for the ‘best’ forward match. Figure adopted from Dorf et al. (2006).

In general, the satellite measurements show systematically higher values below 25 km and a change in profile shape above about 25 km.

4. SUMMARY & CONCLUSION

Observations of the SCIAMACHY instrument are validated by so far 5 deployments of the LPMA /

DOAS payload at high, mid and low-latitudes since 2003. Studies investigating the quality of level 1 (limb radiance and solar irradiance) and scientific level 2 SCIAMACHY data products (O_3 , NO_2 , BrO) are published in a suite of papers: Gurlit et al. (2005), Weidner et al. (2005), Dorf et al. (2006) and Butz et al. (2006). Here, we assess the quality of the operational level 2 processor OL 3.0 for O_3

and NO₂ stratospheric profiles. We found that agreement between the validation dataset and the SCIAMACHY trace gas profiles are greatly improved compared to previous version of the processor. However, the satellite retrievals tend underestimate the balloon borne O₃ profiles in the 20 to 30 km altitude range. For NO₂, overestimation is observed below the concentration maximum around 28 km. Our balloon borne data set is also of value for the validation of other existing satellite measurements of BrO, NO₂ and O₃ (e.g. OMI) or satellite instruments, that intend to measure these trace gases in the future (e.g. GOME-2). Our future validation activities will particularly concentrate on the level 2 SCIAMACHY products OCIO (at high-latitudes) and O₃, NO₂, BrO, IO, OIO obtained at low latitudes and the validation of complementary data products from the ENVISAT MIPAS and GOMOS instruments.

For future validation studies, digital copies of all balloon-borne DOAS related products for all validation flights, can be obtained from the NILU data server (<http://www.nilu.no>), upon signing the data protocol of the ESA sponsored Envisat validation activities. DOAS data is also available on <http://www.iup.uni-heidelberg.de/institut/forschung/groups/atmosphere/stratosphere/>.

Acknowledgement: Support of the project by Bundesministerium für Bildung und Forschung (BMBF) through grants 50EE0017, and 50EE0019, the Deutsche Forschungsgemeinschaft (DFG) Pf 384/3-1 and 384/5-1, and European Union (EU) through grants EVK2-CT-2000-00059 (QUILT) GOCE-CT-2004-505337 (ACCENT), EVK2-CT-2001-00111 (HIBISCUS) and 505390-GOCE-CT-2004 (SCOUT-O3) is highly acknowledged. Additional support came through the European Space Agency by the ESABC project (AO 146, AO 465, AO 629 and AO 694). We thank the technical team of LPMA (Y. Té, P. Jeseck, and V. Ferreira) for the assistance given to perform successfully the balloon flights. We are also grateful to the CNES teams ‘*equipe nacelles pointées*’ and the balloon launching team from Aire-sur-l’Adour / France without which the successful launch of the balloons would have been impossible. We also acknowledge the support given by Physikalisch-Technische Bundesanstalt (PTB) Braunschweig (A. Höpe und P. Sperfeld) in the commissioning of the absolute radiometric standards.

References:

1. Bovensmann, H.: SCIAMACHY: Mission Objectives and Measurement Modes, *J. Atmos. Sci.*, 56, 127 - 150, 1999.
2. Brinksma, E. J., Bracher, A., Lolkema, D. E., Segers, A. J., Boyd, I. S., Bramstedt, K., Claude, H., Godin-Beekmann, S., Hansen, G., Kopp, G., Leblanc, T., McDermid, I. S.,

- Meijer, Y. J., Nakane, H., Parrish, A., von Savigny, C., Strebler, K., Swart, D. P. J., Taha, G., and Piders, A. J. M., Geophysical validation of SCIAMACHY Limb Ozone Profiles, *Atmos. Chem. Phys.*, 6, 197 - 209, 2006.
3. Butz, A., H. Bösch, C. Camy-Peyret, M. Chipperfield, M. Dorf, G. Dufour, K. Grunow, P. Jeseck, S. Kühl, S. Payan, I. Pepin, J. Pukite, A. Rozanov, C. von Savigny, C. Sioris, F. Weidner, K. Pfeilsticker, Inter-comparison of stratospheric O₃ and NO₂ abundances retrieved from balloon-borne direct sun observations and Envisat/SCIAMACHY limb measurements, *Atmos. Chem. Phys.*, 6, 1293 -1314, 2006.
4. Camy-Peyret, C.: Balloon-borne Fourier transform spectroscopy for measurements of atmospheric trace gases, *Spectrochim. Acta*, 51A, 1143 - 1152, 1995.
5. Canty, T., R.S. Salawitch, J.B. Renard, E.D. Reviere, K. Pfeilsticker, M. Dorf, R. Fitzenberger, H. Bösch, R.M. Stimpfle, D.M. Wilmouth, J.G. Anderson, E.C. Richard, D.W. Fahey, R.S. Gao, and T.P. Bui, Analysis of BrO, ClO, and nighttime OCIO in the arctic winter stratosphere, *J. Geophys. Res.*, 110, D01301, doi:10.1029/2004JD005035, 2005.
6. Chipperfield, M. P.: Multiannual simulations with a three-dimensional chemical transport model, *J. Geophys. Res.*, 104, 1781 - 1805, 1999.
7. Dorf, M., H. Bösch, A. Butz, C. Camy-Peyret, M. P. Chipperfield, A. Engel, F. Goutail, K. Grunow, F. Hendrick, S. Hrechanyy, B. Naujokat, J.-P. Pommereau, M. Van Roozendael, C. Sioris, F. Stroh, F. Weidner, and K. Pfeilsticker, Balloon-borne stratospheric BrO measurements: Comparison with Envisat / SCIAMACHY BrO limb profiles, *ACP*, 6, 2483–2501, 2006.
8. Dufour, G., Payan, S., Lefèvre, F., Eremenko, M., Butz, A., Jeseck, P., Té, Y., Pfeilsticker, K., and Camy-Peyret, C.: 4-D comparison method to study the NO_y partitioning in summer polar stratosphere – Influence of aerosol burden, *Atmos. Chem. Phys.*, 5, 919 - 926, 2005.
9. Feng, W., M.P. Chipperfield, M. Dorf and K. Pfeilsticker, Mid-latitude Ozone Changes: Studies with a 3-D CTM Forced by ERA-40 Analyses, *ACPD*, 6, 6695–6722, 2006.
10. Ferlemann, F., Camy-Peyret, C., Fitzenberger, R., Harder, H., Hawat, T., Osterkamp, H., Schneider, M., Perner, D., Platt, U., Vradelis, P., and Pfeilsticker, K.: Stratospheric {BrO} profiles measured at different latitudes and seasons: Instrument description, spectral analysis and profile retrieval, *Geophys. Res. Lett.*, 25, 3847 - 3850, 1998.
11. Ferlemann, F., Bauer, N., Fitzenberger, R., Harder, H., Osterkamp, H., Perner, D., Platt, U., Scheider, M., Vradelis, P., and Pfeilsticker, K.: Differential optical absorption spectroscopy

- instrument for stratospheric balloon-borne trace gas studies, *Appl. Opt.*, 39, 2377 - 2386, 2000.
12. Fontenla, J., White, O. R., Fox, P. A., Avrett, E. H., and Kurucz, R. L.: Calculation of solar irradiances, I. Synthesis of the solar spectrum, *Astrophys. J.*, 518, 480 - 500, 1999.
 13. von Friedeburg, C., Derivation of Trace Gas Information combining Differential Optical Absorption Spectroscopy with Radiative Transfer Modelling, Dissertation, Universität Heidelberg, 2004, <http://www.ub.uni-heidelberg.de/archiv/3758>
 14. Frieler, K., M. Rex, R.J. Salawitch, T. Canty, M. Streibel, R.M Stimpfle, K. Pfeilsticker, M. Dorf, D.K. Weisenstein, S. Godin-Beekmann, P. von der Gathen, Towards a better quantitative understanding of polar stratospheric ozone loss, *Geophys. Res. Lett.*, 22(385), doi:10.1029/2005GL025466, 2006.
 15. von der Gathen, P., Rex, M., Harris, N., Lucic, D., Knudsen, B., Braathen, G., Backer, H. D., Fabian, R., Fast, H., Gil, M., Kyr, E., Mikkelsen, I., Rummukainen, M., Sthelin, J., and Varotsos, C.: Observational evidence for chemical ozone depletion over the Arctic in winter 1991 - 92, *Nature*, 375, 131 - 134, 1995.
 16. Gurlit, W., H. Bösch, H. Bovensmann, J. P. Burrows, A. Butz, C. Camy-Peyret, M. Dorf, K. Gerilowski, A. Lindner, S. Noel, U. Platt, F. Weidner, and K. Pfeilsticker, The UV-A and visible solar irradiance spectrum: Inter-comparison of absolutely calibrated, spectrally medium resolved solar irradiance spectra from balloon-, and satellite-borne measurements, *Atmos. Chem. Phys.*, 5, 1879 - 1890, 2005.
 17. Harder, J., G. Lawrence, G. Rottman, and T. Woods, The Spectral Irradiance Monitor (SIM) for the SORCE Mission, *Proc. SPIE*, 4135, pp. 204 - 214, see: http://lasp.colorado.edu/sorce/presentation/SORCE_Brochure_10_25_FINAL.pdf, 2000.
 18. Hawat, T. M., Camy-Peyret, C., and Torguet, R. J.: Suntracker for atmospheric remote sensing, *SPIE Optical Engineering*, May 1998, 37(05), 1633 - 1642, 1998.
 19. Kurucz, R. L., Furenhild, I., Brault, J., and Testermann, L.: Solar flux atlas from 296 to 1300 nm, National Solar Observatory Atlas No. 1, June 1984, (<ftp://ftp.noao.edu/fts/fluxat1>), 1984.
 20. Langematz, U., Labitzke, K. and Reimer, E.: Synoptic analysis and trajectories during the MAP/GLOBUS campaign 1983, *Planetary and Space Science*, 35, 525 - 538, 1987.
 21. Platt, U. and J. Stutz, *Differential Optical Absorption Spectroscopy, Principle and Applications*, Springer Verlag, Heidelberg, ISBN 3-340-21193-4, 2006.
 22. Rodgers, C. D., *Inverse Methods For Atmospheric Sounding*, World Scientific, Singapore, New Jersey, London, Hong Kong, 2000.
 23. Sierk, B., Richer, A., Rozanov, A., von Savigny, C, Schmoltner, A. M., Buchwitz, M., Bovensmann, H., and Burrows, J. P.: Retrieval and monitoring of atmospheric trace gas concentrations in nadir and limb geometry using the space-borne SCIAMACHY instrument, *Envir. Mon. and Assess.*, Vol. 20, No. 1-3, DOI10.1007/s10661-005-9049-9, 2006.
 24. Sioris C. E., C. S. Haley, C. A. McLinden, C. von Savigny, I. C. McDade, W. F. J. Evans, J. C. McConnell, N. D. Lloyd, E. J. Llewellyn, D. Murtagh, U. Frisk, T. P. Kurosu, K. V. Chance, K. Pfeilsticker, H. Bösch, and F. Weidner, Stratospheric profiles of nitrogen dioxide observed by OSIRIS on the Odin satellite, *J. Geophys. Res.*, 108, NO. D7, 4215, doi: 10.1029/2002JD002672, 2003.
 25. Sioris, C.E., L. J. Kovalenko, C. A. McLinden, R. J. Salawitch, M. Van Roozendaal, F. Goutail, M. Dorf, K. Pfeilsticker, K. Chance, C. von Savigny, X. Liu, T. P. Kurosu, J.-P. Pommereau, H. Bösch, and J. Frerick, Latitudinal and vertical distribution of bromine monoxide in the lower stratosphere from SCIAMACHY limb scattering measurements, *J. Geophys. Res.*, 111 (D14), D14301, doi: 10.1029/2005JD006479, 2006.
 26. Skupin, J., Nöel, S., Wuttke, M. W., Bovensmann, H., and Burrows, J. P.: Calibration of SCIAMACHY in-flight measured irradiances and radiances—first results of level 1 validation, *Proc. of the Envisat Validation Workshop (SP-531)*, ESA Publications Division, 2002.
 27. Thuillier, G., Hersé, M., Simon, P. C., Labs, D., Mandel, H., and Gillotay, D.: Observation of the UV solar irradiance between 200 and 350 nm during the ATLAS-1 mission by the SOLSPEC spectrometer, *Sol. Phys.*, 171, 283 - 302, 1997.
 28. Thuillier, G., Hersé, M., Simon, P. C., Labs, D., Mandel, H., Gillotay, D., and Foujols, T.: The visible solar spectral irradiance from 350 to 850 nm as measured by the SOLSPEC spectrometer during the ATLAS-1 mission, *Sol. Phys.*, 177, 41 - 61, 1998a.
 29. Thuillier, G., Hersé, M., Simon, P. C., Labs, D., Mandel, H., and Gillotay, D.: Solar radiometry and solar spectral irradiance: Observation of the solar spectral irradiance from 200 nm to 870 nm during the ATLAS 1 and ATLAS 2 missions by the SOLSPEC spectrometer, *Metrologia*, 35, 689 - 697, 1998b.
 30. Weidner, F., Development and Application of a Versatile Balloon-Borne DOAS Spectrometer for Skylight Radiance and Atmospheric Trace Gas Profile Measurements, Dissertation, Universität Heidelberg, 2005, <http://www.ub.uni-heidelberg.de/archiv/5503>

31. Weidner, F., H. Bösch, H. Bovensmann, J. P. Burrows, A. Butz, C. Camy-Peyret, M. Dorf, K. Gerilowski, W. Gurlit, U. Platt, C. von Friedeburg, T. Wagner, and K. Pfeilsticker, Balloon-borne Limb profiling of UV/vis skylight radiances, O₃, NO₂, and BrO: Technical set-up and validation of the method, *Atmos. Chem. Phys.*, 5, 1409 - 422, 2005.
32. WMO: Scientific Assessment of Ozone depletion: 2002, World Meteorological Organization Global Ozone Research and Monitoring Project, Report 47, 2003.