UPDATE ON ENVISAT / SCIAMACHY VALIDATION WITH BALLOON-BORNE DOAS INSTRUMENTS: COMPARISON OF O_3, NO_2 AND BRO PROFILES

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ABSTRACT

Here the level 2 product validation (profiles of O_3, NO_2 and BrO inferred by the IUP Bremen) from the Envisat/SCIAMACHY instrument is updated with spectroscopic balloon-borne measurements of the same quantities during 2003 - 2009. Generally, the satellite and the balloon-borne O_3 retrievals agree within 15 to 20 % above 15 km. For NO_2, a good agreement of < 20% is found for altitude > 18 km. For lower altitudes the relative differences increase, reaching about 40% at 15 km. For BrO we extend earlier validation studies of the BrO profile inter-comparison [1, 15]. A fair agreement of typical 20% discrepancies is found for BrO profile measurements between 15 and 25 km at mid and high-latitudes. For inter-comparisons of tropical measurements the discrepancy ranges between 20-50% above 20 km.

1. INTRODUCTION

The SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument onboard the European Environmental Satellite (Envisat) is an UV/visible/near-IR spectrometer designed to measure direct and scattered sunlight in various viewing geometries [2].

Here, we briefly report on the efforts made to validate SCIAMACHY level 2 products by balloon-borne direct sun UV/visible spectroscopy (big DOAS instrument) and limb scattered skylight measurements (mini-DOAS instrument). So far, the big DOAS instrument has been deployed on 8 validation flights of the LPMA/DOAS payload [3], [4], and [5]. Data from 7 LPMA/DOAS flights have already been processed (Tab. 1). Further a mini-DOAS instrument has been deployed on a series of flights with the LPMA/DOAS, LPMA/IASI, MIPAS-B2, and SALOMON-N2 payloads (Tab. 2). Here, we report on data collected during a subset of the balloon flights. The multitude of the balloon flights however covers low, mid and high-latitudes at different seasons, thus providing a unique data set for ENVISAT/SCIAMACHY level 2 validation exercises. Best matches of correlative measurements with the SCIAMACHY satellite instrument are identified using an air mass trajectory modelling technique (section 2.2). For photochemically sensitive gases correction is 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 e.g. [1], [5], [6] and [7].

2. METHOD

The validation studies comprise the following methods and tools.

2.1. Instrumentation and trace gas retrieval:

Our validation method builds on the remote sensing of UV/vis absorbing species from balloon platforms using two different type optical spectrometers (in the following briefly called 'big DOAS spectrometer' [3] and 'mini-DOAS spectrometer' [5]). These spectrometers have been deployed on various azimuth-controlled gondolas (e.g., the LPMA/DOAS, LPMA/IASI, MIPAS and SALOMON-N2) in the

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Table 1: Compendium of LPMA/DOAS observations and coincident Envisat / SCIAMACHY overpasses. BA and SO indicate balloon ascent and solar occultation measurements, respectively.

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 5285, 5 Mar., 07:17	20–30 23–24	−5.1 +15.3	369–496 498–499
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 SO: LPMA, DOAS	5545, 23 Mar., 11:07 5558, 24 Mar., 09:01 5545, 23 Mar., 11:07 5558, 24 Mar., 09:01	18–28 19–29 20–30 17–30	−5.2 +17.4 −6.2 +16.0	268–496 10–495 63–458 256–453
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., 09:51 8421, 10 Oct., 09:20	17–31 25–33	−6.5 +17.2	738–988 547–977
24 Mar. 2004 14:04 - 17:31	Kiruna 67.9°N, 21.1°E	High-lat. spring SZA: 74.5°– 95.3°	BA: DOAS SO: DOAS	10798, 24 Mar., 10:35 10812, 25 Mar., 10:04 10798, 24 Mar., 10:35 10812, 25 Mar., 10:04	12–33 6–16 10–33 10–20	−5.4 +19.9 −6.9 +16.7	371–499 32–485 191–436 301–475
17 June 2005 18:32 - 21:13	Teresina 5.1°S, 42.9°W	Tropical winter SZA: 60.6°– 95.8°	BA: DOAS SO: DOAS	17240, 17 June, 11:53 17255, 18 June, 13:02 17240, 17 June, 11:53 17255, 18 June, 13:02	25–30 5–33 23–32 8–33	−8.1 +18.4 −9.1 +16.2	382–491 6–490 519–971 12–496
27 June 2008 05:22 - 21:18	Teresina 5.1°S, 42.9°W	Tropical winter SZA: 60.4°– 95.7°	SO: DOAS	33058, 26 June, 13:04 33072, 27 June, 12:32 33072, 27 June, 12:34	18-30 12-35 12-38	-19.9 +3.9 +3.9	416-989 540-991 160-543
7/8 Sep 2009 14:53 - 06:05	Kiruna 67.9°N, 21.1°E	High-lat. summer SZA: 76.4°– 94.5°	LPMA/DOAS BA/SO	39326, 7 Sept., 09:57 39326, 7 Sept., 09:59 39340, 8 Sept., 09:25	21-34 9-34 10-34	-6.9 -6.8 +18.1	418-497 219-404 58-382
23 Apr 2011 03:28 - 07:03	Kiruna 67.9°N, 21.1°E	High-lat. spring SZA: 84°– 65°	BA: DOAS	To be detemined			

past 8 years. A deployment of the instruments on such azimuth controlled payloads is necessary, in order to guarantee well-defined observation geometry for solar occultation measurements. Details of the setup and operational performance of the both instruments have already been described previously (e.g., in [4], [3] and [5]). Therefore, only a short description of the instrumental features most important for SCIAMACHY validation is given here.

The big DOAS spectrometer, which is solely deployed on the LPMA/DOAS payload, observes direct sun-light at a moderate spectral resolution in the UV/visible (UV: FWHM = 0.5 nm, visible: FWHM = 1.5 nm) during balloon ascent and balloon float. Details of the instrument are given here [3], and in series of subsequent scientific publications.

The mini-DOAS instrument was deployed on different azimuth-controlled balloon platforms, where it either detected limb scattered skylight (e.g., LPMA/DOAS, MIPAS-B2, LPMA/IASI) received from different tangent heights by scanning actively through the atmosphere at balloon float altitude. So far on one occasion it performed direct sunlight observations from the SALOMON-N2 payload during ascent and at balloon float.

All recorded spectra are analyzed using the Differential Optical Absorption Spectroscopic (DOAS) technique [8] for the absorption of O_3, NO_2, BrO, IO, OIO, and HONO (and possibly in future also of OClO, CH_2O and H_2O) [5]. From the direct sunlight spectra, slant column amounts of the targeted atmospheric absorbers are inferred, using the DOAS approach in the UV/visible [8]. Additionally ray-tracing and radiative transfer modelling is required to correctly account for the light path, which is necessary for the inversion algorithm in order to infer vertical profiles of the measured trace gases [e.g, 9, 16]. Upon trace gas retrieval the measured 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 [9]. For the profile inversion of reactive species (e.g. NO_2 and BrO), a correction based on photochemical modeling is included [6].

In the case of subsequent scans through the atmosphere (e.g. in limb mode), even time series of trace gas profiles can be inferred, which form a rather valuable tool for a direct match of balloon to satellite measurements [e.g., 14].

2.2. Trajectory modelling:

Balloon-borne measurements are inherently restricted by different operational constraints, limiting their flexibility in satellite validation. Accordingly air mass trajectory modelling is used to find the best coincidences between air masses probed by the balloon-borne and the satellite instrument.

Based on the operational analyses of the European Centre for Medium Range Weather Forecasts (ECMWF) the model calculates the movement of the air parcels probed by the balloon instrument on 25 isentropic levels. The parcels movement is calculated in a 10 min time steps and trajectories steps are stored for each hour (e.g. [11]).

These calculations are made 48 hours forward and backward in time to find the best satellite coincidence for comparison.

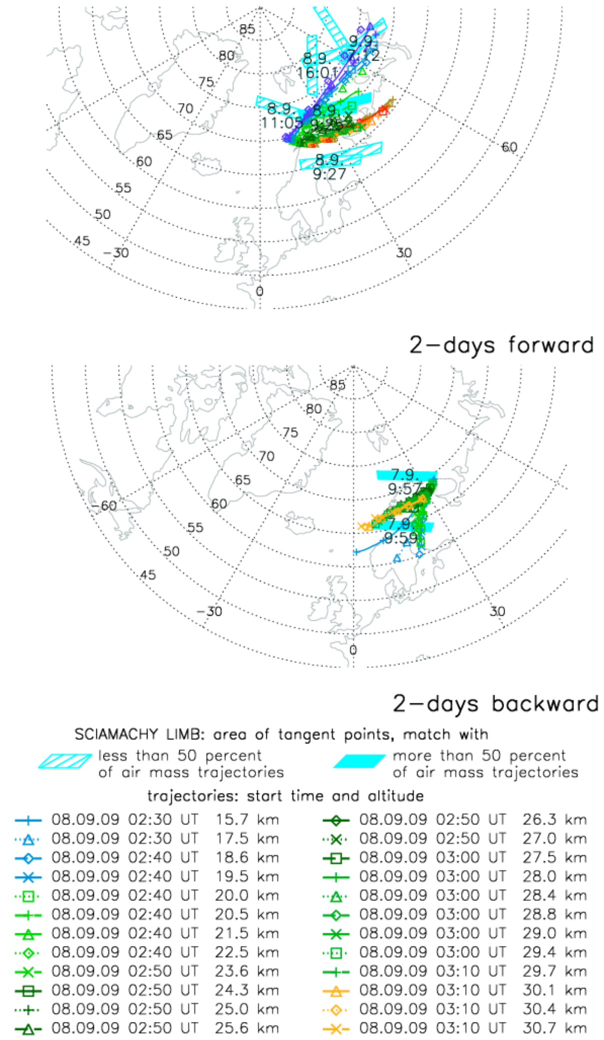

Figure 1. Air mass trajectories modeled for 48 hours forward (upper panel) and backward (lower panel) to the balloon measurements of the LPMA/DOAS payload at Kiruna on Sept. 7th and 8th, 2009. The trajectories are color-coded according to their starting altitude as indicated in the legend. A symbol is 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/shaded rectangles correspond to SCIAMACHY limb observations for which more/less than 50% of the calculated air masses are coincident with the balloon measurements.

The actual geo-locations 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 geo-location 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.

Further, 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. [11]: 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 observations do not fulfill these criteria, the distance criterion is extended up to 1000 km.

2.3. Photochemical modelling:

A 1-D column model is used to reconstruct the diurnal cycle of the targeted species 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 for each height level at 00:00 UT with outputs of the 3-D CTM SLIMCAT [12] model at the adjacent 48 hour model time step closest to the balloon launch site.

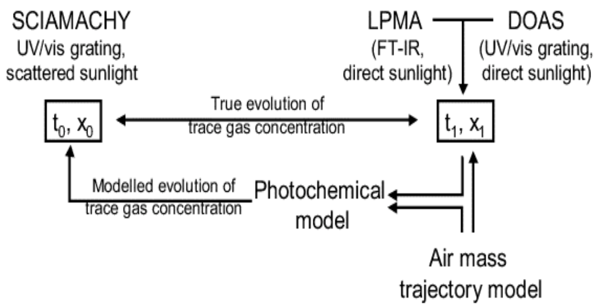

Figure 2. Schematics of the validation strategy: Air mass trajectory modelling is used to find SCIAMACHY observations (time t_0, location x_0) probing the same air masses as the balloon-borne measurements conducted at time t_1 and location x_1. For the validation of photochemically active trace species, the illumination history of the coincident air masses is fed into a photochemical model to reproduce the evolution between satellite and balloon-borne observations and to infer appropriate scaling factors. The Figure is adopted from [6].

The 1-D model is constrained to follow the evolution of the SZA time-line, which is taken from the air mass trajectory calculations. For satellite validation, these measures guarantee that the photochemical evolution of the modelled 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 taken by the remote sensing instruments on SCIAMACHY and on each of the balloon gondola is a composite of changing photochemical conditions (due to changing SZA) along the line-of-sight. Therefore, 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 [6]. A schematic of the overall validation procedure is given in Fig 2.

3. PRODUCTS & RESULTS

Validation of level 2 products:

Here we report on seven LPMA/DOAS validation balloon flights performed since 2003. Four balloon flights were conducted from ESRANGE, Kiruna, Sweden, one from Aire sur l'Adour in southern France and two from Teresina in north eastern Brazil (see Tab. 1). For each balloon flight a satellite coincident measurement is identified before and after the balloon flight using the trajectory matching technique described above (Fig.1). In the following we refer to these coincidences as backward and forward coincidences. For each balloon flight Tab. 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 and NO_2 validation:

Fig. 3 shows an illustrative comparison between SCIAMACHY O_3 profiles inferred by the IUP-Bremen retrievals (version 3.1) and the coinciding LPMA/DOAS balloon-borne observations. Comparisons of O_3 profiles retrieved from the SCIAMACHY limb measurements at IUP Bremen show a good agreement within the entire altitude range. Generally, relative differences between the two data sets are on the order of 15-20%. However, below 15 km, some outliers with too low O_3 concentrations resulting from SCIAMACHY retrievals are observed. Deviations in the lower layers might be due to the lower sensitivity of the satellite retrieval or unaccounted horizontal trace gas variations.

In Fig. 6 relative deviations between the satellite and balloon-borne observations for NO_2 profiles are shown (update of Figure 9 in [6]).

In the 20 km to 30 km altitude range the agreement between the balloon-borne NO_2 profiles and the satellite observations is on the order of ±20% and most often well represented by the combined error bars. Down to 15 km, the relative differences between SCIMACHY and balloon-borne measurements increase with decreasing altitude to about 40% with SCIAMACHY results generally being lower than the balloon-borne data. Below 15 km the agreement gets worse with the relative differences exceeding 50% which is most probably due to a decreasing sensitivity of the SCIAMACHY retrievals at these altitudes. Thus for low altitudes the SCIAMACHY retrieval might depend on the actual parameters, e.g. a priori information. The latter finding is supported by the characteristics of the corresponding averaging kernels (not shown).

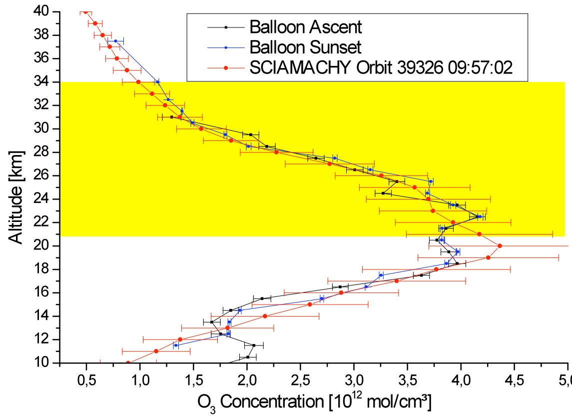

Figure 3. Comparison of O_3 profiles inferred from SCIAMACHY with balloon-borne measurements. The DOAS observations, plotted in black and blue, were conducted at Kiruna on September 7^{th} 2009, during ascent and sunset. The IUP-Bremen Satellite data, shown as orange dots, are from a scan conducted at 09:57 UT that day. The grey area marks the vertical range where according to trajectory modell calculations both instruments probed the same air mass.

3.2. BrO validation:

The BrO profiles retrieved at mid and high-latitudes from SCIAMACHY limb measurements agree mostly within 20% with co-located photochemically [1] corrected balloon-borne observations (Kiruna and Air sur l'Adour launches). The agreement is best between 15 and 25 km. Above 25 km the relative differences are slightly higher reaching about 30% which is most

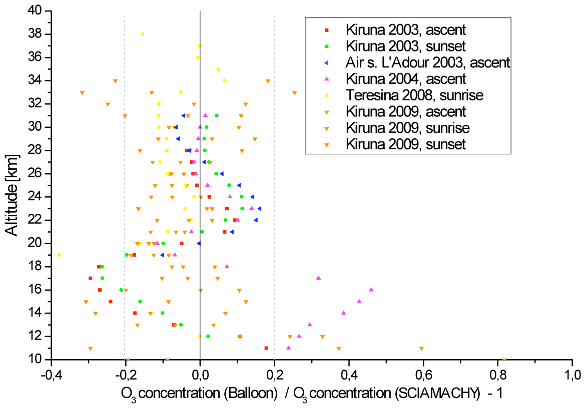

Figure 4. Relative deviations between SCIAMACHY (IUP-Bremen) and LPMA / DOAS O_3 measurements.

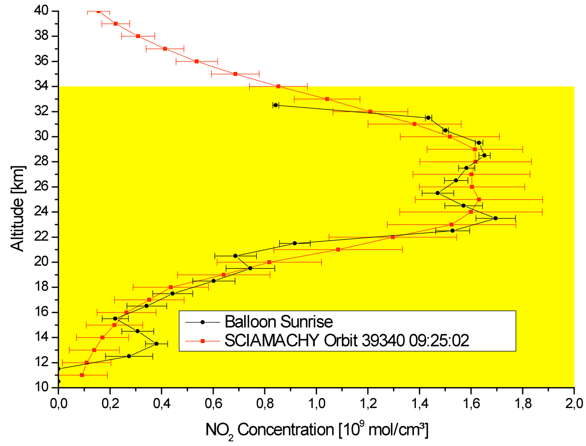

Figure 5. Comparison of NO_2 profiles inferred from SCIAMACHY limb observations with correlative balloon-borne measurements. The DOAS observations, plotted in black, were conducted at Kiruna on September 8th 2009, during sunrise. The IUP-Bremen Satellite data, shown as orange dots, are from a scan conducted at 09:25 UT that day. The yellow area marks the vertical range where according to trajectory modell calculations both instruments probed the same air mass.

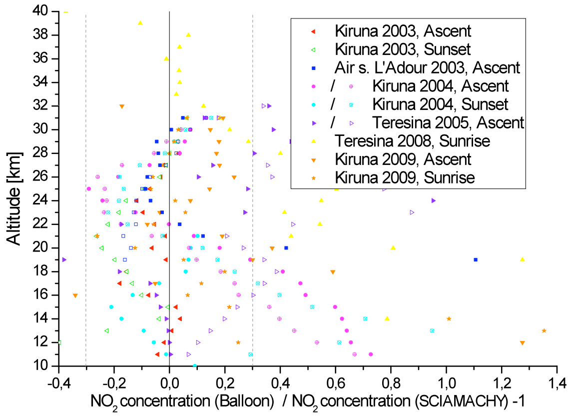

Figure 6. Same as Fig. 4, but for NO_2.

probably due to lower signal to noise ratio of the SCIAMACHY limb spectra at higher tangent heights – see Fig.8. Below 15 km the sensitivity of SCIAMACHY retrievals decreases resulting in a higher relative differences with respect to collocated balloon-borne measurement of up to 50%. For a tropical profile (only one profile for the comparison) originating from the Teresina balloon flight on 17 June 2005, the agreement is worse as compared to other flights with the relative deviation varying from 10 to 50% between 17 and 30

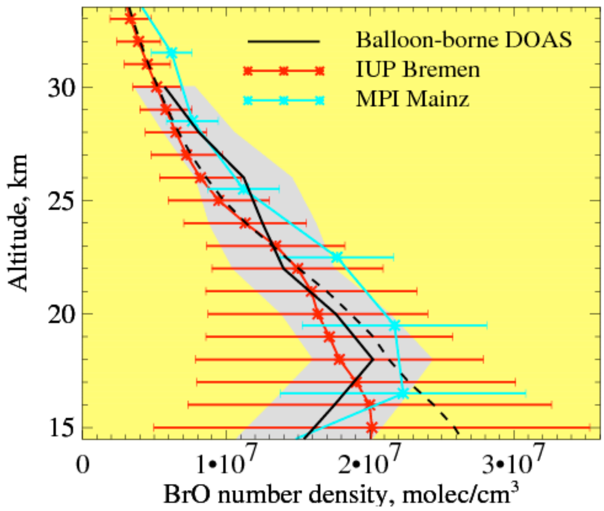

Figure 7. Comparison of BrO Concentrations measured with the Big-DOAS instrument (black) on the flight in September 2009 in Kiruna and SCIAMACHY retrievals of the IUP Bremen (red) and MPI Mainz (green) for the Orbit #39340 (see Tab.1). The Altitude range probed by both instruments is given in yellow.

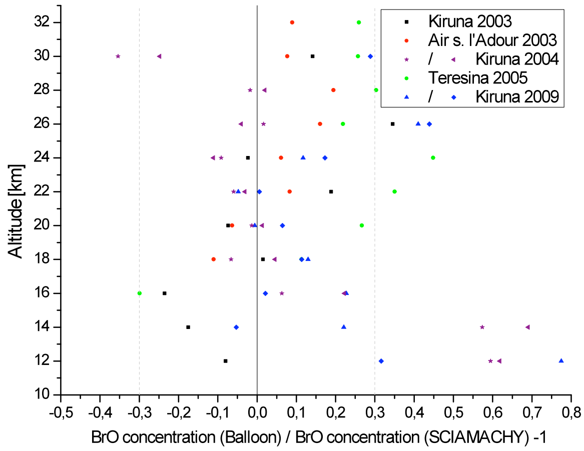

Figure 8. Same as Fig. 4, but for BrO.

km. In this altitude range the SCIAMACHY profiles for both forward and backward match are lower as compared to the comparable balloon-borne observation. Below 17 km the statistical error of both SCIAMACHY

Table 2: Compendium of mini-DOAS observations and payloads.

Date	Time / UT	Location		Geological condition	SZA	Payload
18./19.Aug. 2002	15:15 - 02:38	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, summer	70° - 94°	LPMA/DOAS
04.Mar. 2003	12:55 - 15:25	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, spring	78° - 89°	LPMA/DOAS
23.Mar. 2003	14:47 - 17:35	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, spring	79° - 95°	LPMA/DOAS
09.Oct. 2003	15:39 - 17:09	Aire-sur-l'Adour/ France	43.7°N, 0.3°W	Mid-latitude, fall	66° - 88°	LPMA/DOAS
24.Mar. 2004	13:55 - 17:35	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, spring	72° - 98°	LPMA/DOAS
24.Nov. 2004	18:30 - 21:00	Teresina / Brasil	05.1°S, 42.8°W	Tropical summer	63° - 90°	Ground-based
13.June 2005	09:00 - 17:10	Teresina / Brasil	05.1°S, 42.8°W	Tropical winter	93° - 29°	MIPAS
17.June 2005	18:30 - 21:30	Teresina / Brasil	05.1°S, 42.8°W	Tropical winter	61° - 95°	LPMA/DOAS
30.June 2005	09:00 - 17:00	Teresina / Brasil	05.1°S, 42.8°W	Tropical winter	93° - 29°	LPMA/IASI
01.Mar. 2006	08:00 - 17:22	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, winter	77° - 100°	LPMA/IASI
13.June 2008	08:45 - 13:50	Teresina / Brasil	05.1°S, 42.8°W	Tropical winter	91° - 32°	LPMA/IASI
27.June 2008	05:22 - 21:18	Teresina / Brasil	05.1°S, 42.8°W	Tropical winter	60° - 95°	LPMA/DOAS
10.Mar. 2009	03:34 - 05:10	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, winter	98° - 90°	MIPAS
07./08. Sep 2009	14:53 - 06:05	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, summer	70° - 94°	LPMA/DOAS
30./31. March 2011	03:01 - 07:05	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, spring	94° - 71°	MIPAS
24. April 2011	03:01 - 07:03	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, spring	87° - 66°	LPMA/DOAS
28. April 2011	02:35 - 05:03	Kiruna / Sweden	67.9°N, 21.1°E	High-latitude, spring	85° - 76°	SALOMON-N2

and balloon-borne profiles is too high making the comparison results less meaningful (see [14]).

3.3. Validation with the mini-DOAS instrument:

An advantage of scattered skylight measurements as compared to direct sun measurements results from the potential to record the diurnal variation of the targeted species, rendering photochemical modelling unnecessary in satellite validation (see Figure 9).

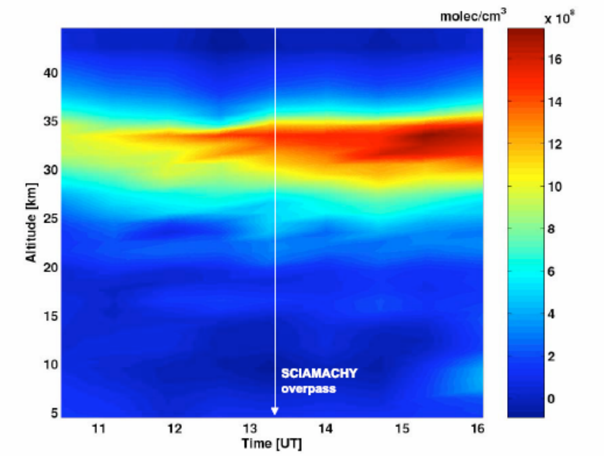

Figure 9. Diurnal variation of NO_2 retrieved from mini-DOAS measurements aboard the LPMA/IASI payload on June 30, 2005. The white arrow marks the time of the SCIAMACHY overpass.

While the altitude resolution of the mini-DOAS and the SCIAMACHY instrument are alike, the sensitivity of the mini-DOAS instrument peaks just below balloon altitude near the concentration maximum of the target species [13].

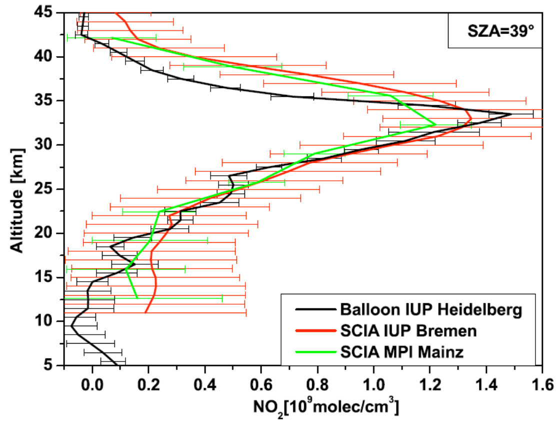

Figure 10. Concentration of NO_2 retrieved from SCIAMACHY measurements in orbit 17427 by the IUP Bremen (red) and from balloon-borne measurements, retrieved by the IUP Heidelberg (black), both at SZA=39° on June 30, 2005. Figure adopted from [13].

4. SUMMARY & CONCLUSION

Results from the 7 LPMA/DOAS and 15 mini-DOAS validation flights:

- Variable good agreement is obtained for level 2 products (BrO, NO_2 and O_3)
- In general comparisons indicate an accuracy of: ± (20 - 30) % for NO_2 and BrO (except for tropical measurements), and ± (15 - 20)% for O_3.
- A BrO comparison study has been made by the University of Bremen for the algorithms and retrieval results from IUP-Bremen, MPI-Mainz, DLR and Harvard-Smithsonian [14].

- Air mass trajectory calculations prove to be an important and powerful tool in satellite validation e.g. for coinciding balloon flights and satellite overpasses planning and for the calculation of the photochemical change of the targeted species.
- mini-DOAS observations are very purposeful for satellite validation, due to reasonable large degrees of information and the potential to monitor the time dependency of radicals, thus rendering photochemical corrections for collocated observations unnecessary.
- Latest validation activities include an LPMA/DOAS and several mini-DOAS flights during the Enriched campaign at Kiruna, Sweden during March/April 2011.
- During the Enriched campaign an adapted mini-DOAS instrument was used for the first time in direct sun observations onboard the Salomon-N2 gondola.

The methods presented here are discussed in detail in [1], [5] and [6]. They are also of value for the validation of other existing satellite measurements of BrO, NO_2 and O_3 (e.g. GOME-2, OMI).

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