GROUND-BASED COMPARISONS OF EARLY SCIAMACHY O_3 AND NO_2 COLUMNS

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ABSTRACT

During the Commissioning Phase of the ENVISAT satellite, the geophysical consistency of early SCIAMACHY ozone and nitrogen dioxide columns was investigated through comparisons with correlative measurements acquired from pole to pole by a variety of ground-based sensors. Based on the outcome of those correlative studies, the present review draws a preliminary quality assessment of the first SCIAMACHY operational products obtained with the data processors SCI_RV 3.53 and SCI_NL 3.51 to 3.53.

1. INTRODUCTION

On March 1, 2002, the third Earth observation satellite platform of ESA, ENVISAT, was launched onto a heliosynchronous polar orbit with 10:00 mean solar local time at descending node. As part of its atmospheric chemistry payload, SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) is a joint project of Germany, The Netherlands and Belgium, aiming at the global measurement of various trace gases in the troposphere and the stratosphere [1]. Among others, the vertical column and distribution of ozone (O_3) and nitrogen dioxide (NO_2) is derived from SCIAMACHY measurements of the solar irradiance and Earth nadir radiance spectra. An extensive validation campaign of ENVISAT has been organised by the ESA Atmospheric Chemistry Validation Team (ACVT) [2] and by the SCIAMACHY Validation and Interpretation Group (SCIAVALIG) [3] through so-called Announcement of Opportunity (AO) projects. A list of AO projects aims at the geophysical validation of SCIAMACHY O_3 and NO_2 columns via correlative studies with ground-based measurements.

The present report summarises the outcome of such ground-based comparisons performed during the Commissioning Phase (CP) of the satellite and discussed during the ENVISAT Validation Workshop held at ESRIN on December 9-13, 2002. Results presented hereafter are based on the data acquired and the work carried out by a large number of teams grouped in several AO projects, namely: AOID 126 (PI: M De Mazière), 158 (J-C. Lambert), 174 (H. Kelder), 179 (R. McKenzie), 191 (T. Blumenstock), 300 (D. De Muer), 331 (J. P. Burrows), 427 (Yu. M. Timofeyev), 429 (E. Kyrö), and 1103 (A. Petritoli). Section 2 describes the correlative database and the SCIAMACHY data sets generated during CP and used in the reported studies. In Section 3 and Section 4, preliminary ground-based comparisons are discussed and conclude to a first quality assessment of early SCIAMACHY O_3 and NO_2 columns, respectively. More details and individual AO contributions are reported elsewhere in the same issue [4-10]. The paper concludes with perspectives for the Main Validation Phase of SCIAMACHY, which extends from September 2002 through September 2003.

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2. DATA SETS

2.1 Correlative Measurements of Total Ozone

Observations of total ozone have been collected from a variety of instruments offering complementary capabilities and, to some extent, independent sources of uncertainty: (i) Dobson [11] and (ii) Brewer [12] ultraviolet spectrophotometers record daytime ozone in the direct sun geometry (weather permitting) or zenith-sky geometry; (iii) DOAS/SAOZ UV-visible spectrometers derive ozone at sunrise and sunset from zenith-scattered sunlight measurements [13,14]; (iv) Russian ultraviolet filter radiometers (M-124 model) [15] use the same geometry as the Dobson; (v) Fourier Transform infrared (FTIR) spectrometers measure daytime ozone in the direct sun geometry [16], a few of them being also capable of moonlight observations [17]; (vi) calibrated onto the Dobson but extending its capabilities towards low solar elevation, multi-channel filter radiometers (GUV) deployed in Norway derive ozone from total irradiance measurements [18]. Most of the instruments contribute to network programmes in the framework of WMO's Global Atmospheric Watch (GAW) and/or the Network for the Detection of Stratospheric Change (NDSC). During WMO- and NDSC-endorsed intercomparison campaigns [19,10], Dobson instruments can be adjusted to agree within 0.3-1%. The overall agreement between the various instrument types generally falls within the 3% range at middle latitudes. At higher latitudes, the enhanced amplitude of several effects (temperature dependence of the absorption cross-sections, atmospheric profile shape effect, internal straylight at low sun elevation, sensitivity to stratospheric aerosols etc.) generates average differences of about ±3-7% varying with the season and other parameters (see e.g. [20], [21], and references in [22]).

2.2 Correlative Measurements of Total Nitrogen Dioxide

Observations of total NO_2 at sunrise and sunset have been collected from a network of about thirty UV-visible DOAS/SAOZ UV-visible spectrometers measuring zenith-scattered sunlight spectra [13,14]. Nine stations have provided daytime NO_2 columns measured by FTIR spectrometers. Due to either the zenith-sky measurement geometry (DOAS/SAOZ) or pressure broadening of the weak NO_2 absorption lines in the lower troposphere (FTIR), both types of measurement are sensitive mainly to stratospheric NO_2. During major field intercomparison campaigns, the agreement between UV-visible spectrometers generally falls within the 5% to 10% range for total NO_2 [13,14]. Long-term comparisons of nearly co-located UV-visible instruments conclude to a mean agreement of 3% in summer and 9% in winter [23]. The precision of single NO_2 measurements performed by FTIR is estimated to range between 6% and 12%. Again, most of the contributing instruments are affiliated with the NDSC.

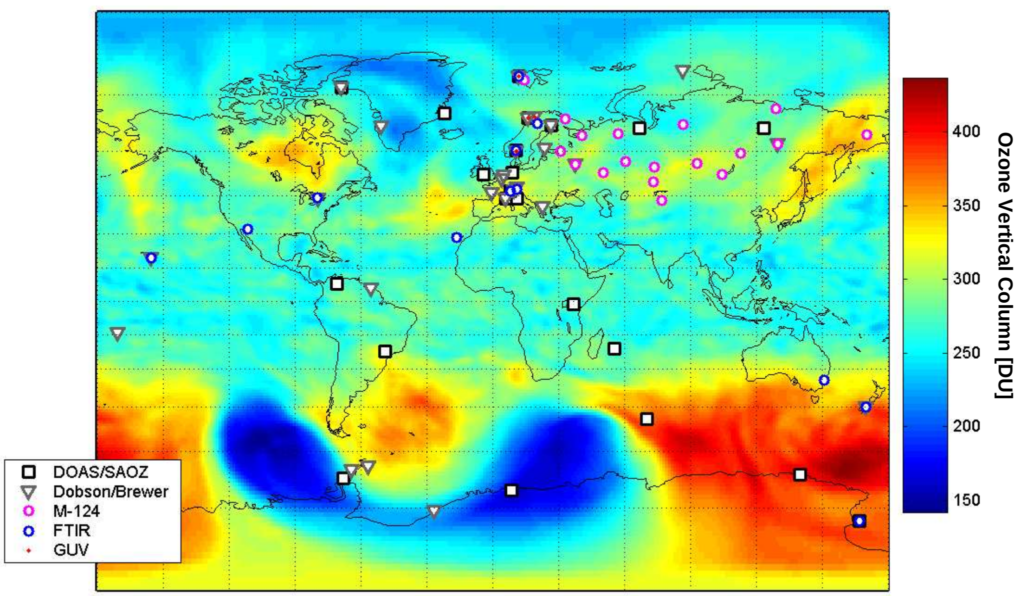

Fig. 1. Contributing total ozone sensors are highlighted on top of the global ozone field measured on September 25, 2002, by the GOME satellite (assimilated GOME total ozone (in DU) generated by KNMI/ESA). Symbols indicate the type of sensor (see text for more details). The figure illustrate clearly the exceptional morphology of the Antarctic ozone hole 2002 due to unusual patterns in the atmospheric circulation surrounding the polar vortex.

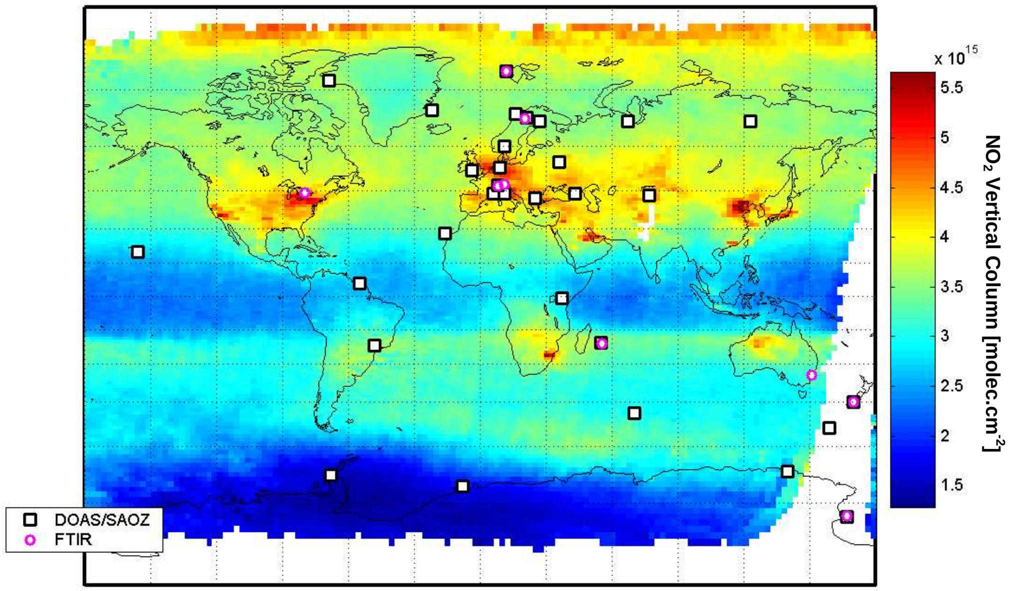

Fig. 2. Contributing total NO_2 sensors are highlighted on top of the September 2000 monthly mean field of total NO_2 derived from GOME satellite measurements (generated by IASB/DLR/ESA). DOAS/SAOZ UV-visible spectrometers appear as squares and FTIR spectrometers as circles.

2.3 SCIAMACHY Operational Data Products

The operational delivery of the Meteo product (SCI_RV) version 3.53 started on September 17, 2002, allowing studies over nearly 2.5 months of those data. The small amount of information available in the Meteo data files – i.e. primarily the ozone column value and its geolocation – limits the field of investigation to studies of time-series and of a possible column dependence. Further studies require additional parameters (solar zenith angle, line-of-sight etc.) reported in the near real-time (NRT) product (SCI_NL), which also includes the nitrogen dioxide data.

During the ENVISAT commissioning phase, four versions of the SCI_NL data product were distributed. Unfortunately, each of those data sets covers a different time period in 2002: (i) a few orbits in August with v3.51; (ii) a few orbits in early September with v3.52; (iii) v3.53 data starting from October; and (iv) one orbit on August 23 processed with v4.0. This composite, sparse data set can be used for preliminary quality checks as reported hereafter but it does not allow comprehensive geophysical validation studies: (a) the amount and/or quality of collocations with ground-based data might be poor, especially from ground-based techniques requiring adequate weather conditions such as FTIR; (b) geophysical validation of important features such as seasonal cycles or Arctic springtime are not possible; (c) few or no collocations exist between the different SCIAMACHY data products. Despite this lack of collocations between the different SCIAMACHY versions, the equivalence between Meteo and NRT products has been tested using ground-based time-series as a standard transfer. Due to minor bugs that will be fixed in future versions, small differences are found between the Meteo and NRT products. Fortunately, those differences seem to occur mainly in the geolocation parameters and have reduced amplitude – e.g. a maximum latitude difference of 0.15°, that is, 17 km. Therefore comparisons carried out with the Meteo and with the NRT products should not reach significantly different conclusions.

So far, no data product from the SCIAMACHY off-line processor (SCI_OL) has been made available for validation.

Among the various groups, SCIAMACHY data have been selected for comparison according to different space/time criteria. For total ozone, differences from one group to another may change the 1s scatter but do not alter the mean difference significantly. The advantage offered by physically based methods when atmospheric variability is high, is illustrated in Fig. 7-a where SCIAMACHY is compared to ground-based zenith-sky data during Antarctic springtime. Compared to classical selection methods involving simple distance criteria only (e.g. all data within a radius of 300 km), SCIAMACHY pixels crossing the more complicated zenith-sky sampled air mass yield a value closer to the mean but a reduced scatter. Nevertheless, for several versions of the SCI_NL product, the amount of available collocations with ground-based data is sometimes so small that cruder selection criteria have to be used.

3. OZONE COLUMN COMPARISONS

3.1 Comparative Study of Successive NRT Versions

To overcome the lack of simultaneous collocations between the different SCIAMACHY data sets, ground-based data can be used as a standard transfer to test the evolution of successive versions of the SCIAMACHY ozone product. Such studies indicate that, from v3.51 to v3.53, the best results might be obtained with v3.53. Fig. 3 illustrates this evolution at the Arctic station of Kiruna, where comparisons with IMK FTIR data conclude to a mean total ozone underestimation of the order of 30% with v3.51, 25% with 3.52, and 6% with 3.53. No statement can be made from the unique v4.0 collocation. The comparison of the three versions 3.5x with Russian M-124 network data, shown in Fig. 4, yield similar conclusions and suggests also a significant reduction of the scatter. This evolution justifies that only results obtained with v3.53 are shown hereafter. Moreover, the relative equivalence established between Meteo and NRT products allows concentrating the ozone study on the Meteo product (SCI_RV v3.53), which offers the longest SCIAMACHY time-series available at present time.

3.2 Pole-to-pole Agreement

From pole to pole, time-series of total ozone values reported by SCIAMACHY and by ground-based instruments have been looked at qualitatively and, data sets permitting, quantitatively. Qualitatively, short-term fluctuations reported by ground-based sensors are reproduced similarly by SCIAMACHY. Other variations such as seasonal cycles cannot be studied with the current data sets. Quantitatively, most of the stations from one polar circle to the other conclude to a 4%-10% general underestimation of the ground-based values by SCIAMACHY. Beyond the polar circles, the mean agreement improves to a few percent. This meridian variation is displayed in Fig. 5.

3.3 Antarctic Ozone Hole 2002

During late September and early October 2002, unusual patterns in the atmospheric circulation surrounding the Antarctic polar vortex led to the separation of a large mass of air from the vortex. In addition to the exceptional morphology it gave to the ozone hole (see Fig. 1), this polar vortex split also decreased the level of chlorine activation and consequently of ozone depletion by raising stratospheric temperatures to sufficiently high to hamper the formation of polar stratospheric clouds. Ground-based studies have investigated in details to what extent SCIAMACHY was able to reproduce this unexpected behaviour. The investigation is based on several Dobson and UV-visible instruments spread around Antarctica and surroundings. The CNRS SAOZ at Kerguelen Island (49°S), a station usually located in the circumpolar belt characterised by high ozone values, reported in late September 2002 somewhat lower ozone values (around 300 DU instead of 400 DU), the station being close to the vortex edge. Simultaneously, in the Antarctic Peninsula,

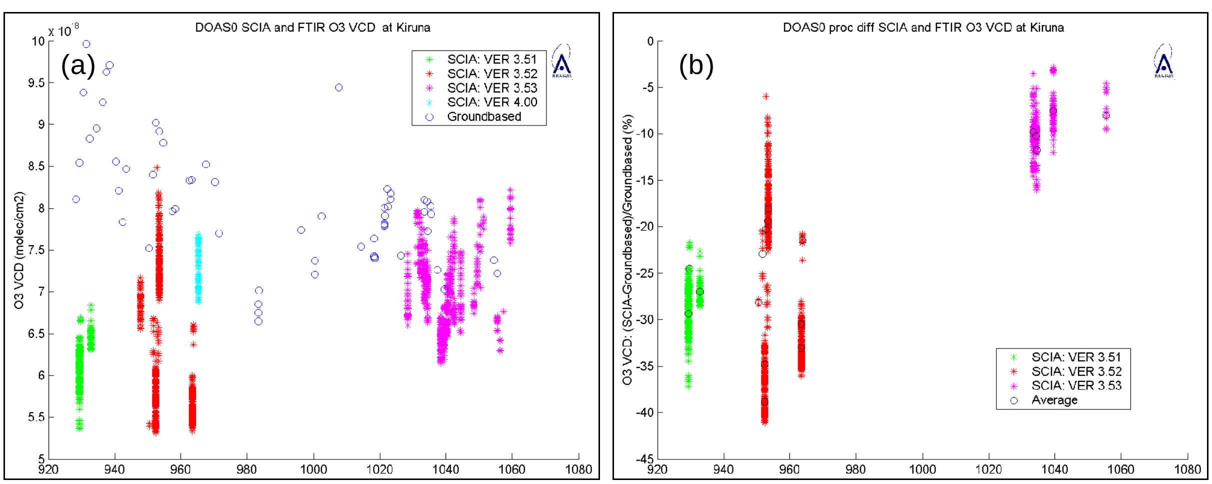

Fig. 3. Confrontation of successive versions of SCIAMACHY NRT total ozone (SCI_NL 3.51, 3.52, 3.53, and 4.0) with respect to ground-based measurements acquired at Kiruna (68°N) from August through November 2002 by the IMK FTIR spectrometer: (a) vertical column amount as a function of the day number since 1 January 2000, and (b) percentage relative difference.

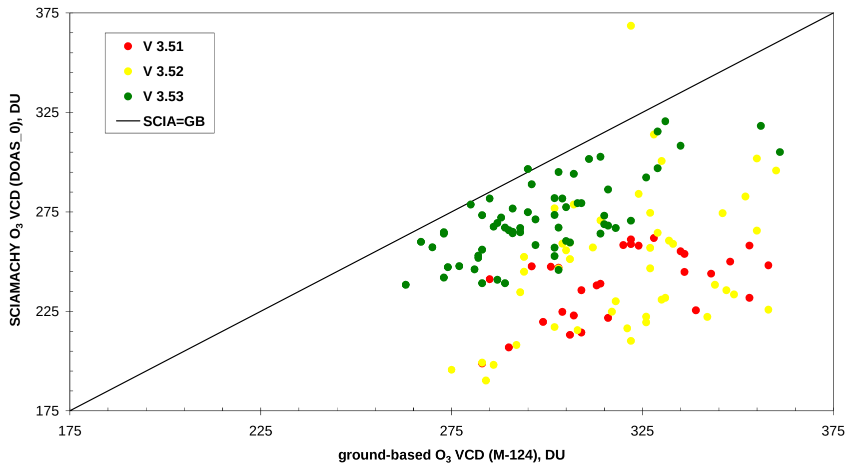

Fig. 4. Confrontation of successive versions of SCIAMACHY NRT total ozone (SCI_NL 3.51, 3.52, and 3.53) with respect to ground-based measurements acquired by the Russian network of M-124 radiometers (43°-78°N / 14°-170°E) from August through September 2002.

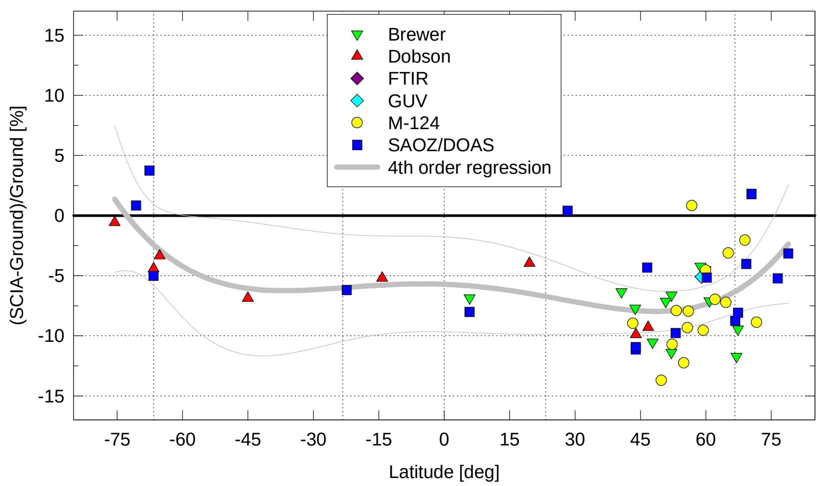

Fig. 5. Mean relative difference between SCIAMACHY Meteo v3.53 and ground-based total ozone as a function of the latitude. A general underestimation of ground-based values by SCIAMACHY vanishes near the poles.

the Dobson operated by KTSU/BAS at Vernadsky (65°S), and the BAS SAOZ operated at Rothera (68°S), provided independent observations of the air mass that separated from the vortex. They also recorded exceptionally high values of 380 DU for the first time in this season. The CNRS SAOZ at Dumont d'Urville (67°S) in Terre Adélie, a station usually alternating between in-vortex and out-of-vortex conditions during spring, was found this time deeply in the circumpolar belt, where it recorded high total ozone values over 400 DU. The high-latitude site of Halley (76°S), located permanently in the polar vortex and where the BAS Dobson reports very low ozone values every spring since the 80s, was now on the vortex edge, in the heart of its rupture.

Despite the unexpected conditions observed this year in Antarctica, the qualitative agreement of SCIAMACHY with ground-based data records looks excellent. Fig. 5 shows average relative differences of a few percent while Fig. 6 and Fig. 7 shows how well short- and mid-term variability is reproduced. From a global point of view, SCIAMACHY and ground-based sensors give a view of dynamical features consistent with those described in the WMO Antarctic Bulletins [24] and in [25].

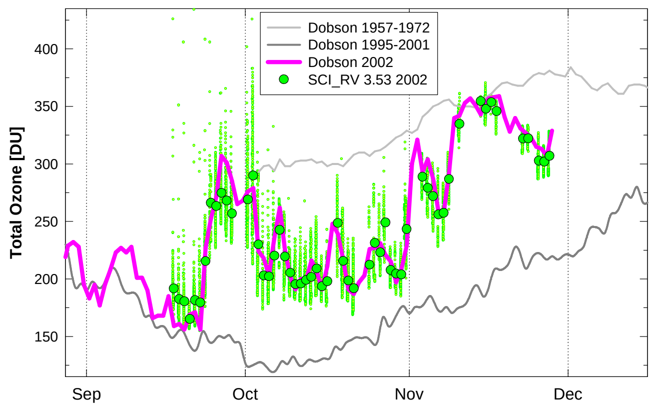

Fig. 6. Vertical column of ozone at the Antarctic station of Halley, as measured by SCIAMACHY in springtime 2002 and the BAS Dobson (i) in 2002, (ii) during the 6 last years, and (iii) during the pre-ozone hole period. Despite the unusual behaviour of the ozone field caused by the early-spring separation of the Antarctic polar vortex, SCIAMACHY captures remarkably the general evolution and all short-term fluctuations reported by the Dobson.

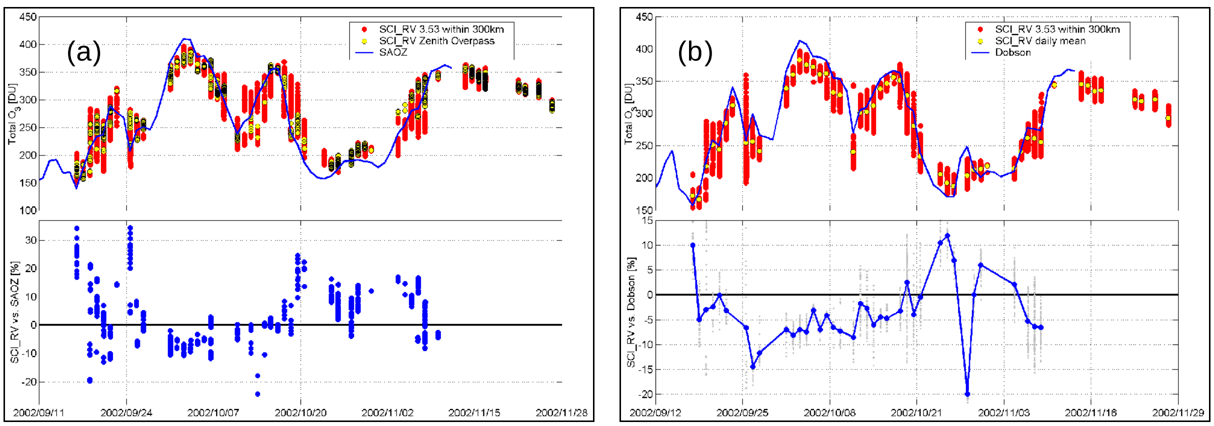

Fig. 7. Comparison of SCIAMACHY v3.53 with ground-based total ozone at two neighbouring NDSC/Antarctic stations: (a) with BAS SAOZ data at Rothera (68°S), and (b) with the KTSU/BAS Dobson data at Vernadsky (65°S). Upper panels: time-series of total ozone observed by SCIAMACHY (two methods of selection) and the ground-based instrument; lower panels: time-series of relative differences. Although the average agreement is excellent, systematic differences of ±15% are frequent. Those differences correlate with the ozone column value. The reduction of the scatter gained by a physically based selection method is well illustrated by the SCIAMACHY/SAOZ comparisons in (a).

Fig. 5 and Fig. 7 show that the average relative difference does not exceed 5% at Antarctic stations, which might be considered as excellent for Antarctic springtime. A closer look at the data suggests that, as expected from known properties of the current SCI_RV retrieval algorithms, the accuracy of individual values varies with several parameters. A major issue consists in the strong overestimation of the lowest ozone column values appearing in Antarctica when the stations are located in the heart of the ozone hole. This is well illustrated in Fig. 7 where the difference between SCIAMACHY and ground-based sensors is found to correlate with the ozone column. The known column dependence between BAS SAOZ and Dobson data can certainly not account for this strong correlation: its effect can be significant on the lowest values but is limited to a few percent at 200 DU. A solar zenith angle dependence is also discernible at low sun elevation, but the available data sets are too sparse to give an accurate description of this dependence. More generally, individual differences of about 20-30% are frequent and the standard deviation of the differences is twice as large as that expected from natural variability. Although computing statistics with such a short and variable data record is hazardous, it is interesting to note that differences are less scattered elsewhere, e.g., at northern middle latitudes where SCIAMACHY underestimates ground-based values by 6% on the average. This suggests that the large scatter of the difference might be linked – at least partly – to the high variability of stratospheric ozone in the studied conditions. Current data sets are not suitable to determine to what extent this apparent link can be attributed to differences in air masses, nevertheless contributions from the current SCIAMACHY algorithms have already been identified.

It also happens that SCIAMACHY reports out-of-range values (negative or beyond 600 DU), and too high values at the end of orbits. Such effects might be caused by errors associated with high solar zenith angle and with the viewing angle, or simply by bugs, as reported e.g. in [26]. Forward and backward scans of SCIAMACHY rarely differ by more than 1% in total ozone, which is acceptable compared to the level of agreement with ground-based data. The very limited information contained in the Meteo data files does not allow further studies of possible errors.

In summary, there is evidence that reliable ozone information is present in the SCIAMACHY Meteo product. It may be used for studies of the SH polar vortex separation of September 2002, provided that end-of-orbit and aberrant data are properly filtered out and that studies concentrate on qualitative aspects of the phenomenon. More quantitative studies of the phenomenon should take into account dependences on the ozone column and the solar zenith angle. The excellent average agreement during Antarctic springtime should not hide the systematic bias observed outside of polar areas.

4. NITROGEN DIOXIDE COLUMN COMPARISONS

4.1 Diurnal Cycle

The major difficulty of NO_2 comparisons arises from the diurnal cycle of this molecule. Controlled by the diurnal variation of solar illumination, photochemical reactions involving the NO_x family (NO, NO_2, NO_3, N_2O_5) lead to the day/night alternance illustrated in Fig. 8-a and characterised by: (i) a sharp decrease of NO_2 and the emergence of NO at sunrise resulting from the fast photolysis of NO_2 when daylight appears, followed by (ii) a quasi-linear, slight increase during daytime combining NO_2/NO photochemical equilibrium and photolysis of their night-time reservoir N_2O_5; (iii) as sun sets, a sharp increase of NO_2 and the disappearance of NO as NO_2 photolysis vanishes; and finally (iv) a slow decrease of NO_2 during night due to its conversion with NO_3 into N_2O_5. Other nitrogen compounds exhibit a small diurnal cycle (e.g. $ClONO_2$ and HNO_3) but their contribution to the cycle of NO_2 is not significant.

Varying with the solar illumination regime and with the vertical distribution of NO_x species and of temperature, the diurnal cycle of NO_2 exhibits marked seasonal and meridian features. Modelling studies carried out at IASB in collaboration with U. Leeds indicate that sunrise values acquired by twilight instruments (SAOZ/DOAS) might be reasonably close to the mid-morning values acquired by SCIAMACHY, that is, within a few 10^{14} molecule.cm^{-2}. Two main exceptions have been identified: (i) polar springtime, where strong changes of temperature and of NO_y partitioning (e.g. resulting from denoxification or denitrification) make the amplitude of the diurnal cycle somewhat large and unpredictable; and (ii) polar day, where the strong NO_2 diurnal cycle of about 0.5-1.5 10^{15} molecule.cm^{-2}, driven only by the photochemical equilibrium between NO and NO_2, makes NO_2 alternating between a minimum around noon and a maximum under midnight sun conditions. The latter regime is illustrated in Fig. 8–b, for which an adjustment factor has been drawn as a function of the SCIAMACHY solar zenith angle, enabling quantitative comparisons.

4.2 Pole-to-pole Agreement

SCIAMACHY v3.53 total NO_2 has been compared to UV-visible and FTIR ground-based data at nearly all stations displayed in Fig. 2. Fig. 9 shows the reasonable level of agreement reached at the low latitude station of Izaña (28°N) on Tenerife Island. When the diurnal cycle is taken into account properly, the three data records are mutually consistent. More globally, the absolute difference between SCIAMACHY and ground-based total NO_2 exhibits a striking meridian structure, as illustrated in Fig. 10. From an average 3-13 10^{14} molecule.cm^{-2} underestimation of ground-based values through the Southern Hemisphere and till the Northern Tropic, the difference rises rapidly towards the Northern middle latitudes to reach a maximum of 37 10^{14} molecule.cm^{-2} around 50°N. Fig. 10 also shows that the station of Izaña is exactly at the limit between positive and negative differences. Such large differences with SCIAMACHY, as well as the pronounced meridian structure, were not observed with GOME GDP 2.4 data, the version which SCI_NL is supposed to rely on. In the Southern Hemisphere, where stations often are characterised by an unpolluted troposphere, the agreement between the GOME and ground-based data records is within a few $\pm 10^{14}$ molecule.cm^{-2}. At several Northern Hemisphere stations, where clean and polluted conditions alternate, a bimodal behaviour appears due to the sensitivity of GOME to tropospheric NO_2: absolute differences of ± 5 10^{14} molecule.cm^{-2} are typical of clean conditions, while tropospheric pollution events produce an average overestimation of ground-based values ranging from 5 10^{14}

molecule.cm^{-2} in summertime to 25 10^{14} molecule.cm^{-2} during wintertime. Differences observed between SCIAMACHY and ground-based total NO_2 exceed by far those GOME results and do not reproduce the same behaviour.

It is known that GOME and SCIAMACHY total NO_2 might differ partly due to differences in absorption cross-sections. The meridian structure highlighted by our study indicates that other major effects must be taken into account. The shape of this structure is symptomatic of nadir air mass factors (AMF) calculated with an atmospheric profile database relying on 2D modelling results. Such models propagate unrealistically high tropospheric NO_2 values through zones featuring NO_x emission sources. This propagation produces underestimated AMFs in the clean parts of the zone, leading to overestimated vertical columns. The meridian structure of SCIAMACHY correlates well with the systematic error introduced by the database used with GOME GDP 2.0. If this matter of fact is verified, it is recommended to replace the GDP 2.0 database by that used in later versions of GOME GDP (2.3 and following). No statement can be made on SCI_NL version 4.0 since preliminary comparisons - also depicted in Fig. 10 - cover only one orbit in a different time period.

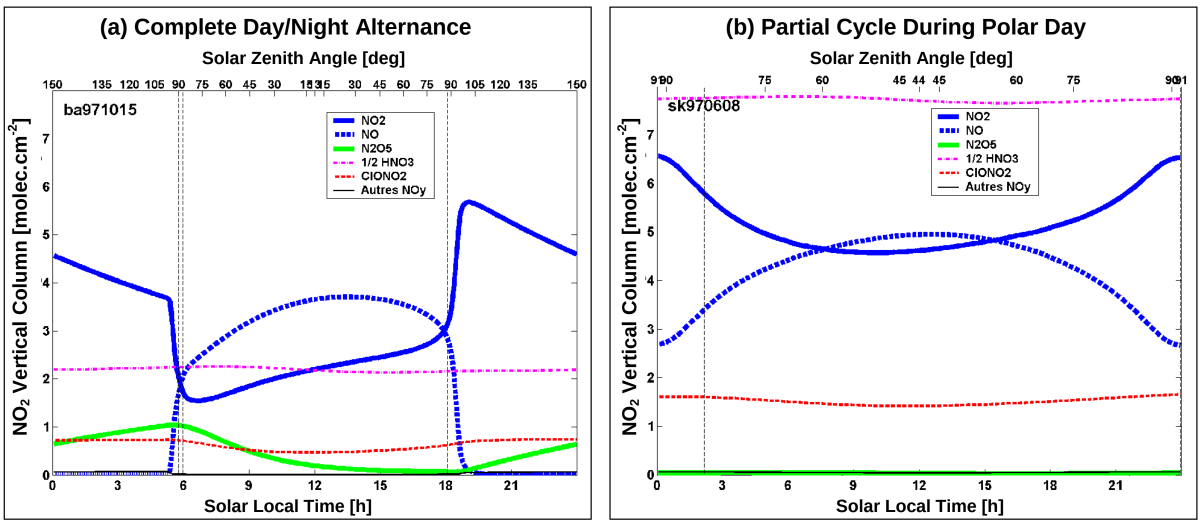

Fig. 8. Diurnal cycle of stratospheric NO_2 and NO_y species as derived from coupled PSCBOX/SLIMCAT modelling results. (a) Complete day/night alternance regime, typical of low and middle latitudes, involving primarily NO_x species (NO_2, NO, NO_3, N_2O_5). (b) Partial cycle in polar summertime, limited to the NO_2/NO equilibrium. In both regimes, the contribution of HNO_3, $ClONO_2$ and others nitrogen compounds is negligible.

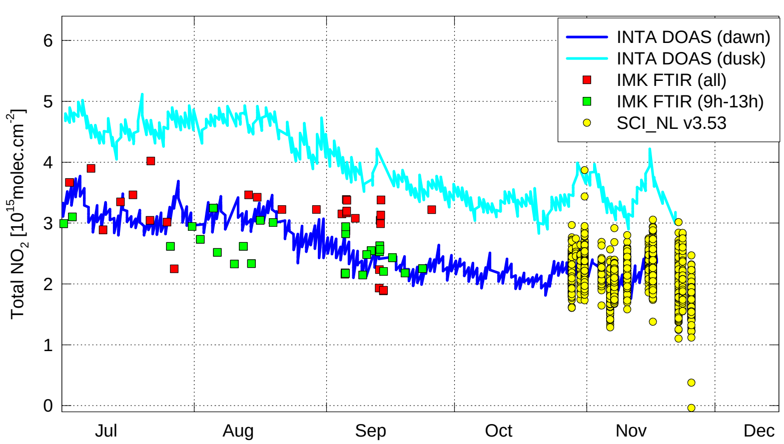

Fig. 9. Vertical column of NO_2 at Izaña (28°N) from July through November 2002, as measured by the INTA DOAS UV-visible spectrometer (average value at dawn and at dusk), the IMK FTIR spectrometer (all daytime data, and also data within 2 hours around the SCIAMACHY overpass), and SCIAMACHY v3.53 (around 10:00 solar local time). Taking the diurnal cycle into account, the three data records are mutually consistent.

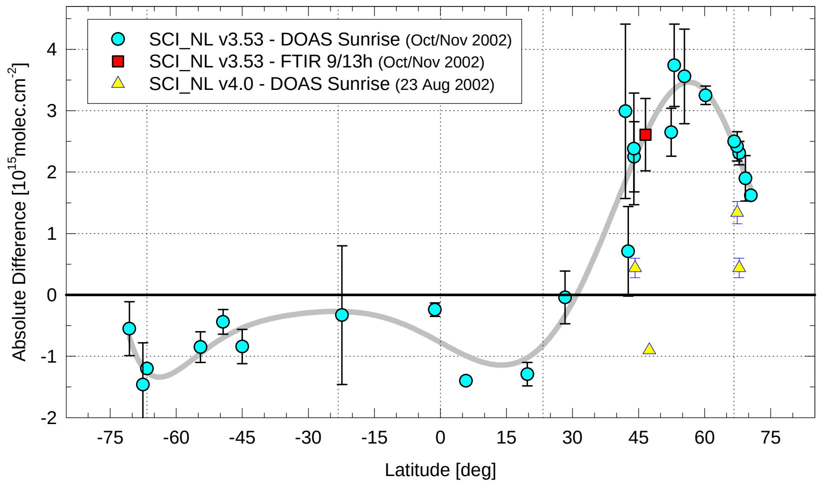

Fig. 10. Meridian variation of the absolute difference between SCIAMACHY v3.53 and ground-based total nitrogen dioxide in October/November 2002. A strong structure appears that might be related to the use of an inadequate atmospheric profile database for the calculation of the SCIAMACHY AMF. The enhanced 1s scatter at the tropical station of Bauru (22°S) results from perturbations linked to the South Atlantic Anomaly. Results obtained with one orbit on August 23, processed with the new SCIAMACHY v4.0, are also displayed.

5. CONCLUSION AND PERSPECTIVES

During the ENVISAT Commissioning Phase, early ozone and nitrogen dioxide column products generated by successive versions v3.5x of the SCIAMACHY level-1-to-2 data processors were tested through comparisons with correlative measurements acquired from pole to pole by complementary types of ground-based sensors. The late delivery of ENVISAT data products, furthermore limited to a part of the Northern fall/Southern spring season, hampered the output of the planned validation effort. Nevertheless, the amount of available collocations with ground-based data was sufficient to demonstrate that SCIAMACHY measurements contain valuable geophysical information. The quantitative agreement is more a concern. For total ozone, version 3.53 of the meteo (SCI_RV) and near real time (SCI_NL) products offers a remarkable qualitative agreement and the closest quantitative agreement with ground-based data. Results related to the exceptional Antarctic ozone hole of 2002 are particularly good. However, compared to ground-based data, SCIAMACHY total ozone also exhibits a systematic bias of -4% to -10% on an average, vanishing near the poles. For total nitrogen dioxide, SCIAMACHY data differ from ground-based measurements by -1 10^{15} molecule.cm^{-2} in the Southern Hemisphere and up to +3.7 10^{15} molecule.cm^{-2} in the Northern Hemisphere. The marked meridian structure of the discrepancy suggests some problems with the atmospheric profile database used for the calculation of air mass factors. For both ozone and nitrogen dioxide columns, the algorithm and processing teams are aware of possible solutions and the operational algorithms are being upgraded. Provided that the operational delivery of ENVISAT gets on and that the entire SCIAMACHY data record acquired during CP is reprocessed with future upgrades of the level-1-to-2 processors, it is anticipated that first geophysical validation results, that is, firm conclusions on the quality and geophysical usability of SCIAMACHY data, could be drawn in the second half of 2003.

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