GEOPHYSICAL VALIDATION OF SCIAMACHY NO_2 VERTICAL COLUMNS: OVERVIEW OF EARLY 2004 RESULTS

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ABSTRACT

Following the recommendations drawn after the Commissioning Phase of the ENVISAT satellite in 2002, SCIAMACHY near real time data processors were upgraded to version 5.01 in early 2004. Before public release of the new SCIAMACHY nitrogen dioxide (NO_2) vertical column data product, several validation teams investigated its improvement and assessed its geophysical consistency by means of correlative studies involving NDSC-affiliated ground-based networks of DOAS UV-visible and FTIR spectrometers and the ERS-2 GOME satellite. In parallel, preliminary SCIAMACHY NO_2 column data products generated by research processors under development at scientific institutes were also tested, using the same correlative data and validation procedures. Digesting the results obtained by a list of validation teams and SCIAMACHY data processing teams, this overview paper draws a preliminary quality assessment of the SCIAMACHY NO_2 column data sets available in spring 2004.

1. INTRODUCTION

On March 1, 2002, the third Earth observation satellite platform of ESA, ENVISAT, was launched onto a heliosynchronous polar orbit crossing the Equator at 10:00 mean solar local time (descending node). As part of its atmospheric chemistry payload, the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) is a joint project of Germany, The Netherlands and Belgium, aiming at the global, continuous measurement of various trace gases in the troposphere and the stratosphere [1]. Among other geophysical data products, the vertical column of 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 through 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. About ten AO projects include the geophysical validation of SCIAMACHY NO_2 column data via correlative studies involving ground-based and satellite measurements.

The present report summarises the outcome of such correlative studies performed one and half years after the Commissioning Phase (CP) of the satellite, consecutive to major upgrades of the operational SCIAMACHY near real-time data processor operated by ESA, and the development of research data processors at scientific institutes. Results were exchanged and discussed among the SCIAMACHY validation community during two meetings: (i) the SCIAMACHY Validation Workshop organised by SCIAVALIG on April 5-6, 2004, at KNMI in De Bilt

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(The Netherlands); and (ii) the second workshop on the Atmospheric Chemistry Validation of ENVISAT (ACVE-2) held at ESA/ESRIN (Frascati, Italy) on May 3-7, 2004. 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), 331 (J.P. Burrows), 427 (Yu.M. Timofeyev), 429 (E. Kyrö), 651 (M. Weber), and 1103 (A. Petritoli). Section 2 describes the correlative database collected during and after CP and used in the reported studies. Section 3 lists the SCIAMACHY data sets delivered to validation scientists for the reported studies, up to March 2004. Section 4 addresses major issues of the validation of NO_2 column data. Section 5 proposes a digest of the comparison results related to the operational data processor and obtained by the different partners involved in the validation campaign. More details and individual contributions are reported elsewhere in this issue. Section 6 shows a first validation of the NO_2 data generated by four different scientific institutes with a preliminary version of their own retrieval algorithm. Section 7 concludes with a tentative quality assessment of the currently available SCIAMACHY NO_2 column data products and with perspectives for the main validation of SCIAMACHY.

2. CORRELATIVE DATA SETS

2.1 Ground-based UV-visible Measurements

Observations of total NO_2 at sunrise and sunset have been collected from a network of nearly 30 UV-visible spectrometers measuring routinely the sunlight scattered from the zenith sky during twilight ([4-7] and references therein). Figure 1 shows their geolocation. The retrieval method, referred to as Differential Optical Absorption Spectroscopy (DOAS), consists of studying the narrow absorption features of NO_2 in the visible part of the spectrum, after removal of the broadband signal associated with Rayleigh and Mie scattering processes. A differential optical thickness is calculated as the logarithm of the ratio between the zenith-sky spectrum and a reference recorded at higher solar elevation. Column densities along the optical path, or apparent slant columns, are derived by an iterative least squares procedure, fitting the observed differential optical thickness with high-pass filtered absorption cross-sections measured in the laboratory and convolved with the instrument slit function. Interfering species like O_3, O_4 and H_2O are fitted simultaneously. Slant columns are converted into vertical columns using an optical path enhancement factor, or so-called air mass factor (AMF). The AMF is calculated with a radiative transfer model assuming vertical distributions of the atmospheric

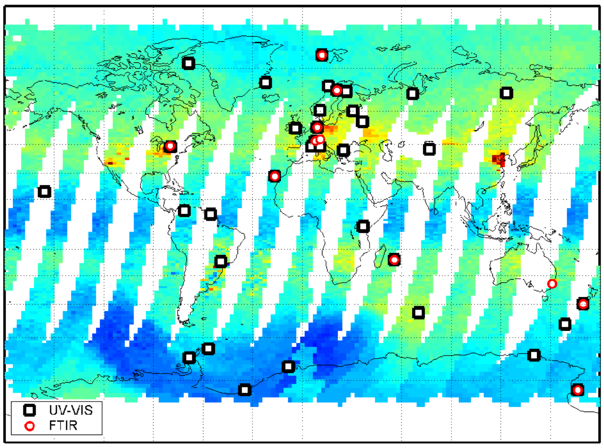

Figure 1 - Contributing ground-based UV-visible and FTIR spectrometers highlighted on top of the NO_2 column field observed by ERS-2 GOME (GDP 3.0) on September 23, 2002, at the beginning of the unexpected split of the Antarctic wintertime polar vortex. Colour scale: from 0.5 10^{15} (dark blue) to 7 10^{15} (dark red) $molec.cm^{-2}$.

parameters controlling the path and the absorption of the solar radiation through the atmosphere, namely, pressure, temperature, NO_2, ozone, aerosols and clouds. The zenith-sky optical path enhancement at twilight makes it sensitive mostly to the stratospheric NO_2 column.

Most of the contributing instruments are certified for the international Network for the Detection of Stratospheric Change (NDSC) [8], a major contributor to WMO's Global Atmospheric Watch (GAW) working under the auspices of United Nations Environment Programme (UNEP). They have to participate to field intercomparison campaigns, during which their agreement on the slant column amount generally falls within the 5% to 10% range [6,7]. Long-term comparisons of nearly co-located slant column measurements at middle latitudes conclude to a mean agreement of 3% in summer and 9% in winter [9]. Despite this good relative agreement, several uncertainties limit the absolute accuracy of the NO_2 slant column and of the AMF, hence, the accuracy of the NO_2 vertical column retrieved by standard procedures adopted within the NDSC: errors of 0-20% associated with the NO_2 absorption cross-sections and their temperature dependence [9,10], errors of ±2-7% associated with rotational Raman scattering [9], uncertainties of ±0-10% associated with seasonal and meridian variations of the NO_2 profile shape [11], bias of up to 4% associated with multiple scattering by aerosols [12], and various effects (scattering, transport and chemical processes) linked to the presence of clouds [13]. The largest uncertainty concerns single measurements corrupted by strong pollution episodes, especially when multiple scattering within clouds, haze or snow showers enhances the optical path. Other sources of errors do not contribute to more than 1% to the error budget [9,12].

2.2 Ground-based FTIR Measurements

Daytime NO_2 column data have been collected from a network of 9 stations equipped with Fourier Transform infrared (FTIR) spectrometers [8]. Contributing instruments are highlighted in Figure 1. On clear-sky days, FTIR instruments record solar absorption spectra at high resolution, encompassing the NO_2 multiplet at 2914.65 cm^{-1}. The NO_2 slant column is derived by an iterative fitting procedure which models the radiative transfer of the infrared sunlight through the atmosphere, taking into account the Earth curvature and atmospheric refraction. The calculation includes only molecular absorption features in selected narrow spectral microwindows. Scattering and absorption by aerosols and by air that have broadband extinction structures are therefore neglected. Data and algorithms intercomparison exercises indicate that the precision of single NO_2 measurements performed by FTIR spectrometry is in the range 6% to 12% [14]. The accuracy is estimated to be about 10% as well. The spectroscopic characteristics of the NO_2 multiplet (weak lines, interfering with strong methane absorptions), make it difficult to distinguish the contribution of the tropospheric NO_2 column to the measured absorption – therefore the retrieved NO_2 column amount is representative chiefly of the stratospheric column.

2.3 GOME Satellite Data

Observations of total NO_2 have been acquired from space since 1995 by Global Ozone Monitoring Experiment (GOME) on board of ESA's second Earth Remote Sensing satellite ERS-2. Figure 1 illustrates the spatial coverage achieved by GOME after one day of operation thanks to its swath width of 960 km and the polar orbit of the satellite. Three different GOME data sets have been used for the present validation campaign. The first one consists of all GOME data taken within the available SCIAMACHY data set period (July to November 2002) and processed routinely by version 3.0 of the operational GOME Data Processor (GDP) established at DLR (Germany) [15]. The second and third ones correspond to the closest two GOME orbits to the so-called SCIAMACHY verification orbits (orbit numbers 2509 and 2510) acquired on August 23, 2002. The first verification orbit starts in Barent Sea, gets through Eastern Europe, scans the African continent from Libya to Congo, and goes throughout the South Atlantic Ocean to reach Antarctica in Halley Bay. The following orbit starts Northward of Spitsbergen, passes over Western Europe and Western Africa, and goes throughout the South Atlantic Ocean to end in Antarctica in Weddell Sea. GOME orbits coincident with SCIAMACHY verification orbits have been processed with GDP 3.0 and also with the retrieval algorithm developed at IUP/Bremen [16,17], yielding the GOME data set hereafter referred to as GOME_IUP. Both GDP and IUP processors rely on the same DOAS technique as that developed for the ground-based UV-visible instruments. Different settings and retrieval codes lead nevertheless to different data products.

3. SCIAMACHY DATA SETS

3.1 Near Real-Time Processor

During ENVISAT CP in 2002, preliminary NO_2 column data products generated by four versions of the near real-time processor (SCI_NL) were distributed to the validation teams. Unfortunately, each of those data sets sampled a different time period of 2002 with only a limited list of orbits: (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. Nevertheless, this composite, sparse data set was useful to conduct the preliminary quality checks reported in [18], which pointed out major problems and led to the improvement of the SCI_NL processor studied hereafter. After CP, in order to test quickly the improvement of the data products after any future SCIAMACHY processor change, a validation reference data set of 3026 SCIAMACHY states – the so-called Master Set (MS) – was selected carefully to provide sufficient and suitable coincidences with the ground-based measurements collected during CP, as well as suitable sampling of the period from July 2002 to mid 2003.

In 2003, NO_2 column data were produced with the successive versions 4.0 and 4.01 of the near real-time processor.

In early 2004, the SCI_NL processor was upgraded to the newly operational version 5.01. Unfortunately, only 1927 MS states were processed and delivered to the validation teams in March 2004. Limited to the period from July to November 2002, this subset is by far not sufficient for a complete validation of all major geophysical features, but it should at least yield indication of the improvement with respect to the previous SCI_NL processor versions. Results are presented in Section 5.

3.2 Off-Line Processor

At the time of this report, too few orbits have been processed with the off-line data processor (SCI_OL) being developed at DLR-Oberpfaffenhofen (Germany). Moreover, those orbits have been selected for verification of the level-1-to-2 retrieval processing chain rather than for validation. Therefore no SCI_OL results have been obtained yet.

3.3 Research Retrieval Algorithms

The development of operational retrieval algorithms relies greatly on the findings of independent retrieval algorithms developed for research purposes by scientific institutes. Untied to the constraints of operational processing, those research algorithms usually are more flexible and they allow detailed investigation of specific retrieval issues. Several institutes have adapted to SCIAMACHY the research algorithms they had developed in the past years for the derivation of NO_2 column data from GOME spectra, based on the same DOAS techniques as that developed for ground-based UV-visible instruments. Ground-based comparisons of NO_2 vertical column products generated by four different institutes will be presented in Section 6 of this overview paper. The description of the different algorithms presented here follows the same order by which they will appear later in the discussion of the comparison results.

3.3.1 IUP

The GOME retrieval algorithm developed by the Environmental Physics Institute of the Bremen University, Germany (hereafter IUP), is described in [16,17]. Further details specific to the adaptation to SCIAMACHY are given elsewhere in this issue [19]. IUP retrieval uses as radiances all level-0 data and "raw" level-1 data available. Dark current is accounted for in level-0 data using "orbital darks" and in level-1 data through "lv1c2pc". The wavelength calibration combines pre-flight settings with calibration based on Fraunhofer atlas. No other calibrations have been applied. The background spectrum used in the slant column fitting is the ASM diffuser spectrum recorded on December 15, 2002. Slant columns are fitted in the 425-450 nm spectral window using the NO_2 absorption cross-sections measured at 243 K by Bogumil *et al.*, (1999). Interfering species are fitted simultaneously using the following absorption cross-sections: Bogumil *et al.* at 243 K for O_3, Greenblat et al. (1990) for O_4, and HITRAN 2000 for H_2O. The Ring effect is accounted for using calculations with SCIATRAN (Vountas *et al.*, 1998). An offset of 2.10^{15} molec.cm^{-2} has been added to all slant columns, determined by comparison with IUP ground-based measurements. Research is ongoing to understand the origin of this offset. Conversion of slant to vertical column amount involves the use of stratospheric AMF calculated by SCIATRAN (full multiple scattering) with the following settings: US Standard profiles of the AFGL reference atmosphere, but without tropospheric content; background aerosol settings from LOWTRAN; and a constant surface albedo of 5%. No cloud treatment is applied in this preliminary version.

3.3.2 SAO

The GOME retrieval algorithm developed by Smithsonian Astrophysical Observatory, Cambridge, Massachusetts (hereafter SAO), is described in [20]. The SCIAMACHY NO_2 column data set generated for this preliminary study is based on level-1 data associated with the 1927 MS states produced with version 5.01 of the operational processors and delivered in Spring 2004. Slant columns are fitted in the 428-452 nm spectral window using the NO_2 absorption cross-sections measured at 294 K by Vandaele *et al.* [10]. Vertical columns are inferred from fitted slant columns using a geometric AMF. No cloud treatment is applied in this preliminary version.

3.3.3 BIRA-IASB

The GOME retrieval algorithm developed by Belgian Institute for Space Aeronomy, Brussels, Belgium (hereafter BIRA-IASB), is described in [21]. Three NO_2 column data sets have been generated, based on three different level-1 data sets: (1) the same raw spectra (hereafter “raw L1”) as used by IUP retrieval; (2) Master Set states level-1 data generated with previous version 4.02 of the operational near real-time processor; and (3) the same Master Set states level-1 data generated with current version 5.01 of the operational near real-time processor, as used also by SAO retrieval. Slant columns are fitted in the 426.5-451.5 nm spectral window (channel 3, cluster 15) using the NO_2 absorption cross-sections measured at 243 K by Bogumil *et al.* (1999). The following cross-sections are also included in the fit: O_3 at 223 K by Bogumil *et al.*, O_4 by Greenblat *et al.* (1990), H_2O from HITRAN 2000, and calculated Ring spectrum with SCIATRAN (Vountas *et al.*, 1998). A radiance spectrum taken over the Indian Ocean, where tropospheric NO_2 is supposed to be almost zero, is used as background spectrum. It is updated every month. A mean slant column value is calculated afterwards for each day in this region and subtracted to all NO_2 slant columns. An offset correction is then applied, corresponding to the stratospheric NO_2 vertical column in this region ($1.5.10^{15}$ molec.cm^{-2}) [22]. AMFs are calculated in pure nadir mode with PS-DISORT [23] using the US Standard NO_2 profile of AFGL Reference Atmosphere [24], background aerosol settings from LOWTRAN, and a constant surface albedo of 5%. No cloud treatment is applied in this preliminary version.

3.3.4 KNMI

The NO_2 column product developed at Royal Netherlands Meteorological Institute, De Bilt (hereafter KNMI), is based on the slant columns generated at

BIRA-IASB from raw L1 data and described in previous Subsection 3.3.3. To account for the temperature dependence of the NO_2 absorption cross-sections, a temperature correction is calculated weighting ECMWF temperature analyses with TM modelling results of the NO_2 vertical distribution. Height-dependent AMF lookup tables are based on calculations with Doubling-Adding KNMI (DAK) radiative transfer model. The NO_2 stratospheric column is deduced from a TM assimilation run of the SCIAMACHY NO_2 slant column data. The assimilated-analysed stratospheric slant column is then subtracted from the retrieved DOAS total slant column provided by BIRA-IASB, resulting in a tropospheric slant column. Then the tropospheric vertical column is retrieved using TM tropospheric model profiles (co-located for each SCIAMACHY pixel individually) and combined with cloud information. The latter consists of cloud fraction and cloud top height derived by the FRESCO algorithm developed by Koelemeijer et al. [25]. The retrieval includes surface albedo values constructed based on a combination of the TOMS-Herman-Celarier-1997 and Koelemeijer-2003 surface reflectivity maps (on a monthly basis). No aerosol correction is applied. This choice is based on the realisation that the cloud retrieval will be influenced by aerosol as well, and is further motivated by the error analysis presented in [26]. The final NO_2 column data product is available publicly on the TEMIS project website [27] with detailed error estimates and kernel information (in HDF format) [28].

4. COMPARISON METHODOLOGY

Three major difficulties hamper the direct comparison of SCIAMACHY NO_2 columns measured at nadir with the aforementioned correlative data: the diurnal cycle of the NO_x family, the natural variability of NO_2, and the sensitivity of nadir observation to tropospheric NO_2. For meaningful comparisons, a methodology must be defined that addresses those issues adequately.

4.1 Diurnal Cycle

The first source of variability of atmospheric NO_2 is the strong diurnal cycle of the NO_x family (NO_x = NO + NO_2 + NO_3 +2 N_2O_5), which consists in a day/night alternance of the NO_2 column. Controlled by the diurnal cycle of solar illumination and by the vertical distribution of NO_x species, of temperature and sometimes of other active species, the diurnal cycle of stratospheric NO_2 exhibits marked seasonal variations and meridian structures. The standard regime at low and middle latitudes is 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. During polar summer, the permanent illumination of the stratosphere does not allow the nighttime formation of N_2O_5. Nevertheless, the variation in solar illumination between noon and midnight is sufficient to modify significantly the photochemical equilibrium between NO and NO_2. As a consequence, the stratospheric column of NO_2 alternates smoothly between a minimum around noon and a maximum under midnight sun conditions, with an amplitude of 0.5 10^{15} to 1.5 10^{15} molecule.cm^{-2}. During polar springtime, strong and/or sudden changes of temperature and of NO_y partitioning (e.g. resulting from denoxification or denitrification processes) can alter unpredictably the amplitude and the shape of the diurnal cycle. The diurnal cycle of tropospheric NO_2 is even less predictable as it depends on as many parameters as the vicinity of emission sources of photochemically and radiatively active species, local transport, surface albedo, the presence of clouds etc.

During slow parts of the diurnal cycle, direct comparison of FTIR or GOME correlative data with SCIAMACHY NO_2 is feasible provided that correlative data are selected within a couple of hours around the SCIAMACHY acquisition time. Comparison near the twilight transients requires more care. In particular, the diurnal cycle hampers the direct comparison of SCIAMACHY data acquired in the mid-morning with ground-based zenith-sky data acquired at dawn and at dusk. Modelling studies carried out at IASB in collaboration with U. Leeds [29,30] indicate that sunrise values might be reasonably close – within a few 10^{14} molecule.cm^{-2} – to the mid-morning values acquired by SCIAMACHY. For the particular polar day regime, modelling studies have led to a photochemical adjustment factor calculated as a function of the SCIAMACHY solar zenith angle (SZA), and enabling quantitative comparisons. The polar spring variability can increase the scatter of direct comparisons and sometimes introduce a bias of a few 10^{14} molecule.cm^{-2} over a period of a few days to several weeks.

4.2 Variability and Gradients

Once the diurnal cycle is taken into account properly, the variability of atmospheric NO_2 is characterised by the superposition of low and high frequency phenomena. Low frequency variability of stratospheric NO_2 consists mainly of meridian gradients modulated by low frequency cycles (e.g., seasonal variation at middle and high latitudes, and semi-annual oscillation and quasi-biennial oscillation at low latitudes). At high latitudes, dynamical and photochemical processes coupled to the polar vortex play also an important role in the medium-range control of wintertime stratospheric NO_2. Low frequency variability of tropospheric NO_2 results from the coupling of emission, transport and photochemical processes. It combines local patterns and regional background modulated by a seasonal cycle. High frequency variability of stratospheric NO_2 is associated mainly with phenomena of dynamical origin. It can be large near the edge of the polar vortex but also very weak in the Tropics. Tropospheric NO_2 can exhibit large short-term fluctuations and small-scale gradients resulting from the complex coupling of many dynamical and photochemical processes.

All the contributing validation teams have adopted their own space/time coincidence criteria to reduce the effects of variability on the comparison. Usually, comparison of SCIAMACHY with FTIR data involves simple criteria based on the distance: SCIAMACHY data are selected within a radius of 200 km to 500 km around the ground-based station. The maximum distance can be tuned as the best compromise between 1s scatter and statistical significance. For the comparison with DOAS UV-visible data, two types of criteria have been used: simple distance criteria, and physically based methods using a ray tracing model to identify the actual air mass probed by the zenith-sky twilight observation. In areas with low variability like the Tropics and the denoxified inner polar vortex, the two types of selection yield comparable results. When variability increases, physically based methods do not alter the mean difference but can reduce the 1s scatter significantly. 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

4.3 Sensitivity to Tropospheric NO_2

In principle, to enhance the detection of subtle stratospheric features, NDSC stations are located in remote areas far away from pollution sources. Practically, GOME validation results teach us that ground-based stations can be classified according to the effect of tropospheric NO_2 on the comparisons.

A first class of stations allows direct quantitative comparisons. It includes extremely isolated sites experiencing nearly no tropospheric NO_2: e.g., all Antarctic stations, Arctic stations in Greenland and Spitsbergen, all existing Southern mid-latitude sites (Kerguelen Islands, New Zealand and Macquarie Islands), and Issyk-Kul in the high isolated mountains of Kyrgyzstan. This first class includes also other remote sites offering similarly low levels of tropospheric NO_2 but experiencing occasionally pollution peaks due to long-range transport from the emission sources: this is the case of several high latitude sites in Scandinavia and of the tropical station of Saint-Denis on Reunion Island. Knowing that NDSC instruments are mostly sensitive to the stratospheric part of the NO_2 total column measured by SCIAMACHY, quantitative validation at those sites is feasible and can rely on straightforward comparisons provided that pollution peaks are filtered out.

A second class of stations consists of sites where tropospheric NO_2 becomes an important part of the vertical column. Some of those sites are located directly in the vicinity of emission sources and face their influence permanently, like Bauru in Brazil, Bremen in Germany and Toronto in Ontario. Other sites located in a relatively clean environment such as the top of high Alpine mountains, are nevertheless under the influence of neighbouring regions with urban and industrial emissions. In principle, the discrepancy between satellite total columns and NDSC stratospheric columns should be a measure of the tropospheric column seen by the satellite. This principle forms the basis of the successful residual technique developed to derive tropospheric column values from total column measurements. Indeed, for this second class of stations, comparisons between GOME and NDSC NO_2 data conclude to the presence of a permanent tropospheric background and to enhanced scatter. A study of the seasonal variation of the background and the correlation of pollution peaks should be good qualitative indicators of the geophysical consistency of SCIAMACHY data. Unfortunately, the validation of SCIAMACHY at present time is limited to such qualitative investigation due to the obvious lack of accurate correlative measurements of the NO_2 tropospheric column. Quantitative validation using NDSC instrumentation can be envisaged in the future as a combination of complementary techniques yielding nearly simultaneously the stratospheric and tropospheric components of the total column. Such an approach requires further developments and generalisation of the multiple axes DOAS technique (MAX-DOAS) and of NO_2 profiling from zenith-sky DOAS measurements, as well as synergy with the existing zenith-sky DOAS technique which provides the reference for the validation of the stratospheric component.

5. NEAR REAL TIME PROCESSOR

5.1 Quantitative Validation of Stratospheric NO_2

Comparison results obtained at NDSC stations belonging to the first class (clean stratospheric sites) are illustrated in parts (a-f) of Figure 2. At middle and high latitude stations of the Southern Hemisphere, the mean absolute difference (b,d) and the 1σ standard deviation

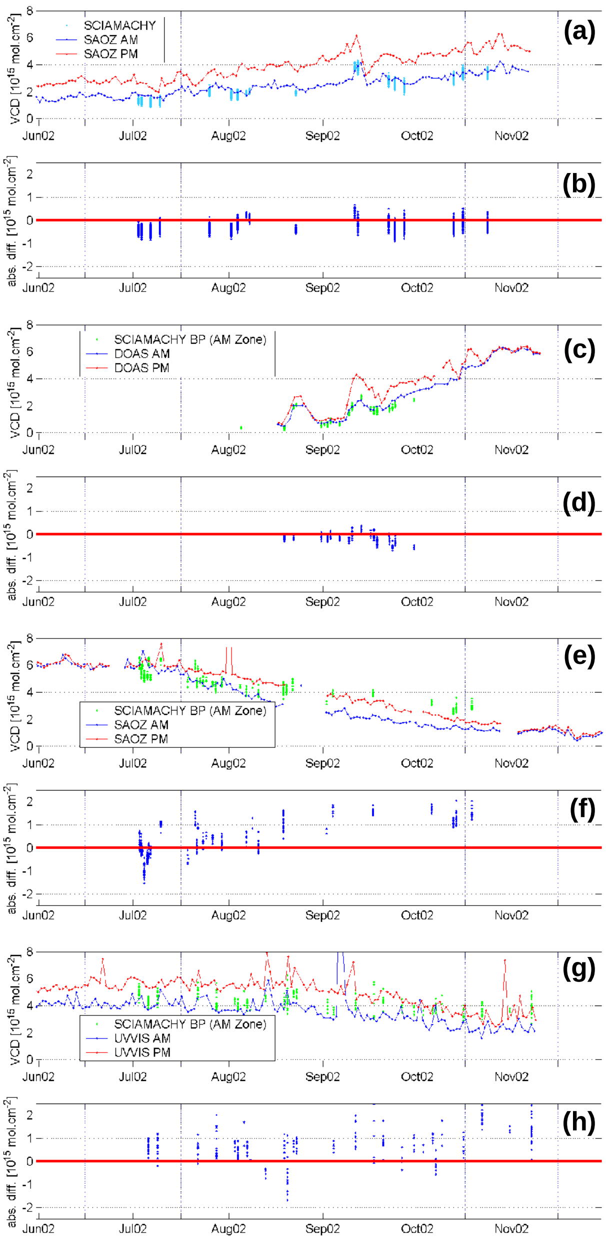

Figure 2 –Vertical column (a,c,e,g) of NO_2 from June to December 2002 as reported by SCIAMACHY v5.01 (mid-morning value) and by NDSC UV-visible spectrometers (sunrise and sunset values), and the absolute difference (b,d,f,h) SCIAMACHY – NDSC sunrise values at the following stations: (a-b) Kerguelen Island, 49°S, 70°E; (c-d) Neumayer, 71°S, 8°W; (e-f) Zhigansk, 67°N, 123°E; and (g-h) Monte Cimone, 44°N, 11°E.

between SCIAMACHY SCI_NL 5.01 and NDSC/UV-visible data do not exceed a few 10^{14} molec.cm^{-2}. SCIAMACHY reproduces the same seasonal variation and shorter-term fluctuations as observed from the ground. The comparison with the UV-visible measurements acquired at the Antarctic station of Neumayer (71°S) by IUP/Heidelberg (c-d), illustrates

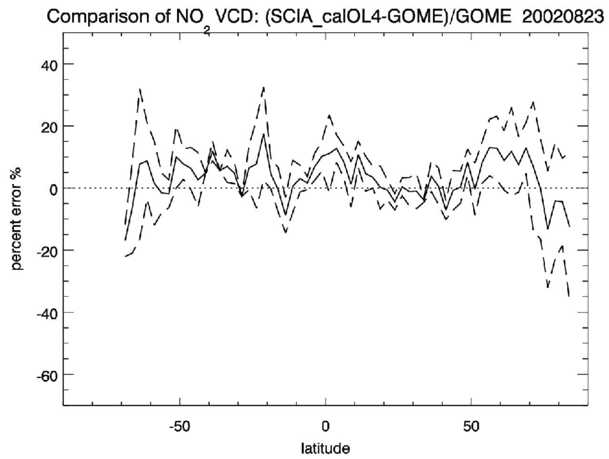

Figure 3 – Percent relative difference in total NO_2 between SCIAMACHY SCI_NL 5.01 and GOME GDP 3.0 along 2 orbits on August 23, 2002, plotted as a function of latitude.

how well SCIAMACHY was able to capture the unexpected split of the Antarctic polar vortex during late September and early October 2002. Unusual patterns in the atmospheric circulation surrounding the Antarctic polar vortex distorted it and made it split, giving an exceptional morphology to the ozone hole and to the associated area where low levels of NO_2 are usually observed during this season (see e.g. Figure 1 showing the morphology on September 23). This event raised stratospheric temperatures sufficiently high to hamper the formation of polar stratospheric clouds wherein NO_2 is converted into reservoirs by heterogeneous processes, decreasing also the level of chlorine activation. The excellent agreement observed at Neumayer is also obtained with correlative observations acquired with the BAS SAOZ instrument at Rothera (68°S) in the Antarctic Peninsula, the CNRS SAOZ instrument at Dumont d'Urville (67°S) in Terre Adélie, the NIWA DOAS spectrometer at Arrival Heights (78°S), and the INTA DOAS spectrometers at Marambio (64°S) and Belgrano (78°S). 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 [31].

While the agreement in Antarctic springtime looks excellent, an unrealistically smooth seasonal variation is reported at several stations in the Arctic. This is illustrated in parts (e-f) of Figure 2 where SCI_NL 5.01 data are confronted to ground-based CNRS/CAO SAOZ data at the East Siberian station of Zhigansk (67°N). The good mean agreement of a few 10^{14} molec.cm^{-2} in summer degrades in September when SCIAMACHY starts reporting higher NO_2 values by 10^{15} to 1.5 10^{15} molec.cm^{-2}. Such an overestimation is also depicted in Figure 8 where SCI_NL 5.01 NO_2 is compared to ground-based FTIR data acquired by IMK/FZK at the Swedish station of Kiruna (68°N). It is interesting to note that SCIAMACHY data retrieved with the aforementioned IUP retrieval algorithm (SCI_IUP), also depicted in Figure 8, offer a much better agreement and do not show this overestimation.

5.2 Effect of Tropospheric Pollution

While a good agreement is reported at stratospheric sites belonging to the first class, a notable discrepancy appears, as expected, over stations belonging to the second class where tropospheric NO_2 becomes important. As discussed in Section 4, this discrepancy is an indication of the difference in sensitivity to the troposphere. The total column detected by SCIAMACHY contains a tropospheric component, which superimposes on the stratospheric column measured by NDSC instruments. At such stations, SCIAMACHY is found to report NO_2 values larger on an average by 0.5 10^{15} to 2.5 10^{15} molec.cm^{-2}. The confrontation of SCIAMACHY with GOME (for the two verification orbits in August 2002), which is supposed to offer the same sensitivity to the troposphere, does not show such large discrepancies: as depicted in Figure 3, the agreement over mid-latitude Europe falls to within the –5% to +10% range.

At Northern mid-latitude stations, the discrepancy between SCI_NL and NDSC data often exhibits a seasonal increase from July to November 2002. Parts (g,h) of Figure 2 show a typical comparison with the UV-visible data acquired by ISAC/CNR at Monte Cimone (44°N) in the Italian Alps. The discrepancy (Figure 2-h) starts at 0.5 10^{15} molec.cm^{-2} in summer and increases to an average value of 1.5 10^{15} molec.cm^{-2} in November. This mean behaviour is consistent with the known seasonal variation of the NO_2 tropospheric content, oscillating between a summertime minimum and a wintertime maximum. But it could also arise partly from a seasonally varying bias just like that observed in the Arctic. Unfortunately, the MS data set available is too scarce to study the correlation of strong pollution episodes with sufficient statistical significance. It is timely to stress once again that no quantitative validation of SCIAMACHY tropospheric NO_2 has been reported so far due to the lack of correlative measurements of the NO_2 tropospheric column.

5.3 Meridian Structure

According to previous subsections, the SCIAMACHY vs. NDSC comparison time series over the period from July to November 2002 at clean sites – that is, where the NO_2 column is essentially of stratospheric origin – do not show any significant dependence on the season. No dependence on the SZA or the column has been detected, neither. The agreement at a single station may be described by mean and standard deviation values. Statistical results for all individual stations may be

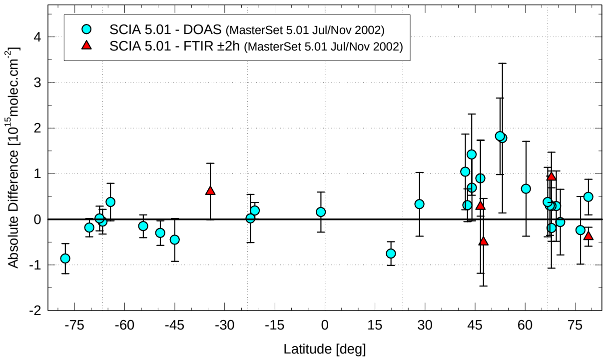

Figure 4 - Meridian variation of the mean absolute difference between near real time total nitrogen dioxide in July/November 2002 as reported by SCIAMACHY SCI_NL 5.01 and by NDSC ground-based spectrometers (SCIA-NDSC).

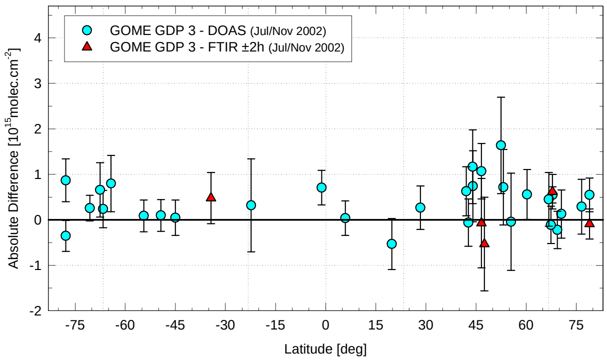

Figure 5 - Meridian variation of the mean absolute difference between total nitrogen dioxide in July/November 2002 as reported by ERS-2 GOME GDP 3.0 and by NDSC ground-based spectrometers.

plotted altogether as a function of the latitude, as in Figure 4. Although not meaningful statistically since corrupted by the seasonal variation of the SCIAMACHY/NDSC agreement, mean and standard deviation values at polluted sites are also plotted for the needs of the discussion in Subsection 5.4. Figure 4 confirms that, for pure stratospheric sites, no meridian bias is found between SCI_NL 5.01 and NDSC NO_2 data. Knowing that no meridian dependence of the GOME vs. NDSC agreement had been reported by detailed validation studies carried out over multi-year data records [Lambert et al. 2002], we have here replaced SCIAMACHY SCI_NL 5.01 total NO_2 by GOME GDP 3.0 total NO_2, and repeated the same exercise, leading to the results presented in Figure 5. To ensure that no compensating errors varying with the season cancel when studying a complete year of data, we have used GOME data only from the same period from July to November 2002 as already explained in Subsection 2.3. Figure 5 does not make any meridian structure appear. The absence of significant meridian bias with respect to NDSC data is also confirmed at least up to 70° of latitude by comparisons between SCIAMACHY and GOME data associated with the two SCIAMACHY verification orbits acquired on August 23, 2002. The two sets of orbits have been processed with the current versions of the operational processing algorithms, SCIAMACHY SCI_NL 5.01 and GOME GDP 3.0, respectively. Depicted in Figure 3 as a function of latitude, the mean relative difference between SCIAMACHY and GOME total NO_2 for those two orbits does not show any striking meridian structure up to 70° of latitude: it falls to within -10% and +15%, with a RMS ranging from 5% to 10%. A peak in the RMS is observed around 20°S, reflecting the influence of the South Atlantic Anomaly (SAA). Beyond 70° of latitude, in both the Arctic and the Antarctic, SCIAMACHY reports systematically lower values by about 10% and the RMS scatter increases up to 20%.

5.4 Effect of Processor Changes Since v3.53

CP validation studies performed in 2002 concentrated on the quality assessment of developmental versions 3.5x of the SCI_NL processor [18]. Figure 6 reminds the pole-to-pole agreement of version 3.53 with NDSC data. Compared to this early version 3.53, the improvement gained with the current version 5.01 is clear: both the negative bias over the Southern Hemisphere and the positive bias at Northern middle latitudes have disappeared in Figure 4. Figure 7 illustrates the effect of the successive processor changes – from v3.53 to v5.01 – at the station of Izaña (Tenerife, 28°N, 16°W). This station, where UV-visible and FTIR spectrometers are operated by INTA and FZK/IMK, respectively, was the only one where the agreement between SCI_NL 3.53 and the ground-based data was acceptable. Versions 4.00 and 4.01 generate systematic biases of 0.5 10^{15} and -1 10^{15} molec.cm^{-2}, respectively. Version 5.01 yields an agreement comparable to that obtained with version 3.53.

6. RESEARCH PROCESSORS

6.1 IUP

Several validation teams have compared to NDSC data the SCIAMACHY NO_2 data set generated by the research processor developed at IUP. All groups report a good agreement over clean sites at middle and high latitudes. Figure 8 illustrates the SCIAMACHY vs. FTIR comparison at the Arctic station of Kiruna. At this station, SCI_IUP and FZK/IMK FTIR NO_2 data are in close quantitative agreement, even in fall 2002 when SCI_NL 5.01 is found to report too high values. The seasonal cycle and day-to-day fluctuations are also captured similarly. A look at the meridian agreement depicted in Figure 9 indicates that SCI_IUP overestimates ground-based values at low latitudes by

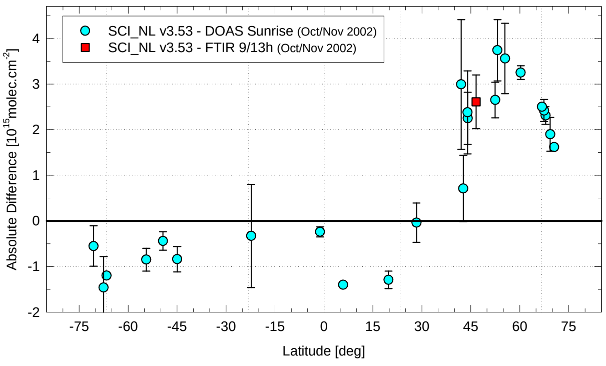

Figure 6 – Same as Figure 4, but with SCIAMACHY generated with near real time processor v3.53.

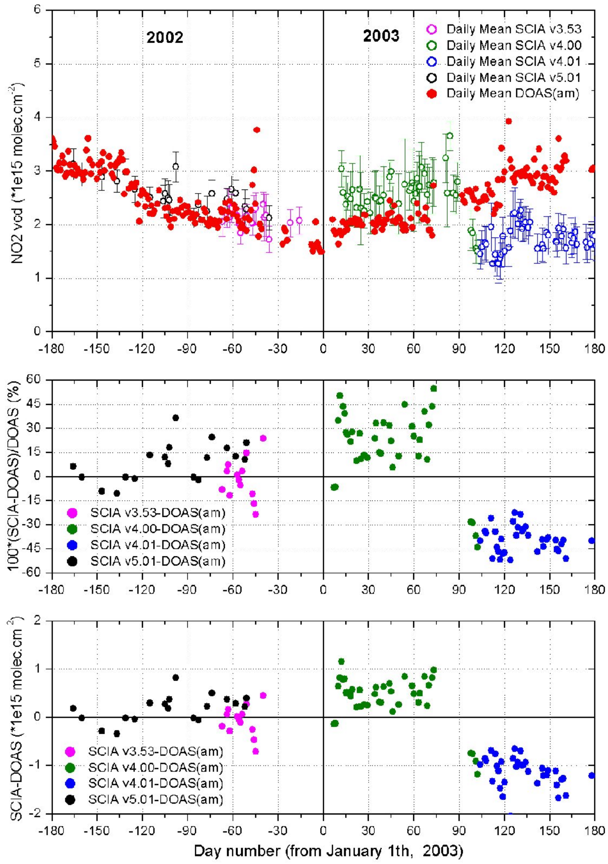

Figure 7 – Agreement between successive versions (3.53, 4.00, 4.01 and 5.01) of the SCIAMACHY near real time NO_2 column and the sunrise ground-based data measured at Izaña (Tenerife, 28°N) with the INTA UV-visible spectrometer: time series of NO_2 vertical column (top), percent relative difference (middle), and absolute difference (bottom).

up to 8 10^{14} molec.cm^{-2}. Such a meridian feature seems also to appear in Figure 10 when comparing SCI_IUP and GOME_IUP data for the two verification orbits, but no firm conclusion may be drawn from only two orbits.

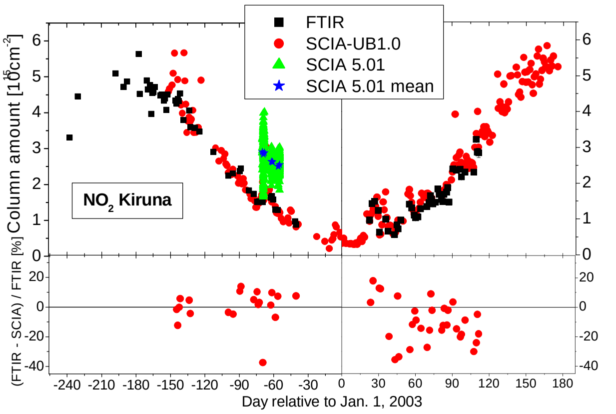

Figure 8 – NO_2 column time-series from spring 2002 to spring 2003 at the Arctic station of Kiruna, as measured by the ground-based FTIR spectrometer operated at the station by IMK/FZK, and as derived from SCIAMACHY data with the IUP algorithm and with version 5.01 of the near real time data processor. FTIR data are selected within 2 hours around the SCIAMACHY overpass.

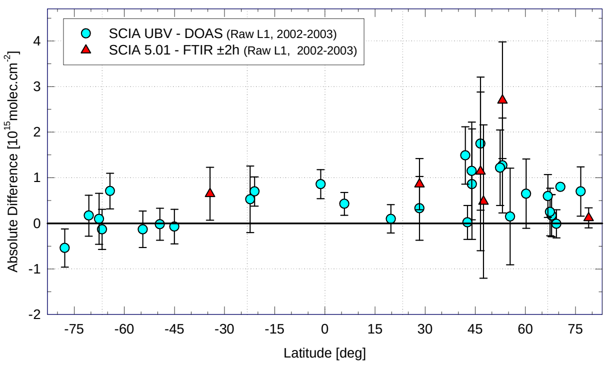

Figure 9 – Same as Figure 4 and Figure 12-c, but with SCIAMACHY NO_2 columns retrieved from raw level-1 data by the research processor developed at IUP, for 2002-2003.

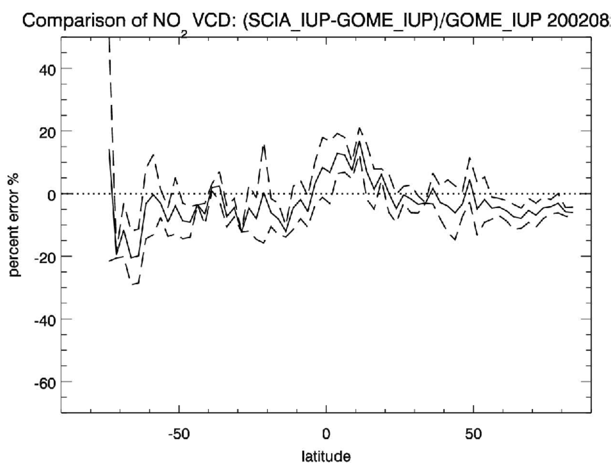

Figure 10 – Percent relative difference between SCIAMACHY and GOME total NO_2 processed by IUP, along 2 orbits on August 23, 2002, plotted as a function of latitude.

The mean relative difference between SCI_IUP and GOME_IUP NO_2 data ranges from –20% to +15%, with a RMS of 2% to 15%. The quantitative comparison between SCI_NL 5.01 and GOME GDP 3.0 data displayed in Figure 3 had reached fairly similar values, but with no marked meridian structure. This suggests that the origin of the meridian distortion would not arise from the IUP processing itself, but from the application of IUP processing to the raw level-1 data used to generate this preliminary SCI_IUP data set. Research is ongoing to solve out this issue. More details are reported in this issue [19].

6.2 SAO

The pole-to-pole comparison between NDSC NO_2 data and the SCIAMACHY data set generated by the retrieval algorithm developed at SAO (SCI_SAO) is depicted in Figure 11. Over stratospheric stations, SCI_SAO reports systematically lower values than the ground-based instruments. This underestimation exhibits a meridian structure: absolute differences start around 0.5 10^{15} molec.cm^{-2} at the polar circles and increase towards the low latitudes. Despite this negative bias, the geophysical content of the SCI_SAO NO_2 data set looks consistent: e.g., seasonal variations are similar to those measured from the ground. Standard deviation values and the absolute difference between clean and polluted are also comparable to those obtained with the other SCIAMACHY data processors. In the preliminary version of the retrieval algorithms used to generate the first-shot data set studied here, at least two simplifications might be the cause of the observed general underestimation: the use of absorption cross-sections at 294 K (which is too high compared to real stratospheric temperatures, and higher than the temperature used for the ground-based retrievals) and the use of a purely geometrical AMF. It is anticipated that the agreement with correlative data will improve with current developments of the SAO processor.

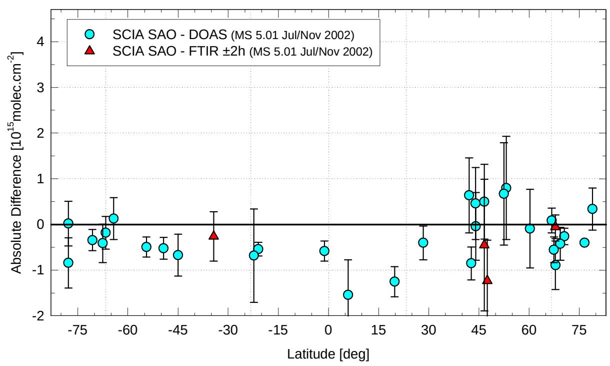

Figure 11 – Same as Figure 4 and Figure 12-a but with SCIAMACHY NO_2 columns processed from MS 5.01 level-1 data by the retrieval algorithm developed at SAO.

6.3 BIRA-IASB

For this exercise, the retrieval algorithms developed at BIRA-IASB have inferred SCIAMACHY NO_2 columns from three different sets of SCIAMACHY level-1 data: (1) the same Master Set v5.01 as used by the near real time processor SCI_NL v5.01; (2) the previous version 4.02 of the Master Set; and (3) raw level-1 data. Except differences justified by the calibration of different level-1 data, other settings have been kept constant. The global agreement with correlative NDSC data is illustrated in Figure 12. Apparently, although the three level-1 data sets yield comparable comparison results, SCIAMACHY NO_2 derived from MS 5.01 level-1 data leads to the best pole-to-pole agreement. For stratospheric sites, the latter ranges from a few 10^{14} to 5 10^{14} molec.cm^{-2}. Larger differences exist at stations where the statistical significance is questionable. No meridian structure is observed.

6.4 KNMI

The pole-to-pole comparison between NDSC NO_2 data and the SCIAMACHY data set generated at KNMI is depicted in Figure 13. The KNMI data set is based on the same level-1 data as used by IUP (Subsection 6.1, Figure 9) and BIRA-IASB (Subsection 6.3, Figure 12). Although using the slant columns retrieved by BIRA-IASB, the temperature correction and the major difference in the conversion to vertical columns produce significantly different comparison results. The main difference is observed at polluted sites of the Northern middle latitudes where the discrepancy between SCIAMACHY and NDSC data reaches now values as high as 3.5 10^{15} molec.cm^{-2}. According to modelling results and (unfortunately non correlated) airborne measurements, such high values would be better estimates of the tropospheric NO_2 column. Such a difference between KNMI retrievals and the others is supposed to come mostly from the use of tropospheric AMF. GOME and SCIAMACHY total NO_2 retrievals using a pure stratospheric AMF are known to underestimate the vertical column in the presence of tropospheric NO_2. Usually limited to 10% to 30%, this underestimation can reach a factor of two under extreme conditions [32]. Another likely source of discrepancy, explaining the difference in behaviour at the stratospheric reference stations of the southern middle latitudes, comes from the use of a correction factor to account for the temperature dependence of the NO_2 absorption cross-sections. DAK RTM-computed AMFs approach geometrical AMF values in the clean Southern mid-latitude case. However, the total AMF used in the retrieval is the product of this quasi-geometrical AMF by the temperature correction based on ECMWF analyses. Studies indicate that stratospheric temperatures are typically about 15K lower than the temperature of the

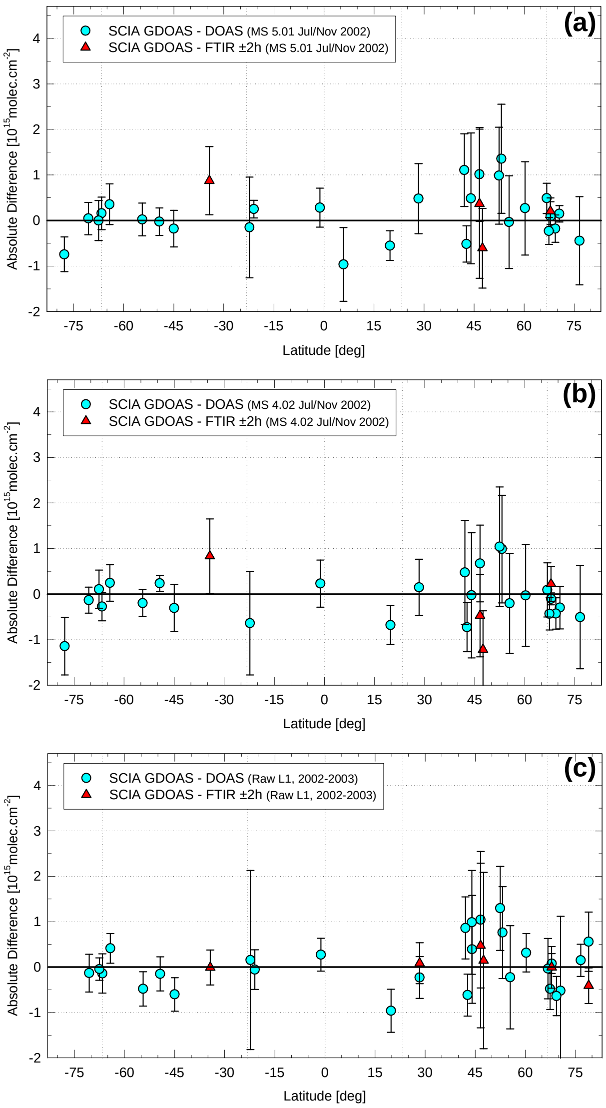

Figure 12 – Same as Figure 4, but with SCIAMACHY NO_2 columns retrieved by the retrieval algorithm developed at BIRA-IASB from: (a) MS 5.01 level-1 data; (b) MS 4.01 level-1 data; and (c) raw level-1 data.

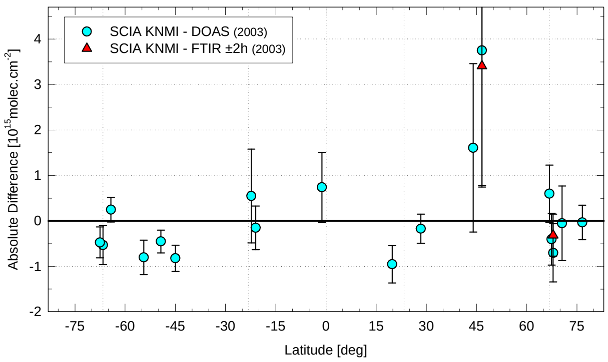

Figure 13 – Same as Figure 4 and Figure 12-c (2003 time-series), but with SCIAMACHY NO_2 slant columns retrieved from raw level-1 data by the research processor developed at BIRA-IASB, converted into vertical columns by KNMI.

cross-sections used in the DOAS fit (243K, SCIA PFM). The typical NO_2 correction applied for such a temperature difference is about -5%, which corresponds to an increase in the reported AMF of 5%. A large part of the observed difference between KNMI and BIRA-IASB total NO_2 could be explained by this temperature effect. Further quantitative interpretation of the results depicted in Figure 13 should take into account how the temperature effect is dealt with in the retrieval of ground-based NDSC data. An important issue that validation should also address is how to distinguish between the information content coming directly from the SCIAMACHY measurement and the contribution of the modelling results used in the retrieval.

7. CONCLUSION AND PERSPECTIVES

In early 2004, several validation teams carried out preliminary comparisons of available SCIAMACHY NO_2 column data with correlative measurements from NDSC-affiliated ground-based networks (mostly stratospheric columns) and from the ERS-2 GOME satellite (total column). The late delivery of ENVISAT operational data products, furthermore limited to an irregular sampling of Northern summer-fall/Southern winter-spring seasons, hampered the output of the planned validation effort. Nevertheless, the amount of available collocations with ground-based data was sufficient to demonstrate that, since the ENVISAT Commissioning Phase in 2002, NO_2 column data products generated by the SCIAMACHY level-1-to-2 data near real-time processor (SCI_NL) have improved. Over Southern Hemisphere stations with low tropospheric NO_2 levels, current version SCI_NL 5.01 provides vertical column data in good qualitative and quantitative agreement with NDSC and GOME observations. A major concern is the systematic bias observed in the Arctic where the good agreement in summertime degrades dramatically in fall, SCIAMACHY reporting too high values by up to 1.5 10^{15} molec.cm^{-2}. Over polluted areas, the difference between SCIAMACHY total columns and NDSC stratospheric columns seems to follow the expected seasonal variation of tropospheric NO_2, but at the time being it is hazardous to conclude whether this behaviour is not partly due to the same overestimation problem as reported in the Arctic. The current SCI_NL 5.01 data set is too limited to verify whether this season-dependent overestimation exists also in the Southern Hemisphere.

SCIAMACHY NO_2 column data sets generated by preliminary versions of the retrieval algorithms developed at BIRA-IASB, IUP, KNMI and SAO, have also been tested against ground-based network data. Despite the premature character of this study, first results are encouraging: all data sets capture major geophysical signals appropriately. As expected, differences in the retrieval settings and in the simplifications adopted for this exercise can produce differences between the data products, like meridian structures and systematic differences. Those differences raise interesting issues to be addressed in future developments of the operational and research processors.

We have already mentioned the limitations of the SCI_NL 5.01 data set delivered to the validation scientists and, consequently, the limitations of our current conclusions about this retrieval algorithm. An additional limitation is that the seven SCIAMACHY NO_2 data sets studied here are based on significantly different time periods, time/space sampling, level-1 data, cross-sections etc. (see Section 3). Although there is an instinctive temptation to rank mutually the retrieval algorithms, it is imperative to realise that the conditions are by far not met to carry out an impartial, rigorous comparative study.

Provided that the operational delivery of ENVISAT gets on and that at least one continuous year of SCIAMACHY data is reprocessed with the current version and with future upgrades of the level-1-to-2 processors, it is anticipated that firm geophysical validation results, that is, consolidated conclusions on the quality and geophysical usability of SCIAMACHY NO_2 stratospheric columns, could be drawn in the second half of 2004. Quantitative validation over polluted areas requires the development of new methods, e.g. combining NDSC-like observations with other geometries more sensitive to the troposphere.

8. ACKNOWLEDGMENTS

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