

VALIDATION OF SCIAMACHY OL 3.0 NO₂ PROFILES AND COLUMNS USING GROUND-BASED DOAS PROFILING

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ABSTRACT

A profiling algorithm based on the Optimal Estimation Method (OEM) has been developed at IASB-BIRA in order to provide low vertical resolution stratospheric NO₂ profiles from ground-based zenith-sky DOAS observations. This algorithm has been applied to observational data from the NDACC (Network for the Detection of Atmospheric Composition Change, formerly NDSC) stations of Harestua (60°N, 11°E) and Reunion Island (21°S, 55°E). The reliability of our NO₂ retrievals has been verified through comparisons with correlative satellite (HALOE and POAM III) and balloon (SAOZ and LPMA/DOAS) observations.

Here, stratospheric NO₂ profiles and columns retrieved at Harestua and Reunion Island are used to validate the SCIAMACHY NO₂ operational products (version OL 3.0). Both limb profiles and nadir columns are included in our validation study. It should be noted that comparisons are performed in the same photochemical conditions owing to the inclusion of a stacked box photochemical model in the forward model of our profiling algorithm.

1. INTRODUCTION

The profiling technique applied to ground-based DOAS (Differential Optical Absorption Spectroscopy) measurements offers new perspectives in the use of ground-based UV-visible networks such as the NDACC (Network for the Detection of Atmospheric Composition Change, formerly NDSC). With this technique, based on the dependence of mean scattering height with solar zenith angle (SZA), not only columns but also low resolution vertical profiles are made available for the purpose of satellite data validation. So far, profiling from ground-based UV-visible observations has been mainly used for the retrieval of stratospheric NO₂ profiles (e.g., [1,2] and references therein). Recently, [3] have also applied this technique to combined zenith-sky and direct sun UV-visible observations in order to infer time-series of tropospheric and stratospheric BrO vertical column densities (VCDs) from the retrieved profiles.

At IASB-BIRA, a profiling algorithm has been developed in order to retrieve stratospheric NO₂ profiles

from ground-based zenith-sky UV-visible observations [2]. In the present study, this algorithm is applied to observational data from the NDACC stations of Harestua (60°N, 11°E) and Reunion Island (21°S, 55°E) and the retrieved profiles and corresponding columns are used to validate the SCIAMACHY NO₂ operational products (limb profiles and nadir columns) version off-line (OL) 3.0.

2. GROUND-BASED UV-VIS OBSERVATIONS

At Harestua, two zenith-sky UV-visible spectrometers have been continuously operated by IASB-BIRA since 1998. The instrument and the data analysis have been validated through several NDACC campaigns (Lauder 1992, OHP 1996, and Andøya 2003). In case of NO₂, the DOAS fitting window used is 425-450 nm and the other settings of the DOAS analysis can be found in [2].

At Reunion Island, ground-based multi-axis (MAX-) DOAS observations have been performed from July 2004 till July 2005. Only zenith data are used here and the DOAS settings for NO₂ are similar to those used at Harestua.

3. IASB-BIRA NO₂ PROFILING ALGORITHM

The IASB-BIRA NO₂ profiling algorithm is described in detail in [2]. It is based on the Rodgers Optimal Estimation method (OEM, [4]) and the forward model consists in the radiative transfer model UVspec/DISORT [5] coupled to the IASB-BIRA stacked box photochemical model PSCBOX. Both models have been validated through intercomparison exercises ([6,7]). The inclusion of a photochemical model in the retrieval algorithm allows to reproduce the effect of the rapid variation of the NO₂ concentration during twilight. It also makes possible profile retrieval at any SZA. An important step in the OEM method is the choice of the a priori profile. Here, the a priori NO₂ profile is the output of the stacked box photochemical model PSCBOX daily initialized with SLIMCAT 3D-CTM chemical and meteorological fields [8]. It should be noted that the reliability of our NO₂ retrievals has been verified through comparisons with correlative satellite (HALOE and POAM III) and balloon (SAOZ and LPMA/DOAS) observations [2].

The averaging kernels (AVKs) matrix A is the key parameter in the characterization of the retrieval. The shape of the AVKs can give a rough estimate of the vertical resolution and the trace of A is the number of degrees of freedom, which gives an estimate of the number of independent pieces of information contained in the measurements. A typical example of ground-based UV-vis NO_2 AVKs, obtained for the Harestua 12/07/2004 sunrise retrieval, is shown in Fig. 1.

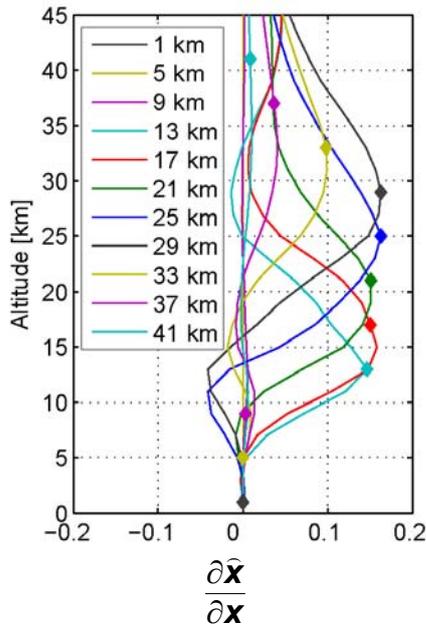


Figure 1. Typical example of ground-based NO_2 averaging kernels. Plain diamonds indicate the altitude at which each averaging kernel should peak in an ideal case.

From the examination of these AVKs, it is found that the vertical resolution is 8-10 km at best and the value of the trace of A is 2.5, so there are about 2.5 independent pieces of information in our ground-based DOAS NO_2 observations.

4. SCIAMACHY LIMB NO_2 PROFILES VALIDATION

4.1. Coincidence criteria

For the comparison at Harestua, the maximum distance between the SCIAMACHY NO_2 profiles and the station has been fixed to 750 km for late spring, summer, and early fall, and to 500 km for winter, early spring, and late fall in order to avoid as much as possible coincident events with large PV difference between SCIAMACHY and ground-based UV-vis measurements. Concerning the temporal criterium, ground-based UV-vis NO_2 profiles have been retrieved at the SZAs corresponding to the SCIAMACHY limb NO_2 profiles using the stacked box photochemical model PSCBOX included in

the profiling algorithm. Photochemical conditions are therefore similar for both SCIAMACHY and ground-based UV-vis profiles. When applying these criteria, 23 coincident events (morning conditions) have been selected for comparison. At Reunion Island, only two coincidences have been found using as spatial criterium a distance of maximum 1000 km between SCIAMACHY NO_2 profiles and the station.

4.2. Results

Figs. 2 and 3 show comparisons between mean SCIAMACHY limb and ground-based UV-vis NO_2 profiles at Harestua and Reunion Island, respectively.

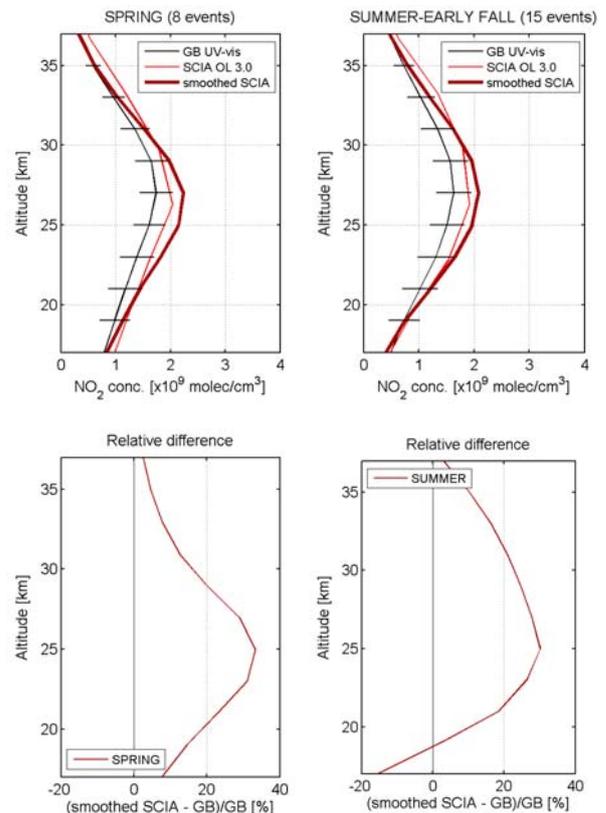


Figure 2. Comparison between mean SCIAMACHY limb and ground-based UV-vis NO_2 profiles at Harestua for spring and summer-early fall conditions. The relative differences appear in the lower plots. The relative differences appear in the lower plots. The error bars in the upper plots represent the sum of the systematic and random errors on the mean ground-based UV-vis profiles.

At Harestua, SCIAMACHY overestimates the ground-based NO_2 profiles, except below 18 km in summer-early fall where an underestimation is obtained. For both spring and summer-early fall conditions, a mean relative difference value of +16 % is found in the 17-37 km altitude range, with a maximum difference value of about 30% at 25 km.

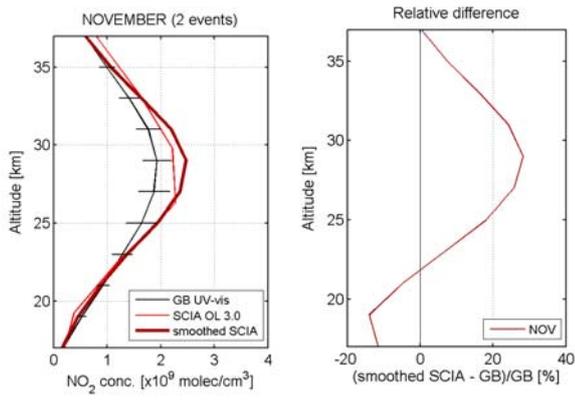


Figure 3. Comparison between mean SCIAMACHY limb and ground-based UV-vis NO₂ profiles at Reunion Island. The error bars in the left plot represent the sum of the systematic and random errors on the mean ground-based UV-vis profiles.

At Reunion Island, SCIAMACHY also overestimates the ground-based UV-vis profile, except below 22 km

of altitude where an underestimation is found. The mean relative difference in the 17-37 km altitude range is +9% with a maximum overestimation of 28% at 29 km. It should be noted that these results are not statistically significant since only two coincident events have been found for the comparison at this station.

In order to minimize the effect on the comparison of the smoothing error associated to the ground-based measurements (see Fig. 1), we have also compared partial columns. Fig. 4 shows a comparison of NO₂ partial columns calculated by integrating the SCIAMACHY limb and ground-based UV-vis profiles in the 16-35 km altitude range. 16-35 km roughly corresponds to the common altitude range where both SCIAMACHY and ground-based UV-vis measurements are sensitive enough to the vertical distribution of NO₂. A mean overestimation by SCIAMACHY of +0.35 x 10¹⁵ molec/cm² is found, which corresponds to relative difference values ranging from +5% in late spring-early summer (large NO₂ columns) to +30 % in winter-early spring and late summer-fall (moderate

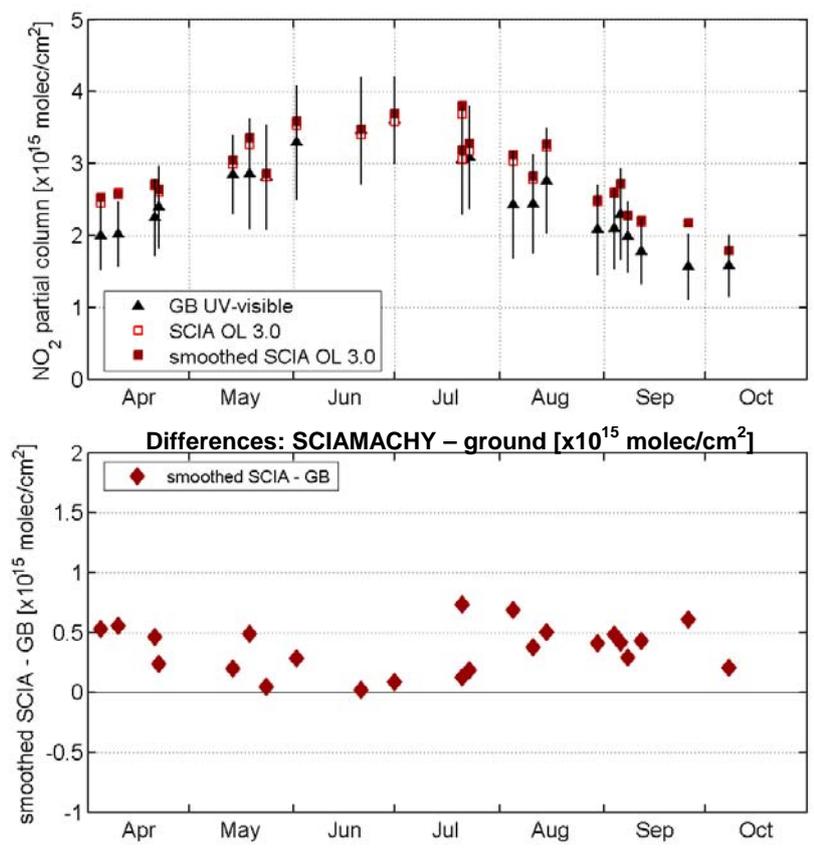


Figure 4. Comparison of the 16-35 km partial columns calculated from the SCIAMACHY limb and ground-based UV-vis NO₂ profiles at Harestua (2002-2005 period). The error bars on the ground-based UV-vis partial columns have been estimated from the total retrieval errors on the ground-based UV-vis profiles. The absolute differences appear on the lower plot.

and small NO_2 columns). However, these differences are not significant since SCIAMACHY partial columns are generally within the error bars associated to the ground-based UV-vis partial columns. These error bars have been estimated from the total retrieval error on ground-based UV-vis profiles. Despite the limited number of coincident events in late spring-early summer, a small seasonality seems to be present in the absolute difference, with smaller values in late spring-early summer and larger values in winter-early spring and late summer-fall.

5. SCIAMACHY NADIR NO_2 COLUMNS VALIDATION

5.1. Column comparison

SCIAMACHY nadir NO_2 vertical column densities (VCDs) have been compared here to ground-based NO_2 VCDs calculated by integrating NO_2 profiles retrieved at the SCIAMACHY overpass time. Fig. 5 shows time-series of SCIAMACHY and ground-based UV-vis NO_2 VCDs at Harestua. Concerning the SCIAMACHY data,

all pixels that within 300 km around the station have been selected for the comparison. Sunrise ground-based NO_2 VCDs from the NDACC database (which are the SCDs at 90°SZA divided by appropriate AMF) have been also included in the comparison. These data have been photochemically corrected in order to take into account the time difference between the SCIAMACHY and ground-based UV-vis measurements ($\sim 10\text{h}$ local for SCIAMACHY and around 90°SZA for ground-based UV-vis observations). A photochemical correction has been calculated for each day using the stacked box photochemical model PSCBOX daily initialized with chemical and meteorological fields from the SLIMCAT 3D-CTM. As can be seen from Fig. 5, the consistency between both ground-based UV-vis data sets is very good. Concerning the agreement between SCIAMACHY and ground-based UV-vis NO_2 VCDs, a mean overestimation by SCIAMACHY of $+0.65 \times 10^{15} \text{ molec/cm}^2$ is obtained for the 2002-2005 period. This corresponds to relative differences values ranging from +10% in late spring-early summer to +35% or

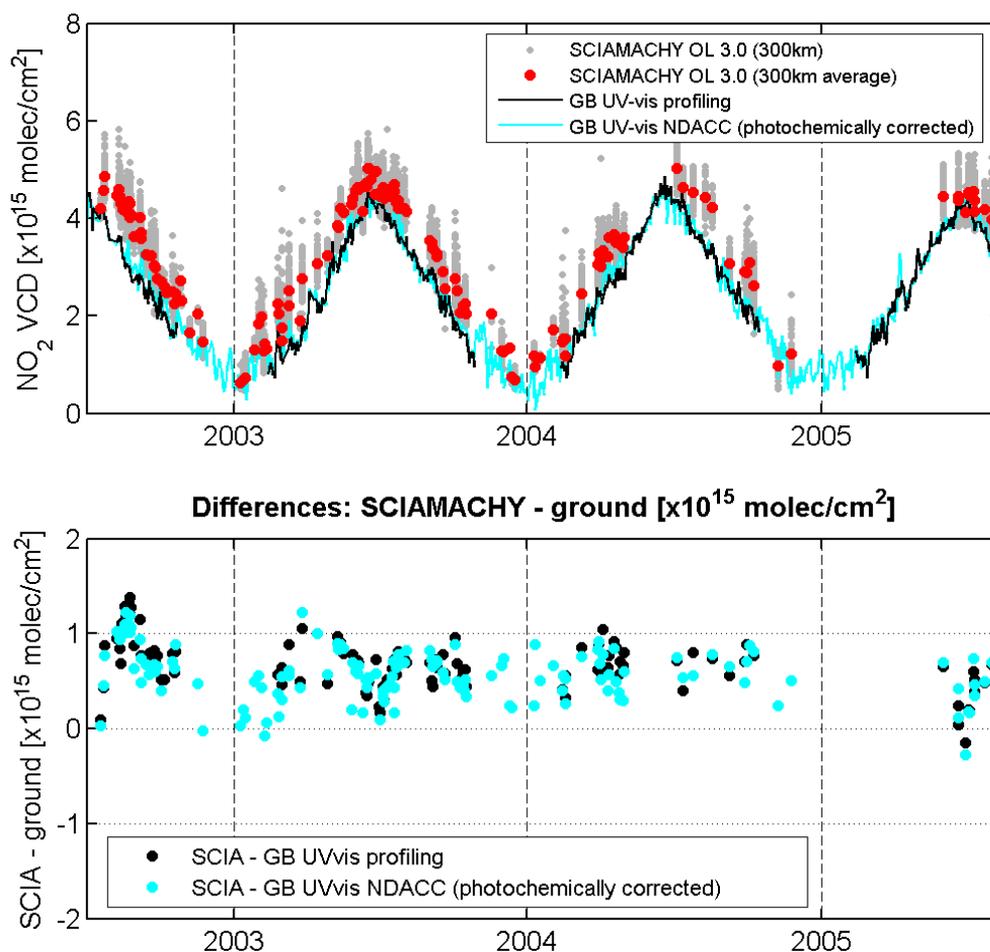


Figure 5. Comparison between SCIAMACHY nadir and ground-based UV-vis NO_2 VCDs at Harestua.

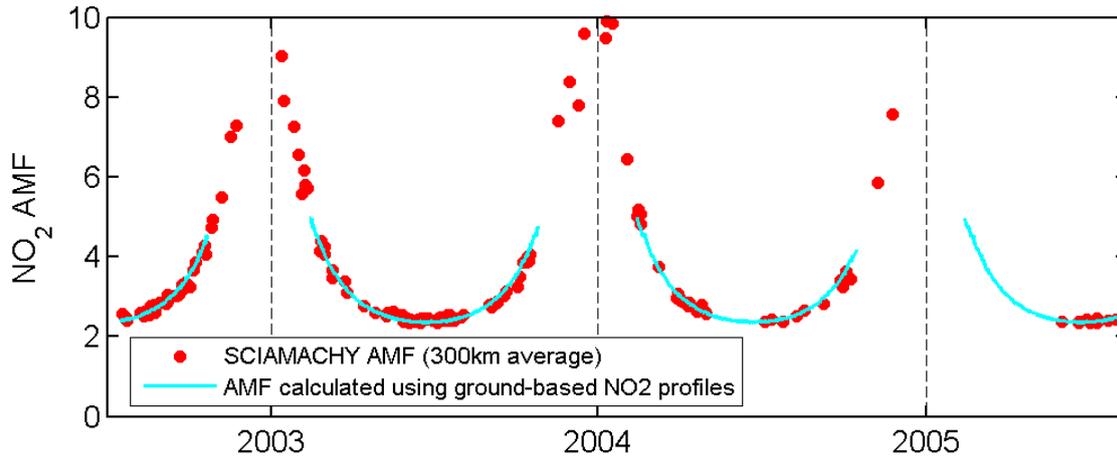


Figure 6. Comparison for the Harestua station between SCIAMACHY NO_2 AMFs and nadir NO_2 AMFs calculated using the radiative transfer model UVspec/DISORT with the ground-based UV-vis NO_2 profiles as input.

more in winter-early spring and late summer-fall. The absolute difference seems also to vary with season with smaller absolute difference values from mid-June to mid-July than for the rest of the year (see comparison for 2003 in Fig. 5). It should be noted that, given the accuracy of their comparison method, Lambert et al. [9] have found similar results in their pole-to-pole validation of SCIAMACHY nadir NO_2 columns using the NDACC/UV-vis network.

5.2. NO_2 AMF verification

NO_2 air mass factors (AMFs) used in the OL 3.0 processor to derive VCDs from the SCIAMACHY NO_2 slant column densities have been verified for the Harestua station (non-polluted site). In order to achieve that, SCIAMACHY NO_2 AMFs have been compared to nadir NO_2 AMFs calculated using the radiative transfer model UVspec/DISORT [6] with the ground-based UV-vis NO_2 profiles retrieved at the SCIAMACHY overpass time as input. For these calculations, the wavelength was fixed to 437 nm. As can be seen in Fig. 6, a very good agreement is obtained between both NO_2 AMF sets.

6. CONCLUSIONS

SCIAMACHY NO_2 OL 3.0 operational products (limb profiles and nadir columns) have been compared to profiles (and corresponding integrated vertical columns) retrieved from ground-based zenith-sky UV-vis observations at Harestua (60°N , 11°E) and Reunion Island (21°S , 55°E). A good overall agreement has been found, given the accuracy of the ground-based UV-vis profiling technique. Mean profiles comparison results show that SCIAMACHY overestimates ground-based UV-vis profiles in the 17-37 km altitude range by 16% and 9% in average at Harestua and Reunion Island, respectively. It should be noted that the statistics of the

comparison at the Reunion Island station is very poor (only 2 coincident events). Partial columns calculated by integrating SCIAMACHY and ground-based UV-vis NO_2 profiles in the 16-35 km altitude range have been also compared at Harestua. It is found that SCIAMACHY overestimates the ground-based partial columns by 0.35×10^{15} molec/cm². However, this difference is not significant since SCIAMACHY partial columns are generally within the error bars associated to the ground-based UV-vis columns.

Concerning the comparison between SCIAMACHY nadir and ground-based NO_2 total columns at Harestua, a mean overestimation by SCIAMACHY of $+0.65 \times 10^{15}$ molec/cm² is found for the 2002-2005 period. This is consistent with the results obtained by Lambert et al. [9] in their pole-to-pole validation of SCIAMACHY nadir NO_2 columns using the NDACC/UV-vis network. A very good consistency has also been found between the NO_2 AMFs used in the OL 3.0 processor and those calculated using the radiative transfer model UVspec/DISORT initialized with ground-based UV-vis NO_2 profiles retrieved at the SCIAMACHY overpass time.

7. Acknowledgements

This work was supported by Prodex Belgium. We wish also to thank M. P. Chipperfield (University of Leeds) for providing us with SLIMCAT data and ESA and DLR for extending the SCIAMACHY validation reference set.

8. REFERENCES

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