ON THE USE OF THE MAXDOAS TECHNIQUE FOR THE VALIDATION OF TROPOSPHERIC NO_2 COLUMN MEASUREMENTS FROM SATELLITE

Gaia Pinardi[1], F. Hendrick[1], K. Clémer[1], J.C. Lambert[1], J. Bai[2], and M. Van Roozendael[1]

(1) BIRA/IASB, Av Circulaire 3, B-1180 Brussels, Belgium
(2) Institute of Atmospheric Physics, Chinese Academy of Sciences, Beichenxilu 40, Beijing, China

Abstract

The MultiAxis DOAS (MAXDOAS) technique has been recently developed as a new remote sensing tool for the monitoring of tropospheric pollutants by means of the differential optical absorption spectroscopy (DOAS) method (Heckel et al., 2005, Hönninger et al., 2004). In contrast to zenith-sky DOAS instruments which have been commonly used over the last decades for stratospheric monitoring and satellite validation, e.g. as part of the Network for the Detection of Atmospheric Composition Change (NDACC), MAXDOAS instruments are designed to allow the quasi simultaneous observation of the scattered sun light in a range of different line-of-sight (LOS) directions from the horizon to the zenith, which leads to increased sensitivity towards atmospheric absorbers present close to the surface. Through adequate retrieval process, the near-surface concentration of atmospheric pollutants like NO_2 can be determined, as well as their integrated tropospheric and stratospheric column abundances. Owing to these capabilities, the MAXDOAS technique represents a very promising technique for the validation of tropospheric NO_2 column measurements that have been retrieved from UV-Visible nadir sounders such as GOME, SCIAMACHY, OMI and more recently GOME-2. In this work, we summarize the experience acquired at BIRA-IASB with tropospheric NO_2 validation using the MAXDOAS technique. Results from the DANDELIONS (Dutch Aerosol and Nitrogen Dioxide Experiments for validation of OMI and SCIAMACHY) campaigns, from recent observations performed in China nearby Beijing as well as from longer-term measurements performed at the Observatoire de Haute Provence (OHP), in Southern France (44°N, 5.7°E) are discussed. Strengths and limitations of the MAXDOAS technique for satellite validation are highlighted and illustrations of achievements recently obtained are given with particular emphasis on the GOME-2 instrument.

1. THE GEOMETRICAL APPROXIMATION APPLIED TO MAXDOAS DATA

NO_2 tropospheric vertical column densities (VCD) are retrieved from MAXDOAS measurements performed in Cabauw (The Netherlands), Observatoire de Haute Provence (OHP, France) and Beijing (China). The vertical columns are obtained from the differential slant column densities (DSCD) using a simple geometrical approximation. This approach is based on two assumptions: first that the stratospheric absorption is similar in the horizon-viewing and zenith-sky direction of the same scan, and therefore cancels when subtracting the zenith slant column from the off-axis column (see Figure 1), and second, that the NO_2 layer is located below the mean last scattering altitude of the photons, so that for the 2 highest off-axis viewing angles (generally around 15° and 30°), the tropospheric AMF can be reduced to his geometrical enhancement along the line-of-sight (LOS) ($AMF_{geom} = \sin^{-1}(LOS)$). The tropospheric columns are thus retrieved as:

$$VC_{geom} = \frac{DSCD^{tropo}}{AMF^{tropo}} = \frac{SCD_{off} - SCD_{zen}}{\sin^{-1}(LOS) - 1} \qquad (1)$$

In order to ensure the validity of this approximation, the results from the 30° and 15° elevation angles are compared and only measurements agreeing within 20% are taken into account. This check guarantees that the geometric light path enhancement is a good approximation in the boundary layer and excludes measurement greatly affected by clouds or horizontal inhomogeneities.

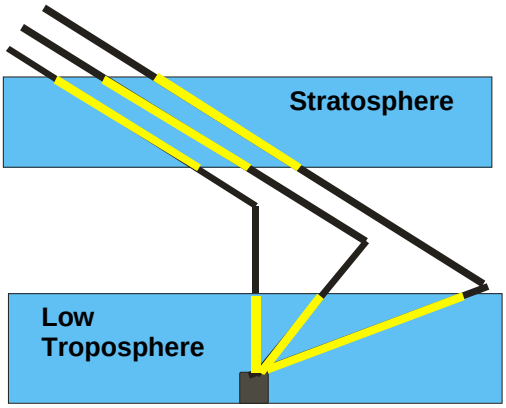

Figure 1: **Sketch of the MAXDOAS geometry and the conditions for the application of the geometrical approximation: last scattering altitude above the NO_2 layer and same stratospheric path for consecutives zenith and off-axis elevations.**

Sensitivity tests have been performed with the UVspec/DISORT radiative transfer model (RTM, Mayer and Kylling, 2005; Hendrick et al., 2006) to estimate the error done using this approximation. Simulations have been performed for several aerosol conditions, geometries (solar zenith angles (SZA) from 20° to 80° and relative azimuths from 0° to 180°), albedo and NO_2 profiles. The aerosols are characterized by their asymmetry factor (g=0.68), a very small absorption (single scattering albedo ~ 1) and a constant extinction in the first layer, between the surface and 1 km. Different aerosol loadings are tested, by varying the aerosol optical depth (AOD). The results of the baseline simulations at 440nm for an albedo of 5% and an NO_2 profile constant in the first km and zero above are compared to results of the geometrical approximation in figure 2. The results are expressed as the relative error on the tropospheric vertical column when applying the geometrical AMF instead of appropriate radiative transfer calculations to the $DSCD^{tropo}$:

$$Error = \frac{VC_{geom} - VC_{RTM}}{VC_{RTM}} = \frac{AMF_{RTM}^{tropo}}{AMF_{geom}^{tropo}} - 1 \qquad (2)$$

Figure 2 shows that the columns obtained from 30° elevation angles can overestimate or underestimate, depending on the geometry and the aerosols, while the column retrieved from 15° elevation (not shown) over-estimate at every azimuth and SZA. Large errors occur for small azimuth angles (up to ±40°), while a minimum error is obtained around 80°-90° azimuth. The plain lines are the columns that are retained when applying the 20% criterion between data retrieved at 15° and 30° elevation angles. This allows the elimination of most of the critical situations. More details on the error characterization can be found in Pinardi et al. (2008b).

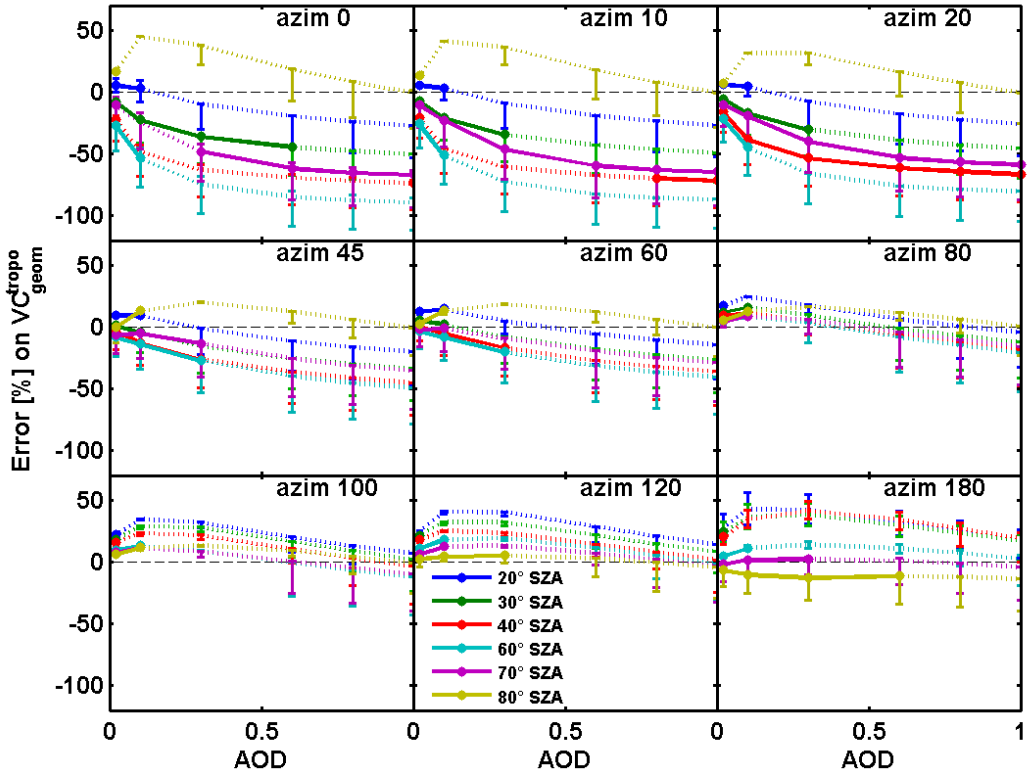

Figure 2: **Error in percent on the retrieval of NO_2 tropospheric VCD using the geometrical approximation, for different aerosol optical depths (AOD). Every subplot represents a different relative azimuth angle (from 0° to 180°) and several solar zenith angles (SZA) are plotted with different colours. The dotted lines illustrate the errors for the baseline assumptions simulations, the error bars resume the different simulations tested (albedo of 10%, NO_2 and aerosol profiles constant up to 0.5km or 2km, with the possibility of free tropospheric and stratospheric NO_2 content), while the plain lines are the errors when keeping only measurements where the columns retrieved at 30° and 15° elevation agrees within 20%.**

2. TROPOSPHERIC NO_2 RETRIEVAL FROM SATELLITES

Tropospheric NO_2 can be measured from satellite since 1995 with the launch of the Global Ozone Monitoring Experiment (GOME) instrument aboard ERS-2, and followed by SCIAMACHY (Scanning Imaging Absorption Chartography) aboard ENVISAT, the Ozone Monitoring Experiment (OMI) aboard AURA and more recently with the GOME-2 instrument aboard MetOp. All these satellite instruments are grating spectrometers collecting light back-scattered by the surface and the atmosphere with a nadir viewing geometry and provide, among others products, total and tropospheric NO_2 columns. In the followings, we focus on SCIAMACHY, OMI and GOME-2. These three instruments differ in their temporal and spatial resolution. Their overpass time is around 10h, 13h30 and 9h30 respectively, with a spatial resolution of 60x30km², 13x24km² and 80x40km², respectively, and with a global coverage achieved every 6 days for SCIAMACHY and every day for OMI and GOME-2.

The retrieval of tropospheric NO_2 from these measurements is based on a residual technique that involves three steps. First, the total NO_2 slant column densities are retrieved from the spectra by applying the DOAS technique. Secondly, stratospheric content is estimated and subtracted, and finally, the tropospheric slant columns are converted into vertical columns by applying tropospheric AMF. Several retrieval algorithms exist, developed by different groups, which differ in at least one of the steps that lead to the tropospheric vertical columns. The main differences between the retrievals of interest in this study are summarized in Table 1.

	GOME-2 (DLR)	**OMI (SP)**	**OMI (NRT)**	**SCIAMACHY (TEMIS)**	**SCIAMACHY (IUP Bremen)**
Reference	GOME-2 ATBD	Bucsela et al. 2006	Boersma et al. 2007	Blond et al. 2007; van der A et al. 2006	Richter et al. 2005
SCD retrieval	DOAS retrieval within 425-450nm (GDP 4.2)	DOAS retrieval within 405-465 nm		DOAS retrieval within 426.3-451.3nm	DOAS retrieval within 425-450nm
Stratospheric	Spatial masking/	Spatial masking/	Assimilated NO_2 stratospheric SCD		Stratospheric

correction	smoothing of the polluted NO_2 field	smoothing of the polluted NO_2 field	with the TM4 chemistry-transport model		reference sector over Pacific
AMF calculation	LIDORT RTM	TOMRAD RTM	DAK RTM		SCIATRAN RTM
NO2 a-priori profile	Monthly mean profiles (MOZART-2)	Pollutes/unpolluted scenarios (GeosCHEM)	Daily profiles (TM4)		Monthly mean profiles (MOZART-2)
Cloud treatment	Correction based on OCRA/ ROCCIN cloud retrieval scheme	Correction based on OMI operational O_2-O_2 cloud retrieval scheme		Correction based on FRESCO cloud retrieval scheme	Screening based on cloud fraction
Aerosols	Implicitly corrected by cloud treatment				Scenarios (LOWTRAN)
Albedo	GOME/TOMS database	GOME database	GOME/TOMS database		

Table 1: **Main differences between the different satellite tropospheric NO_2 retrievals.**

3. TROPOSPHERIC NO_2 VALIDATION

3.1 Cabauw (52°N, 5°E)

MAXDOAS measurements have been performed by BIRA-IASB, University of Bremen and University of Heidelberg during the DANDELIONS campaigns (Brinksma et al. 2008; http://www.knmi.nl/omi/research/validation/dandelions) in May-July 2005 and in September 2006. Tropospheric NO_2 VCDs are obtained from the MAXDOAS applying the geometrical approximation and are compared to OMI and SCIAMACHY tropospheric NO_2. For OMI, both the standard and the near-real-time products are compared, while for SCIAMACHY we focus on Bremen and TEMIS (Tropospheric Emission Monitoring Internet Service, http://www.temis.nl) retrievals. More details can be found in Pinardi et al. (2008b).

The ground-based data are averaged around ±1h of the satellite overpass time and the closest point of the cloud free data (CF<20%) within 50km for OMI and 200km (over land) for SCIAMACHY are selected.
Results of the comparisons are summarized in Figure 3 and more details can be found in Pinardi et al. (2008b). Very good agreement is obtained, with correlations from 0.62 to 0.74 and regression slopes from 0.82 to 0.97 (±0.11 to ±0.33). The standard (SP) and the near-real-time (NRT) OMI products agree within 1.6% while for SCIAMACHY, larger differences between the two products are found. On average over both campaigns, OMI SP slightly underestimates the ground-based data (-1.1%) while the NRT columns are within 0.5%. SCIAMACHY Bremen tends to underestimate the ground-based data (-7%) while the TEMIS data are ~18% higher.
One main issue highlighted during this comparison in Cabauw, is the large scatter of the data, most likely related to effect of local NO_2 inhomogeneities.

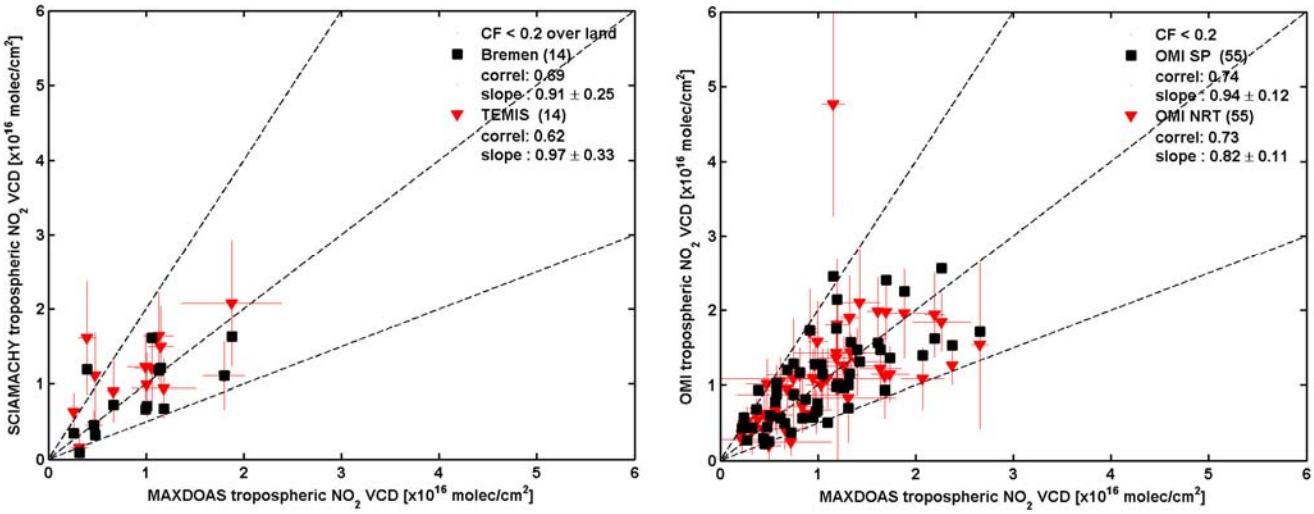

Figure 3: **Correlation plot of OMI and SCIAMACHY tropospheric NO_2 and ground-based MAXDOAS dataset for both DANDELIONS campaigns. The results of an orthogonal regression and the correlation coefficients are included in the legend.**

3.2 OHP (44°N, 5.7°E)

Within the context of the O3MSAF CDOP project (http://o3msaf.fmi.fi), BIRA-IASB MAXDOAS measurements performed in OHP since June 2007 have been used to test and set up a method for the comparison/validation of GOME-2 tropospheric NO_2 data retrieved by DLR. This comparison is a first attempt of tropospheric NO_2 validation for GOME-2, covering a complete year. More details can be found in the O3M-SAF validation technical note (Pinardi et al., 2008a).

Ground-based data are interpolated around ±1h of the satellite overpass time and only GOME-2 cloud free data (CF<20%) within 100km are used for the comparison. Figure 4 shows that the pollution episodes are captured by GOME-2 and the quantitative comparison are very encouraging, with a correlation coefficient of 0.65 and a linear regression slope of 1. A relatively large scatter characterize the comparison, but similar results are also found for other satellites products (e.g., OMI SP), pointing to the large differences in the NO_2 field sampled by the satellite and the ground-based instrument. Moreover, OHP usually alternates between clear unpolluted conditions with low NO_2 content, and some pollution episodes (mainly transport from polluted European regions). In case of low tropospheric NO_2 conditions, the GOME-2 observation is essentially a measurement of the noise of the retrieval.

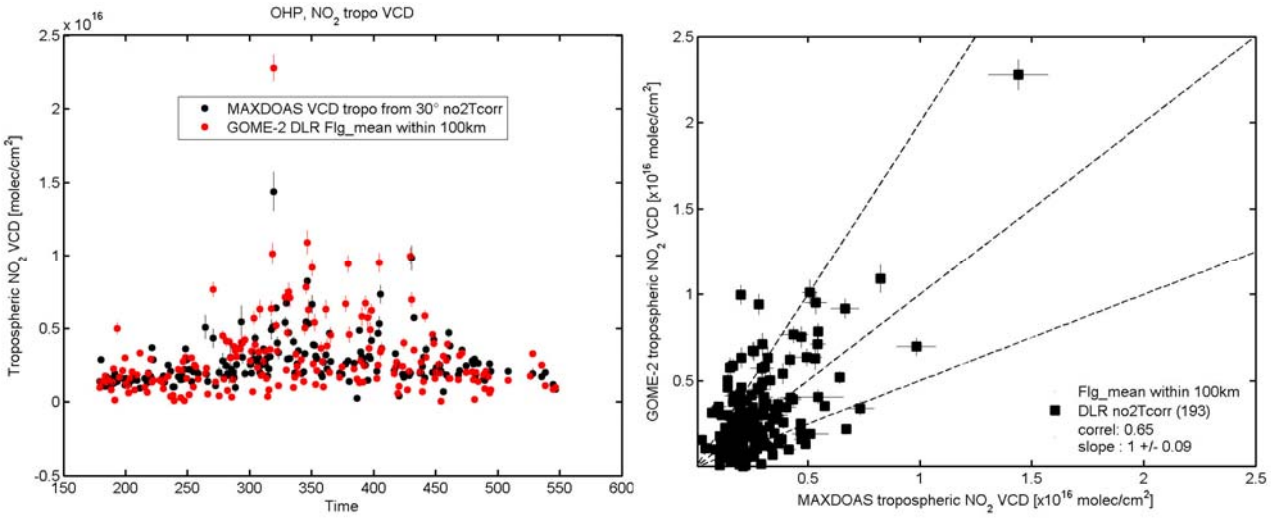

Figure 4: **Time series and scatter plot of the MAXDOAS and the GOME-2 DLR tropospheric NO_2 VCD (mean value of all the pixels within 100km around OHP). The error bars correspond to the estimation of the error done on the MAXDOAS when using the geometrical approximation (as described in section** Error! Reference source not found.**). Statistical information as the correlation coefficient and the slope of a linear regression analysis are included in the scatter plot legend.**

3.3 Beijing (40°N, 116.3°E)

Since June 2008, BIRA-IASB operates a MAXDOAS in Beijing centre, within the framework of the AMFIC project (Air Quality Monitoring and Forecasting in China, http://www.amfic.eu/index.php).
The ground-based MAXDOAS data are averaged around ±1h of the OMI overpass time and are compared to the NRT mean cloud free data (CF<20%) over 100km around Beijing.
Figure 5 shows the behaviour of the measured tropospheric NO_2 since June. It should be noted than since the 20th of July, severe limitations have been imposed by the Chinese government in order to try to decrease the pollution during the Olympic Games period (from 8 to 24 August, http://news.bbc.co.uk /2/hi/asia-pacific/7498198.stm). A large day to day variation is found in the measurements, and no definitive conclusion can be drawn at this stage, on the outcome of the adopted limitations.
The MAXDOAS measurements have been also used to validate emissions estimates from the CHIMERE model. This model was used for air quality previsions during the Olympic period. Figure 6 shows the differences in the diurnal cycle of NO_2, as predicted by the model and as measured by MAXDOAS and OMI NRT.

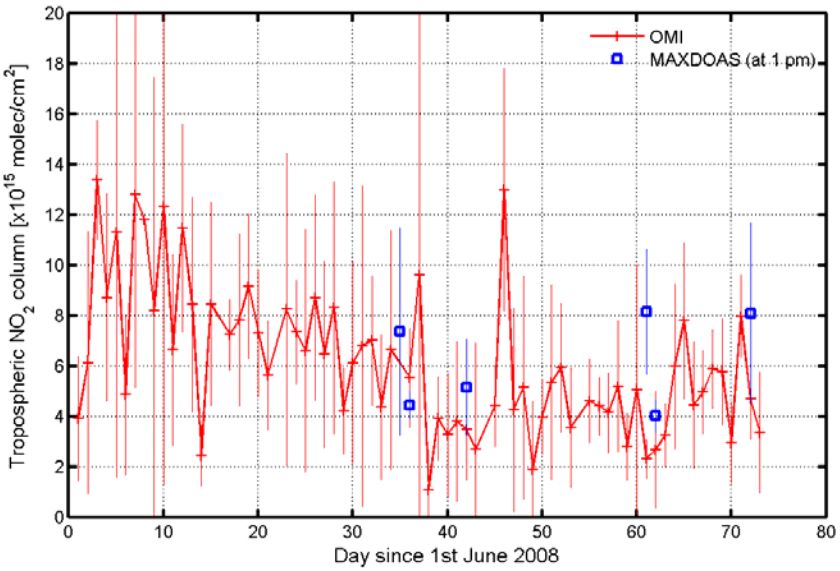

***Figure 5*: Tropospheric NO_2 over Beijing measured by OMI (TEMIS product) and by BIRA-IASB MAXDOAS.**

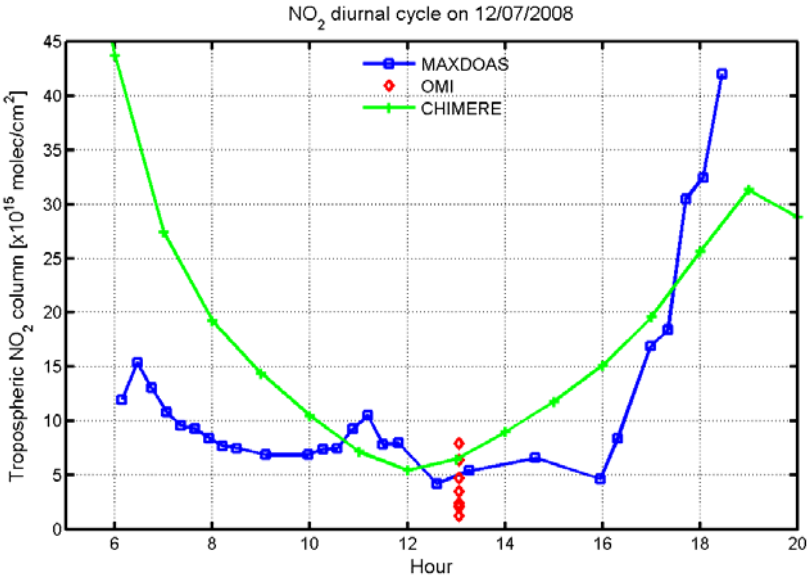

***Figure 6*: Diurnal cycle of NO_2 from the CHIMERE model and MAXDOAS data.**

4. CONCLUSIONS AND FUTURE WORK

The geometrical approximation has been used to infer tropospheric NO_2 VCD from MAXDOAS measurements performed by BIRA-IASB at different locations (Cabauw, OHP and Beijing). Radiative transfer calculations have been performed for several geometrical settings, aerosols loadings and NO_2 vertical distributions, in order to assess the errors related to the use of this approximation. Examples of tropospheric NO_2 validation have been presented at the three locations for several satellites (SCIAMACHY, OMI and GOME-2).

A large scatter due to the different NO_2 field sampled by satellite and by the MAXDOAS data (including spatial inhomogeneities and temporal variations) is pointed out in the comparisons. Efforts are needed to improve the spatial co-location, and this can be achieved e.g., using multi-azimuth instruments and/or spatial array of MAXDOAS instruments. To reduce the difference in the probed air-masses, the use of trajectory analyses and local air-quality modelling is envisaged for the future, in order to analyze the impact of NO_2 field gradients on the comparison results.
Application of the BIRA-IASB MAXDOAS aerosol/NO_2 profile retrieval tool to DANDELIONS and Beijing data is under progress, with the aim of (1) comparing with other ground-based profiles

available during DANDELIONS and validate the profiling tool, and (2) validating tropospheric a priori profiles used in satellite retrievals.

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