OVERVIEW OF THE VALIDATION OF GOME-2 TOTAL AND TROPOSPHERIC NO_2 COLUMNS

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Abstract

In the framework of EUMETSAT's Satellite Application Facility on Ozone and Atmospheric Chemistry Monitoring (O3M-SAF) nitrogen dioxide (NO_2) total column and tropospheric column data are generated operationally from MetOp-A GOME-2 measurements. Different parameters are needed to derive the final NO_2 column product: the slant column density along the optical path, the fractional cloud cover and the cloud top pressure, the geometrical enhancement factor (AMF), and the NO_2 stratospheric reference to be subtracted from the total column to obtain the tropospheric column.
The validation of GOME-2 GDP 4.4 NO_2 data in an operational environment is at the developmental stage. To ensure meaningful and continuous quality assessment of GOME-2 NO_2 data products, BIRA-IASB, DLR and RMI have developed an end-to-end validation approach, which consists in performing the verification and validation of critical individual components of the level-1-to-2 retrieval chain. This approach uses other established retrieval facilities, a set of correlative observations performed by complementary ground-based instruments, measurements from other satellites (e.g. SCIAMACHY), and modelling support (CHIMERE model).
The end-to-end validation process of GOME-2 NO_2 data is illustrated by means of representative results at pilot stations (e.g., over OHP, France, and over Beijing, China) and operational perspectives are discussed.

1. GOME-2 GDP4.4 NO_2 RETRIEVAL

The GOME-2 instrument, onboard MetOp-A platform since October 2006, is a grating spectrometer collecting the nadir sun light back-scattered at the surface and by the atmosphere (Munro et al. 2006). GOME-2 is characterized by a spatial resolution of 80x40km², global coverage in 1.5 day and an equatorial overpass time at 9h30 in the descending node.
The retrieval of tropospheric NO_2 is based on a residual technique that involves three steps: $V_t = (S - M_s V_s)/M_t$. First, the total NO_2 slant column densities (S) are retrieved from the spectra by applying the DOAS technique. Secondly, stratospheric content (V_s) are estimated and subtracted, and finally, the tropospheric slant columns are converted into vertical columns by applying tropospheric AMF (M_t) (Boersma et al., 2004). In the following, we mainly focus on the GDP 4.4 NO_2 retrieval (Valks et al., 2010), performed operationally at DLR in the context of the O3M-SAF (http://o3msaf.fmi.fi/). In this product, after a first calculation of the total columns assuming no tropospheric pollution and pure stratospheric AMF, a second calculation is performed for polluted cases resulting in a tropospheric NO2 column (V_t) and a improved total column that is corrected for the tropospheric component. It should be noted that several retrieval algorithms exist, developed by different groups, which differ in at least one of the steps that lead to the tropospheric vertical columns. For example, different methods for the separation of the troposphere/stratosphere have been proposed (Leue et al., 2001; Wenig et al., 2004; Bucsela et al., 2006), as well as different choices for the calculation of the tropospheric AMF. The main differences between the retrievals of interest in this study are listed and compared to GDP 4.4 in Table 1.

	GOME-2 (GDP 4.4)	**GOME-2 (TEMIS)**	**SCIAMACHY (TEMIS)**
Reference	Valks et al. 2010	van der A et al. 2010	Blond et al. 2007; Boersma et al. 2004
Slant column retrieval	DOAS retrieval within 425-450nm (irradiance reference)	DOAS retrieval within 425-450nm (irradiance reference)	DOAS retrieval within 426.5-451.5nm (earthshine reference)

Stratospheric correction	Spatial filtering and masking of the polluted NO_2 field using MOZART-2 model	Assimilated NO_2 stratospheric SCD with the TM4 chemistry-transport model	Assimilated NO_2 stratospheric SCD with the TM4 chemistry-transport model
AMF calculation	LIDORT RTM	DAK RTM	DAK RTM
NO_2 a-priori profile	Monthly mean profiles (MOZART-2)	Daily profiles (TM4)	Daily profiles (TM4)
Cloud treatment	Correction based on OCRA/ ROCCIN cloud retrieval scheme	Correction based on FRESCO cloud retrieval scheme	Correction based on FRESCO+ cloud retrieval scheme
Aerosols	Implicitly corrected by cloud treatment		
Albedo	GOME/TOMS database		

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

2. THE END-TO-END VALIDATION APPROACH

The validation of GOME-2 GDP 4.4 NO_2 columns in the O3M-SAF context (http://o3msaf.fmi.fi/valreps.html) has been set up as an end-to-end approach, consisting in the validation of each component of the retrieval, as recommended by Reference Protocols and Guidelines (CDRH 2002, CEOS 2004, Lambert et al. 2009). This approach is useful in that it allows hidden compensating errors to be unraveled. Here we focus on the verification and comparison of:

- Slant columns S, by testing the operational algorithm on other datasets (e.g., GOME and GOME-2) and correlating it to other state-of-the-art scientific algorithms;
- Stratospheric vertical columns V_s, by comparing with correlative ground-based measurements from the NDACC network (both in unpolluted and polluted conditions) and with other satellite data;
- Tropospheric vertical columns V_t, by direct comparison with other satellite data and with MAXDOAS measurements.

3. END-TO-END VALIDATION AT THE OHP STATION

The NDACC station at Observatoire de Haute Provence (OHP, 44°N, 5.7°E) is an interesting pilot site for the study of tropospheric NO_2, as it alternates between clean air conditions and situations where it is influenced by polluted air masses transported from source regions. MAXDOAS measurements are available since 2005 and in the context of the O3M-SAF CDOP project (http://o3msaf.fmi.fi) the data from June 2007 have been used to test and set up a method for the comparison/validation of GOME-2 GDP 4.4 tropospheric NO_2 (Pinardi et al., 2008b).

3.1 Comparison of NO_2 slant columns from different satellite sensors

Figure 1 shows a comparison between time series of monthly mean slant columns measured from January 2007 until July 2010 within 300 km around OHP by different satellites: GOME (GDP 4.1), SCIAMACHY (TEMIS) and GOME-2 (GDP 4.4 and TEMIS products). As can be seen there is a large variability in the slant column data, however the GDP 4.4 product appears to be in good agreement with the other datasets, considering the instrumental and retrieval differences.

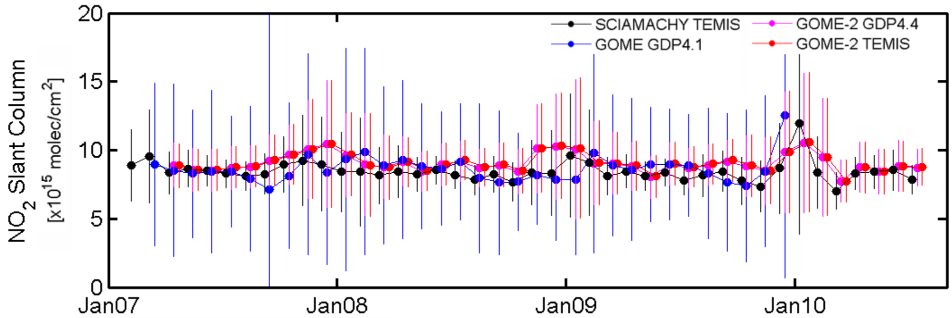

Figure 1: **Comparison of the time series of NO_2 slant column data measured by GOME (GDP 4.1), SCIAMACHY (TEMIS) and GOME-2 (GDP 4.4 and TEMIS) between January 2007 and July 2010. The dots represent the monthly means of all the pixels within a radius of 300km around OHP and the error bars represent the variability (one sigma standard deviations).**

3.2 Comparison of the total and stratospheric NO_2 columns

Figure 2 shows the total initial vertical columns (computed with stratospheric AMFs, and thus “uncorrected“ for tropospheric pollution) and the corresponding stratospheric columns above OHP, for different cloud selections. Results are binned according to fractional cloud fractions considering all GOME-2 pixels within 300km of OHP, and for the time-period from March 2007 until July 2010. The left hand plot corresponds to a selection of high clouds (cloud top pressure, CTP, smaller than 400hPa) while the other plot corresponds to lower clouds (CTP higher than 400hPa). As expected, the stratospheric content is similar in both plots, while the total uncorrected column strongly depends on the bulk altitude of the clouds. High clouds effectively mask the signal from surface NO_2 while in case of low-lying clouds the satellite observations remain sensitive to the NO_2 in the free troposphere even for fully cloudy pixels.

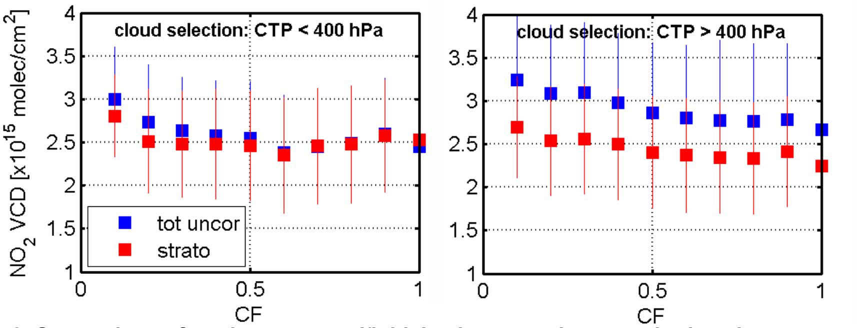

Figure 2: **Comparison of total uncorrected/initial columns and stratospheric columns around OHP. The squares correspond to averages in bins of 0.25x10^{16} molec/cm² and the error bars to the variability of all pixels in time and space. The left subplot is focusing only on pixels with a cloud top pressure (CTP) smaller that 400hPa while the right subplot is focusing on pixels with a CTP higher than 400hPa.**

Twilight sunrise zenith sky light (ZSL) data, mostly sensitive to stratospheric NO_2, have been used to validate satellite NO_2 columns (Lambert et al. 2004, Lambert 2006, Ionov et al. 2008, Celarier et al. 2008). Here we use stratospheric NO_2 columns derived from zenith-sky measurements performed at sunrise between 87°-91° SZA. Zenith-sky AMFs are accurately determined using a-priori climatological stratospheric NO_2 profiles (Lambert et al., 1999) similar to those used in the satellite evaluations. Figure 3 gives an overview of the comparison of the monthly mean stratospheric columns retrieved between January 2007 and July 2010 from satellite data (SCIAMACHY (TEMIS) and GOME-2 (GDP 4.4 and TEMIS)) and from coincident ground-based ZSL measurements.

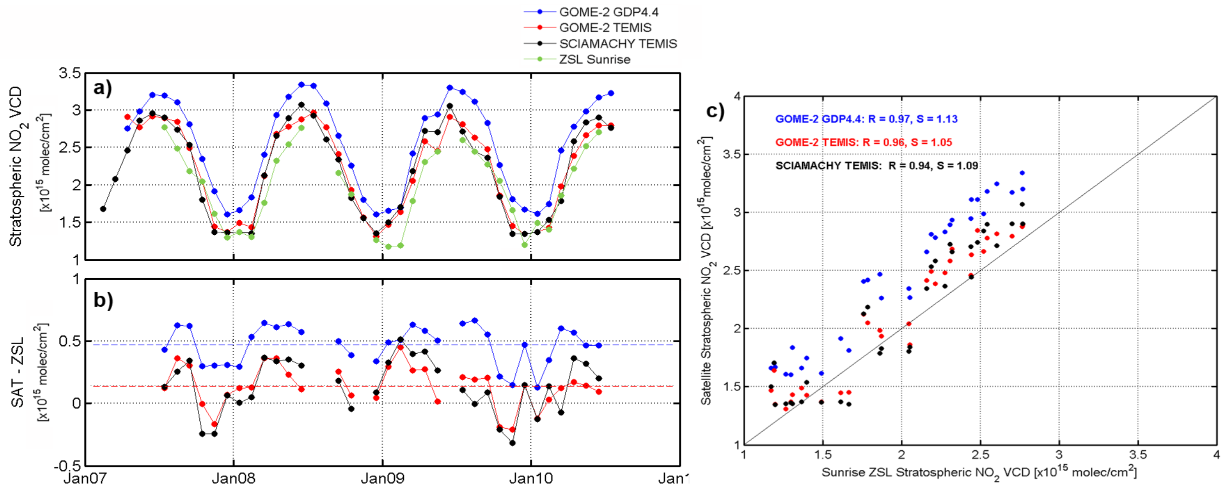

***Figure 3*:** **Time series and scatter plot of the monthly mean stratospheric NO_2 columns measured by GOME-2 (GDP 4.4 and TEMIS), SCIAMACHY (TEMIS) within 300km of OHP and ground-based ZSL, between January 2007 and July 2010. a) shows the raw time-series, b) the time-series of the absolute differences between the satellite and the ground-based ZSL sunrise measurements (dotted lines represent the mean difference values), and c) shows the scatter plot of the satellite as a function of the ZSL, with the correlation coefficient R and the slope S of the linear regression line are given in the legend.**

GOME-2 GDP 4.4 is in good agreement with the other datasets: good correlation (R = 0.97), similar seasonal variation, however slightly higher values are reported in comparison to the ZSL data (~4.8x10^{14} molec/cm² and slope S = 1.13). Part of the difference might be related to the spatial filtering approach used in GDP4.4 to infer the stratospheric correction. Stratospheric NO_2 columns derived in this way might include a residual tropospheric NO_2 content from oceanic origin, while the assimilation technique used in the TEMIS product (see Table 1) would be less sensitive to this effect. Similar findings were obtained e.g. by Lamsal et al. (2010) who found differences of about $1x10^{15}$ molec/cm² between the two OMI products (SP vs DOMINO) that also use similar different methods for the stratosphere separation.

3.2 Comparison of the tropospheric NO_2 columns

MAXDOAS data are increasingly exploited to validate satellite tropospheric NO_2 columns (Brinksma et al. 2008, Celarier et al. 2008, Irie et al. 2008). Here, we use tropospheric vertical columns obtained from MAXDOAS differential slant column densities (DSCD) and, for the AMF calculation, a simple geometrical approximation as described in Brinksma et al. 2008 and Pinardi et al. 2008a. Sensitivity tests have been performed to estimate the error resulting from this approximation (Pinardi et al. 2008a). These show that NO_2 columns obtained from 30° elevation angles can either overestimate or underestimate the true columns (depending on the measurement geometry and on the aerosol load). An overview of the NO_2 columns retrieved at OHP between July 2007 and July 2010 is given in the upper panel of figure 4. In the same figure, the lower panel represents the error estimated for each measurement point, the time of the day being colour coded as in Pinardi et al. (2008b). The difference in the error done using this approximation for the different geometries and time of the day is important; the mean error is around 8.5% when considering all the MAXDOAS points (plain line), and of only 3.1% when restricting MAXDOAS points to the satellite overpass time (dotted line).

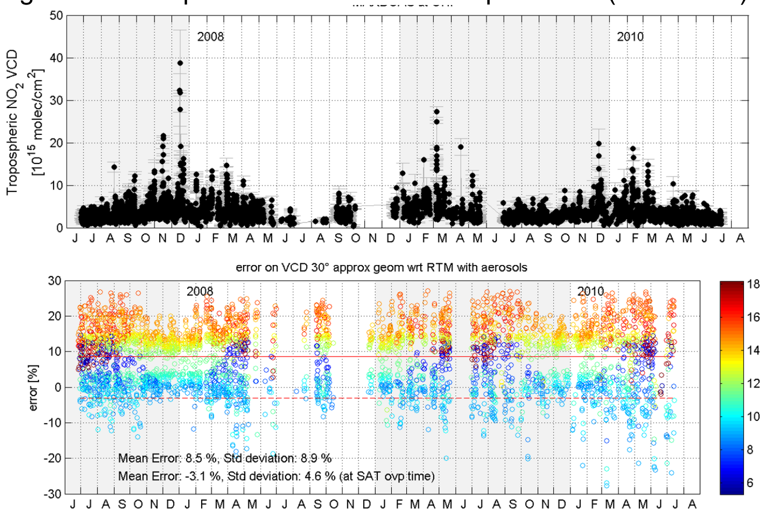

Figure 4: **Time series of MAXDOAS data at OHP from June 2007 to July 2010. The upper panel shows the tropospheric NO_2 column derived from the 30° elevation angle. The lower panel displays corresponding percent errors on the geometrical AMFs, with a colour code for the different hours of the day. The mean error value and its standard deviation are also given, for the whole time-series and when considering only points around the satellite overpass time.**

For the comparison with the satellites, ground-based data are extracted within a time window of ±1h around the GOME-2 overpass time and only cloud free satellite data (CF<20%) within 100km are used. Figure 5 presents an overview of the monthly mean tropospheric columns retrieved between July 2007 and July 2010 from GOME-2 GDP 4.4 and ground-based MAXDOAS. One can see that pollution episodes are well captured by GOME-2 (a). Also the comparison of monthly averaged columns (b) shows consistent seasonal variations, with high NO_2 in winter and low NO_2 in summer. Quantitatively speaking, both data sets are found to agree pretty well with a correlation coefficient of 0.82 and a linear regression slope of 0.96. Finally, comparing GOME-2 GDP 4.4 tropospheric columns to other satellite datasets, a good agreement is found with SCIAMACHY and GOME-2 columns from the TEMIS product as demonstrated in Figure 6.

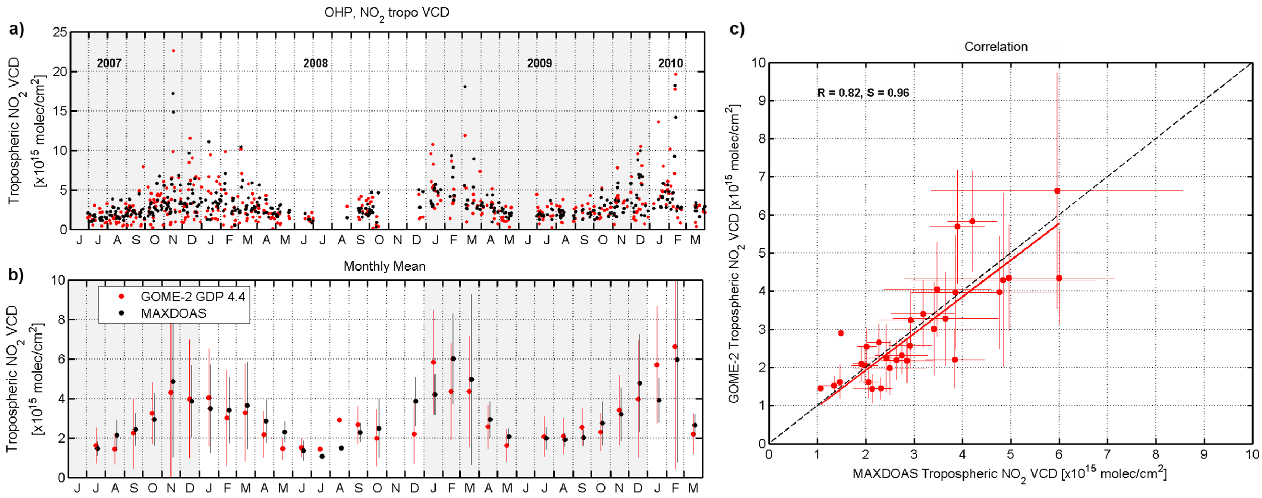

Figure 5: **Time series and scatter plot of the MAXDOAS and the GOME-2 GDP4.4 tropospheric NO_2 VCD above OHP. a) MAXDOAS data at satellite overpass time and satellite points within 100km of OHP, b) monthly mean values, c) correlation plot of the monthly means, with information on correlation (R) and slope (S) of a linear regression fit.**

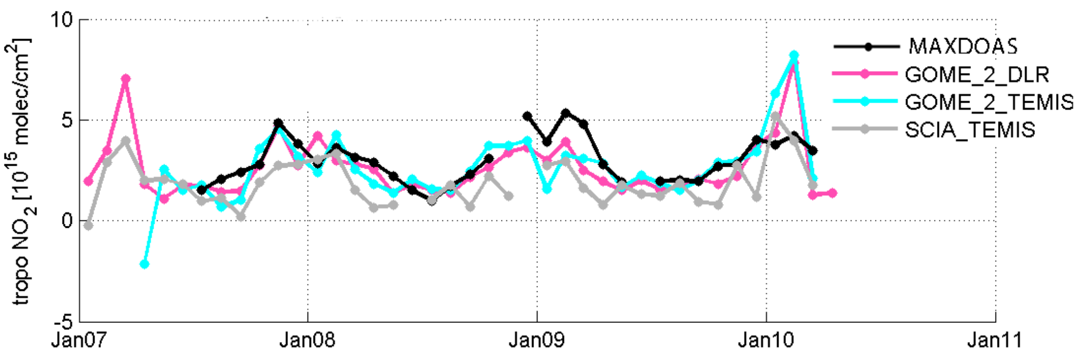

Figure 6**: Time series of the monthly mean MAXDOAS and satellite tropospheric NO_2 columns above OHP between January 2007 and March 2010. For the satellite points, the mean of all cloud-free pixels within 50km of OHP is considered.**

4. PRELIMINARY VALIDATION AT THE BEIJING STATION

BIRA-IASB has been running a MAXDOAS instrument in Beijing (40°N, 116.3°E) from June 2008 to April 2009 in the framework of the AMFIC project (Air Quality Monitoring and Forecasting in China, http://www.amfic.eu/index.php). Algorithms have been developed and demonstrated to derive vertical profiles of aerosols and trace gases (Clémer et al. 2010), including tropospheric NO_2 columns and profiles. Here we present first attempts to use this data set for the validation of GOME-2.

The ground-based MAXDOAS data are averaged around ±1h of the satellite overpass time and are compared to cloud free data (CF<20%) within 50km around Beijing, for different satellite and NO_2 products. Figure 7 shows the scatter plots between MAXDOAS and GOME-2 data, for the GDP 4.4 product on the left and for the TEMIS product on the right. It can be seen that both GOME-2 products correlate well with the ground-based data (correlation coefficients R between 0.81 and 0.91) but that GDP4.4 is much smaller than the MAXDOAS data (slope of the linear fit S of 0.5). Part of the difference might be related to the different sensitivity of each technique to the horizontal distribution of emissions. One expects the MAXDOAS measurements being more sensitive to local pollution peaks, while these will tend to be smeared out in the satellite pixel. However part of the discrepancy might also be related to shortcomings or uncertainties in the applied satellite retrieval settings, as suggested by the different results obtained comparing MAXDOAS results with the TEMIS product.

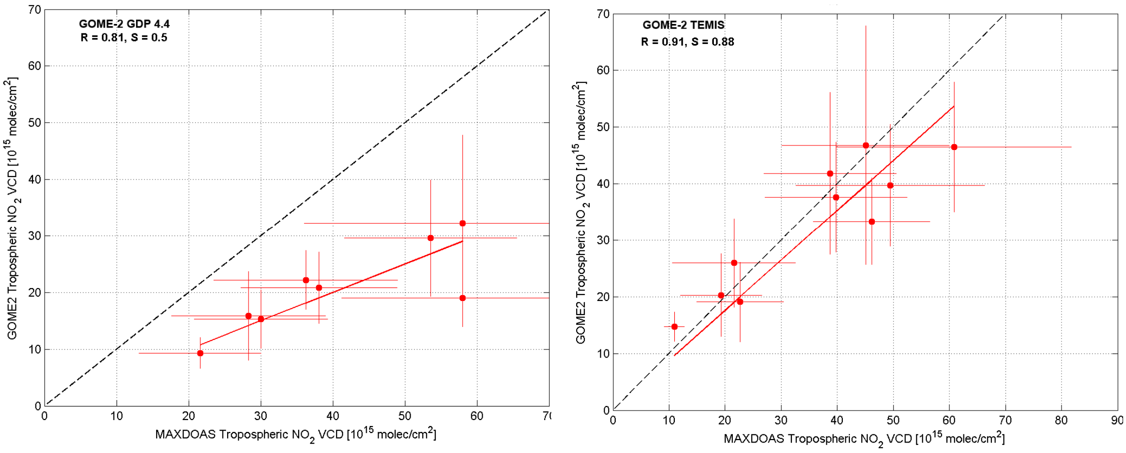

Figure 7: **Correlation plots of tropospheric NO_2 over Beijing measured by BIRA MAXDOAS and by GOME-2 (GDP 4.4 and TEMIS product).**

In Figure 8, time series of monthly averaged tropospheric NO_2 columns derived from different satellite retrievals are compared to MAXDOAS results. The discrepancies between the satellite data are to be related to the cloud treatment (OCRA/ROCCIN vs FRESCO) and to the retrieval choices for the tropospheric AMF calculation. E.g. a-priori NO_2 profile shapes are prescribed using monthly MOZART-2 profiles in the GDP4.4 algorithm, while the TEMIS algorithm uses daily TM4 profiles. Hains et al. (2010) have shown that the uncertainties on the tropospheric AMF due to profile shape errors are of the order of 15% in average. However profile shape uncertainties will typically be much larger in a highly polluted area like Beijing than in a rural or semi-rural location like OHP.

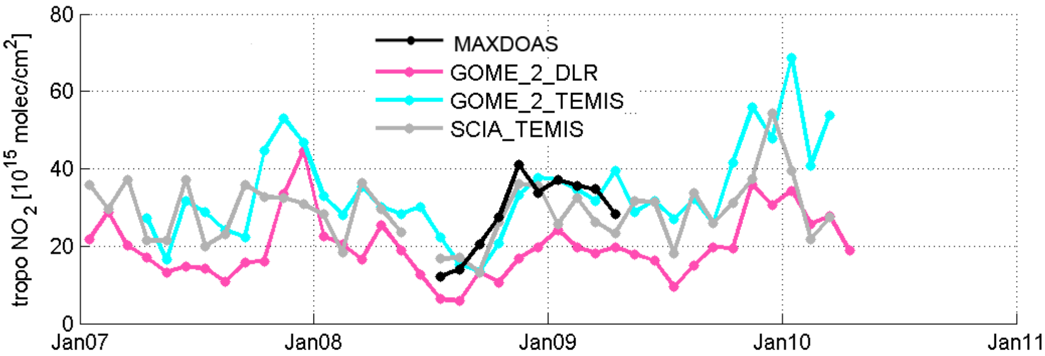

Figure 8: **Time series of the monthly mean MAXDOAS and satellite tropospheric NO_2 columns around Beijing between January 2007 and March 2010. For the satellite points, the mean of all cloud-free pixels within 50km of Beijing is considered.**

4. CONCLUSIONS AND FUTURE WORK

Validation results addressing stratospheric and tropospheric GDP 4.4 NO_2 columns are regularly updated within the CDOP project, confirming a good global agreement with ground-based correlative data sets. A pole to pole validation against the NDACC UVVis network (Pinardi et al., 2008b) shows that GOME-2 data meets the target requirements in the Northern Hemisphere, but that a systematic underestimation by about $0.6x10^{15}$ molec/cm² is observed in the Southern middle latitudes. No degradation of the product is seen in the comparisons, although inspection of the time-evolution of fitting residuals reveals a clear degradation mostly evident over the Pacific region.
In this paper we illustrate the end-to-end validation of GOME-2 NO_2 GDP 4.4 making use of a complete set of correlative observations available at the OHP station. Results are very encouraging. The GDP 4.4 stratospheric columns show a small bias of about $4x10^{14}$ molec/cm², while the tropospheric columns agree well with the other datasets. Extending the study to the more polluted site of Beijing, larger differences are found between the different satellite data products. A detailed study on tropospheric NO_2 product is in progress, exploring the causes for such discrepancies (Yu et al., in preparation). In this context, tests using MAXDOAS profile information as an input for satellite retrievals are on-going. Moreover, case studies using CHIMERE regional modeling are started with the

aim to account for the different horizontal sensitivities between satellite observations and correlative data sets such as MAXDOAS and in-situ surface concentration measurements.

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