

# Monitoring Changes in Tropospheric Constitution from Space

A contribution to subproject ACCENT-TROPOSAT-2 (AT2), Task Group 1

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## Summary

Data from four UV/visible instruments, GOME, SCIAMACHY, GOME-2, and OMI has been analysed for tropospheric NO<sub>2</sub> columns. The data have been compared for consistency with a focus on the GOME-2 data which have become available only recently and the time evolution above China.

GOME-2 tropospheric NO<sub>2</sub> fields derived using similar settings as for SCIAMACHY show a high degree of consistency with SCIAMACHY data. The agreement is further improved if only coincident measurements are used for the comparison. As result of the shorter integration times, GOME-2 retrievals are slightly noisier than SCIAMACHY retrievals unless a larger fitting window is employed. GOME-2 and SCIAMACHY data agree very well above China, indicating a continuing increase in NO<sub>2</sub> column amounts. OMI data for the same region show a similar trend but slightly lower values if the same retrieval settings are used, the difference in absolute values probably indicating a diurnal cycle in tropospheric NO<sub>2</sub> burden.

This study complements the work reported in the previous two years where improvements of the SO<sub>2</sub> retrieval over volcanic scenes were discussed, the first GOME-2 SO<sub>2</sub> retrievals were presented and a combined set of GOME and SCIAMACHY SO<sub>2</sub> columns was used to investigate the evolution of SO<sub>2</sub> over China.

## Introduction

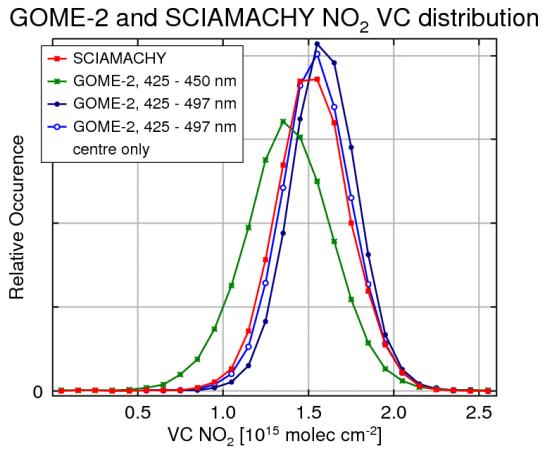
Tropospheric NO<sub>x</sub> is a key species for ozone formation and acidification of rain and aquatic eco-systems. It is mainly emitted in the form of NO from anthropogenic activities (all combustion processes) but to a lesser degree also from biogenic sources in soils and through lightning.

Remote sensing measurements of NO<sub>2</sub> from space are possible using the structured absorption features of NO<sub>2</sub> in the visible spectral region. Satellite measurements have the advantage of providing global data sets which facilitate monitoring of pollution, investigation of transport processes and estimation of source strengths.

## Scientific activities

### *Retrievals of GOME-2 NO<sub>2</sub> columns*

On October 19, 2006, the first of a series of three GOME-2 instruments was launched into orbit on MetOp-a. The GOME-2 instrument is very similar to GOME but provides better spatial coverage (global coverage every 1.5 days instead of every 3 days) and better spatial resolution (40 x 80 km<sup>2</sup> instead of 40 x 320 km<sup>2</sup>). Compared to SCIAMACHY, the spatial resolution is somewhat poorer but the spatial coverage is much improved.



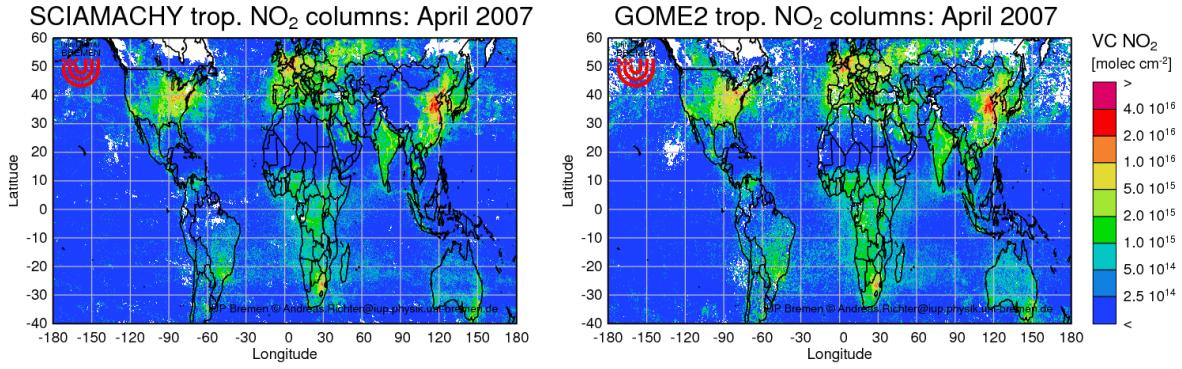
**Fig. 1 Frequency distribution of GOME-2 and SCIAMACHY NO<sub>2</sub> columns over the tropical Pacific (180°E (+-20°), 0°N (+-10°)) in September 2007.** The underlying assumption is that NO<sub>2</sub> slant columns should be constant in this area and the width of the distribution is mainly given by the noise in the data. As can be seen, the GOME-2 data have a broader distribution (larger noise) if the same fitting window is used as for SCIAMACHY but comparable noise if the larger spectral region is employed.

Using the settings developed for GOME and SCIAMACHY, NO<sub>2</sub> columns have been retrieved from GOME-2 data at the University of Bremen. While the overall agreement between GOME-2 and SCIAMACHY data is good, there is clearly more scatter in the GOME-2 data than in SCIAMACHY measurements. This can probably be explained by a combination of several effects including the shorter integration time needed to have good spatial resolution at a wide swath and the use of correlated sampling techniques in SCIAMACHY detectors but not in GOME-2. As larger noise reduces the quality of GOME-2 measurements, an attempt was made to improve the SNR by extending the fitting window from the traditional 425 – 450 nm region up to 497 nm. As shown in Fig. 1, the scatter of NO<sub>2</sub> slant columns retrieved over the tropical Pacific can in fact be reduced to the level seen in SCIAMACHY measurements with the larger fitting window. In addition, the small offset between the two data sets is further reduced although the reasons for this are not entirely clear. Unfortunately, using the larger window also introduces a scan angle dependency in the data at least for some lv1 data versions, probably related to interference with not fully corrected polarisation dependency of the GOME-2 instrument. Therefore, reprocessing of the full lv1 data set with the latest processor version will have to be awaited before a final statement can be made on the applicability of the larger fitting window. All of the results shown in the following sections are therefore based on the traditional fitting window.

## Scientific results and highlights

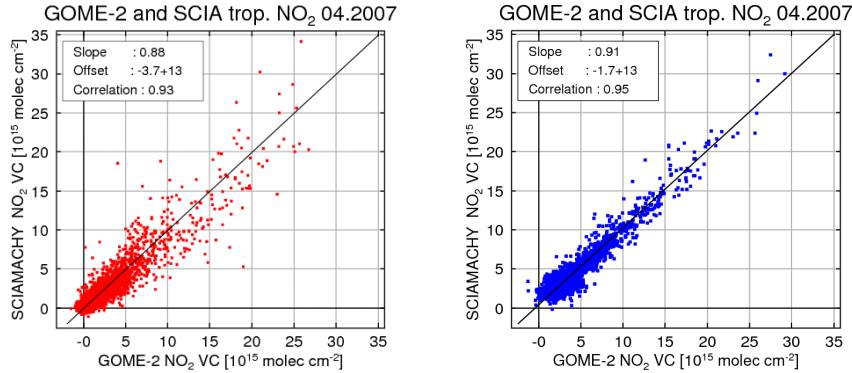
### *Comparison of GOME and SCIAMACHY measurements*

One of the main advantages of satellite measurements is the availability of global data and long data sets. However, to fully exploit this potential, time series from different platforms have to be combined. Therefore, tropospheric NO<sub>2</sub> columns retrieved from SCIAMACHY and GOME-2 measurements have been compared to investigate their consistency. All settings with the exception of cross-sections which are adapted to the instrument resolution have been kept identical in this inter-comparison. While this minimises differences introduced from different processing, good agreement between the two data sets does not necessarily also imply good absolute accuracy as systematic errors will be similar in the two products.



**Fig. 2:** Tropospheric NO<sub>2</sub> columns for April 2007 retrieved from SCIAMACHY measurements (left) and GOME-2 data (right). White areas are data gaps over regions with persistent cloud cover or snow and ice. The different sampling of the two instruments is not taken into account.

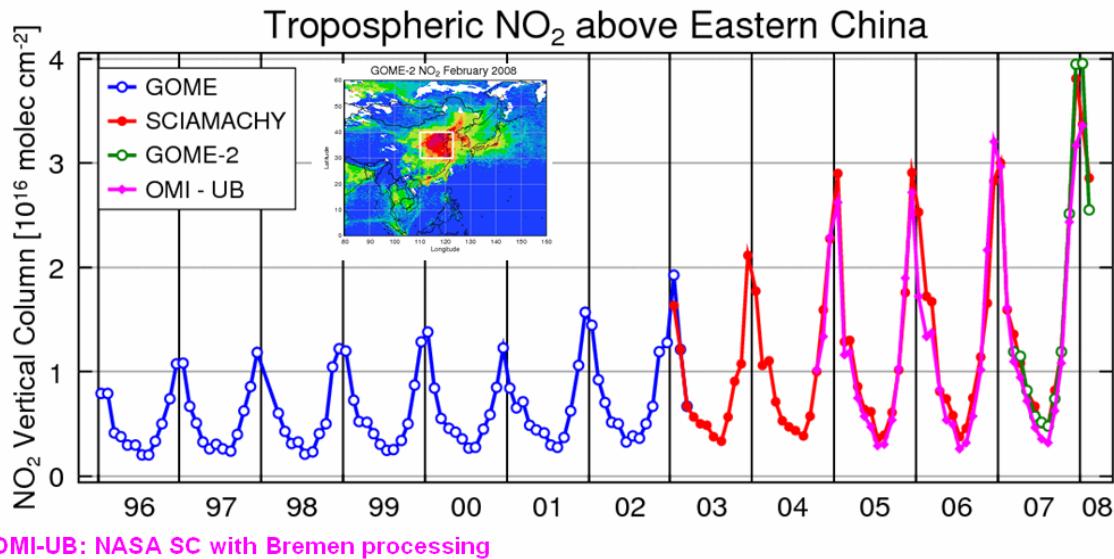
As shown in Fig. 2 for April 2007, the agreement is very good at first glance. A more quantitative comparison is shown in Fig. 3, where the same data is displayed in a scatter plot after binning in  $1^\circ \times 1^\circ$  cells. While the correlation is excellent, there is scatter of the order of several  $10^{15}$  molec  $\text{cm}^{-2}$  and also lower values in GOME-2 measurements. If only those measurements are used for which both instruments have data on the same day, the scatter is strongly reduced but GOME-2 data is still lower by 9%. This highlights the importance of sampling for the data set in particular in the case of SCIAMACHY which has global coverage at the equator in 6 days. What the origin for the apparent low bias of the GOME-2 data is has not yet fully been understood. As discussed in the previous section, changing to a larger fitting window increases the NO<sub>2</sub> columns slightly but this can not fully remove the difference for regions with large tropospheric NO<sub>2</sub> columns.



**Fig. 3:** Scatter plot of GOME-2 and SCIAMACHY tropospheric NO<sub>2</sub> columns for April 2007. Data have been binned on a  $1^\circ \times 1^\circ$  grid. In the left panel, all data have been used for both instruments while on the right hand side, only those data are used, for which both instruments had a measurement on the same day.

### Comparison of tropospheric NO<sub>2</sub> columns over China

One region of prime interest for tropospheric measurements from space is the rapidly developing part of China. Using the GOME and SCIAMACHY data sets, a strong and consistent upward trend has been detected for NO<sub>2</sub>, probably because of increasing emissions of NO<sub>x</sub> from industry, power generation and transportation (Richter *et al.*, 2005, van der A *et al.*, 2006). In Fig. 4, data from GOME-2 and OMI are included in this time series.



**Fig. 4:** Time series of tropospheric NO<sub>2</sub> columns above East Central China for GOME, SCIAMACHY, OMI and GOME-2. The consistency between the data sets is very good, indicating a continuing increase in NO<sub>2</sub> columns. Lower OMI values in summer are in agreement with results of previous studies.

For this region with very large signals, GOME-2 and SCIAMACHY measurements agree nearly perfectly. This gives further confirmation for the SCIAMACHY observations, in particular as no attempt was made to correct GOME-2 data for the much better sampling (nearly daily coverage of the area). This indicates that even at their low sampling rate, SCIAMACHY data can be considered to be representative for long-term averages over China.

Interestingly, OMI data also match the other two time series quite well. The OMI data shown here are based on the operational slant columns provided by NASA which have then been analysed using the reference sector method and airmass factors identical to the other data sets. There is some indication for lower OMI data in summer, in agreement with a diurnal variation in NO<sub>x</sub> emissions () but the effect is relatively small on an annual average.

All three data sets agree in that NO<sub>2</sub> columns are still increasing over this area in spite of measures taken to reduce specific NO<sub>x</sub> emissions, probably because of continuing growth in the number of cars and production of cement and energy (*Zhang et al., 2007*). In how far changes in aerosol loading are also relevant through their impact on the light path is still a subject of further investigations.

## References

- Boersma, K. F., D. J. Jacob, H. J. Eskes, R. W. Pinder, J. Wang, and R. J. van der A, Intercomparison of SCIAMACHY and OMI tropospheric NO<sub>2</sub> columns: observing the diurnal evolution of chemistry and emissions from space, *J. Geophys. Res.*, **113**, D16S26, doi:10.1029/2007JD008816, 2008
- Richter, A., Burrows, J. P., Nüß, H., Granier, C., Niemeier, U., Increase in tropospheric nitrogen dioxide over China observed from space, *Nature*, **437**, 129-132, doi: 10.1038/nature04092, 2005
- van der A, R. J., D. H. M. U. Peters, H. Eskes, K. F. Boersma, M. Van Roozendael, I. De Smedt, and H. M. Kelder, Detection of the trend and seasonal variation in tropospheric NO<sub>2</sub> over China, *J. Geophys. Res.*, **111**, D12317, doi:10.1029/2005JD006594, 2006
- Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter, A., Burrows, J. P., Uno, I., Jang, C. J., Chen, D., Yao, Z., Lei, Y., NO<sub>x</sub> emission trends for China, 1995–2004: The view from the ground and the view from space, *J. Geophys. Res.*, **112**, D22306, doi:10.1029/2007JD008684., 2007

### **Recent Publications related to AT2 work in the refereed literature**

- Myrookefalitakis, S., Vrekoussis, M., Tsigaridis, K., Wittrock, F., Richter, A., Brühl, C., Volkamer, R., Burrows, J.-P. Kanakidou, M., The influence of natural and anthropogenic secondary sources on the glyoxal global distribution, *Atmos. Chem. Phys.*, **8**, 4965-4981, 2008
- Franke, K., Richter, A., Bovensmann, H., Eyring, V., Jöckel, P., and J. P. Burrows, Ship emitted NO<sub>2</sub> in the Indian Ocean: comparison of model results with satellite data, *Atmos. Chem. Phys.*, **8**, 15997-16025, 2008
- Lee, C., Richter, A., Weber, M., Burrows, J. P., SO<sub>2</sub> retrieval from SCIAMACHY using the weighting function DOAS (WFDODAS) technique: comparison with standard DOAS retrieval, *Atmos. Chem. Phys. Discuss.*, **8**, 10817-10839, 2008
- Celarier, E. A., Brinksma, E. J., Gleason, J. F., Veefkind, J. P., Cede, A., Herman, J. R., Ionov, D., Goutail, F., Pommereau, J.-P., Lambert, J.-C., van Roozendael, M., Pinardi, G., Wittrock, F., Schönhardt, A., Richter, A., Ibrahim, O. W., Wagner, T., Bojkov, B., Mount, G., Spinei, E., Chen, C. M., Pongetti, T. J., Sander, S. P., Bucsela, E. J., Wenig, M. O., Swart, D. P. J., Volten, H., Kroon, M., and Levelt, P. F., Validation of Ozone Monitoring Instrument nitrogen dioxide columns, *J. Geophys. Res.*, **113**, D15S15, doi:10.1029/2007JD008908, 2008
- Gil, M. Yela, M., Gunn, L. N., Richter, A., Alonso, I., Chipperfield, M. P., Cuevas, E., Iglesias, J., M. Navarro, M., Puentedura, O., Rodríguez, S., NO<sub>2</sub> climatology in the northern subtropical region: diurnal, seasonal and interannual variability, *Atmos. Chem. Phys.*, **8**, 1635-1648, 2008
- Brinksma, E., G. Pinardi, R. Braak, H. Volten, A. Richter, A. Schönhardt, M. Van Roozendael, C. Fayt, C. Hermans, R. Dirksen, T. Vlemmix, A.J.C Berkhouw, D.P.J. Swart, H. Oetjen, F. Wittrock, T. Wagner, O. W. Ibrahim, G. de Leeuw, M. Moerman, L. Curier, E. A. Celarier, W. H. Knap, J. P. Veefkind, H.J. Eskes, M. Allaart, R. Rothe, A. J. M. Piters, and P. Levelt (2008), The 2005 and 2006 DANDELIONS NO<sub>2</sub> and Aerosol Intercomparison Campaigns, *J. Geophys. Res.*, **113**, D16S46, doi:10.1029/2007JD008808.
- Lee, C., Richter, A., Lee, H., Kim, Y. J., Burrows, J. P., Lee, Y. G., Choi, B.C., Impact of transport of sulfur dioxide from the Asian continent on the air quality over Korea during May 2005, *Atmospheric Environment*, **42**, 1461–1475, 2008
- Schönhardt, A., Richter, A., Wittrock, F., Kirk, H., Oetjen, H., Roscoe, H. K. and Burrows, J. P., Observations of iodine monoxide (IO) columns from satellite, *Atmos. Chem. Phys.*, **8**, 637-653, 2008
- Konovalov, I., Beekmann, M., Burrows, J. P., Richter, A., Satellite measurement based estimates of decadal changes in European nitrogen oxides emissions, *Atmos. Chem. Phys. Discuss.*, **8**, 2013-2059, 2008
- Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter, A., Burrows, J. P., Uno, I., Jang, C. J., Chen, D., Yao, Z., Lei, Y., NO<sub>x</sub> emission trends for China, 1995–2004: The view from the ground and the view from space, *J. Geophys. Res.*, **112**, D22306, doi:10.1029/2007JD008684., 2007
- He, Y., Uno, I., Wang, Z., Ohara, T., Sugimoto, N., Shimizu, A., Richter, A., Burrows, J. P., Variations of the increasing trend of tropospheric NO<sub>2</sub> over central east China during the past decade, *Atmospheric Environment*, **41**, 4865–4876, 2007

### **Recent Publications related to AT2 work in the conference literature**

- A. Richter et al. Long-term evolution of satellite derived tropospheric NO<sub>2</sub> fields, *Quadrennial Ozone Symposium, Tromsoe, June / July 2008*
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- J. Leitão et al., Impact of aerosol on the NO<sub>2</sub> airmass factors used for satellite retrievals, *EGU – General Assembly 2008, Vienna, Austria, 13th to 19th April 2008*
- J. Leitão et al., Sensitivity of satellite NO<sub>2</sub> retrievals to aerosols, *4th DOAS International Workshop 2008 for Environmental Research and Monitoring, Hefei, China, 31st March to 2nd April 2008*
- A. Richter et al., Tropospheric Composition Change observed from Space, *2nd ACCENT Symposium, Urbino, Italy, July 23 - 27, 2007*
- M. Vrekoussis et al., Global observations of Glyoxal (CHOCHO) from space, *M. Vrekoussis et al., Aerosols - Properties, Processes and Climate (APPC), An ESF - INTROP conference, Crete, Greece, April 22-24, 2007*
- M. Vrekoussis et al., Long-term measurements of glyoxal (CHOCHO) and formaldehyde (HCHO) from space, *EGU meeting, April 2007*
- C. Lee et al., Impact of transport of sulfur dioxide from the Asian continent on air quality over Korea in May 2005, *EGU meeting, April 2007*
- A. Schönhardt et al., First observations of iodine oxide columns from satellite, *EGU meeting, April 2007*
- A. Richter et al., How large is the influence of tropospheric signals on SCIAMACHY limb measurements?, *DPG Frühjahrstagung, March 2007*

A. Schönhardt et al., MAXDOAS measurements from the DANDELIONS campaigns, *DPG Frühjahrstagung*,  
March 2007