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 the two UV/visible instruments GOME and SCIAMACHY has been analysed for tropospheric NO_2 columns. The data have been compared for consistency in the time period of overlapping measurements, and good general agreement was found. Two effects have been identified that need to be accounted for to avoid inaccuracies when comparing tropospheric NO2 measurements from different platforms: spatial resolution and spatial sampling.

Data from the SCIAMACHY instrument were used to identify NO_x emissions from international shipping and to estimate the total emissions which are in good agreement with recent bottom up inventories.

The long time series from GOME was used to study changes of NO_2 columns in particular over China, and a significant increase has been observed which probably is related to increasing emission.

Introduction

Nitrogen oxides (NO_x) play an important role in tropospheric chemistry. They are a key catalyst in tropospheric ozone formation, and also contribute to acid rain and radiative forcing. Sources of NO_x are both natural (soil emissions, wild fires, lightning) and anthropogenic (mainly fossil fuel combustion and biomass burning).

Starting with the GOME instrument, a series of space-borne UV/visible nadir viewing spectrometers will be operated throughout the next years which provide data that can be used to retrieve tropospheric NO_2 columns. These instruments will create a data base of measurements eventually spanning several decades of atmospheric observations. The resulting data set should be a valuable tool to asses changes in atmospheric constitution, and could be used for monitoring the effects of anthropogenic activities and changes in emission patterns.

The key to a combined use of measurements from different instruments is the creation of a consistent and continuous time series. Therefore, comparison of parallel measurements and investigation of the impact of instrument specific parameters is the first step towards a useful data set.

Scientific activities

Analysis of SCIAMACHY data

The retrieval of tropospheric NO2 measurements from SCIAMACHY follows closely the approach taken for GOME [Richter and Burrows, 2002] with the improvements discussed in [Nüß, 2005] and [Richter et al., 2005]. Two main differences still exist between the two data analyses. The first is the lack of correction for longitudinal inhomogeneities in the stratospheric NO2 fields, which is based on SLIMCAT [Chipperfield et al., 1999] model data in the GOME data analysis but is not yet performed on SCIAMACHY data. The second difference is the cloud fraction criterion used, which is 20 % cloud fraction from FRESCO [Koelemeijer et al. 2001] in the case of GOME and a simple intensity threshold for SCIAMACHY. FRESCO cloud fractions for SCIAMACHY are available from KNMI, but only for the subset of data for which lv1 products have been generated. As a large part of the SCIAMACHY NO_2 data used in this project are based on lv0 data, using FRESCO data is not yet an option. However, the intensity threshold was selected to be comparable to a FRESCO cloud fraction of 20%.

Using this SCIAMACHY NO_2 data retrieval, all available lv0 and lv1 data from August 2002 to present have been analysed and are available as images on the internet (www.doas-bremen.de).

Comparison of GOME and SCIAMACHY measurements

Parallel measurements of GOME and SCIAMACHY are available for the time period from August 2002 to June 2003. After that point, GOME coverage is much reduced as the result of a failure of the last tape recorder on ERS-2, and although a substantial number of GOME measurements over polluted regions in the Northern hemisphere is still available, the lack of measurements over the Pacific necessitates scaling assumptions that make is a less independent product than the original data set.

In the first year of this project, the effects of spatial resolution and spatial sampling have been studied and first direct comparisons been performed.

Resolution effects

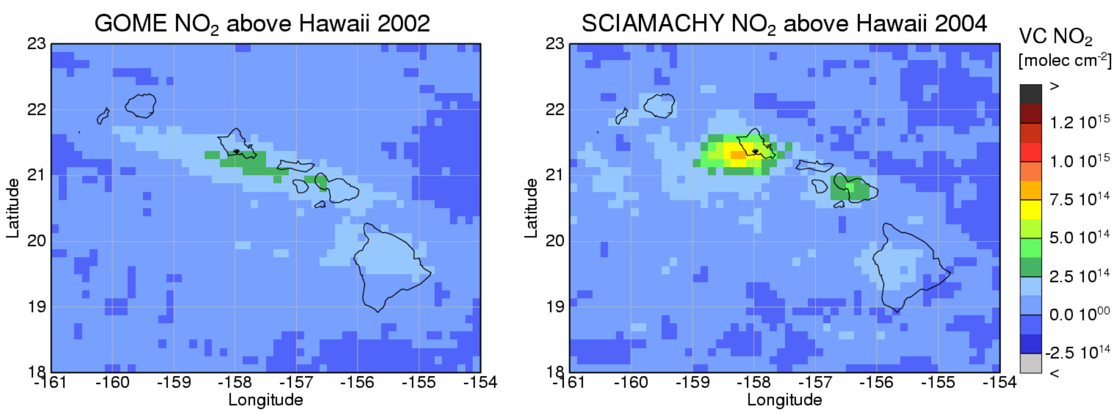

Figure RICHTER_PI_1: Annual averages of tropospheric NO_2 over Hawaii. GOEM data are for 2002, SCIAMACHY data for 2004. The apparent shape of the NO_2 plume in the GOME data is related to the GOME pixel which is 320 km wide and slightly slanted.

Improved spatial resolution has a number of important consequences for tropospheric product. First of all, sources of short lived species can be identified with better accuracy. Secondly, local hot spots such as point sources which might be lost through spatial averaging can be identified and thirdly the probability for cloud free scenes increases with decreasing pixel size. As an example, annual averages over Hawaii are shown in Figure RICHTER_PI_1 for GOME (320 x 40 km^2) and SCIAMACHY (60 x 30 km^2) measurements. Clearly the much more consistent spatial distribution and the higher local values can be seen. It is also interesting to note that the approach used in several studies to estimate NO_2 lifetime from the decay in a downwind plume will provide different answers when applied to the two data sets.

Sampling effects

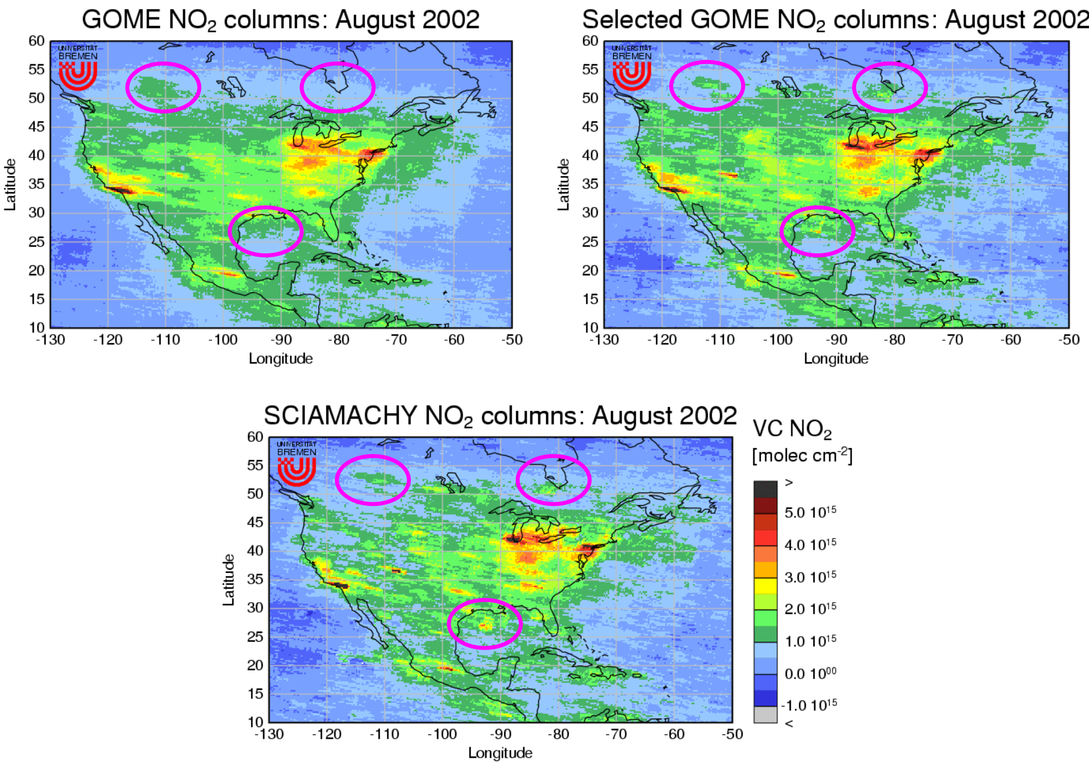

Figure RICHTER_PI_02: GOME and SCIAMACHY monthly averages of tropospheric NO_2 for August 2002. The top left panel is the average using all available GOME measurements, the top right images shows the results if only those GOME data for which a corresponding SCIAMACHY measurement exists are used. The improvement in agreement with the SCIAMACHY measurements is apparent in many locations, some of which are marked.

Over most parts of the world, low earth orbit (LEO) satellites take one measurement per day or less. In the case of GOME, global coverage at the equator is achieved every 3 days, while for SCIAMACHY it takes 6 days as a result of the limb-nadir chess board pattern of measurements. If cloud screening is applied to the data as is necessary for most tropospheric products, the number of measurements per location used in a monthly average is often small, and differences in sampling of two instruments might introduce significant differences in the mean values obtained. As an example, monthly averages are shown in Figure RICHTER_PI_02 for tropospheric NO_2 over North America in August 2002 for both GOME and SCIAMACHY. As can be seen, the average using all GOME data differs in several places from the average obtained when only GOME data with matching SCIAMACHY data are used. Overall the agreement improves substantially when consistent data sampling is used.

This is relevant for cross-validation of instruments, but also has important consequences for satellite measurements of tropospheric species in general. Clearly, temporal sampling of current instruments is not sufficient to resolve even large spatial structures in the NO_2 fields, and much better coverage is needed to retrieve a fully representative NO_2 distribution. This can be obtained by a large number of identical LEO satellites or more simply by observations from geostationary orbit.

Correlation

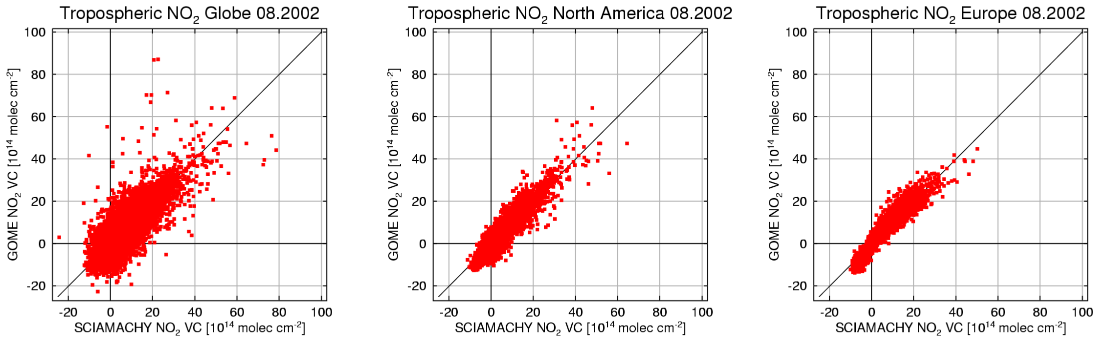

Figure RICHTER_PI_03: Scatter plots of GOME and SCIAMACHY tropospheric NO2 columns for August 2004 for all data (left), North America (middle) and Europe (right). Data have been gridded on a 1 x 1 degree grid for comparison. The scatter of values is significant, but relatively small over areas with large anthropogenic emissions. The differences are probably mainly related to clouds and the difference in time of emasurement.

The results of a direct correlation of GOME and SCIAMACHY tropospheric columns on a 1 x 1 degree latitude longitude grid are shown in Figure RICHTER_PI_03. GOME data have already been selected for matching SCIAMACHY pixels as discussed above. For the global data set, the scatter is substantial and in part originates from measurements over water and at higher southern latitudes. Over areas with strong anthropogenic pollution, the agreement between the two instruments is good.

Scientific results and highlights

Detection of ship emissions in SCIAMACHY NO_2 measurements

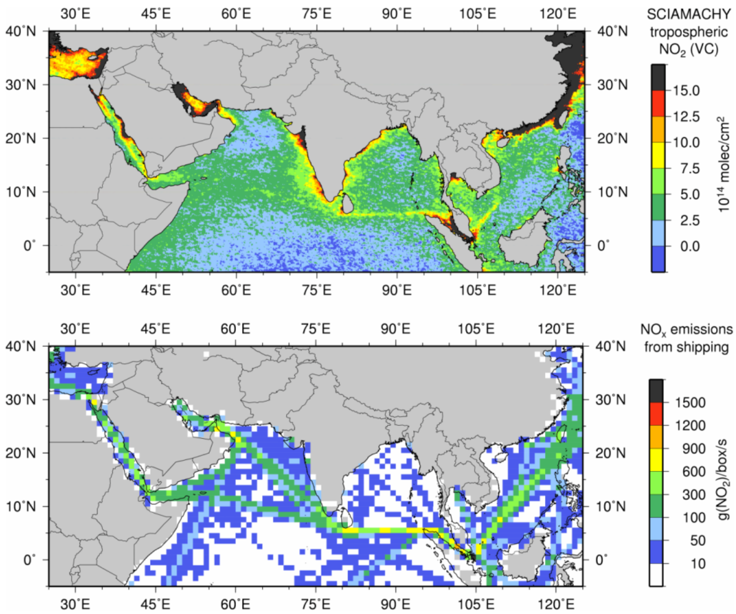

Figure RICHTER_PI_04: NO_x signature of shipping in the Red Sea. Top: Tropospheric NO_2 columns derived from SCIAMACHY data from August 2002 to April 2004 , bottom Estimated distribution of NO_x emissions by shipping in the same region. Figure from Richter et al., 2004.

Measurements from SCIAMACHY have been analyzed for tropospheric NO_2 signatures of shipping emissions. Clear indication for NO_2 produced from ship emissions has been found over the Red Sea and along the main shipping lane to the southern tip of India, to Indonesia and north towards China and Japan where the signal is lost (Figure RICHTER_PI_04). Using simple assumptions for the NO_x loss, emission strengths were estimated and compared to an updated ship emission inventory. Good agreement was found in the spatial distribution and the absolute values for the Red Sea agree within a factor of 2, but larger discrepancies exist in other areas. Although the fluxes calculated still have large uncertainties, the results highlight the importance of ship emissions for the marine boundary layer and at the same time demonstrate the potential of satellite observations. Details of the study can be found in [Richter et al., 2004].

Observation of NO_2 column changes in GOME measurements

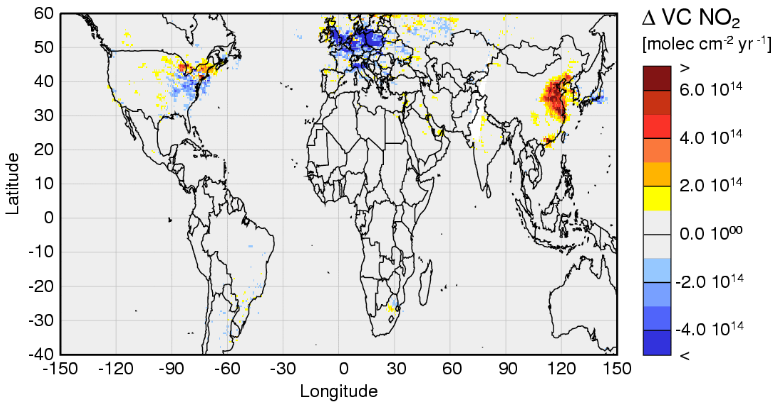

Figure RICHTER_PI_05: The gradient obtained from a linear regression of the annual averages of tropospheric GOME NO_2 columns, retrieved close to 10.30 am LT from 1996 to 2002. Reductions in NO_2 are observed over Europe and the Central East Coast of the United States, while large increases are evident over China. Figure from Richter et al., 2005.

Whereas NO_x concentrations in many industrialised countries are expected to decrease, as a result of emission control and technical improvements in fossil fuel combustion, rapid economic development in parts of Asia has the potential to increase significantly the emissions of NO_x. In order to identify possible changes in NO_x emissions by their corresponding changes in NO_2 concentrations, the tropospheric column amounts of NO_2 retrieved from GOME and SCIAMACHY have been studied over the years 1996-2004. The results show substantial reductions over some areas of Europe and the USA but a highly significant and accelerating increase of about 50% over the industrial areas of China. An overview of the results is shown in Figure RICHTER_PI_05, where the slope of a linear fit through all GOME measurements from 1996 to 2002 is shown. While in most areas the changes are smaller than the estimated uncertainty of about 1 10^{14} molec/cm^2 per year large increases are readily seen over China, while NO2 over Europe and parts of the US has a downward tendency. More details are given in [Richter et al., 2005].

Future outlook

In the next year, data analysis of SCIAMACHY measurements will continue and the sources of the still existing scatter between GOME and SCIAMACHY measurements will be analysed. OMI measurements will also become available shortly and will be included in the comparisons.

One focus of work in the near future is improving the validation of GOME and SCIAMACHY NO_2 columns by both airborne and ground-based measurements.

References

Chipperfield, M. P.: Multiannual Simulations with a Three-Dimensional Chemical Transport Model, *J. Geophys. Res.*, **104**, 1781–1805, (1999).

Koelemeijer, R. B. A., Stammes, P., Hovenier, J. W. & de Haan, J. F., A fast method for retrieval of cloud parameters using oxygen A band measurements from the Global Ozone Monitoring Experiment, *J. Geophys. Res.,* **106**, 3475-3490, (2001).

Nüß, J. H.: An improved tropospheric NO_2 retrieval for GOME and SCIAMACHY, PhD Thesis, University of Bremen, (in German), (2005).

Richter, A. and J.P. Burrows, Retrieval of Tropospheric NO_2 from GOME Measurements, *Adv. Space Res.*, **29(11)** ,1673-1683, 2002 .

Richter, A., V. Eyring, J. P. Burrows, H. Bovensmann, A. Lauer, B. Sierk, and P. J. Crutzen, Satellite Measurements of NO_2 from International Shipping Emissions, *Geophys. Res. Lett.*, **31**, L23110, doi:10.1029/2004GL020822, (2004).

Richter, A., Burrows, J. P., Nüß, H., Granier, C., Niemeier, U., Significant increase in nitrogen dioxide levels over China observed from space, *submitted to Nature*, (2005).