Integrated analysis of long-range pollution transport to mid- and high- latitudes over Europe using model simulations, satellite observations, and aircraft measurements (INTAS)

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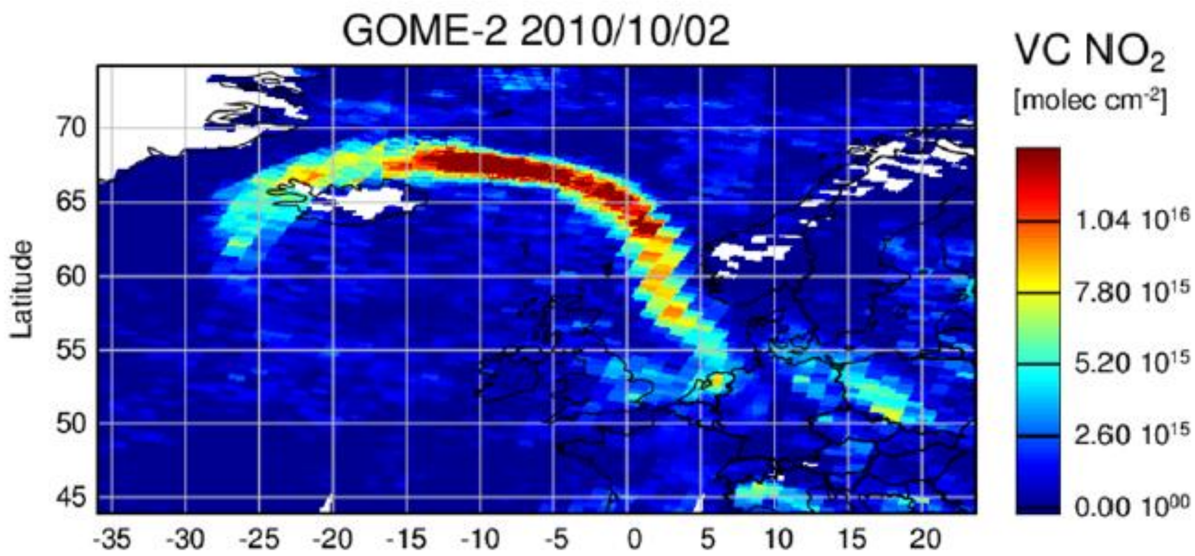

1 General Information

1.1 DFG reference number

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1.2 Applicant

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1.4 Topic of the project

Integrated analysis of long-range pollution transport to mid- and high- latitudes over Europe using model simulations, satellite observations, and aircraft measurements (INTAS)

1.5 Period covered by the report and overall funding period

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1.6 Peer reviewed publications

Zien, A. W., Richter, A., Hilboll, A., Blechschmidt, A.-M., and Burrows, J. P.: Systematic analysis of tropospheric NO_2 long-range transport events detected in GOME-2 satellite data, *Atmos. Chem. Phys.*, **14**, 7367-7396, doi:10.5194/acp-14-7367-2014, 2014.

1.7 Posters

Zien, A.; Richter, A.; Hilboll, A., Burrows, J.P., Systematic quantitative analysis of NO2 long-range transport events and comparison to model data, *AGU Fall meeting, San Francisco, USA, December 2012*

Zien, A., Richter, A., Hilboll, A., Burrows, J.P., Comparison of NO2 long-range transport events in GOME-2 observations and CTM simulations, *EGU General Assembly, Vienna, Austria, April 2012*

Leitão, J., Hilboll, A., Richter, A., Zien, A., Burrows, J.P., Aerosol Effects on Satellite Observations of NO2 Pollution, *3rd Urbino Sympsium, Urbino, Italy, 13 - 16 September 2011*

Richter, A., Wittrock, F., Hilboll, A., Leitão, J, Zien, A., Vrekoussis, M., Burrows, J.P., Long-term observations of pollution from space, *IUGG 2011, Melbourne, Australia*, 28 June - 7 July 2011

Richter, A., Hilboll, A., Zien, A., Burrows, J.P., Cloud effects in satellite observed tropospheric NO2, *EGU General Assembly, Vienna, Austria,* April 2011

Richter, A., Leitão, J., Hilboll, A., Zien, A., Burrows, J.P., Satellite observations of biomass burning NO2, *DPG Spring meeting, Dresden, Germany,* March 2011

Zien, A., Richter, A., Hilboll, A., Burrows, J.P., Remote sensing trace gas observations by satellite instruments over bright surfaces, *DPG Spring meeting, Dresden, Germany,* March 2011

Zien, A., Richter, A., Hilboll, A., Burrows, J.P., Sensitivity of satellite observations over bright and cloudy scenes, *COSPAR 38th scientific assembly, Bremen, Germany*, 18 - 25 July 2010

Richter, A., Hilboll, A., Zien, A., Burrows, J.P., GOME-2 satellite observations of NOx emissions from ships, *EGU General Assembly 2010, Vienna, Austria,* 2nd to 7th May 2010

Richter, A., Hilboll, A., Zien, A., Burrows, J.P., GOME-2 satellite observations of NOx emissions from ships, *DPG Spring meeting, Hannover, Germany*, March 2010

Zien, A., Richter, A., Hilboll, A., Burrows, J.P., Cloud effects on tropospheric NO2 measurements from satellite, A. Zien, *DPG Spring meeting, Hannover, Germany*, March 2010

All posters can be downloaded at http://www.doas-bremen.de/poster_gallery.htm.

2 Final Progress Report

2.1 Project's initial questions and objectives

The initial main objective of the project was to investigate the import of pollution from North America to Europe using a combination of in-situ measurements, models, and satellite data. The focus was on identification and analysis of different pollution sources and transport pathways and their effects on air quality and radiative forcing.

More specifically, the IUP Bremen objectives within the project were to

- Provide satellite data for an integrated analysis with aircraft observations and modeling for investigations of LRT of pollution;
- Develop improvements of SCIAMACHY retrievals for conditions with elevated pollution plumes and clouds, in particular over ice and snow;
- To use modelled vertical distributions as a priori for SCIAMACHY data analysis
- To validate SCIAMACHY retrievals using airborne observations

2.2 Project developments

In the first phase of the project, an attempt was made to collect in-situ airborne data for NO_2 long-range transport (LRT) events which can be used for satellite validation. It soon became clear that there are very few data sets of actual NO_2 measurements available, and even when using NO measurements and applying steady state or model predicted NO_2 / NOx ratios, there are basically no vertical profiles through LRT events available that can be used for validation purposes.

A second problem was encountered when comparing NO_2 columns during LRT events identified in SCIAMACHY measurements to data from 3d-CTMs (first GEOS-chem, later the MACC global forecast and reanalysis model runs). While there are similarities of the spatial structures observed on some days, the spatial mismatch is often large, and in many cases, the modelled and observed LRT events do not match. This is also true for clouds which are often present during LRT events and which are not well represented in models, in particular in global models with low spatial resolution.

Based on these findings, the focus of the project was shifted to using mainly satellite data without model input and unfortunately also without external validation. As at that time GOME-2 data had become available which provide better spatial coverage than SCIAMACHY, it was decided to use GOME-2 observations. As one of the benefits of satellite data is their global nature, the study area was not limited to Europe but included the full globe. In order to evaluate the large data set, an objective and automatic detection of LRT plumes is needed.

Initial analysis of GOME-2 data showed, that many of the LRT events which are apparent in the data occur for cloudy conditions. These data are not used in the standard satellite data products, and NO_2 a priori profiles and air mass factors normally used cannot be applied.

Considering the above mentioned points, the revised project objectives were

- Development of an objective LRT plume identification algorithm working on GOME-2 (and other) satellite data
- Development of appropriate cloud treatment and air mass factors
- Statistical analysis of the resulting data set

2.3 *Main results*

2.3.1 Development of an objective LRT plume identification algorithm working on GOME-2 (and other) satellite data

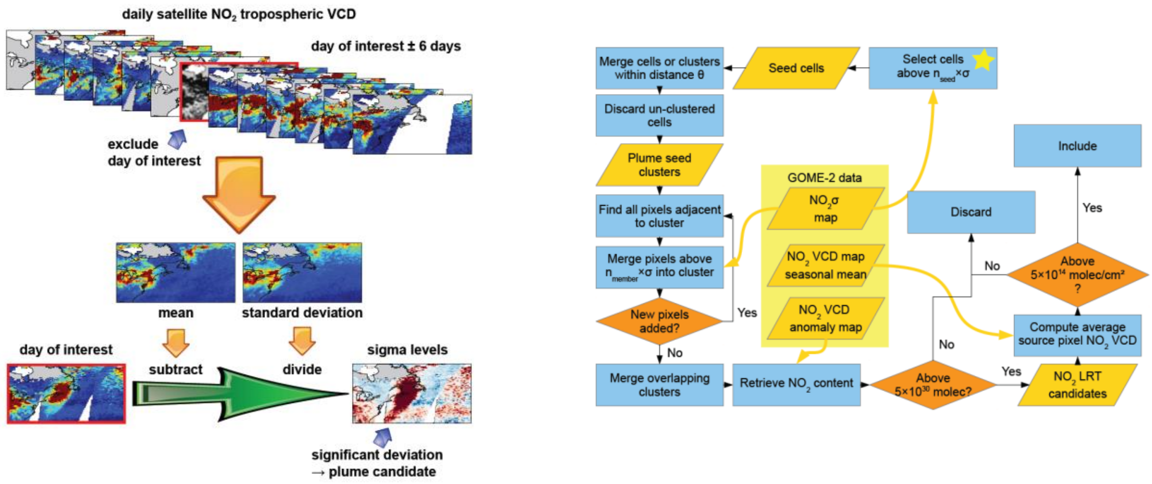


Figure 1: Overview of the steps performed to identify long-range transport events. Left: initial identification of an LRT candidate. Right: Flow chart of steps performed to identify and accept / reject LRT candidates

The LRT plume identification algorithm developed in this work (Zien et al., 2014) is based on the analysis of consecutive days of satellite (here GOME-2) NO_2 column fields. It can be subdivided into several steps (see Figure 1):

1. Identification of candidates for LRT events in a single gridded day of measurements
2. Clustering of all cells associated with this event
3. Filtering of clusters using thresholds and air mass origin

A cell in a daily NO_2 image is identified as a candidate for LRT if its NO_2 column is higher than the sliding mean over +/- n_{days} (excluding the current day) by a value which exceeds the standard deviation by a factor of n_{seed}. This classification is based on the assumption, that LRT events are short term anomalies in the daily NO_2 observations. This is a good assumption over the oceans only and therefore pixels over land are excluded from the analysis. To reject outliers, only those pixels having other LRT candidates in a vicinity of ΔΘ are used. Starting from these seed cells, neighbouring cells having also enhanced values (by a factor of n_{member} which can be smaller than n_{seed}) are clustered with the seed cells to combine all high values into one plume. Only plumes containing a minimum number of $n_{min, molec}$ NO_2 molecules are considered. For our analysis, we chose the following constants: n_{days} = ±6, n_{seed} = 3, n_{member} = 2, ΔΘ = 1.0°, and $n_{min, molec}$ = $5 \times 10_{30}$ molecules.

Once the LRT cells have been identified and clustered, backward trajectories using the NOAA HYSPLIT model are initiated at altitudes between 1000 and 6000m in 500m steps to identify possible source regions. For each altitude, the 120 h backward trajectories are calculated and the number of steps is counted for which the backward trajectory is at or below 1000m, the assumed BL height. The altitude with the largest number of hits is selected as most probable height of the NO_2 plume. Finally, the "climatological" NO_2 column for all the source pixels of this altitude is determined based on the long-term average of all GOME-2 measurements between 2007 and 2001, and only if it exceeds a threshold of 5×10^{14} molec cm^{-2}, a plume is classified as long range transport of pollution event.

This limitation to known source regions is applied to reduce the number of false positives, for example from lightning or from imperfect removal of stratospheric variations in the satellite data. As it is based on GOME-2 data, all regions acting regularly as source regions will be accepted, including both anthropogenic and biomass burning sources. However, exceptional events might be excluded by this filter.

A practical example of the detection, clustering and source region identification is shown in Figure 2 for an NO_2 export event from North America. As this algorithm is fully objective and works without user interaction, it could be applied to the full 5 year time series of GOME-2. Results are discussed in section 2.3.3.

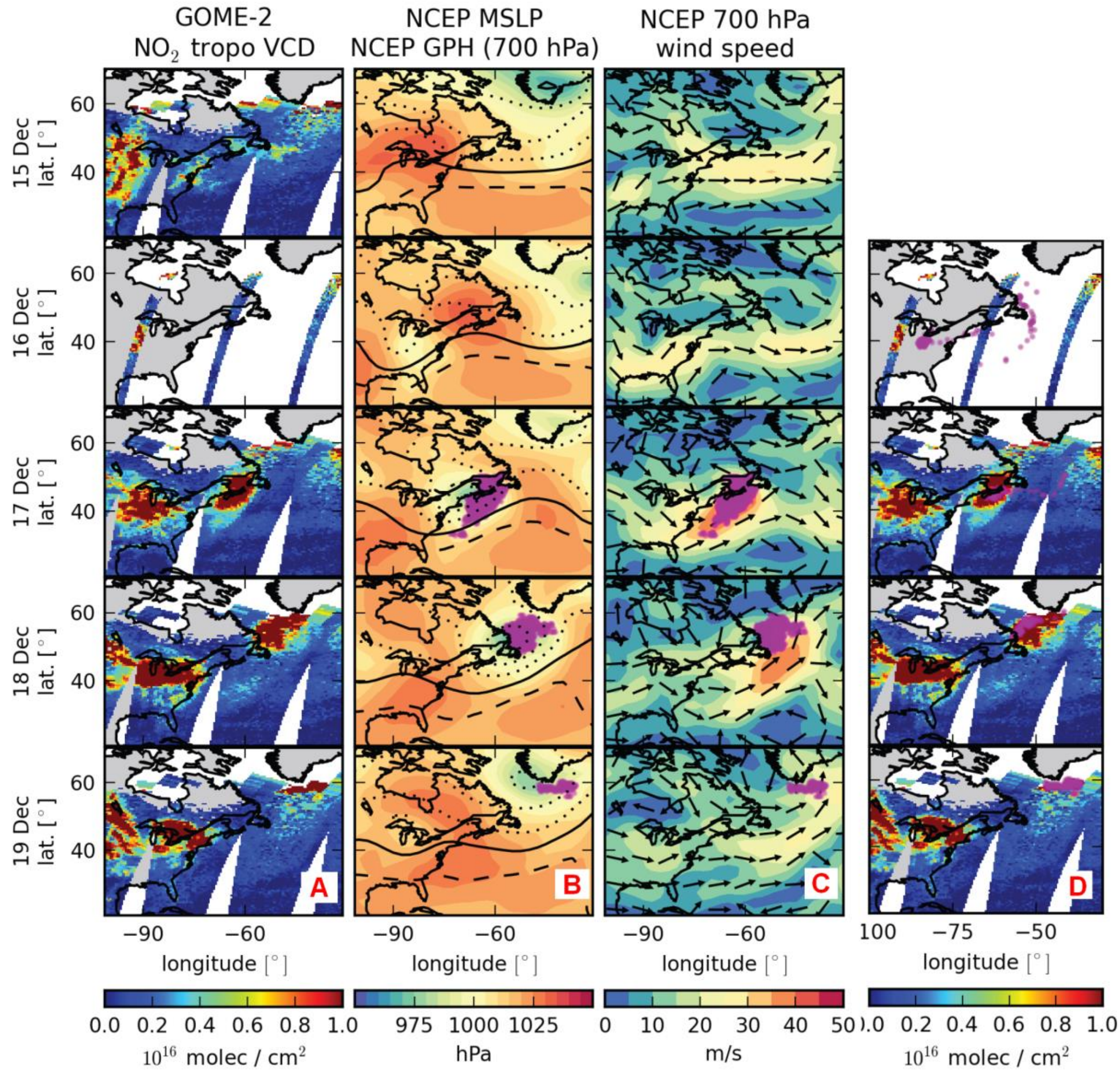


Figure 2: Time series of the days preceding and during a long-range transport event over the North Atlantic on 17 to 19 December 2007. The locations of satellite pixels identified as belonging to the long-range transport plume are indicated by purple circles in the two centre columns. Shown are (A) the GOME-2 NO_2 tropospheric vertical column density, (B) the NCEP DOE AMIP-II (National Centers for Environmental Prediction Atmospheric Model Intercomparison Project II) reanalysis mean sea-level pressure (colours) and geopotential height at 700hPa (contours) and (C) horizontal wind speeds at 700hPa (amplitude and direction). For geopotential height, the solid line denotes 3km, dashed/dotted indicate higher/lower geopotential height in steps of 125m. A low pressure system is quickly evolving into a cyclone. It elevates an NO_2 plume – as seen in its back trajectories – and transports it towards Greenland. IN panel (D), purple circles indicate the locations of the back trajectories of the plume from 19 December 2007 at the respective dates. The plume detected on 19 December 2007 is only partially visible due to polar night. This becomes evident when comparing the back trajectories on earlier dates with the observed NO2 vertical column densities.

2.3.2 Development of appropriate cloud treatment and air mass factors

Initial analysis of NO_2 LRT events in GOME-2 data showed, that many of them are linked to cloudy situations. Therefore, cloud screening, the standard approach used at IUP Bremen in the retrieval of tropospheric NO_2 is not applicable here. In order to quantify the NO_2 amount in cloudy situations, assumptions need to be made on the NO_2 vertical profile but also on the vertical cloud profile. Originally, it was planned to use data from atmospheric models for this purpose. However, already for the first few examples, model and measurements disagreed strongly in position and size of the NO_2 columns when using cloud and NO_2 a priori from the models (first GEOS-chem, then MACC reanalysis). Regional models have the potential to perform better in such situations, but they do not provide global data. Therefore, it was decided to use simple assumptions in the analysis and to investigate the uncertainty introduced by these assumptions.

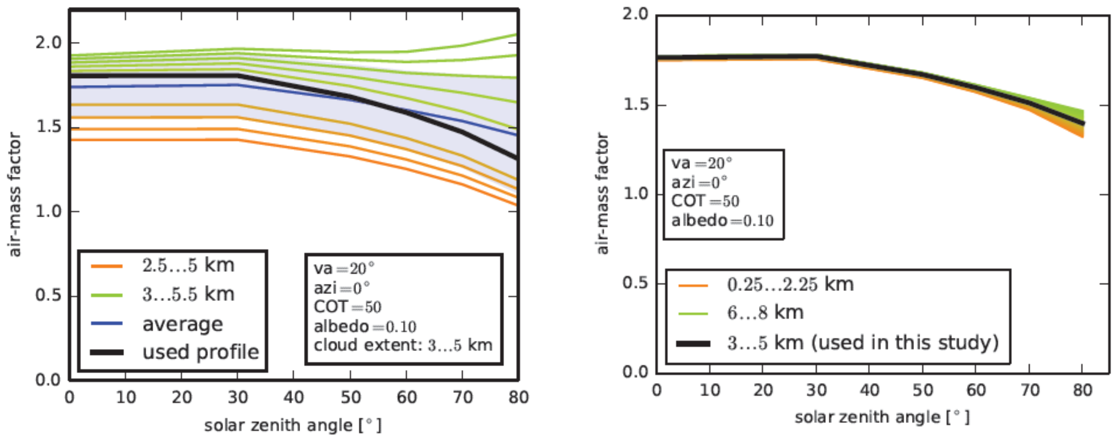


Figure 3: Left: Illustration of the air-mass factor dependency on the solar zenith angle for varying NO_2 profiles and an invariant cloud profile. The black line shows the profile used for this study. The ensemble average and the standard deviation are indicated in blue. Right: sample air-mass factor dependency on solar zenith angle for NO_2 fully mixed inside clouds at varying altitudes. The black line indicates the cloud and NO_2 profile used in this study. A cloud vertical extent of 3. . .5km, a surface albedo a = 0.1, a viewing angle va = 20°, relative azimuth angle azi = 0° (forward direction) and cloud optical thickness COT = 50 were used.

The assumptions made are

1. NO_2 is well mixed through the cloud
2. There is no NO_2 below or above the cloud
3. Outside the cloudy area, the vertical profile of the NO_2 is the same as inside the cloud
4. All clouds extend from 3 – 5 km

While these assumptions are certainly not always valid, they are at least simple and easy to evaluate. In order to investigate the uncertainties introduced by these assumptions, a series of sensitivity tests have been performed. For example, the NO_2 profile has been stretched to extend to above or below the cloud (see Figure 3a), resulting in changes of the AMF by +/- 15%. To some extent, the (poorly visible) NO_2 below the cloud is compensated by the amplification of any signal from NO_2 above the cloud, at least at solar zenith angles below 70°. Another test is shown in Figure 3b, where the change in AMF is displayed that results from varying the altitude level at which the cloud is positioned. Here, changes are of the order of less than 3%, indicating that cloud altitude is not the main source of uncertainty.

How realistic the assumption of NO_2 being well mixed within the cloud is cannot be judged as basically no NO_2 vertical profiles have been measured in such situations. In Figure 4, an example is shown from the MACC reanalysis model (Inness et al., 2013), where GOME-2 observations and model NO_2 distribution are similar during an LRT event. As can be seen, the three vertical profiles taken at different points in the plume are quite different, but in each case the NO_2 is well mixed within the cloud and follows the development of the cloud vertical extension. In contrast to the assumptions made in our analysis, the model predicts that enhanced NO_2 is present down to the surface, which will decrease the air mass factor and lead to an underestimation of the total NO_2 amount in the cloud. However, these model results have to be interpreted with care as they depend on details of the parameterisation of vertical mixing and sub-grid processes such as convection. We take this comparison as indication that the assumptions made are not completely unrealistic.

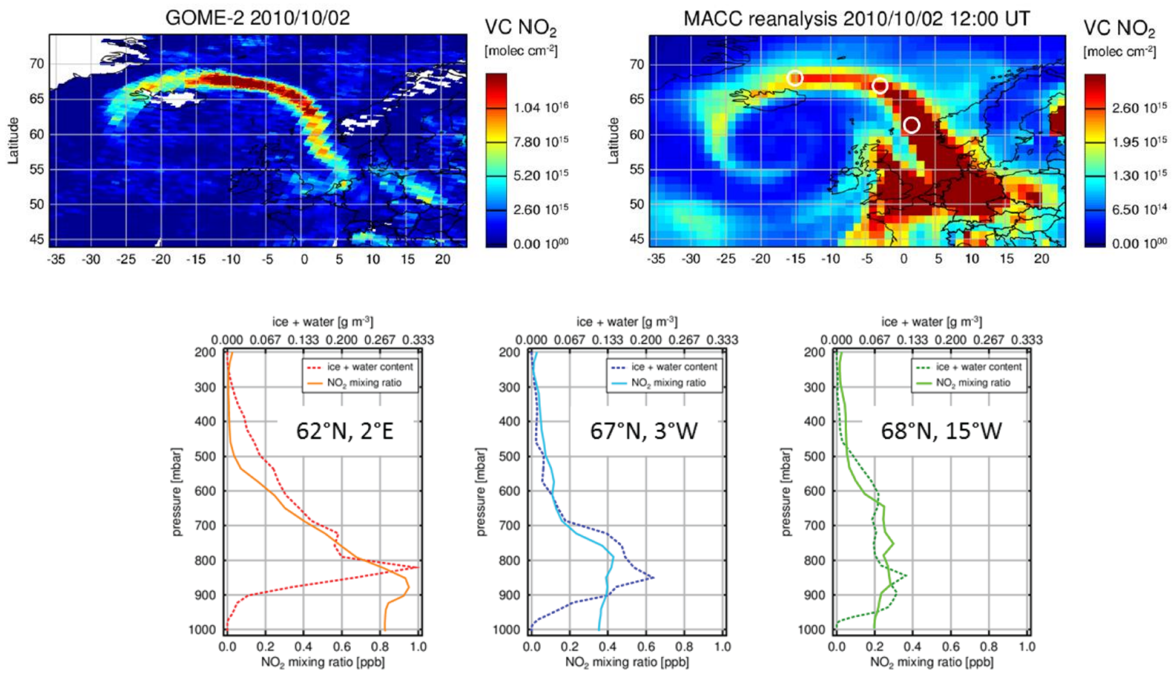


Figure 4: Vertical profiles of NO_2 and cloud ice and water for a LRT example in the MACC reanalysis data. Top left: GOME-2 NO_2 columns, top right: model columns (note change in colour scale). Bottom: three examples of vertical profiles for the locations identified by circles in the model data

In the standard IUP tropospheric NO_2 analysis, the FRESCO+ cloud fraction product is used for cloud screening. Here, we developed a simple cloud fraction algorithm which is based on the intensity in the NO_2 fitting region. As in other cloud algorithms, the surface reflectivity is taken from a climatology, here from MERIS (Popp et al., 2011) and measured reflectivites are compared to values modelled by SCIATRAN (Rozanov et al., 2014) for clear and cloudy situations. The results have been compared to FRESCO+-values obtained from the TEMIS data base (www.temis.nl), and overall, excellent agreement was found. However, there are some clear advantages of the new algorithm:

1. The intensity weights for clear and cloudy pixel are available at the appropriate wavelength for the NO_2 retrieval

2. There are no unrealistic high cloud fractions over deserts (probably as result of the high resolution MERIS reflectivities used),

3. There are less artefacts due to sun glint on oceans (effect is smaller at 450 nm than at 760 nm as used in FRESCO+)

4. The dependence on scan position is smaller, probably due to a mistake in the lookup-tables used in the current FRESCO+ implementation (Y. Huan, personal communication).

In spite of these differences, the effect of choice of cloud fraction algorithm on the global results turned out to be very small.

2.3.3 Statistical analysis of the LRT data set

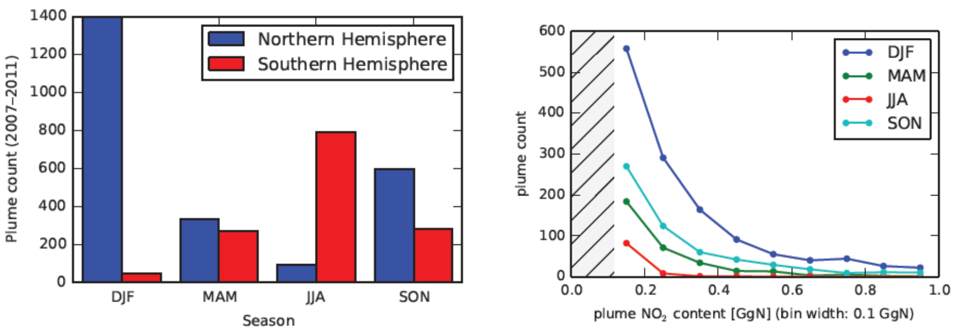


Figure 5: Left: Number of verified long-range transport plumes found by the detection algorithm in 2007–2011. There is a strong seasonality in both hemispheres, with a strong peak in local winter. Right: NO_2 content of long-range transport events in the Northern Hemisphere, for different seasons. Note the lower mass limit of 5×10^{30} molecules (hatched area, corresponding to 0.12 Gg N).

The cloud fraction algorithm and clear and cloudy sky air mass factors as described in section 2.3.2 have been applied to GOME-2 data over the oceans and the plume identification algorithm as described in section 2.3.1 was run for the GOME-2 data set from 2007 – 2011. A total of 3808 verified NO_2 long-range transport events (out of 8626 candidate events) was found, mostly in the northern hemispheric winter. The seasonal distribution in the two hemispheres is shown in Figure 5a. The smallest number of LRT events is found in the respective summers, and overall there are many more events in the northern hemisphere than in the southern hemisphere. As shown in Figure 5b, the mass distribution for the detected LRT events is in good approximation exponential with most plumes being rather small. The largest plumes are found in winter as might be expected from both increased frontal activities and increased NO_2 lifetime. The distributions of both, the seasonality of detections and the size distribution are stable over the entire time period evaluated. Analysis of the most probable plume heights retrieved from the backward trajectories (not shown) revealed the expected patterns – low plume heights on the first day, an increase for the second and third day and little change after that. More than half the plumes remain below 2 km.

When browsing through the GOME-2 NO_2 data manually, long-range transport events tend to appear in particular regions and follow particular paths. With the derived data set, this can be verified in a quantitative manner. In order to visualise these regions, daily maps of NO_2 from identified LRT plumes were created and added up over the full data set. For normalisation, they were then divided by the total number of GOME-2 measurements in the respective cell. The result is shown in Figure 6, separated by season.

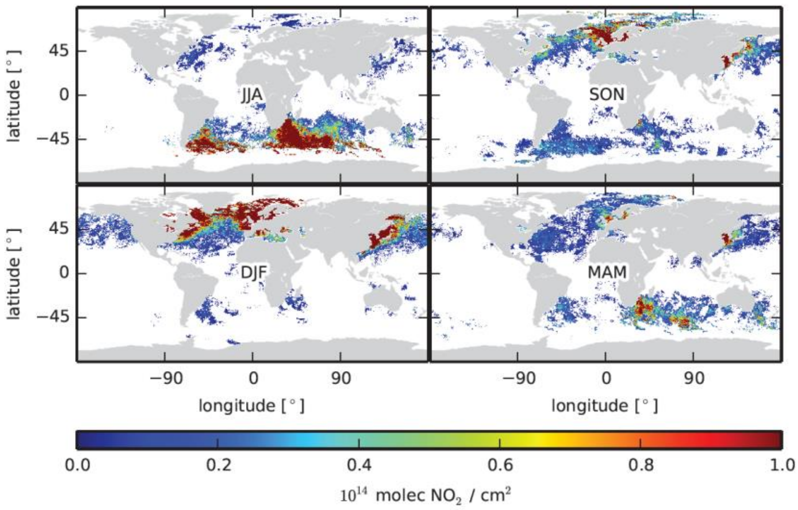


Figure 6: Seasonal maps of the mean vertical column density of NO_2 observed in plumes associated with long-range transport events. Note that vertical column densities near Europe are always higher than columns near North America – at least partially due to its special geography.

Several hotspot areas can be identified: European waters, the North Atlantic, the outflow regions of South Africa and South America and to a lesser degree also the outflow region of East Asia. This is in agreement with previous publications discussing the effects of long range transport of pollution, for example for North America (Stohl et al., 2003), South Africa (Spichtinger et al., 2001, Wenig et al., 2003) or Eckhardt et al. (2003) for Europe. Again, the strong seasonality is apparent indicating that this type of LRT is most effective in winter / fall, probably because of meteorology favouring frontal systems and the lower sun increasing NO_2 lifetime thereby extending the transport range of NO_2. It is also clear that no such events are find at low latitudes which is a combination of less pollution hotspots and different meteorology.

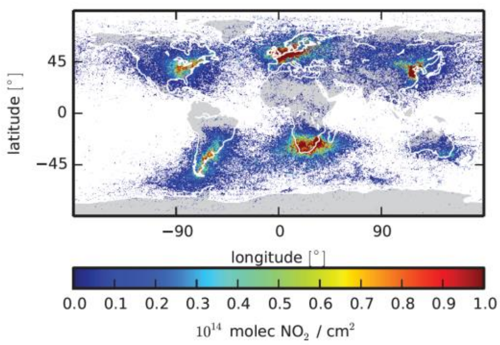


Figure 7: Regional contribution to average NO_2 vertical column densities observed in long-range transport plumes. Sources of most long-range transport events are clearly visible. The high scatter originates from the low resolution of meteorological data used for the back trajectories and uncertainties in the determination of the most likely back trajectory.

As outlined in section 2.3.1, the algorithm determines a source area from which the LRT event originates using back-trajectories. With this information, the contribution of different regions to the NO_2 in LRT events can be calculated. This is shown in Figure 7, highlighting again the expected pollution hot-spots (Europe, Eastern US, China, South Africa). It is interesting to note the relatively small contribution of China when compared to the very high NO_2 columns observed there, and also the absence of signals from the Western US. The disproportionately high contribution from South Africa is linked to the unique position of the Highveld industrial area where coal fired power plants produce large amounts of NO_2 at a high altitude, facilitating frequent injection into the free troposphere and subsequent LRT.

As can be seen in the figure, there also is a significant degree of scatter in the source appointment. This is created by the uncertainties in backward trajectories which over time leads to a spreading of the tracked air parcels. Use of higher resolution meteorological fields would probably reduce this effect but such data was not available during the project.

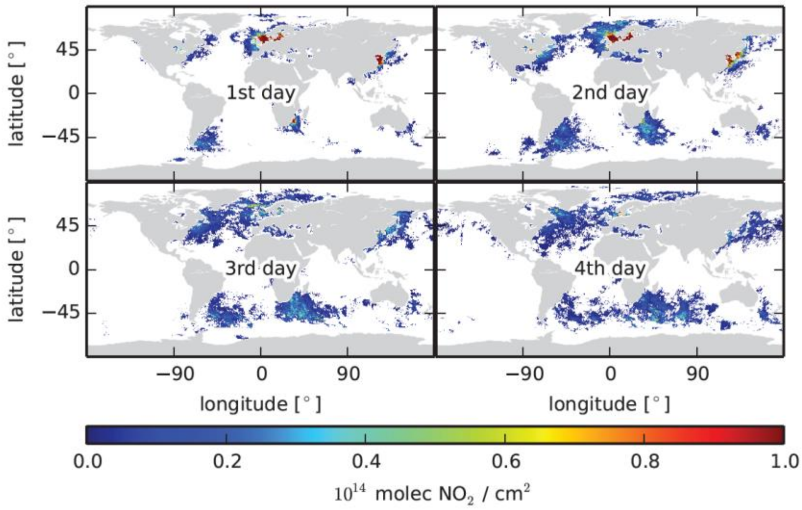
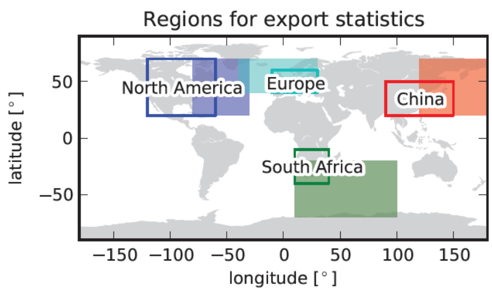


Figure 8: Map of the mean NO_2 vertical column density observed in plumes of long-range transport events, binned by the age of plumes since emission at observation time.

Another interesting piece of information that is provided by the analysis is the evolution of the LRT plumes over time. In Figure 8, the mean NO_2 distribution from LRT events is shown using only data from 1, 2, 3, or 4 days after the estimated emission time. As expected, the distance between emission point and NO_2 plume increases over time in the direction of transport. However, the relative contributions of short-term and long-term plumes varies between regions, with China and Europe having a very large contribution from young LRT events. This is probably linked to the average lifetime of transport events but also to the geography. In Europe, a multitude of sources surrounds the Baltic Sea and North Sea, providing many opportunities for the observation of young LRT plumes. During transport, these plumes can then be moved over the continent again and are then lost in the approach used in this study. In China, transport on the Pacific often returns towards the continent and while it might continue there, it cannot be detect with our method.

To come to a quantitative estimate of the NO_2 resulting from emissions in different source regions, f the NO_2 content of all the plumes in the respective region was summed up and normalize by the number of days of observation. This yields the total yearly export of NO_2 from the continent onto the ocean.

The data in Figure 9 show, that the strongest emitter of plumes is China, followed by Europe. However, the plumes from China do not last as long and do not form such a prominent route. It appears that South Africa experiences the opposite effect: here, plumes follow a very stable and visible route over the open ocean. Therefore, small absolute emissions are being strongly represented in the data set.



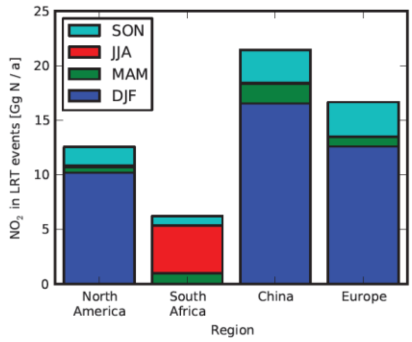


Figure 9: Left: Regions used for the statistical study. Only plumes are considered that were observed over the ocean in the filled rectangle and were found over land-masses in the open rectangle 24 h earlier. Right: Total yearly NO_2 content in long-range transport plumes emitted in the regions shown in left for different seasons.

The total NO_2 outflow of these four regions amounts to more than 50 GgN/a in long-range transport events, which are exported to the ocean and remote regions. This is slightly more than a per mil of the estimated global yearly NOx emission rate of 43 TgN/a. This might appear to be a small fraction, but it constitutes a significant amount for such an unstable gas – especially, as our back tracing algorithm does not take the decay of NO_2 from emission to observation of the plume into account. The main conclusion from this study therefore is, that long range transport of NO_2 is not a rare event but rather a common situation which was so far not appreciated enough in satellite data as transport is often linked to cloudy situations which are routinely excluded from tropospheric satellite NO_2 data analysis.

2.4 Future directions

There are several possible extensions and applications of the algorithm and analysis developed in this project:

1. The rough estimates made for the calculation of air mass factors should be compared to results obtained when employing a priori data from regional models which are expected to provide a better representation of the NO_2 and cloud profiles during LRT events.

2. The analysis should be extended to later GOME-2 years and data from the second GOME-2 instrument. It would also be interesting to include OMI data which would provide information on the evolution of the plumes by adding an afternoon observation. However, additional work is needed to combine data from different sensors having different spatial resolution and observation geometry.

3. The analysis could be applied to model data. In fact, first application to MACC reanalysis data showed a lot of similarities to the results from GOME-2 in the northern hemisphere but much less LRT events in the southern hemisphere. More analysis is needed to understand this result.

4. The detection algorithm could also be applied for the automated identification of other plumes, for example SO_2 from volcanoes or aerosols from biomass burning events.

5. In order to better quantify the NO_2 transport statistics here in terms of NOx or NOy transport, results should be combined with models providing the partitioning of NOx and

NOy within the plume. However, this requires good agreement between modelled and observed LRT events in space and time.

2.5 Exploitation of results

The results of the project are not deemed economically valuable and exploitation in the form of joint ventures or patents is not anticipated.

2.6 Project contributors

This project part was executed at the University of Bremen by Achim Zien (PhD student), Dr. Andreas Richter (project leader) and Prof. Dr. John Burrows (Head of Institute). Individual aspects of the project were supported by Dr. Anne Blechschmidt (meteorology, MACC model) and Dr. Andreas Hilboll (statistical data analysis, GOME-2 data analysis).

The work was performed in co-operation with

- Dr. Hans Schlager, DLR, Institute of Atmospheric Physics (in situ airborne NOx data)
- Dr. Bastien Sauvage, Laboratoire d'Aérologie, Observatoire Midi Pyrenees, France (GEOS-chem model data)

2.7 Qualification of young researchers

During the project, Achim Zien worked on his PhD project which will be submitted by the end of 2014.

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3 Summary

In this project, satellite observations of nitrogen dioxide in the atmosphere have been used to investigate the occurrence of long range transport of pollution over the oceans. Such export of pollution is important as it affects air quality in clean regions, changes background levels of pollutants in large parts of the world and is relevant for international conventions such as LRTAP. Nitrogen dioxide is a tracer of pollution which is mainly produced in the combustion of fossil fuels but also by biomass burning and lightning. As the atmospheric lifetime of NO_2 is short, long range transport is only possible at high wind speed, preferably at low solar irradiation (mid and high latitudes in fall and winter).

In standard satellite products of tropospheric NO_2 there is little evidence for long range transport. This is due to the fact that such transport is often linked to the presence of clouds, and cloudy data is usually excluded from the satellite data sets as in such cases, the instrument does not have an unobstructed view to the surface where most of the pollution is located. Therefore, in this study all NO_2 data from the European GOME-2 satellite instrument have been used and a simplified treatment of the effect of clouds on the detection sensitivity has been developed. It assumes that in long range transport events in the presence of clouds, the NO_2 is well mixed within the cloud. This assumption is supported by some case studies on CO measurements in the atmosphere and NO_2 data from atmospheric models.

Using measurements from several days, long range transport events can be identified in the satellite data using image processing techniques and the assumptions that a) NO_2 plumes from transport are short lived and can therefore be identified by evaluating deviations from the mean values and b) that they are contiguous in space and c) that they can be traced back to regions with elevated NO_2 values. An algorithm based on these principles has been developed and implemented, and a multi-annual data set of GOME-2 measurements has been evaluated, identifying nearly 4000 individual NO_2 transport events over oceans.

Using this data set, a statistical evaluation of NO_2 long range transport events could be performed. The results show, that the main regions affected by NO_2 from long range transport are between the US and Europe, in the outflow of China and East of South America and South Africa. In all regions, most events are observed in fall and winter. For Europe and China, mainly short lived events are observed as NO_2 plumes are often rapidly transported back over the continent where they cannot be detected by the algorithm. While from South Africa and the Eastern US many well defined transport events can be traced in the satellite data, the quantitative NO_2 export is largest from China, followed by Europe. In total, an NO_2 outflow of 50 GgN/a is computed for the four main NO_2 export regions which is small in comparison to total NOx emissions but significant for atmospheric chemistry downwind of the US, Europe, China and South Africa.

The data set created provides a first estimate of global NO_2 transport over the oceans and can be used to evaluate the ability of current atmospheric models to simulate pollution export from the continents. Further refinements with data from other satellite instruments and more detailed modelling of measurement sensitivity in cloudy situations could reduce the uncertainties of the estimates made.