Improvement of GOME NO_2 Retrieval

J. H. Nüß[1], A. Richter[1], B.-M. Sinnhuber[1], F. Wittrock[1], J. P. Burrows[1], U. Niemeier[2], C. Granier[2], J.-F. Müller[3], N. Savage[4], K. S. Law[4], J. A. Pyle[4], J. G. J. Olivier[5], R. Koelmeijer[6]

[1]Institute of Environmental Physics, University of Bremen, Bremen, Germany (Hendrik.Nuess@iup.physik.uni-bremen.de)
[2]Max Planck Institute for Meteorology, Hamburg, Germany, also at Service d'Aeronomie, Paris, France
[3]Belgian Institute for Space Aeronomy, Brussels, Belgium, [4]University of Cambridge, Cambridge, UK
[5]National Institute of Public Health and Environment, Bilthoven, Netherlands; [6]Space Reseach Organization Netherlands, Utrecht, Netherlands

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Introduction

Tropospheric NO_x has its main sources in emissions from the soil, fires, lightning, transport and industry. It plays an important role in the formation of tropospheric ozone and together with SO_2 it is the main cause of acid rain.

The *Global Ozone Monitoring Experiment* (GOME) is a UV/visible spectrometer on board of the European satellite ERS-2. GOME is a 4 channel double monochromator covering the wavelength range of 230 - 800 nm with a spectral resolution of 0.2 - 0.4 nm. ERS-2 was launched into a polar sun-synchronous orbit in April 1995. With a ground pixel size of 40 x 320 km^2 (40 x 960 km^2) GOME reaches global coverage at the equator within 3 days. The main objective of GOME is the global measurement of ozone columns, but other trace gases such as NO_2, SO_2, HCHO, BrO and OClO can be retrieved from the spectra as well.

NO_2 Retrieval from GOME

Using the *Differential Optical Absorption Spectroscopy* (DOAS) technique, NO_2 is retrieved from GOME spectra in the wavelength range 425 - 450 nm. Only data of pixels with less than 10% cloud cover are taken into account.

The result of the fit is the total slant column (SC).

Subtration of the stratosperic NO_2 amound yields the tropospheric SC. The stratosperic NO_2 amound is derived from data of the stratospheric 3D chemistry and transport model SLIMCAT[4]: On the assumption that the sector at the longitude 180°-190° is virtually free of tropospheric NO_2, the SLIMCAT data are scaled zonally to the GOME measurements in this sector.

The tropospheric SC is converted to a total vertical column (VC) using the radiative transfer model SCIATRAN[1]. The conversion depends on the vertical profiles of NO_2 for each pixel. The profiles are unknown, therefore they are taken from the 3D tropospheric chemical transport models MOZART[2], TOMCAT[4] and IMAGES. The output of SCIATRAN is the airmass factor (AMF), the ratio between SC and VC.

Airmass Factors

The sensitivity of the GOME retrieval depends on the height of the absorber within the atmosphere. Therefore the AMF_i of a given layer i is a function of the height. The AMF of the total column depends on the concentration profile of the absorber, not on the total concentration. The very high concentration near the ground over anthropogenic sources leads to small AMF, whereas the NO_2 above biogenic sources is mostly located in the free troposphere and causes larger AMF.

As the AMF depends strongly on the solar zenith angle (SZA), this variable must be taken into account for the retrieval. The height dependency of th AMF increases with the SZA.

The AMF depends also on the aerosol type: The optical thick urban aerosol absorbs photons and causes small AMF from ground up to 3 km. The rural and maritime aerosols are reflecting light, which increases the albedo of lower layers. This causes a larger AMF for layers above 500 m since the sensitivity depends on the number of backscattered photons.

The computation time for the AMFs for one day on the grid of MOZART (8192 pixel) with SCIATRAN is approx. 2.5 days on a 0.8 GHz PC. To facilitate an efficient, i. e. fast retrieval the 2D airmass factor scheme was implemented.

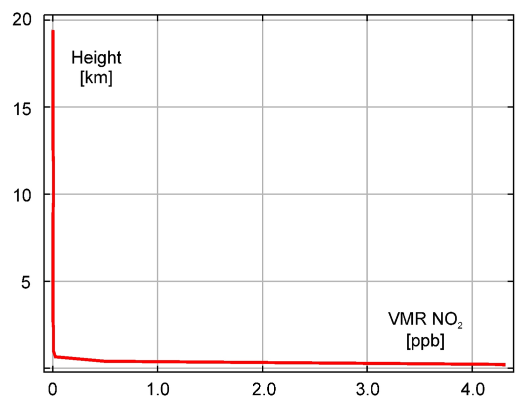
The basic idea is to substitute the radiative transfer calculation by summing precalculated AMF_i for different height layers weighted by the concentration of NO_2 V_{ci}:

$$AMF = {}_i V_{Ci} \cdot AMF_i / {}_i V_{Ci}$$

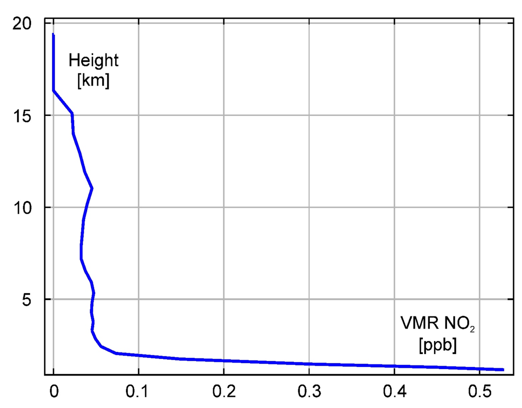
It is assumed that the atmosphere is optically thin for NO_2, i. e. the radiative transfer through the layers is independent.

The AMF_i values for layers of a height of 100 m from 0 km - 20 km above sea level are precalculated. To account for the surface height dependence of the reflectivity of the atmosphere below each layer there is one individual set of AMF_i for each ground height between 0 km - 9 km in steps of 100 m.

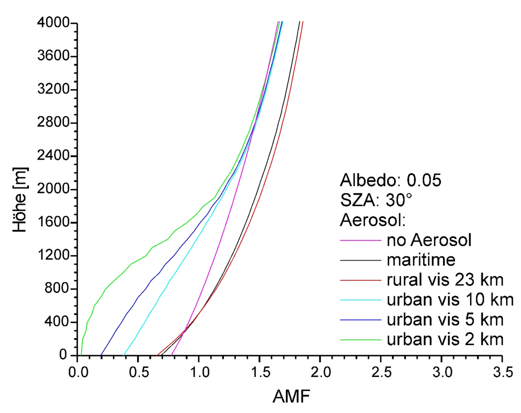
For each day an individual global AMF map is approximated. A comparison between a full SCIATRAN calculation and the 2D AMF approximation for one day at the resolution of MOZART shows a RMS < 3%. The computation time of the 2D AMF approximation is approx. 22s/day on the same PC.

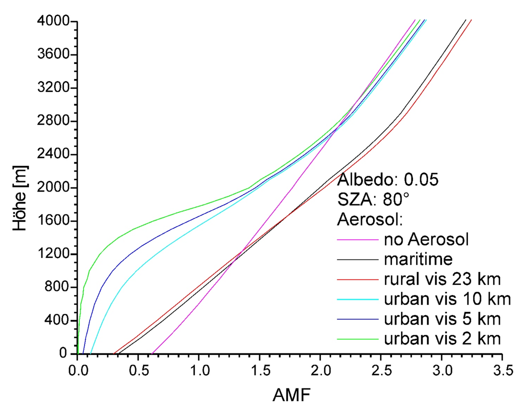


NO_2 distribution over anthropogenic sources



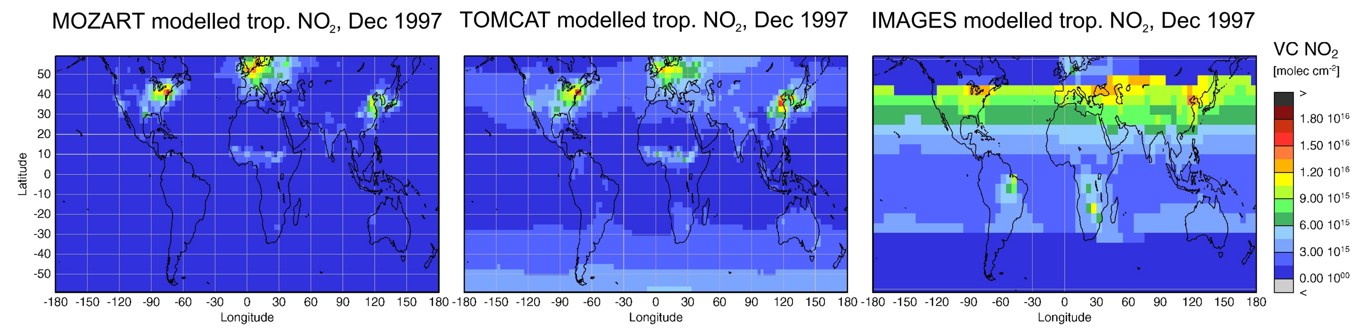
NO_2 distribution over biogenic sources





High NO_2 conentrations near the ground over anthropogenic source regions (pic. 1) and small sensitivity near the ground at high SZA (pic. 4) during winter leeds to small total AMF

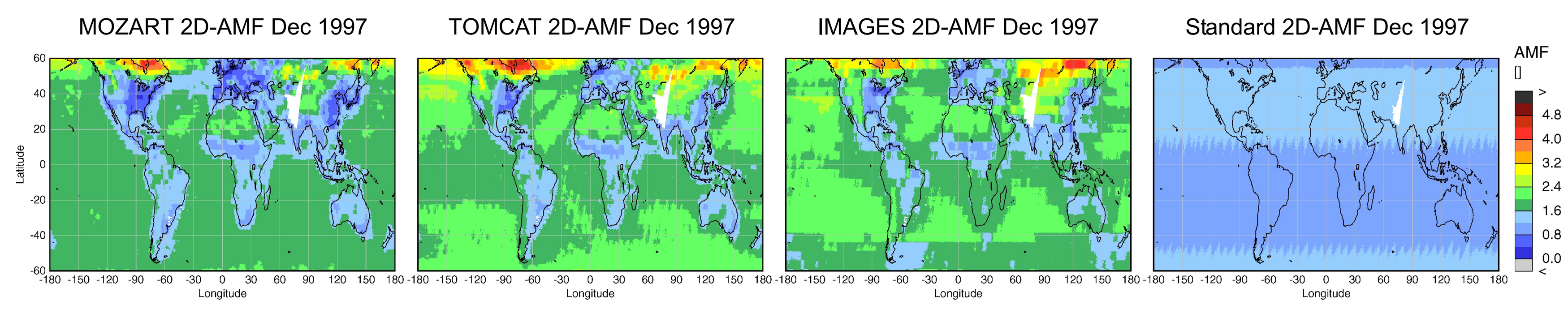
Model Comparison



The distribution pattern of the NO_2 concentrations in MOZART and TOMCAT are mostly alike, but MOZART shows the highest values above Europe and, compared with TOMCAT and IMAGES, lower values above Asia. The emissions over Africa from TOMCAT and MOZART match fine, nevertheless MOZART shows in contrast to IMAGES and TOMCAT virtually no emissions above South America.

The different patterns of MOZART on the one hand and of TOMCAT and IMAGES on the other hand are caused by different emission scenarios.

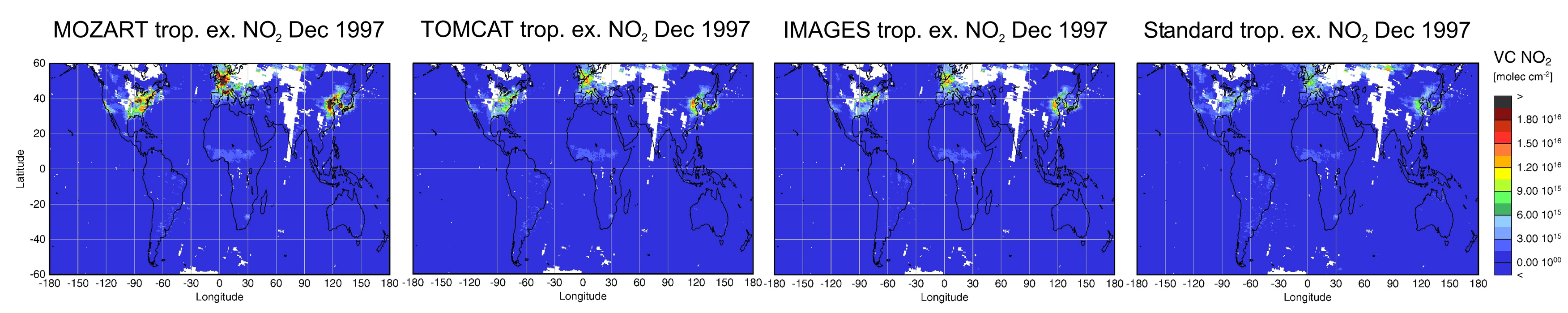
2D Airmass Factor Comparison



The values of the block AMF, which are based on the shape of the modelled NO_2 profiles, are almost in every case higher than those of the Standard-AMF. An exception to that are the values above anthropogenic source regions: High concentrations near the ground combined with a low sensitivity of the measurements for low layers especially for an urban aerosol leads to small AMF values.

The influence of the shape of the profie on the AMF is clearly visible over the oceans: For all AMF above the ocean a maritime aerosol is applied. The legible increasments of the model derived AMF are based on the shape of the absorber profile in which the mean amound of NO_2 is assumed in higher layers and with that in a region of higher sensitivity of the measurement.

Retrieval Comparison



Above anthropogenic source regions the useage of AMF based on MOZART lead to three times higher tropospheric VC in comparision with the standard retrieval. The AMF based on TOMCAT and IMAGES causes a doubling of the VC compared to the standard retrieval. This large enhancement is caused by the high SZA during winter.

The influence on the absolute NO_2 values above biogenic sources is small due to the small source strength and the small difference between the standard AMF and the block AMF.

The modelled tropospheric VC of NO_2 over Asia derived from MOZART are smaller than the ones derived from TOMCAT or IMAGES. For the retrieved VC it´s the other way round.

In contradiction to the modelled data the retrieval based on AMF derived from TOMCAT shows higher values above North America compared to the values of the tropospheric NO_2 VC above Europe.

That indicates that the AMF depends on the shape of the NO_2 profiles instead of the absolute NO_2 concentrations predicted by the models.

Consequently the retrieval of TOMCAT and IMAGES looks very similar since both models are based on the same emisson scenario.

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