Determining Tropospheric Constituent Columns from UV/visible Nadir Satellite Measurements

TROP-20

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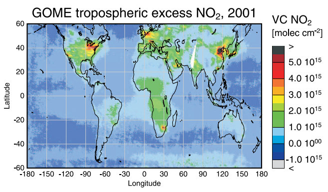
Introduction

Tropospheric chemistry and in particular the impact of anthropogenic activities on atmospheric constitution is one of the key research areas of the coming years. Many of the processes studied turn out to be not only of local and regional interest, but do have global implications. Therefore, a growing need for global data sets of tropospheric constituents is developing, that can only be satisfied by satellite measurements.
The new generation of UV/visible space borne spectrometers such as GOME, SCIAMACHY and in the future OMI and GEOSCIA already contribute to the field by providing global maps of tropospheric species such as O_3, NO_2, BrO, HCHO, H_2O and SO_2. However, for those species with significant concentrations in the stratosphere, the tropospheric and stratospheric contributions to the observed signal have to be separated. In this study, several approaches to achieve this have been developed and applied to the retrieval of tropospheric NO_2 from GOME measurements.

A Reference Sector Method

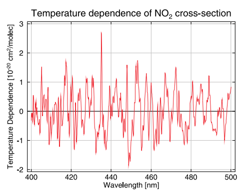
The standard method for the separation of the tropospheric and stratospheric contributions to the measurement is the reference sector method. When using this approach, two basic assumptions are made:
1) the stratospheric concentration field of the absorber depends only on latitude, but not on longitude, and 2) in a certain region (for example around the date line) the tropospheric columns are negligible therefore and measurements in this region give the stratospheric column as a function of latitude.
For each day, the data from the reference sector are subtracted from all measurements, and the remaining "excess" column is attributed to the tropospheric burden.
While this method is simple to apply and yields reasonable results in most cases, it can not be used in high latitudes in spring when stratospheric dynamics lead to inhomogeneous stratospheric fields or in the case, when no clean background region exists, where the tropospheric column is known to be small.

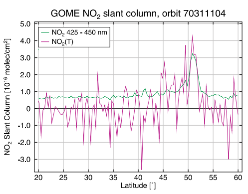


An example for the tropospheric excess NO_2 column as derived using the reference sector method is shown in the figure above for GOME data from the year 2001. Some of the structures at high latitudes are artefacts resulting from inhomogeneous stratospheric distributions in spring. Please note, that negative values as seen over clean ocean regions are to be expected both from longitudinal variations in stratospheric and tropospheric background.

B Temperature Method

Usually, temperatures in the stratosphere are much lower than in the troposphere, in particular in the boundary layer. Many molecules have temperature dependent absorption cross-sections, and these differences can in principle be used to distinguish between absorption in the warm troposphere and absorption in the cold stratosphere. This method has successfully been used for ground-based measurements to correct the impact of tropospheric NO_2 on the retrieval of stratospheric NO_2 columns.
As shown in the upper figure, the absorption cross-section of NO_2 has a very structured temperature dependence, making it a good candidate for the application of the method. Unfortunately, the most pronounced of these features correlate with instrumental features of GOME, and retrieval can be performed on a subset of the lines only. Therefore, up to now only very qualitative retrievals have been possible, showing that a temperature signal is in the measurements, but clearly too noisy to be used for a quantitative retrieval (see lower figure). This should improve significantly for other instruments.





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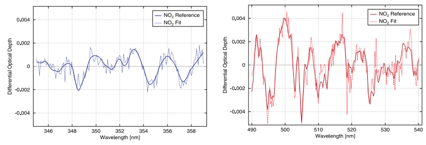
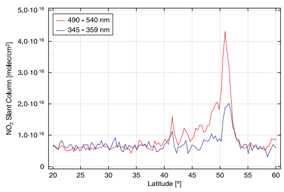
C Wavelength Method

As a result of the increase in Rayleigh scattering cross-section, photons in the UV have a much larger scattering probability than in the visible. Therefore, the proportion of photons observed from space that have actually probed the boundary layer is decreasing with decreasing wavelength.

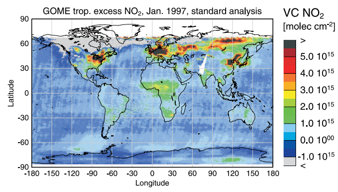


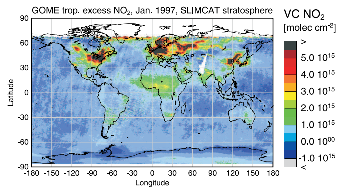
For species that absorb in a broad wavelength range (i.e. NO_2), columns can be retrieved in different wavelength regions as shown in the upper two figures. For a stratospheric absorber, the retrieved column depends only weakly on the wavelength region, whereas for an absorber in the boundary layer, the UV retrieval yields much smaller columns as shown in the lower plot for a GOME orbit passing over polluted parts of Germany. While this shows the feasability of application of the method to GOME NO_2 measurements, results still have larger errors than in the reference sector method.



D Model Method

State of the art chemical transport models such as SLIMCAT can provide a very good estimate of the stratospheric NO_2 amounts. In principle, the SLIMCAT values can be converted to the expected stratospheric columns and then subtracted from the measurements to yield the tropospheric columns. However, in practice it turns out that relatively small uncertainties in the absolute amount of NO_2 in model or measurement can have a large impact on the retrieved tropospheric columns. Therefore, the model output has to be scaled to the measurements over the reference sector (see section A), using only the longitudinal variation and not the absolute value of the stratospheric NO_2.
As shown in the figures, most of the artefacts at high latitudes (upper panel) are removed when the model output is used for the stratospheric correction, significantly improving the retrieval at high latitudes.





Summary and Conclusions

Tropospheric columns of a number of trace species can be retrieved from UV/visible nadir measurements from space as has been shown for GOME data. The standard method of separation between stratospheric and tropospheric absorptions is the reference sector method that yields good results under most circumstances. In the case of NO2, it can be improved by using SLIMCAT model output to account for the longitudinal variation of the stratospheric column.
It has been shown, that the wavelength dependence of the tropospheric absorption can also be used to retrieve the tropospheric amount, but currently errors are still larger than for the reference sector method.
The temperature dependence of the cross-section does also give some height information, but for GOME data errors still are too large, partly for instrumental reasons.
For the new (SCIAMACHY) and upcoming (OMI, GEOSCIA) instruments, errors should decrease significantly, and combination of the methods should lead to more accurate tropospheric products from space borne observations.

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see also: www.iup.physik.uni-bremen.de

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