A MULTI WAVELENGTH APPROACH TO THE RETRIEVAL OF TROPOSPHERIC NO_2 FROM GOME MEASUREMENTS

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INTRODUCTION

Nitrogen oxides play an important role in the chemistry of earth's atmosphere. In the troposphere, they provide the only chemical source for ozone, arguably the most important radical in the lower atmosphere. Nitrogen oxides are central in the formation of photochemical smog in the industrialised countries and also in regions with intense biomass burning. In addition to its relevance to tropospheric chemistry, the NO_2 radical may also contribute locally to radiative forcing in the troposphere.

Today, nitrogen oxides are mainly released by human activities, in particular burning for heating, industry or transportation and the burning of biomass. Natural sources are soil emissions and lightning and also forest and savannah fires, although the latter may partly be anthropogenic. With ever increasing population and rapid industrialisation of the developing countries, strategies for a reduction in nitrogen oxide emissions become more and more important. Such strategies need to be based on reliable measurements of the NO_x flux from the different sources and regions, preferably continuously and on a global scale.

In this study, a novel approach to the determination of tropospheric NO_2 columns from nadir viewing UV/vis satellite measurements has been developed. First, possible methods for the separation of stratospheric and tropospheric signals are discussed in view to NO_2 measurements with GOME. Then a new technique based on the wavelength dependence of the Rayleigh scattering cross-section is presented, and first applications to measurements above Europe are shown. Finally, remaining problems of the method and future developments are discussed.

THE GOME INSTRUMENT

The Global Ozone Monitoring Experiment (GOME) [2] is a grating pseudo double monochromator covering the spectral range of 240 to 790 nm with a spectral resolution of 0.2 – 0.4 nm. GOME observes the light scattered back from the atmosphere and reflected on the ground in near nadir viewing geometry. The ground pixel size is 320 km across-track and 40 km along track for the three forward scans and 960×40 km^2 for the back scan. Global coverage is achieved within three days at the equator and within one day at 67°. Once per day, an extraterrestrial solar spectrum is measured via a diffusor plate. The GOME instrument has been launched on the European ERS-2 satellite in April 1995 and is operational since July of that year. ERS-2 is in a sun-synchronous polar orbit with an equator crossing time of 10:30 UT. As a result of this orbit, GOME measurements in low and middle latitudes are always taken in the late morning (local time).

The main target of GOME is the determination of the global stratospheric ozone field. However, the broad spectral coverage and the relatively good spectral resolution also enables the retrieval of a number of other trace gases with structured absorptions in the UV and visible wavelength range. Under clear sky conditions, visible and near UV radiation reaches the surface, and provides GOME with a unique sensitivity towards tropospheric absorbers. In particular, GOME measurements have been used to study tropospheric SO_2 [3], BrO [21, 17], HCHO [10, 20] and NO_2 [11, 15].

SEPARATION OF STRATOSPHERIC AND TROPOSPHERIC ABSORPTIONS

The GOME instrument observes the earth in nadir viewing geometry. Photons registered by the detectors have either been reflected on the ground or scattered by atmospheric particles. The ratio between reflected and scattered photons depends strongly on surface albedo, wavelength and atmospheric aerosol loading. In general, scattering is more important at shorter wavelengths owing to the increase in Rayleigh scattering cross-section and the simultaneous decrease in surface albedo.

In a cloud free atmosphere, the spectra measured by GOME contain absorptions from both stratospheric and tropospheric trace species. In the case of NO_2, varying amounts are present in the stratosphere, the free troposphere and the boundary layer. If the tropospheric column is the quantity of interest, the contributions from different heights have to be separated using additional information, preferably from the GOME measurements themselves.

In principle. several methods can be used to distinguish between tropospheric and stratospheric absorptions from space borne UV/vis observations, each having its own advantages and problems.

Reference Sector Method

The reference sector method is based on two assumptions: that there exists a region, where tropospheric contributions to the measurement can be neglected, and that the stratospheric contribution is longitudinally homogeneous. To derive the tropospheric column, the background value determined at the same latitude but in the clean longitude range is subtracted from the measured total column. In the case of NO_2, air-borne measurements [19] and examination of the GOME data have shown, that the Pacific region can be used to determine a clean background value. The assumption of zonal homogeneity is justified by the fact, that stratospheric NO_2 concentrations are mainly driven by photolysis of the reservoir substances N_2O_5 and HNO_3. In contrast to O_3, dynamics are important only at high latitudes and mainly in spring, when temperature distributions in the vortex regions are asymmetrical.

The advantage of the reference sector method is its simplicity, the main drawback is the fact, that variations in the stratospheric NO_2 can not be distinguished from variations in the tropospheric columns. This is in particular the case in winter and spring at northern mid and high latitudes.

Modified versions of the reference sector method have been used to derive tropical tropospheric ozone from TOMS measurements [8] and GOME data [10]. For the retrieval of tropospheric NO_2 from GOME measurements, it is the standard method [11, 15]. In the work of [11], the method has been modified by using data measured over all oceans to create interpolated maps of stratospheric NO_2. This improves the representation of the stratospheric fields in the case of dynamical variations, but can lead to artefacts if NO_2 is present for example over the Southern Atlantic.

Cloud Selection Method

Clouds can also be used to distinguish between tropospheric and stratospheric absorptions. For a satellite based instrument, clouds effectively block the view to the parts of the troposphere below cloud top height. In contrast to ground-based zenith-sky measurements, light path enhancement by multiple scattering in the cloud is important for thin clouds or aerosol layers only [9, 6]. By comparing measurements taken at different cloud cover, the column below the cloud can be extrapolated.

The main problem with this method is, that measurements taken on different days are compared, and that both stratospheric and tropospheric concentrations need to be constant in time for a simple interpretation of a correlation plot between cloud cover and NO_2 column. Also, as a result of the large ground-pixel size of the GOME instrument, most measurements are taken at partial cloud cover and only very few data are available for clear sky or completely overcast scenarios. It has to be noted, that the latter problem also affects all other methods described in this section.

Temperature Method

Another information on the distribution of NO_2 between troposphere and stratosphere comes from the temperature dependence of the absorption cross-section of NO_2. At stratospheric temperatures, the differential absorption cross-section is smaller than at room temperature, and also changes in shape even at the spectral resolution of GOME [1]. This difference has been used to separate cold stratospheric from warm tropospheric NO_2 contributions in ground-based zenith-sky measurements [16], but up to now has not been successfully applied to GOME measurements. The main problems are residual spectral structures from the polarisation dependence of the instrument and the lower signal to noise ratio of the spectra when compared to ground-based measurements, where many spectra can be co-added without loss of spatial resolution.

Two Instrument Method

If the stratospheric column is measured by an other instrument, then subtraction of this value from the column measured by GOME will yield the tropospheric column. Such an approach has been used to derive tropospheric ozone from TOMS and SAGE measurements [4] and HALOE measurements could be used for NO_2. However, The number of HALOE profiles is limited in space and time, and the photochemical changes between the GOME measurements near noon and

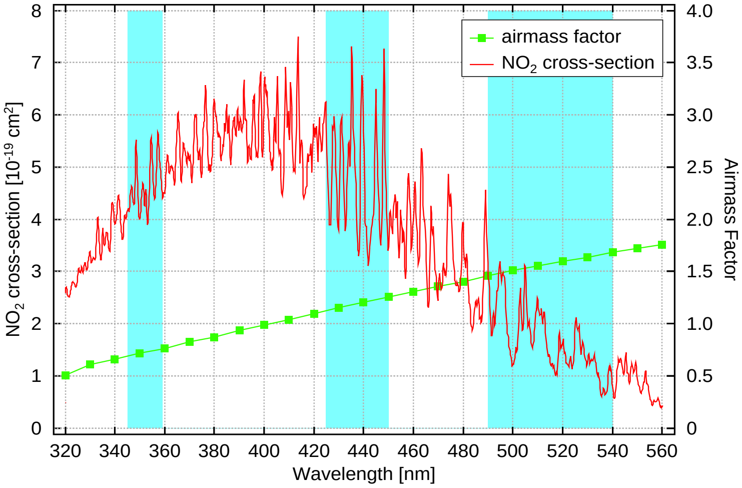

Fig. 1. Wavelength dependence of the airmass factor for an absorber situated in the boundary layer. For the calculations, a homogeneously mixed absorber in the lowermost 1.5 km was assumed, the albedo set to 0.05 and a maritime aerosol used. Also shown is the absorption cross-section of NO_2 and the three wavelength windows used for the retrieval as discussed in the text.

the occultation measurements from HALOE necessitate the use of a chemical model to calculate the stratospheric column for the time of the GOME overpass. The use of two measurements, one for the total column and one for the stratospheric part will be used with the SCIAMACHY instrument, where nadir measurements alternate with limb profile measurements that can be integrated to yield the stratospheric column.

Model Method

As alternative to measurements from a second instrument, the stratospheric column can also be taken from the results of a global chemical transport model. This approach depends crucially on the accuracy of the model results and will have to be investigated further.

Wavelength Method

In the wavelength method, the wavelength dependence of the penetration depth of photons is used to derive information on the vertical distribution of the absorber. Several factors influence the penetration depth of photons: the scattering probability that is increasing with decreasing wavelength as a result of the wavelength dependence of the Rayleigh cross-section, the strong increase of ozone absorption in the UV, and the variation of surface albedo. For ozone, stratospheric vertical profiles with moderate height resolution can be retrieved from nadir measurements between 280 and 340 nm [7] by comparison with the results of forward models. In principle, the increase in ozone optical thickness below 320 nm provides profile information for all absorbers in this wavelength region. However, in practice the strong ozone absorption structures interfere with the retrieval of other, weaker absorbers such as NO_2, SO_2, or BrO and no useful information can be deduced on these trace gases.

At wavelengths between 340 and 550 nm, the atmosphere is optically thin and most photons reach the troposphere. However, in the lowermost layers the importance of scattering increases and depending on wavelength, a substantial fraction of photons is scattered before reaching the surface. In combination with the fact, that surface albedo is very small in the UV, this results in a wavelength dependence of the sensitivity of nadir viewing instruments towards absorptions in the boundary layer.

In the case of NO_2, the structures in the absorption cross-section enable DOAS retrievals anywhere between 340 and 600 nm, with an optimum in the 425–450 nm window used for the operational NO_2 product [5]. Over this wavelength region, the sensitivity towards an absorber situated in the lowermost 1.5 km increases by a factor of two as illustrated in Figure 1. The quantity shown in the graph is the airmass factor, which is defined as the ratio between the measured slant column and the vertical column, the integral of the absorber over height. Airmass factors are derived with radiative transport calculations, and depend on wavelength, solar zenith angle, viewing angle, the vertical profile of the absorber,

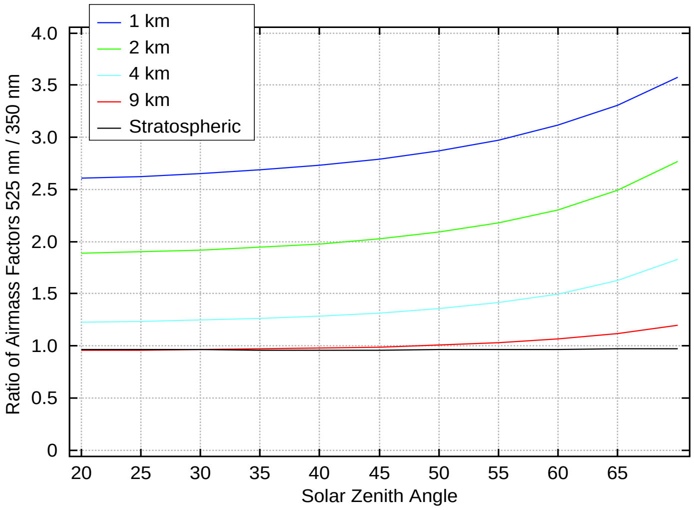

Fig. 2. Wavelength dependence of the airmass factor for a 1 km layer in the troposphere at different altitudes. Shown is the ratio of the airmass factor at 525 and 350 nm. Also plotted is the same ratio but for a stratospheric profile (US standard atmosphere).

the density profile, the surface albedo and the aerosol loading. In this study, all airmass factors have been calculated using the GOMETRAN model [18].

How large the variation of the airmass factors is with wavelength, depends critically on the height where the bulk of the absorber is situated. In Figure 2, the ratio of the airmass factors at 525 and 350 nm is shown as a function of height for a uniformly mixed layer of 1km thickness. As can be seen, the sensitivity of the wavelength method is largest for absorbers in the boundary layer, and decreases rapidly with height. Also shown is the ratio for a stratospheric profile (US standard atmosphere with all tropospheric parts removed). This ratio is in good approximation unity at all solar zenith angles.

Another critical factor is surface albedo. In the case of a reflecting surface such as ice, the largest part of the signal detected by an satellite instrument originates from the surface at all wavelengths, and little height information can be retrieved. Similarly, clouds or even reflecting aerosols enhance the reflected part of the signal and reduce the wavelength dependence of the sensitivity.

APPLICATION TO GOME DATA

Data Analysis

NO_2 columns at different wavelengths have been derived with the Differential Optical Absorption Method DOAS [12]. Calibrated GOME lv1 earth-shine and solar measurements were analysed using the software tools developed for ground-based zenith-sky measurements [14]. In the selection of the two fitting windows, a compromise had to be found between an optimum in difference in the sensitivity towards the troposphere and good NO_2 detection accuracies. After a series of tests, the windows 345–359 nm and 490–540 nm were selected. Below 345 nm, NO_2 retrieval is noisy and subject to interference by ozone absorption, above 540 nm water vapour absorptions interfere strongly with the NO_2 fit. In Figure 4, an example is shown for a simultaneous fit of NO_2 during a pollution event. The NO_2 signature is clearly identified in both fitting windows, albeit with different columns as expected. The resulting columns for this event are shown in Figure 3, nicely illustrating the difference in sensitivity towards tropospheric absorptions. However, from the graph it is also clear, that in particular the UV retrieval is subject to noise that will introduce errors in the tropospheric results.

From the NO_2 columns retrieved at the two wavelengths, the tropospheric column can easily be determined. Assuming that the measured slant column SC is composed by a stratospheric column SC_{strat} and a tropospheric contribution SC_{trop}, and that the corresponding airmass factors are AMF_{strat} and AMF_{trop}, then SC can be written as

$$SC^i = \mathrm{VC}_{\mathrm{strat}}\mathrm{AMF}^i_{\mathrm{strat}} + \mathrm{VC}_{\mathrm{trop}}\mathrm{AMF}^i_{\mathrm{trop}} \qquad (1)$$

where VC are the vertical columns and the index i denotes the wavelength interval. With the assumption, that the stratospheric airmass factor is similar at both wavelengths, the tropospheric vertical column can be expressed by the difference

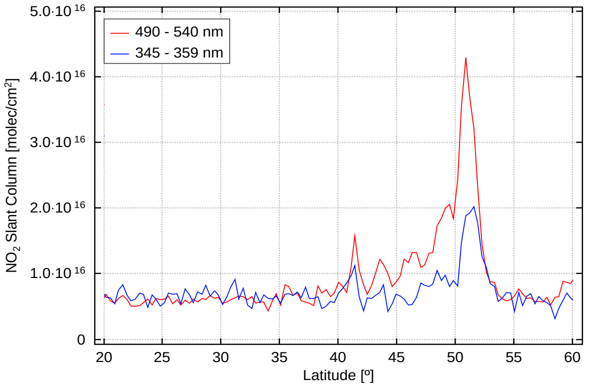

Fig. 3. NO$_2$ slant columns at different wavelengths for a tropospheric pollution event over central Europe (orbit 703311104). For clarity, only the centre scan is included. For the part of the orbit showing the enhanced NO$_2$ values, the ICFA algorithm indicates cloud free conditions.

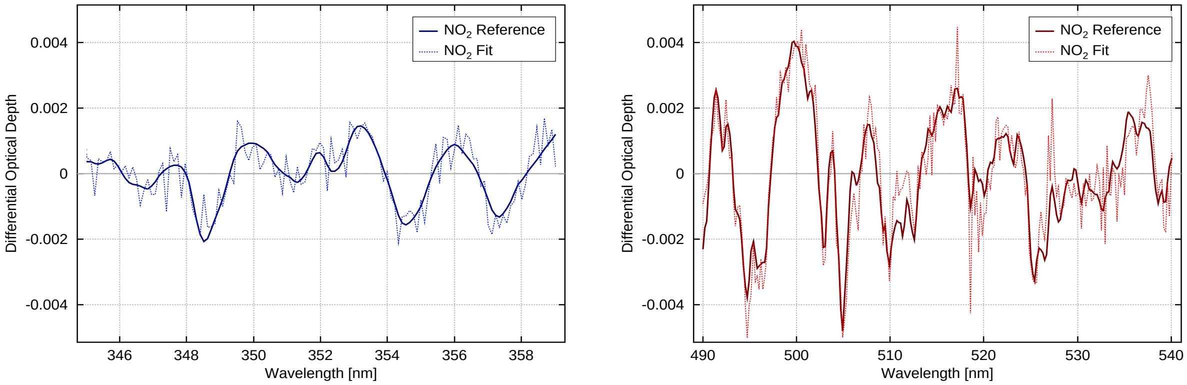

Fig. 4. NO$_2$ fit results for the maximum of the tropospheric pollution event shown in Figure 3 (pixel #547). To give an indication of the quality of the fit, the scaled laboratory cross-section is shown together with the results from the fit, after removing all contributions with the exception of NO$_2$. The slant columns corresponding to the fits are 1.910^{16} molec/cm^2 in the UV window and 4.310^{16} molec/cm^2 in the visible.

of the measured slant columns and the two tropospheric airmass factors:

$$\mathrm{VC}_{\mathrm{trop}} = (SC^{\mathrm{vis}} - SC^{\mathrm{uv}})/(\mathrm{AMF}^{\mathrm{vis}}_{\mathrm{trop}} - \mathrm{AMF}^{\mathrm{uv}}_{\mathrm{trop}}) \tag{2}$$

GOME NO2 above Bremen

As a first application, GOME data in a 500 km radius around Bremen (53°N, 9°E) have been analysed for 1997. In order to avoid problems with clouds, only ground pixels with an ICFA value below 0.15 have been included in the analysis. The ICFA values are part of the operational GOME data product, and give a fractional cloud cover derived from the O$_2$ band in combination with a cloud top height climatology. Although the cloud fraction determined by ICFA is not necessarily correct in a quantitative sense, it does give a good qualitative representation of the cloud cover in a given ground pixel.

In Figure 5, daily averages of the NO$_2$ slant columns are shown for both fitting windows. As can be seen, the visible fit yields larger values on most days, and particulary so during episodes of enhanced NO$_2$. This indicates, that above Bremen there is both a more or less constant tropospheric background and superimposed there are events of strongly enhanced tropospheric NO$_2$, in particular in fall and winter, when emissions are largest and frequent inversions confine pollutants in the boundary layer.

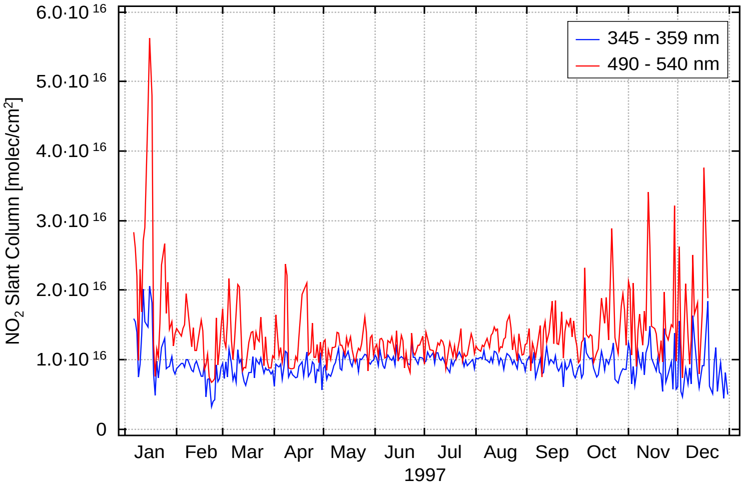

Fig. 5. Daily averaged NO_2 slant columns in a 500 km radius around Bremen, 53° N for the two wavelength windows.

GOME NO2 above Europe

In Figure 6, the results of the analysis for all GOME measurements above Europe in February and March 1997 are shown. Again, only data with little cloud contamination (ICFA < 0.15) have been included. In order to optimise the spatial resolution, only the three forward scans were used and no smoothing or filtering was applied. For the airmass factors, a surface albedo of 0.05 was assumed in the visible, and 0.02 in the UV. As in the sensitivity studies shown in Figures 1 and 2, a maritime aerosol was used although that might not be appropriate in the polluted parts of Europe.

Two observations can be made from the graph: There is a consistent tropospheric NO_2 background of about 210^{15} molec/cm^2 over the continent, and superimposed on this are large areas of strong tropospheric pollution in central Europe. The largest values are 1.310^{16} molec/cm^2 in the two month average, roughly equivalent to 3.5 ppb of NO_2 if a 1.5 km well mixed boundary layer is assumed. These values agree well with the results of in-situ measurements and model calculations, but are somewhat larger than the columns obtained in previous studies using the reference sector method on GOME data [11, 15]. The reason for this difference in the magnitude of the tropospheric NO_2 columns derived with different methods from the same data set are not yet fully understood and will have to be investigated in more detail.

Limitations of the Method

Several factors limit the applicability of the wavelength method to a larger set of GOME measurements. One important point is, that the difference in tropospheric airmass factors used in the analysis is only valid under clear sky conditions. As a result of the large foot print of the GOME instrument, the number of truly cloud free pixels is small, and most measurements are contaminated by residual clouds. As for all other methods available to derive tropospheric columns from nadir measurements, this leads to a systematic underestimation of the tropospheric column.

Also, changes in surface albedo, aerosol loading or vertical NO_2 profile can lead to significant changes in the airmass factors. This can in principle be improved by introducing additional information into the analysis; however, the necessary data are not readily available and often have large uncertainties in themselves.

A more technical limitation is the quality of the NO_2 retrieval in the two fitting windows. Compared to the standard wavelength range 425–450 nm, the spectral regions used here are less advantageous with respect to signal to noise ratio, magnitude of the differential absorption structures and interference by other absorbers. As a result, the retrieved columns are subject to noise, which is further amplified by the fact that two instead of one fit are needed to determine the tropospheric columns. While random noise can be reduced by averaging over time and space, systematic errors are more difficult to treat. Unfortunately, examination of the results of a large number of fits shows, that systematic rather than random contributions dominate the residuals. Possible error sources are GOME calibration problems and geophysical processes not corrected for in the analysis.

One very prominent residual structure could be identified as the absorption in liquid water. As it turned out, absorption in surface water can clearly be seen in the spectra at 515 nm whenever measurements are taken at clear conditions over

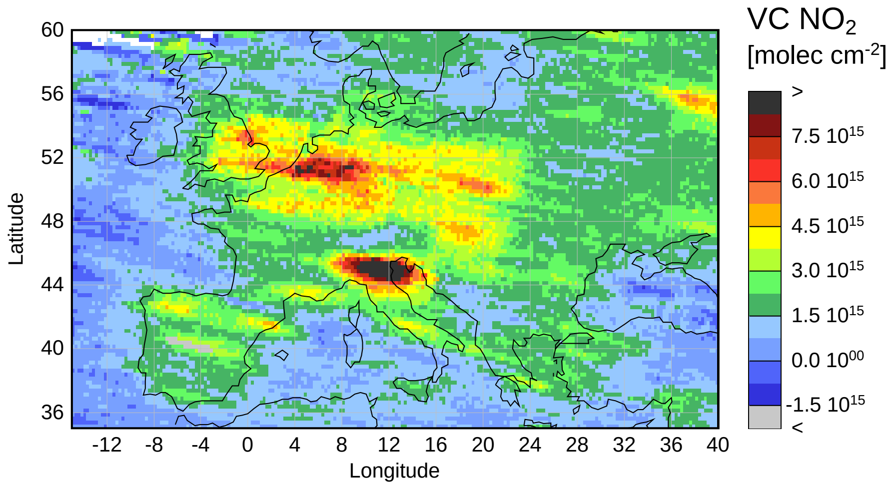

Fig. 6. Vertical tropospheric columns above Europe for February and March 1997 as derived with the wavelength method. Localised pollution areas can be identified as well as a general background of enhanced NO_2 over the continent.

water. This is illustrated in Figure 7, where the fitting coefficient of the liquid water absorption cross-section [13] is shown for all clear pixels of March 1997. All oceans and lakes feature large absorptions, the continents do not. Some of the structures in the oceans are related to water turbidity as measured by SEAWIFS, and could potentially be of interest to ocean science. However, in the context of tropospheric NO_2 retrieval, liquid water absorption is a interfering effect, and has to be corrected. Unfortunately, the available cross-sections of liquid water all are at much lower spectral resolution than GOME data, and also are noisier than acceptable for a good fit. Because of this and probably other systematic errors in the fits, the wavelength method can currently only be reliably applied to GOME NO_2 measurements at mid-latitude regions and over continents. It is hoped that further improvements in the analysis will lead to more robust data sets and allow the creation of global maps.

SUMMARY AND OUTLOOK

In this study, a novel approach has been developed to derive tropospheric NO_2 columns from UV/visible nadir measurements from space. The method is based on the wavelength dependence of Rayleigh scattering and surface albedo that combine to decrease the sensitivity of nadir measurements towards tropospheric absorptions in the UV. By comparing slant columns retrieved with the DOAS method in different spectral windows, vertical tropospheric columns can be determined for trace species with differential absorption structures at these wavelengths. The method is most sensitive towards absorptions in the boundary layer and depends on low surface albedo (no snow or ice).

The method has been applied to the retrieval of NO_2 from GOME measurements, and good qualitative agreement has been found with the results of the independent reference sector method. Some unsolved problems remain in the analysis of the GOME data that are related to instrument calibration problems and probably also to insufficient treatment of scattering effects in both atmosphere and on the surface. As one example, it has been shown, that absorption in liquid water can not be neglected for clear pixels over water in the visible spectral range. Future improvements in the analysis should increase the accuracy of tropospheric NO_2 columns derived with the new method and extend the applicability to low latitudes.

With the new generation of improved space borne UV/visible instruments such as SCIAMACHY on ENVISAT, better signal to noise ratios and smaller ground pixels will be possible. This should further improve the accuracy of the tropospheric columns derived with the wavelength method, possibly even to the point where a coarse profile could be retrieved. In particular, if reliable ozone retrieval in the Chappuis bands becomes feasible with the new instruments, then the method could also be applied to tropospheric ozone retrieval, extending the ozone profiles from the stratosphere down to the ground.

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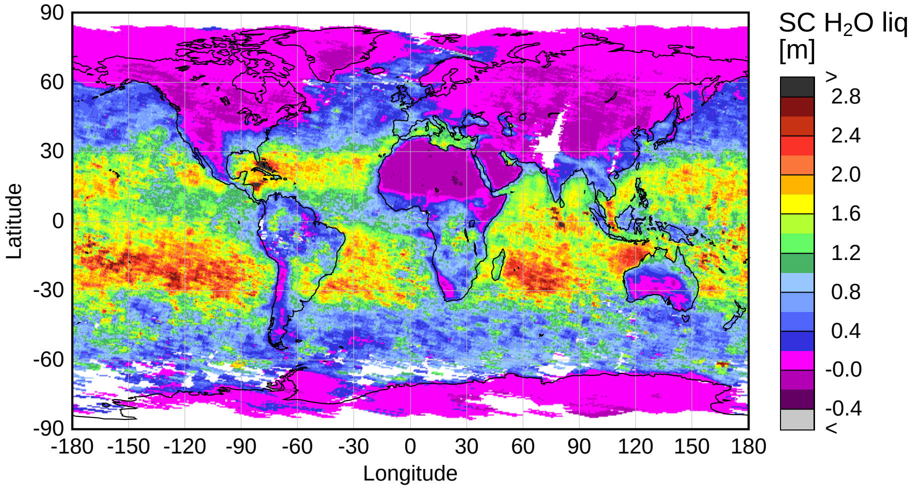

Fig. 7. Fitting coefficient of the liquid water absorption cross section used in the 490–540 nm window. The average over all clear pixels (ICFA < 0.15) in March 1997 is shown. The negative values over continents are indicative of some interference between the DOAS polynomial and the rather broad band absorption in liquid water. While the absolute values are uncertain, the *difference* between water and land surfaces is striking.

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