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RETRIEVAL AND MONITORING OF ATMOSPHERIC TRACE GAS CONCENTRATIONS IN NADIR AND LIMB GEOMETRY USING THE SPACE-BORNE SCIAMACHY INSTRUMENT

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Abstract. The Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) onboard the European Envisat spacecraft performs continuous spectral observations of reflected, scattered and transmitted sunlight in various observation geometries. A unique feature of SCIAMACHY is the capability of probing the atmosphere in three different observation geometries: The nadir, limb, and occultation measurement modes. In nadir mode, column densities of trace gases are retrieved with a spatial resolution of typically 30×60 km using the Differential Optical Absorption Spectroscopy (DOAS) technique (Platt and Perner, 1983). Alternating with the nadir measurement, vertical profiles of absorber concentration in the stratosphere are derived in limb and occultation. In this paper we present an overview over some applications of SCIAMACHY data in space-based monitoring of atmospheric pollution. The DOAS algorithms for the retrieval of total column amounts from nadir spectra are briefly described and case studies of pollution events are presented. We also illustrate the technique used to derive stratospheric concentration profiles from limb observations and show comparisons with other remote sensing systems. Special emphasis will be given to techniques, which take advantage of SCIAMACHY's different viewing geometries. In particular, we will discuss the potential and limits of strategies to infer tropospheric abundances of O_3 and NO_2.

Keywords: optical absorption spectroscopy, satellite remote sensing, tropospheric trace gases

1. Introduction

The space-borne Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) was launched into orbit in March 2002 on board of the European Envisat spacecraft. The spectrometer performs continuous measurements of transmitted, reflected and scattered sunlight in the ultraviolet, visible, and shortwave infrared wavelength region (220–2380 nm) at moderate resolution (0.2–1.5 nm). The optical system allows for light detection in various viewing directions. In down-looking (nadir) mode, the instrument scans the region underneath the spacecraft by detecting upwelling solar radiation that has been transmitted and scattered in the atmosphere and reflected at the Earth's surface. In limb mode, two mirrors direct the sunlight scattered by the atmosphere near the horizon (in flight-direction) into the spectrometer. During a limb scan the atmosphere is probed vertically by changing the tangent height in discrete steps from the surface up to about 100 km. Attenuation processes such as scattering and molecular absorption determine the intensity of the detected radiation at characteristic wavelengths. The radiance measurements therefore contain information on concentration and distribution of a multitude of trace gases showing spectral absorption features in the observed wavelength interval, such as NO_2, O_3, SO_2, BrO, OClO, CO, CO_2, CH_4, and H_2O (Bovensmann *et al.*, 1999). The choice of the analysis technique employed to retrieve this information depends on the target species and the viewing geometry under which the spectral measurements have been acquired. In this study we describe some of the concepts behind the data analysis techniques for SCIAMACHY measurements. The main focus of this paper is to demonstrate, that measurements in nadir and limb observation geometry can be combined to separate stratospheric and tropospheric concentrations of molecular absorbers. The application of these concepts is demonstrated for nitrogen dioxide (NO_2) and ozone (O_3).

2. Analysis of Nadir Spectra: The DOAS Technique

The retrieval of vertically integrated trace gas amounts from nadir measurements is based on the Differential Optical Absorption Spectroscopy (DOAS) technique (Platt and Perner, 1983). Various implementations of this technique for different observation platforms (ground-based, airborne and satellite) have been developed. They make use of a differential absorption signal with respect to a baseline or background absorption, which in case of satellite observations is usually provided by an extraterrestrial solar spectrum. The spectral features due to all absorption and scattering processes are imprinted on the ratio of the nadir earthshine and the solar spectrum. In the DOAS approach, the contribution of molecular (Rayleigh) and aerosol scattering to the attenuation of sunlight, which varies slowly with wavelength, is separated from the higher frequency signal due to molecular absorption. This is achieved by a least-squares fit of the DOAS equation, which is derived from Beer's law, to the intensities I_j measured by the detector pixel j and normalized to the corresponding extraterrestrial intensity I_{0j}:

$$\ln\left(\frac{I_{0j}}{I_j}\right) = \sum_n SCD_{nj} \cdot \sigma_{nj} + P(\lambda_j)$$

The fit parameters determined in the least-squares analysis are the slant column densities (SCD_n) for each trace gas n absorbing in the spectral window and the coefficients of a polynomial P in wavelength λ. The latter accounts for attenuation processes owing to Rayleigh and Mie scattering, which are assumed to be smooth and slowly varying in the considered spectral interval. The retrieval of each trace gas using the above equation requires knowledge of the individual molecular absorption cross sections σ_n, which have been measured in laboratory calibration measurements using the SCIAMACHY instrument (Bogumil *et al.*, 2003).

The SCDs retrieved in the above outlined analysis represent the effective amount of molecules encountered by photons along their average propagation path. The slant column amount strongly depends on geometrical and geophysical conditions described by a multitude of parameters, such as solar zenith angle, line-of-sight direction, vertical absorber profile, cloud fraction, albedo and aerosol loading. For monitoring of ozone and nitrogen dioxide concentrations the desired quantity is the vertical column density (*VCD*). The needed link between the retrieved *SCD* and the vertical column is the airmass factor, simply defined as their ratio $AMF = SCD/VCD$. As this quantity cannot be retrieved from the observed data, its computation requires some assumptions on the atmospheric conditions during the measurement. We use the radiative transfer model (RTM) CDI (Rozanov *et al*., 2001) to compute effective light paths by accounting for all of the above mentioned parameters, thereby calculating *AMF*s under satellite viewing geometries.

One advantage of the two-step approach of retrieving SCDs and forward calculating *AMF*s is that for optically thin absorbers different parts of the atmosphere can be treated separately. In practice, AMFs are often computed independently for the troposphere and the stratosphere. For the latter, a priori information on the vertical distribution of the target species are taken from climatology or, as will be described in the subsequent section, by actual measurements of the absorber profile from limb spectra. For the troposphere, one has to rely on photochemical models and external meteorological information to come up with realistic estimates of relative absorber distributions needed in the AMF calculation. The establishment of AMF climatologies for satellite observations is an ongoing research activity and is likely to be improved in the future. An accuracy analysis of space-borne DOAS retrievals of tropospheric NO_2 is presented e.g. in Boersma *et al.* (2004).

3. Analysis of Limb Spectra

The derivation of stratospheric trace gas concentrations from measurements of the scattered solar radiation in limb viewing geometry is achieved by two different analysis techniques for the molecular species in the focus of this paper – NO_2 and O_3. Both techniques employ the optimal estimation method (Rodgers, 1990) and require a sophisticated RTM. Unlike the DOAS approach, however, the RTM calculations are not decoupled from the analysis with the *AMF* concept, but rather an integrated part of the iterative analysis algorithm. The retrieval algorithms for stratospheric NO_2 and ozone have been described in detail by Savigny *et al*. (2003, 2004) and will only briefly be reviewed here.

NO_2 limb spectra are analyzed in the wavelength window 420–490 nm, which is significantly wider than the interval used for processing the nadir spectra (425–450 nm). The extension of the fit window to longer wavelengths increases the penetration depth of the scattered light into the atmosphere, pushing down the lower altitude limit of the profile retrieval to approx. 15 km. The retrieval is performed using ratios of limb spectra in a selected tangent height region to a limb measurement at a reference tangent height, at which the NO_2 concentration is assumed to be negligible. Limb spectra in the altitude range between 15 and 40 km are divided by the spectrum acquired at the reference tangent height of 46 km. This preparation step largely removes spectral features induced by instrumental effects that are common in all observations. Similar to the DOAS approach, a low-order polynomial is subtracted from the logarithm of measured and modeled limb radiances at each tangent height to remove broadband attenuation components from the differential NO_2 structure. An iterative optimal estimation procedure is then employed to estimate a concentration profile that minimizes the differences between the measured and modeled limb radiances. Each iterative step involves the forward calculation of all limb spectra included in the analysis as well as the weighting functions, defined as the partial derivatives of the radiances with respect to the NO_2 concentration at the tangent height). A typical set of retrieved limb profiles above a nadir ground state is depicted in the left panel of Figure 1. The four curves correspond to different azimuth angles of the limb scan, demonstrating the relative homogeneity of the NO_2 field over the nadir swath width of 960 km.

The retrieval method employed to infer ozone profiles follows the method developed by McPeters *et al.* (2000) and Flittner *et al.* (2000). A fundamental difference to the above outlined spectral fitting procedure is that measurements at only three wavelengths are taken into account in the data analysis. The method ex-

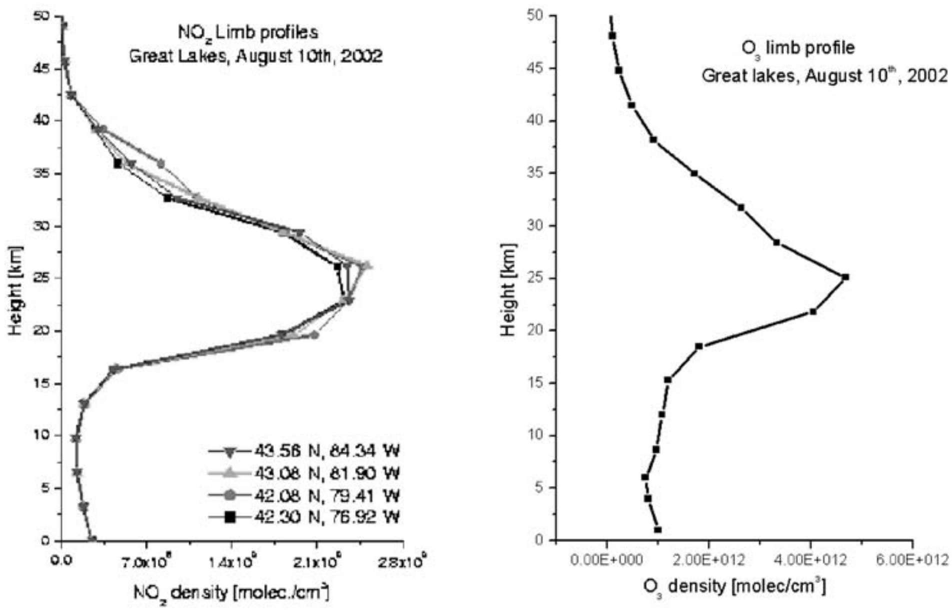

Figure 1. Vertical concentration profiles of NO_2 (left) and O_3 (right) inferred from SCIAMACHY limb radiance measurements.

ploits the differential absorption between the center ($\lambda_1 = 600$ nm) and the wings ($\lambda_2 = 525$ nm/$\lambda_3 = 675$ nm) of the Chappuis absorption bands of ozone. Limb radiance profiles $I(\lambda, TH)$ at these wavelengths are normalized with respect to a reference tangent height of $TH_{\text{ref}} = 43$ km: $I_N(\lambda, TH) = I(\lambda, TH)/I(\lambda, TH_{\text{ref}})$. The normalized limb radiance profiles are then combined to the Chappuis retrieval vector

$$y(TH) = \frac{I_N(\lambda_1, TH)}{\sqrt{I_N(\lambda_2 TH)} \times I_N(\lambda_3, TH)}$$

which is fed into a non-linear Newtonian iteration version of optimal estimation (OE) driving the spherical radiative transfer model SCIARAYS (Kaiser, 2001; Kaiser and Burrows, 2003). Limiting data in the analysis to only three spectral points greatly reduces computing times compared with the full spectral retrieval methods used for the NO_2 profiles, which could in principle be also employed in the Chappuis band of ozone. An altitude range from about 15 to 35–40 km is accessible with the technique outlined above. Below 15 km the line of sight optical depth becomes so large, that these altitudes cannot be “seen” from space in limb geometry, and above 35–40 km the absorption in the Chappuis bands becomes too weak. An example of stratospheric ozone profiles derived from SCIAMACHY limb measurements is shown in the right panel of Figure 1.

4. Retrieval of Tropospheric Trace Columns

In the following we will focus on the retrieval of tropospheric columns of NO_2 and O_3 from SCIAMACHY observations. Significant amounts of both molecular species reside in the stratosphere, playing an important role in the chemistry, radiative transfer and energy budget. However, the two gases are also of particular interest in studying tropospheric pollution, both caused by natural and anthropogenic sources. Soil emissions, industrial burning processes, biomass burning and lightning determine tropospheric NO_X concentrations. It is also one of the most important ozone precursors and locally contributes to radiative forcing.

The results for tropospheric trace gas columns presented in the following are filtered to mask out cloud contaminated pixels. These are identified by applying a empirically determined threshold value to the detected intensity (in counts per second) of the ground scene pixels.

4.1. Reference sector method (RSM)

It has been demonstrated by the Global Ozone Monitoring Experiment (GOME) onboard the ERS-1 satellite that space-borne spectrometers can observe trace gases in the troposphere (Burrows *et al.*, 1999). Different strategies have been developed to separate the stratospheric and the tropospheric components of the absorption signal. Velders *et al*. (2001) performed 3D model calculations of the stratospheric NO_2 field and subtracted this modeled component from total columns observed by the GOME instrument. Leue *et al*. (2001) used DOAS retrievals above clouds over oceans to determine the stratospheric columns. They applied an image processing method to interpolate it along the corresponding latitude band. The fundamental assumption behind this approach is the longitudinal homogeneity of the stratospheric NO_2 layer. This assumption is generally justified as the NO_2 concentration in the atmosphere is predominantly determined by photolysis of the reservoirs and therefore mainly a function of solar zenith angle (SZA). For satellite measurements from a sun-synchronous orbit, spectra at any given latitude are observed under the same SZA on a given day. This condition is exploited in the reference sector method (RSM) (Richter and Burrows, 2002), where retrieved *SCD*s in a longitudinal range over a relatively clean area are subtracted from the corresponding measurements at equal latitude. The resulting excess slant column over polluted regions is interpreted as the tropospheric contribution, which can be transformed into a vertical column by applying a tropospheric *AMF*. As an example Figure 2 shows the tropospheric column amounts of NO_2 over China, Korea and Japan as inferred from SCIAMACHY observations using the RSM method. In the plot, which represents a monthly average for February 2004, the industrial centers of this region are clearly identifiable. Most prominent is a large plume of NO_2 off wind the Shanghai area carried out to the Yellow Sea.

For most atmospheric scenarios, the RSM technique has proven to yield plausible results. However, it is bound to fail in conditions under which the assumption of longitudinal homogeneity of the stratospheric NO_2 field is no longer justified. Such conditions clearly occur close to the polar vortex and during major changes in

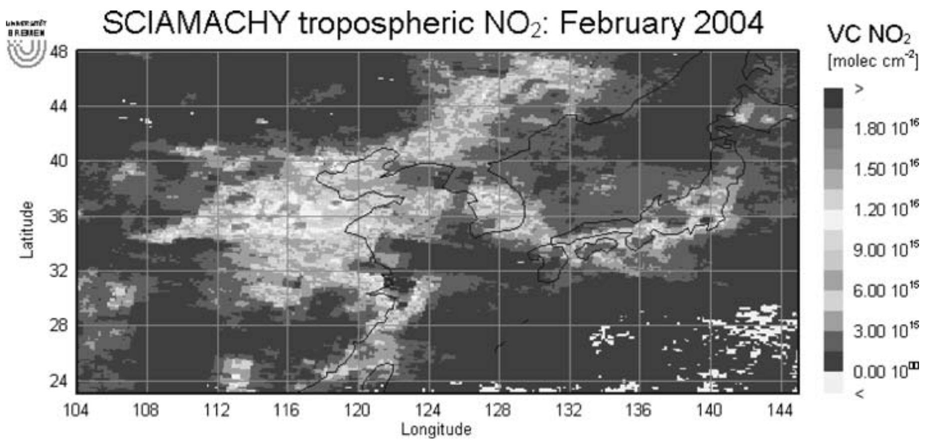

Figure 2. Monthly average of tropospheric NO_2 columns over China, Korea and Japan, derived using the LNM method. The industrial sources of NO_2 emission are clearly visible.

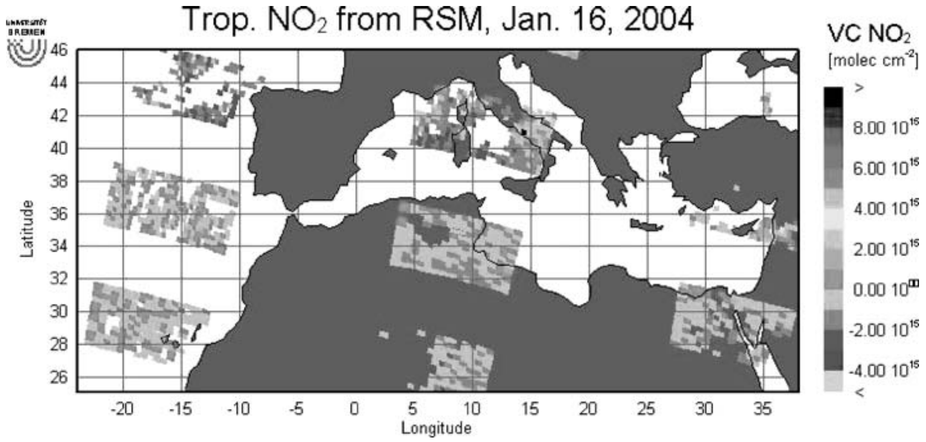

Figure 3. Tropospheric NO_2 columns derived from the RSM method. The technique yields large areas with strong negative bias due to inhomogeneities of the stratospheric NO_2 fields with respect to the reference sector.

stratospheric dynamics. In mid-latitudes, the RSM regularly yields negative tropospheric columns in winter and spring, when stratospheric NO_2 in polar regions is strongly reduced and zonal asymmetries in stratospheric temperatures lead to large zonal gradients. Such a situation is shown in Figure 3, which shows tropospheric *VCD* from RSM analysis of SCIAMACHY data over the Mediterranean on January 16th, 2004. The color scale is chosen to display ground scene pixels with negative values of *VCD* in blue. As can be seen from the plot, the RSM yields unphysical values with a large negative bias over wide areas, which are likely to result from low stratospheric NO_2 columns owing to lower temperatures than over the Atlantic reference sector (20°–30°W). As will be shown in the subsequent section, the incorporation of stratospheric profile information from limb analysis significantly improves the results under such conditions of stratospheric inhomogeneity with respect to the reference sector (see Figure 5).

For ozone, the applicability of the RSM technique is far more limited than for NO_2, as longitudinal homogeneity can only to some degree be assumed in tropical regions. Its lifetime is long enough to enable the introduction of inhomogeneities of the stratospheric ozone field by transport processes. Also, the maximum of the vertical concentration profile is lower for O_3 than for NO_2, making ozone much more sensitive to changes in the tropopause height. Due to these reasons, the requirements for tropospheric ozone retrieval using RSM are rarely met outside the tropics. This is demonstrated by Figure 4, which shows the August 2003 average of RSM derived tropospheric O_3 VCD. While the tropical regions show expected ozone concentration patterns (e.g. due to biomass burning in Africa), the RSM technique yields unphysical values for higher latitudes. In the subsequent section, we will demonstrate that a combination of limb- and nadir observations, which

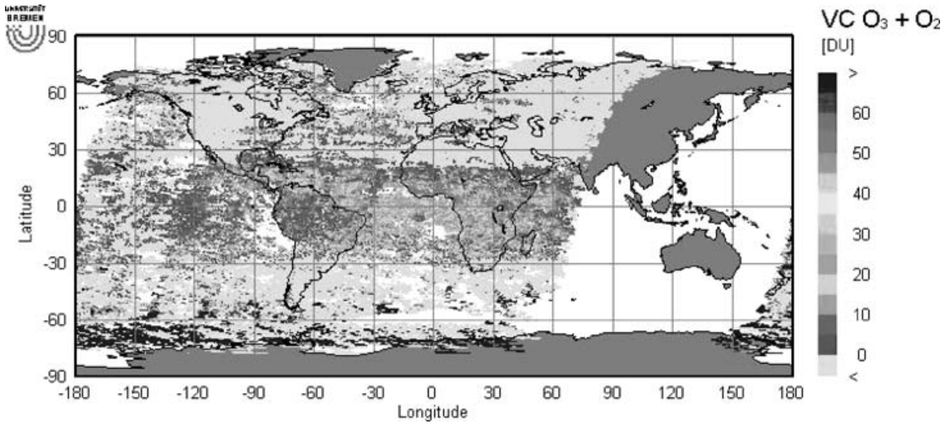

Figure 4. Tropospheric O_3 columns derived using the RSM approach. The method only yields plausible results in tropical regions, where the condition of longitudinal homogeneity is met. Throughout the higher latitudes, the results are negative, indicated by gray color.

is a unique feature of the SCIAMACHY instrument, can be used to characterize stratospheric inhomogeneity and thus extend the ability to derive tropospheric NO_2 and O_3.

4.2. Combination of SCIAMACHY limb and nadir measurements

Improving the ability to derive tropospheric concentrations of trace gas species by limb- and nadir measurements was one of the reasons for implementing the limb-nadir matching mode of the SCIAMACHY instrument. The spectrometer continuously alternates between limb-and nadir observation geometry. The viewing angles for the acquisition of limb spectra were adjusted in such a way that the tangent points are as close as possible over the region, which is covered by a nadir state measured 7 minutes later. This allows to infer vertical stratospheric concentration profiles directly over the region of the nadir measurement. Integrating these profiles from the tropopause upwards yields the stratospheric *VCD* above the target area.

In our implementation of the limb-nadir matching (LNM) technique, we compute tropopause heights from meteorological model data provided by the European Center for Medium Range Weather Forecast (ECMWF). These data comprise three-dimensional model calculations of pressure and temperature on a latitude/longitude grid of 1° resolution and 60 height levels up to about 60 km. From these data, tropopause heights can be computed using various different criteria. We used a combination of two concepts to define the boundary between troposphere and stratosphere: For the tropics (±20° latitude from the equator) we applied a temperature gradient criterion, which defines the tropopause at the point where the condition $dT/dH > -2$ K/km (where T and H denote temperature and altitude, respectively) is fulfilled. In mid-latitudes, we compute the potential vorticity (PV) from the ECMWF data and set a threshold value of PV $= 3.5 \cdot 10^6$ $Km^2/(sec\ kg)$ to define the tropopause. In the transition region between the two regimes ($\pm 20°$–$30°$ latitude range) both criteria are used and weighted with the distance from the regime boundaries. With this scheme, a global tropopause map is computed from the ECMWF data. In preparation for the application to SCIAMACHY measurements, we average the tropopause values within the distinct nadir states. This in turn yields the lower boundary for the numerical integration of the corresponding NO_2 limb profiles, and thus the stratospheric vertical column density.

As the DOAS analysis of the nadir spectra yields the total *SCD*s for the ground scene pixels (see Equation 1), the stratospheric *VCD* has to be mapped to nadir observation geometry by transforming it to a corresponding stratospheric *SCD*. With AMF_{strat} denoting the stratospheric component of the airmass factor, we can form the difference

$$SCD_{\mathrm{trop}} = SCD_{\mathrm{total,nadir}} - VCD_{\mathrm{strat,limb}} \times AMF_{\mathrm{strat,nadir}}$$

to obtain the tropospheric slant column density $SCD_{\mathrm{trop.}}$

In the computation of AMF_{strat}, we can once more make use of the distribution profiles derived from limb observation. This is done by running a RTM calculation with CDI, using the inferred stratospheric NO_2 profiles as input information, and setting the NO_2 concentration in the height layers below the tropopause to zero. In this way, AMF_{strat} is calculated for all nadir measurements considering the observation geometry (given by SZA and line-of-sight (LOS) of the measurement) and the chosen wavelength window for the DOAS analysis of the nadir spectra. The stratospheric *VCD* is scaled by AMF_{strat}, and subtracted from the total *SCD*. The resulting slant tropospheric column can then be transformed into VCD_{trop} by an estimate of the tropospheric airmass factor AMF_{trop}. The latter is again obtained from an RTM calculation, in which assumptions on the tropospheric absorber profile and other factors (see Section 2) have to be introduced. It has been shown, that the uncertainties in albedo, cloud fraction and aerosol loading, needed in the computation of AMF_{trop} introduce errors of up to 50% in VCD_{trop} (Boersma *et al.*, 2004). However, this study focuses on the impact of improved knowledge of stratospheric NO_2 and O_3 from limb measurements, enabling qualitatively useful retrievals in problematic conditions and regions outlined above. Therefore, in our comparisons of the LNM technique with the RSM method, we use the same values for AMF_{trop} derived from a standard tropospheric NO_2 profile.

A significant improvement achieved by using limb profiles from the optimal estimation analysis outlined in Section 3 to characterize the stratosphere is shown in Figure 5, which presents the LNM results for the RSM example shown in Figure 3. As can be seen in the plot, the unphysical negative values for tropospheric NO_2 columns have disappeared. However, the tropospheric NO_2 columns over the Sahara (about 3×10^{15} molec/cm^2) seem rather high, which is probably due to using standard airmass factors over high albedo surfaces. Exaggerated total nadir

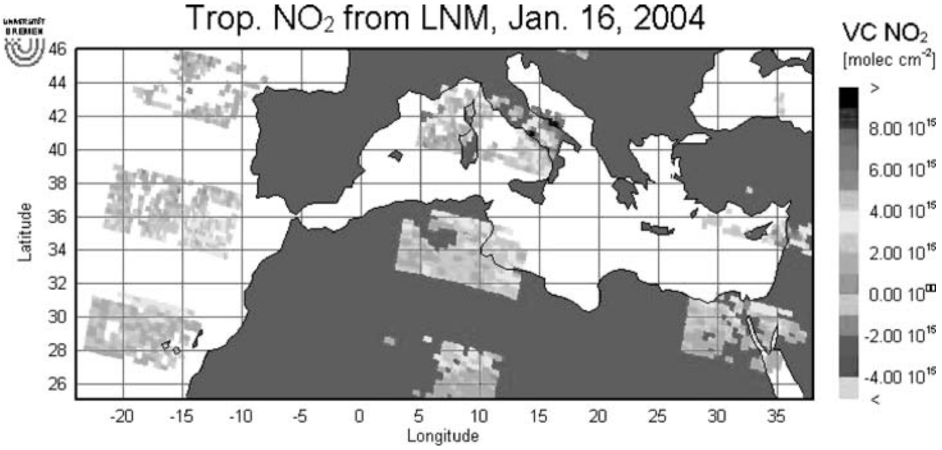

Figure 5. Tropospheric NO_2 columns derived from the combination of SCIAMACHY limb and nadir measurements. Negative values due to stratospheric inhomogeneity shown in Figure 5 do not show up in the LNM results.

columns over snow and white sand are a known problem of our standard global DOAS analysis. A second comparison is shown in Figure 6, which shows RSM and LNM derived tropospheric NO_2 over Southwestern Europe and the Mediterranean Sea on February 7, 2003. As in the previous example, the RSM approach (upper panel) yields negative differences between the reference sector and the target area, indicated by the gray ground scene pixels. These artifacts, which are probably caused by zonal asymmetries in stratospheric temperatures, do not show up in the LNM results (lower panel), as they incorporate information on the stratospheric NO_2 field from limb measurements. The improvement of tropospheric trace gas retrieval by combined limb-nadir analysis might enable a more quantitative assessment of emission events. A closer look at the SCIAMACHY nadir state west of the French coast near the English Channel (north of the Iberian peninsula) reveals elevated NO_2 concentrations along a line, which corresponds to one of the worlds most busy ship routes. Studying NO_x emissions along ship tracks is an ambitious current research objective, which is likely to benefit from improved retrieval techniques for tropospheric trace gases distributions.

The advantage of including measurements of absorber distribution in the stratosphere by combining limb- and nadir observations is even more evident when applying the technique to ozone. While the requirement of longitudinal homogeneity is satisfied for NO_2 in most cases, it is rarely met for ozone at higher latitudes. The LNM approach can therefore be seen as a way to extend our ability to separate and monitor tropospheric ozone to higher latitudes. Figure 7 shows a global picture of VCD_{trop} derived from limb-nadir matching of SCIAMACHY data averaged over the entire dataset for August 2003. The plot incorporates the results from more than 3500 limb profile soundings and their corresponding nadir states. Using LNM, no negative values of VCD_{trop} are obtained in mid-latitudes, as was the case in the

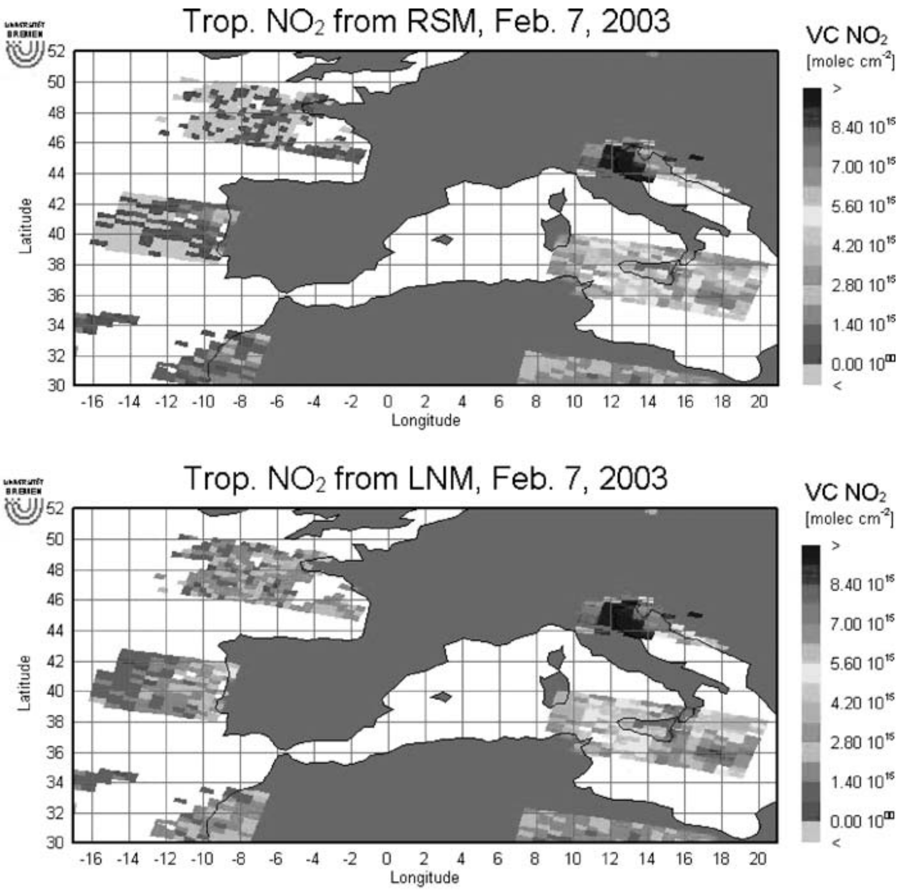

Figure 6. Tropospheric NO_2 columns measured by SCIAMACHY on February 7, 2003 and derived by RSM (upper panel) and LNM (lower panel), respectively.

corresponding plot showing the results of the RSM technique for the same period (see Figure 4). However it should be kept in mind that SCIAMACHY retrievals of ozone can exhibit large errors of estimated 50%. As 90% of the total ozone observed in nadir geometry resides in the stratosphere, the accuracy of LNM derived VCD_{trop} is particularly sensitive to the uncertainty in stratospheric column determination from limb measurements. A quantitative interpretation of these preliminary results is not undertaken here, since a multitude of influencing effects has to be analyzed in ongoing research. However, it can be stated that the LNM technique yields plausible results throughout the investigated period and region, without unphysically large or negative columns, even at higher latitudes. The improvement is evident by comparison with Figure 4, which shows the corresponding RSM results. A comparison with radiosonde soundings and independent remote sensing techniques is currently performed to validate and assess the quality of LNM retrievals.

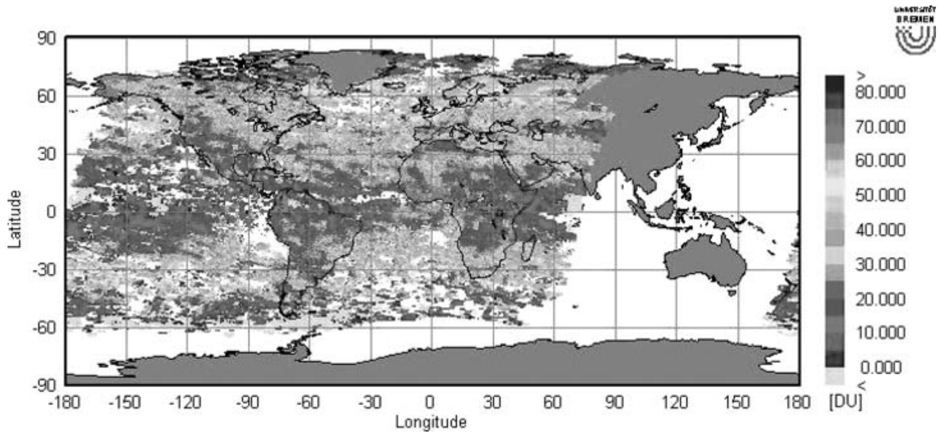

Figure 7. Tropospheric *VCD* for ozone from SCIAMACHY measurements in August 2003. The results have been obtained by applying the LNM technique with more than 3500 stratospheric ozone profiles from limb measurements and averaging over the observation period. No data were available over East-Asia and Australia, the smaller gaps result from applying the could mask to the ground scene pixels.

5. Summary and Outlook

We have shown several examples of space-borne remote sensing of trace gas species using the SCIAMACHY instrument. The concepts of various data analysis strategies have been presented with special attention to their applicability to separate stratospheric and tropospheric distributions of NO_2 and O_3. Case studies showing the limitations of deriving tropospheric columns from the traditionally used reference sector method have been presented. The newly developed limb-nadir matching approach was described, which involves a combination of stratospheric profile information from SCIAMACHY limb observations and total slant columns from DOAS analysis of nadir spectra, as well as tropopause calculations from meteorological model data. The LNM technique was shown to yield plausible results for tropospheric NO_2 and O_3 columns under conditions of stratospheric inhomogeneity, which cause the RSM method to fail. While further improvement is expected from an optimized characterization of tropospheric airmass factors, it can be concluded that combining limb- and nadir spectroscopic measurements from SCIAMACHY will significantly extend our ability to detect and monitor global tropospheric trace gas concentrations.

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