MAX-DOAS Aerosol Corrected Tropospheric Vertical Columns of Nitrogen Dioxide

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ABSTRACT

Nitrogen Dioxide (NO_2) is an important component of air pollution. It is observed on a global scale by satellite instruments such as GOME, SCIAMACHY and OMI. Worldwide there are only few observation sites where nitrogen dioxide tropospheric columns are monitored on an operational basis. For satellite validation it is essential to perform these ground based measurements with good global and seasonal coverage preferably in a network of similar and inter-calibrated instruments. Since November 2007 multi-axis differential optical absorption spectroscopy (MAX-DOAS) observations of NO_2 are performed on an operational basis at the Royal Netherlands Meteorological Institute (KNMI). For this a Mini MAX-DOAS instrument is used. This relatively low cost instrument could be a candidate for a global network as mentioned above. We will explain the tropospheric column retrieval algorithm which includes a correction for aerosols. The Doubling Adding radiative transfer model is used to create look up tables for the Air Mass Factors as a function of radiation intensity which depends on aerosol optical thickness (AOT) and boundary layer height. In addition we will show results from several studies based on the observations. First: a comparison of retrieved AOT and direct sun observations of AOT from the CESAR site in Cabauw, The Netherlands. Second: an analysis of the retrieved tropospheric vertical columns of NO_2 in this highly polluted region. Third: comparison with SCIAMACHY and OMI tropospheric NO_2 columns. Finally: first results from the CINDI inter-comparison campaign for NO_2 measuring instruments. This campaign has taken place in Cabauw, in June and July 2009. Many different groups have participate in this campaign which had resulted in a variety of simultaneous in-situ and remote sensing observations of NO_2, other trace gases and aerosols.

INTRODUCTION

Differential Optical Absorption Spectroscopy (DOAS) [1] is a powerful method to retrieve information on atmospheric trace gases from high spectral resolution UV/VIS remote sensing observations. Depending on the observation strategy information can be inferred on either stratospheric or tropospheric species. The Multi Axis DOAS (MAX-DOAS) technique [2] is especially suited for observations of trace gas absorptions in the boundary layer. Although first MAX-DOAS measurements were made in the 1990s there is still no satisfying standardized method to convert the measured differential slant columns (DSC) in tropospheric vertical columns (TVC) or concentrations. The DSC expresses the *difference* in absorption along the light path for the two viewing directions: the off-axis direction and the zenith direction. If one knows the air mass factors (a quantity proportional to the effective photon path) for the two viewing directions then the TVC can be determined from the ratio of the DSC to the difference in air mass factors (DAMF). Since this effective photon path is a function of many parameters such as the solar position, observation geometry, clouds, surface albedo, horizontal and vertical distributions of aerosols, etc., there are many unknowns that should in principle be retrieved from the measurements along with the trace gas absorptions to have a reliable retrieval of the TVC. Radiative transfer simulations help to quantify the uncertainty in the retrieval caused by inaccurate knowledge of each of the parameters. These simulations show that under cloud free conditions the main uncertainty in the effective light path comes from aerosols. We have applied a method to estimate AOT and to combine this with a look up table of AOT depending air mass factors. The results give confidence that this approach is useful under cloud free conditions – i.e. the most relevant conditions for satellite inter-comparison. A major advantage of this approach is that it provides a self-consistency check. Vertical columns are derived independently from the differential slant columns in each viewing direction. The spread in these vertical columns can be interpreted as a measure of the accuracy of the retrieval.

MEASUREMENTS

The instrument used in this study is a so called Mini MAX-DOAS, produced by the German company Hoffmann GmbH. Spectrometer and telescope are both contained in one metal box which is mounted on the axis of a stepper motor to which allows to rotate in the vertical plane. Stabilizing the temperature by cooling is made possible by a Peltier element on top of the box. Incoming light is focused by a lens (F = 40 mm) on the entrance of an optical fiber which is connected at the other end to the spectrometer: Ocean Optics, USB2000, crossed Czerny-Turner type spectrometer with a Sony ILX511 CCD detector (2048 pixels) and a wavelength range from 290 to 433 nm. The field of view (FOV) of the instrument is around 0.35° (equivalent width). In this study the instrument was located on the roof of the KNMI building in De Bilt, The Netherlands (52.101 N, 5.178 E). The azimuth viewing direction of the instrument is fixed at 46° N which was chosen because of trees and buildings surrounding the site and since the sun is out of the field of view

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throughout almost all seasons. Spectra were recorded at the 0°, 2°, 4°, 8°, 16°, 30° and 90° elevation viewing angles with an integration time of 30 seconds for each elevation resulting in a total of around five minutes per cycle. Observations were done for solar zenith angles smaller than 85°.

DOAS analysis of spectra

NO_2 DSC are derived from the measured spectra with the Qdoas software developed at the Belgian Institute for Space and Aeronomy (IASB/BIRA) (Qdoas is the platform independent version of Windoas [3]). For each spectrum the nearest (in time) zenith spectrum was selected as a reference. The cross sections of NO_2 and O_3 were included in the DOAS fitting routine as well as the Ring cross section. The fitting interval was 415 to 429 nm.

Relative Intensity Observations

Although the instrument has no radiometric calibration, there is a method other than DOAS to derive information on atmospheric constituents. This is the observation of so called relative intensity. In this work intensity observations refer to the number of CCD-pixel counts averaged over a certain spectral interval (426-429 nm). Relative intensity is the ratio of this value determined from a spectral measurement in a certain viewing direction to a zenith sky measurement. In the absence of clouds relative intensities in the visible are mainly influenced by Rayleigh and aerosol scattering and absorption. Since Rayleigh scattering is quantitatively well known, relative intensity observations contain much information on aerosols.

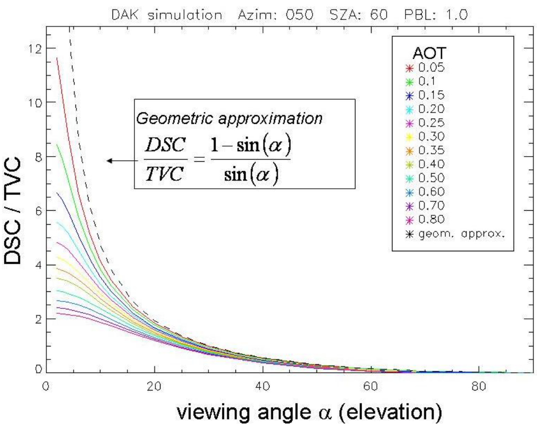

Figure 1.

RADIATIVE TRANSFER SIMULATIONS

To study the effect of various parameters (aerosol optical thickness, single scattering albedo, asymmetry parameter, boundary layer height) on simulated DSC and relative intensities we have used the Doubling Adding KNMI line by line radiative transfer model [4]: DAK version 3.1.2. This version includes a pseudo spherical correction which accounts for the curvature of the earth before scattering occurs for the first time. The sensitivity studies show that knowledge of AOT and boundary layer height is essential to simulate the DAMF (figure 1). Since relative intensity observations are especially sensitive to AOT (figure 2), these observations can in principle be used to retrieve this unknown. Polarization needs to be taken into account here as was shown by the sensitivity study and comparison with actual measurements. Furthermore the radiative transfer studies demonstrate that the altitude dependent sensitivity to NO_2 peaks in the lower troposphere especially for the low elevations.

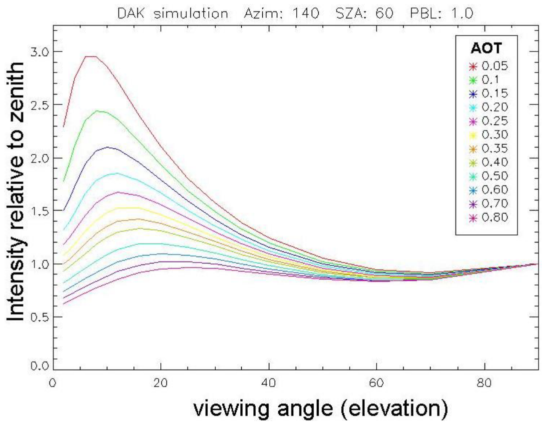

Figure 2.

TROPOSPHERIC COLUMN RETRIEVAL

A TVC can in principle be derived from the DSC in each elevation viewing direction independently provided one knows the right DAMF for each elevation (α). A default approach to derive the DAMF is described in [2]. This method is also referred to as the geometrical approximation:

$$\frac{DSC}{TVC} = \frac{1-\sin(\alpha)}{\sin(\alpha)} \tag{1}$$

Model simulations show that this approximation is accurate to within 20% for the 30° elevation but much less accurate for the lower viewing angles. In our study the DAMF was modeled for each viewing direction, solar position, and AOT as the ratio of the simulated DSC to the model input TVC, which resulted in a DAMF lookup table (assuming climatologic values for the other parameters to which the retrieval is less sensitive). The same was done for relative intensities. The relative intensity lookup table is used to retrieve the AOT from the relative intensity measurements. Knowing this AOT, a DAMF could be selected from the DAMF lookup table. In this way a TVC is derived for each viewing angle separately. Finally an average TVC is computed from the 4°, 8°, and 16° elevations. The lower viewing angles were not included because those are very sensitive to any misalignment of the instrument in the vertical plane. The 30° elevation was not included since this measurement has a much more local character in terms of horizontal distances. Since the purpose of this study is to compare with satellite observations, it is preferable to have observations that are representative for a relatively large horizontal distance. The difference between the maximum and the minimum TVC derived from this set of three elevations is defined to be the uncertainty in the average TVC.

AOT RETRIEVAL

As an intermediate step in the TVC retrieval the AOT is retrieved for each viewing angle. Similar to the TVC, an average AOT can be determined and an uncertainty (figure 3).

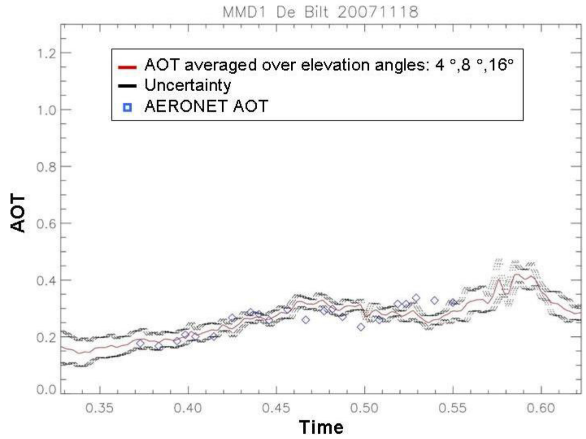

Figure 3.

A comparison is made of this AOT with AERONET cimel sun photometer measurements at Cabauw (25 km distance), see figure 4. This comparison shows a fairly good agreement, keeping in mind the difference between the measurement methods: scattered sunlight versus direct sun observations. This gives some confidence in the AOT retrieval based on the MAX-DOAS relative intensity observations.

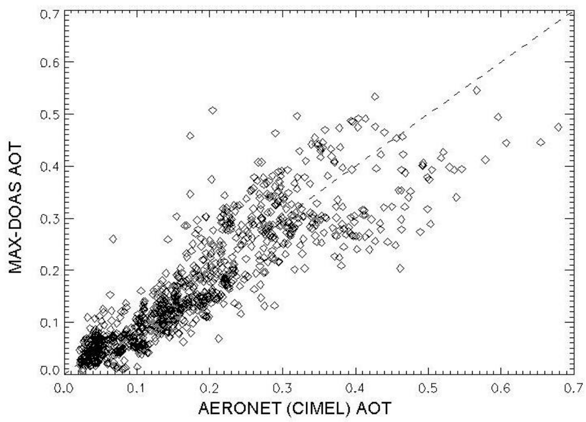

Figure 4.

COMPARISON WITH SATELLITE OBSERVATIONS

A comparison the ground location. Since SCIAMACHY has less overpass data in the same period due to the larger pixels size (which increases t was made with satellite observations of the NO_2 TVC products of OMI and SCIAMACHY [5,6]. For OMI overpass-data only those pixels were selected that were on top of the ground location. Since SCIAMACHY has less overpass data in the same period due to the larger pixels size (which increases the chance of cloud contamination in a pixel) and the alternating limb and nadir viewing modes, a different criterion was used to select the satellite data: all pixels whose center is within 40 kilometers of the ground location were selected. Selection of ground based measurements was done based on a maximum in the uncertainty in the average TVC.

The results show that putting relatively strong constraints on the uncertainty of the selected ground based observations significantly improves the bias in the comparison with OMI (figure 5 & 6). The drawback is the loss of data points to compare. For SCIAMACHY there are yet too few coincident observations to draw any conclusions.

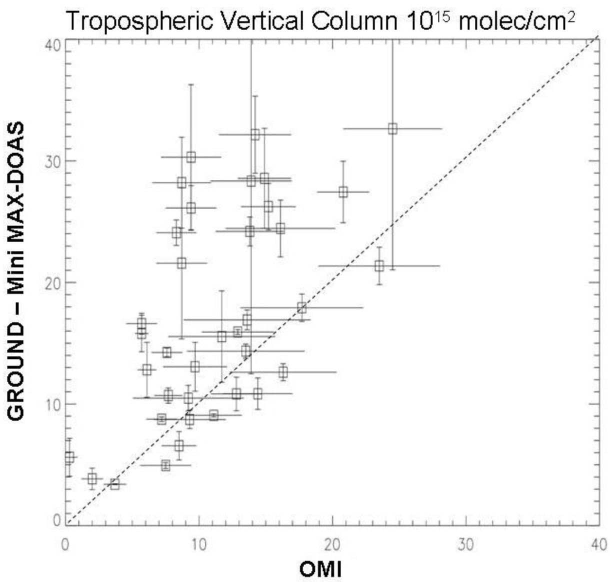

Figure 5.

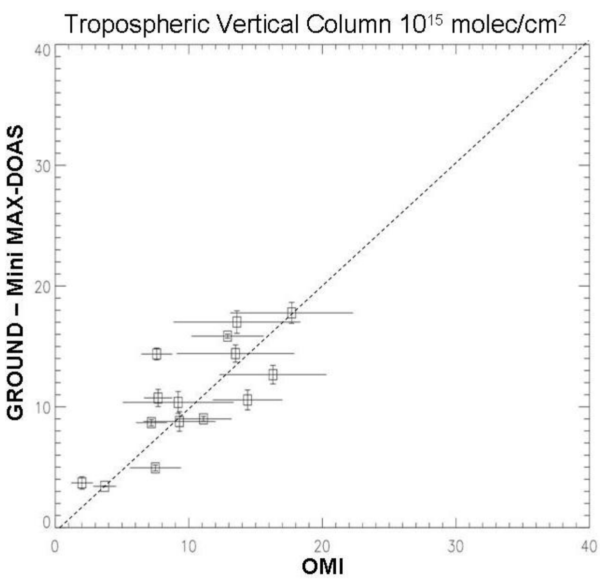

Figure 6.

DISCUSSION

The geometrical approximation mentioned above provides a fairly accurate way to convert DSC to TVC making use of only the 30° elevation. The method proposed here is more complex and more sensitive to any form of clouds. Despite this, there are certain advantages compared to the geometrical approximation method. First is that it convincingly takes into account the effect of aerosols on the retrieval. In the comparison of ground based and satellite observations it is important to give an estimate of effects influencing their relative bias. Aerosols are not taken into account in the geometrical approximation, nor is the solar position. The use of low elevation angles is a second advantage. Using the low elevation angles with the higher DAMF in the lowest layer of the troposphere makes the observation sensitive to NO_2 at larger horizontal distances. Some, but not all, of the disadvantages of the retrieval method applied here (e.g. sensitivity to clouds, many assumed fixed parameters) are counterbalanced by the fact that the error estimate gives a way to assess if or if not the retrieval was successful. In the case of measurements affected by clouds the error on the averaged TVC will be large since the DAMF lookup-table is created for cloud free conditions. Future studies will focus on estimating the boundary layer height. Additional information may come from O_2-O_2 DSC measurements. Model simulations have indicated that like relative intensity, absorption by O_2-O_2 is sensitive to both AOT and boundary layer height. Combined use of those two quantities may lead to improved retrievals, hopefully leading to more TVC retrievals with a low uncertainty based on the same set of observations. Estimation of a boundary layer height allows in addition to convert the TVC to tropospheric concentrations, a quantity of more use for inter-comparison with other types of NO_2 measurements such as *in-situ*.

CONCLUSIONS

(1) MAX-DOAS observations of NO_2 TVC are very sensitive to aerosols, especially the viewing angles below 20°.

(2) We have applied a new method to derive TVC of NO_2 from ground based MAX-DOAS observations taking into account the effect of aerosols on the effective light path. This method relies on the effect of aerosols on relative intensity observations and can only be applied under conditions without any clouds. The method is relatively easy to implement, it can be applied at many wavelengths and it provides a way to estimate the error. Furthermore the low viewing angles can be used which has the advantage that those are more representative to horizontal distances at the order of typical satellite pixels.

(3) AOT is derived as an intermediate product. There is a relatively good agreement with the AERONET direct sun observations of AOT.

(4) A drawback of this method is that it is only successful under cloud free conditions.

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