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AIR MASS FACTOR CALCULATIONS FOR GOME MEASUREMENTS OF LIGHTNING-PRODUCED NO_2

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ABSTRACT

The quantification of lightning-produced nitrogen oxide is difficult as the NO_x-measurement must be concurrent with the thunderstorm. Also the interpretation of the measurements is complicated by the inhomogeneity of the clouds. Currently, the global NO_x-production by lightning is estimated to be 2-20 TgN/a. This study analyses the possibility of the detection of NO_2-production by thunderstorms via satellite. Lightning can be detected by NASA/NASDA-satellite project Lightning Imaging Sensor (LIS). The European satellite spectrometer Global Ozone Monitoring Experiment (GOME) can be used for the coincident NO_2-measurements. For a quantitative analysis of NO_2 measurements, the air mass factor (AMF) is needed. AMF are calculated using the radiative transfer model SCIATRAN and indicate the sensitivity of GOME for NO_2 produced by flashes. The results of the AMF calculations show, that the sensitivity of GOME is largest at the top of a thundercloud, where most of the flash produced NO_x is released. The measurement is insensitive to NO_2 under the thundercloud. For a quantitative analysis, the height of the cloud is needed, but the dependencies of the sensitivity to the height of the NO_2 outside the cloud and the cloud particles density are negligible.

INTRODUCTION

Every second 20 – 100 lightning discharges occur globally in thunderclouds [Christian, 1998]. About 70%-90% of the lightning is intracloud (IC) [Mackerras 1998], and much of the lightning-produced NO_x finds its way to the top of the cloud [DeCaria et al., 2000]. As early as 1827 von Liebig suggested that the nitrogen oxides NO and NO_2 (NO_x) are produced by lightning in large quantities [von Liebig 1827]. The physics of lightning as well as the role of the lightning chemistry in the nitrogen cycle is not fully understood today. At air temperatures above 2000 K, molecular oxygen breaks down into oxygen atoms that initiate the Zel'dovitch mechanism [Zel'dovitch 1966]

Dissociation	$O_2 + M$	$O + O + M$
Zel'dovich mechanism	$O + N_2$	$NO + N$
	$N + O_2$	$NO + O$

The recombination of N and O atoms can form as much as 8% by volume NO [Chameides 1979]. NO produced in lightning can be converted to NO_2 by reaction with O_3; in a matter of minutes, NO and NO_2 reach a steady state determined by the NO_2 photolysis rate coefficient and the temperature dependant rate constant for NO + O_3 [Wang et al., 1998; DeCaria et al., 2000; Stith et al., 1999]. NO_2 can be converted to HNO_3 via reaction with OH, but the reaction is slow. It therefore is possible to measure enhanced NO_2 values hours or even days after the lightning stroke.

Currently, the global NO_x-production by thunderstorms is roughly estimated to be 0.3-22 Tg(N)/a [Huntrieser 1998]. Precise knowledge of the atmospheric NO_x amounts is important as NO_x plays a key role in the formation of tropospheric ozone. Also, NO_2 absorbs in the wavelength range around 400nm and under polluted conditions might locally contribute significantly to radiative forcing in the lower troposphere [Solomon 1999]. Direct measurements of NO_X production by flashes are difficult, as they must be concurrent with the thunderstorm which is problematic for air borne sensors. Also, the number of measurements is necessarily limited, and all current estimates are based on extrapolation of a few local measurements to a global scale. Satellite measurements of NO_2 could fill this gap if they could be linked to individual lightning events. In one previous study, UARS measurements have been used to correlate enhanced NO_2 values with climatological lightning frequencies, but only on a statistical basis [Zhang 2000]. By combining measurements of the Global Ozone Measuring Experiment (GOME) and data from lightning detecting sensors such as the Lightning Imaging Sensor (LIS), direct evidence for NO_2 production from lightning could be gained. In this study, the sensitivity of GOME towards NO_2 from lightning is evaluated, focusing on the changes in radiative transfer introduced by a large thunderstorm cloud observed from a space borne UV/vis sensor.

SATELLITE MEASUREMENTS AND RETRIEVAL OF NO_2 IN THE TROPOSPHERE

Global detection of lightning can be provided by the satellites Lightning Imaging Sensor (LIS) [Christian 1999] and Optical Transient Detector (OTD) [Christian 1996]. The space sensor LIS is a CCD sensor carried by the American Japanese satellite TRMM, launched on at the 28th of Nov. 1997 The instrument scans the latitude range from 35°N to 35°S with a 90% efficiency for lightning detection and provides near to continuous observations of lightning in the tropics and subtropics.

Measurements from the space instrument Global Ozone Monitoring Experiment [Burrows 1999] can be used for the retrieval of NO_2. In April 1995 the nadir viewing GOME spectrometer mounted on the European satellite ERS-2 was launched in a polar, sun-synchronous orbit with an equatorial crossing time of 10.30am local time. GOME scans solar backscatter spectra between 240 and 790 nm with a spectral resolution of 0.2 - 0.4 nm. Each of the three forward scans covers a ground pixel of 320 km x 40 km. The measured spectra can be used for the retrieval of a number of trace gases including O_3, NO_2, H_2O, BrO, OClO, HCHO and SO_2 [Burrows 1999]. The retrieval method is based on the Differential Optical Absorption Spectroscopy (DOAS) [Noxon 1975, Platt/Perner 1984]. For the spectral fitting of the trace gas NO_2 the wavelength window of 425 – 450 nm can be used. The DOAS retrieval results in slant column densities (SCD), which are the integrated column of the absorber along the light path through the atmosphere. The SCD includes both the tropospheric and the stratospheric columns.

To separate the tropospheric from the stratospheric contribution, the reference sector method can be used as described in [Richter 2001]. In this method the stratospheric proportion of the total column is estimated by the total column measured over an unpolluted region such as the Pacific Ocean at the same latitude. The resulting tropospheric slant columns (TSCD) depend mainly on the solar zenith angle (SZA), the vertical profile of the trace gas and – if present – the characteristics of clouds. For a quantitative analysis of NO_2 production by lightning it is necessary to correct for the length of the light path and to compute the tropospheric vertical column densities (TVCD). This is achieved by division by the air mass factor (AMF) [Solomon, 1987]:

$$\text{AMF} = \frac{\text{Slant Column Density (TSCD)}}{\text{Vertical Column Density (TVCD)}}$$

The AMF is a measure of the sensitivity of a measurement towards NO_2, larger values indicating higher sensitivity. In this study, AMF have been computed for a number of different scenarios and are used to characterize the dependence of the measurement sensitivity on cloud parameters and the height of the NO_2 layer. In Figure 1, the relevant light paths contributing to the signal at the satellite are shown for both clear-sky and cloudy situations. For clear sky conditions, single scattered or reflected photons are the dominant contributions to the intensity measured at the top of the atmosphere. Therefore, the AMF mainly depends on SZA and surface albedo.

In the cloudy scenario, the cloud layer determines the signal as shown in Figure 1b. The single scattering contribution above the cloud (1) is similar to that for the clear-sky situation. Reflection at the cloud top (2) and multiple scattering inside the cloud (3) depend strongly on the cloud properties. Single scattering below the cloud (4) and reflection at the surface (5) are again similar to the clear sky scenario, but the number of photons originating in that region that are actually detected at the satellite is much smaller as a result of the cloud layer. In contrast to ground-based measurements, where all photons have passed through the cloud and light path length enhancement in the cloud is very important [Winterrath 1999], the signal measured at the satellite is dominated by reflection at the cloud top.

For inhomogeneous clouds or broken cloud cover, additional light paths are possible both vertically and horizontally, increasing the complexity of the radiative transfer. However, for the large tropical thunderstorm clouds discussed in this study, these effects can be neglected.

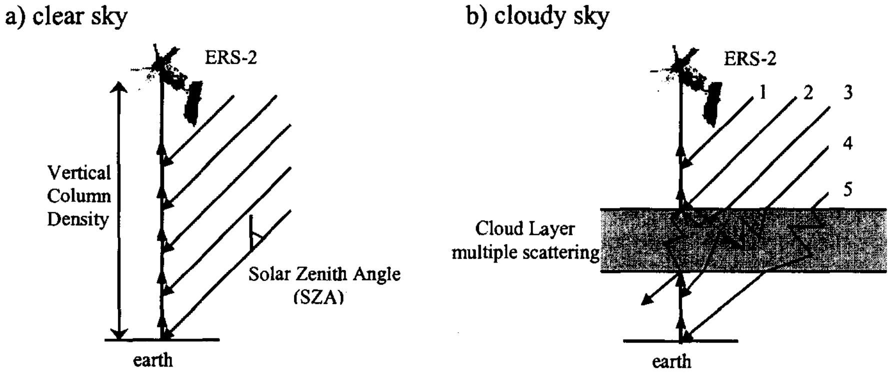

Fig. 1. a) A clear sky scenario is dominated by single scattering and the surface reflection.
b) A scenario with a cloud layer. The light paths to be considered in this case are single scattering above the cloud (1), reflection at the top of cloud (2), multiple scattering inside the cloud (3), single scattering below the cloud (4) and reflection at the surface (5).

AIR MASS FACTOR CALCULATIONS FOR NO_2 PRODUCTION BY LIGHTNING

In this study, air mass factors have been calculated using the radiative transport model SCIATRAN V1.0 developed at the IUP Bremen [Buchwitz 2000]. SCIATRAN is based on monochromatic radiative transport equations for a plane parallel vertically inhomogeneous atmosphere. It is an extension of the radiative transport model GOMETRAN [Rozanov, 1997]. For the calculation of AMF SCIATRAN uses a pseudo spherical approximation. The parameterization of the cumulonimbus cloud type I (Cb I) is based on Stephens [Stephens 1979] and Fouquart [Fouquart 1994] classification [Kurosu 1997]. A tropical scenario has been assumed for the profiles of pressure and temperature. All computations have been performed at a wavelength of 435 nm, representative for the wavelength region used in the NO_2 analysis. In principle, all results depend on wavelength, but for the small spectral window used in the DOAS analysis, this can be neglected.

To quantify the influence of different atmospheric parameters such as position of the NO_2 layer, the position of the cloud and cloud microphysical parameters on the sensitivity of satellite nadir measurements of NO_2 in thunderstorms, air mass factors have been calculated for different heights of a 1 km thick NO_2 layer, different cloud heights and thickness and different cloud particle densities.

In Figure 2, the dependence of the AMF on the height of a NO_2 layer in a thunderstorm cloud is shown. A 1km thick layer of enhanced NO_2 (5ppb) was shifted from the bottom of the cloud to high above the cloud. For comparison, a stratospheric AMF based on a standard NO_2 profile is also shown. As can be seen, the height of

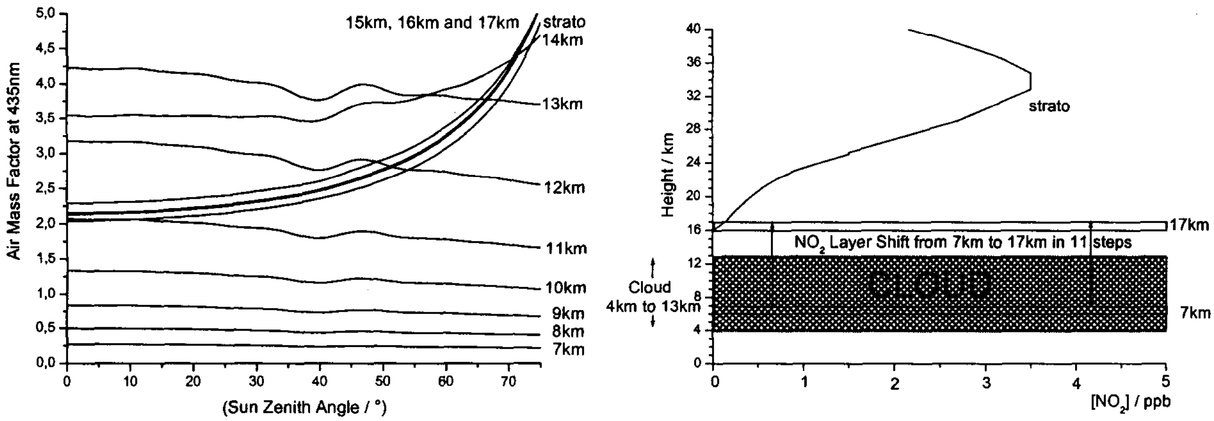

Fig. 2. Air mass factors for NO_2 in a thunderstorm cloud and NO_2 profiles used. The figure shows the AMF calculations for NO_2 at 435 nm. The Cb I cloud range used is 4 to 13 km. A 1km layer of 5 ppb NO_2 was shifted through the troposphere from 7 to 17km in 11 steps. Also one calculation is made with a stratospheric NO_2 profile for comparison. The calculations show that the sensitivity of the satellite nadir viewing geometry for lightning produced NO_2 increases with altitude inside the cloud, but decrease above the cloud (see text for details).

the enhanced NO_2 concentration is an important factor for the AMF. Inside the cloud, the sensitivity of a satellite sensor for lightning produced NO_2 increases with the altitude of the NO_2 layer. For the small solar zenith angles (SZA) typical for GOME measurements in the tropics, the highest sensitivity is displayed at the top of the cloud (see Figure 3). This can be explained by the increase of the optical path length caused by multiple scattering inside the cloud. The decrease in sensitivity towards the ground is a result of the decreasing number of photons that reach that depth and still make it back to the top of the atmosphere. For a NO_2 layer inside the cloud, the AMF is nearly independent of the SZA, in contrast to the AMF outside the cloud, which increases towards larger SZA. The small wavelike structure observed at about 40° - 50° SZA is a result of the Mie phase function of the cloud droplets. For a thunderstorm cloud, the satellite measurements have no sensitivity towards the atmosphere below the cloud. Above the cloud, the sensitivity decreases to the stratospheric limit.

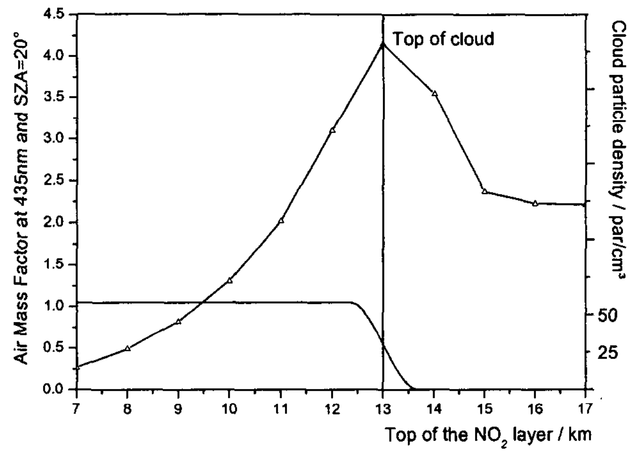

Fig. 3. Air mass factor as a function of NO_2 layer height (cross section of figure 2 at 20° solar zenith angle). The AMF is largest for a NO_2 layer close to the cloud top. Therefore, at SZA 20° the satellite geometry is most sensitive for enhanced NO_2 at the top of the cloud. The smooth variation of cloud particles at the cloud edge, needed for the cloud parameterization in SCIATRAN, is shown in the lower curve.

The smooth change between the two regimes (cloud / non-cloud) is a result of the cloud parameterization in SCIATRAN. To avoid numerical instabilities, the cloud edges extend over 41 sub layers over which the cloud particle density is smoothly reduced from in-cloud values to zero (Figure 3). Therefore, at the cloud top, the cloud is thinner and sensitivity to NO_2 is strongly enhanced by multiple scattering. As a result, the AMF values at 13 and 14km NO_2 layer are special in that they are dominated by the transition zone and might not be representative for a real thunderstorm cloud.

Another important parameter for the AMF is the height of the cloud. In Figure 4, the height of a 1km thick NO_2 layer is varied for different cloud scenarios. In the first case, the top of the NO_2 layer is also the top of cloud, in the second one the NO_2 layer is situated 1km above the cloud, and in the last scenario, no cloud is present. For these case studies, the cloud has a thickness of only 5km, which is not realistic for a thunderstorm but should be enough to simulate a thick cloud. A larger cloud would have limited the range of the possible top of the NO_2 layers. As shown in the figure, the sensitivity towards a NO_2 layer in the top of the cloud increases mutely with increasing cloud top height, probably as a result of decreasing density and therefore increasing importance of light path enhancement. In contrast, the sensitivity towards a layer above the cloud decreases with cloud top height. For high altitudes, it is approaching the sensitivity to a similar NO_2 layer in a clear sky scenario, which is slightly increasing with altitude. This result shows, that the sensitivity towards a NO_2 layer is actually increased if a cloud is situated below it, an effect similar to that of an albedo enhancement in a clear sky scenario. The curves show also, that the AMF are nearly independent of the height of the NO_2 layer, but depend on the position relative to the cloud (Figure 3).

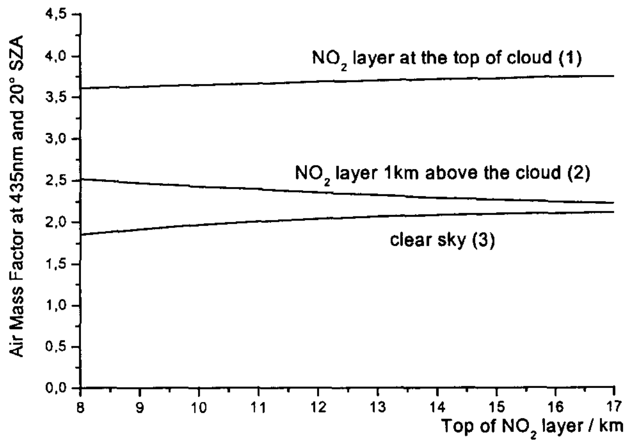

Fig. 4. The dependence of air mass factor on the height of a 1km thick NO_2 layer for different cloud scenarios. The solar zenith angle is 20°. In curve (1) the top of the NO_2 layer is also the top of cloud. In curve (2) the NO_2 layer is 1km above the cloud and the NO_2 layer is shifting through a clear sky scenario in curve (3). The curves show, that the AMF is nearly independent of the height of the NO_2 layer for each scenario, but the AMF vary with the scenario

In a last case study, the importance of the cloud particle density (CPD) has been examined. In SCIATRAN, the CPD is the density of Mie scattering centers per volume and is constant throughout the cloud. The influence of the CPD on the AMF is shown in Figure 5. In this scenario, the cloud extends from 4km to 13km and a 5ppb NO_2 layer is situated between 12km and 13km, the region of the largest sensitivity (see Figure 2). The CPD is varied by approximately 100% around the Fouquart [Fouquart 1994] classification CPD = 58 par/cm³. In the region of CPD<25 par/cm³ the cloud is unrealistically optically thin, showing the transition from a clear sky to a cloudy situation. In the range above 25 par/cm³ the AMF decreases slightly with higher CPD as a result of the increasing absorption inside the cloud, but over the range of realistic values, the dependence is rather small.

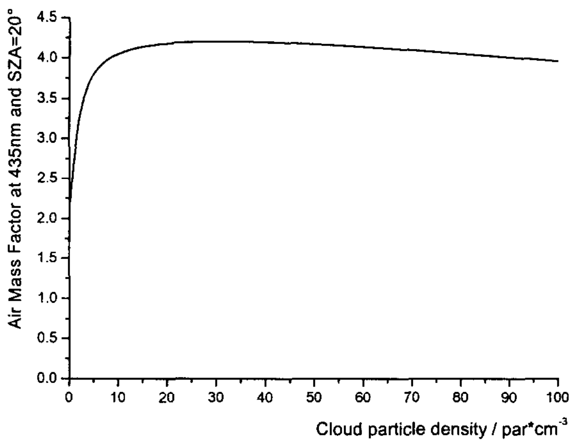

Fig. 5. The dependence of Air Mass Factor (AMF) on the cloud particle density (CPD) for a thundercloud (4km - 13km) and a NO_2 layer of 5ppb between 12km and 13km. In the region of CPD<25 par/cm³ the transition from clear sky to cloudy sky dominates the AMF. Above this region the AMF is depends only weakly on CPD.

To summarize, the satellite nadir viewing geometry is well suited for the measurement of the NO_2 concentration in the upper range of a thunderstorm cloud and above the cloud, in contrast to ground-based DOAS measurements, that are most sensitive to NO_2 in the lower cloud layers and the boundary layer [Winterrath 1999]. Assuming the region of enhanced NO_2 concentrations is situated at the top of the cloud, the AMF is strongly dependent on the height of the NO_2 layer above the cloud top. For the AMF calculation, it is not important to know the precise CPD of the thundercloud or the exact height of enhanced NO_2 above the cloud, as both parameters do not have a large impact on the sensitivity of the measurement.

CONCLUSION

In this study, air mass factors have been calculated for nadir viewing satellite observations of NO_2 in and above a tropical thunderstorm cloud. The results show, that such a measurement has a good sensitivity towards NO_2 above the cloud and in the uppermost layers of the cloud. The sensitivity decreases rapidly towards the lower parts of the cloud and is negligible for NO_2 below the cloud. Roughly 70% of the lightning discharges occur at the top of the cloud. Therefore, satellite measurements from for example GOME should be able to observe a large part of the lightning produced NO_2 with good sensitivity. For a quantitative analysis, the cloud top height must be known and some assumptions must be made on the distribution of NO_2 relative to the cloud top, but the exact cloud particle density is not important. This is important, as cloud top height can be determined from the GOME measurements themselves, but not the CPD.

For a global budget of lightning produced NO_x, GOME NO_2 measurements will have to be linked to coincident measurements of lightning discharges for example from LIS, and a longer time series has to be evaluated. Also, the partitioning between NO_2 and NO has to be taken into account, in particular as the photolysis rates are strongly enhanced above clouds and tend to reduce the NO_2/NO_x ratio under these circumstances.

In summary, GOME is well qualified to detect lightning produced NO_2 from large thunderstorms at the subtropics and the tropics. For the smaller thunderstorms in mid latitudes, future satellite instruments such as SCIAMACHY [Noel 2001] and OMI, with provide better spatial resolution are more appropriate. Another problem of the GOME measurements is the time of measurement (10:30AM local time), that coincides with the diurnal minimum in global lightning strokes. This unfortunate coincidence could be overcome by future instruments that are not in a low earth orbit, but in geostationary orbits as proposed for GeoSCIA [Bovensmann 2001].

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