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Vertical profiles of lightning-produced NO_2 enhancements in the upper troposphere observed by OSIRIS

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Abstract

The purpose of this study is to perform a global search of the upper troposphere ($z \geq 10$ km) for enhancements of nitrogen dioxide and determine their sources. We have searched two years (May 2003–May 2005) of OSIRIS (Optical Spectrograph and Infrared Imager System) operational NO_2 data (version 2.3/2.4) to find large enhancements in the observations by comparing concentrations with those predicted by a photochemical model and by identifying local maxima in NO_2 volume mixing ratio. We find that lightning is the main production mechanism responsible for the large enhancements in OSIRIS NO_2 observations as expected. Similar patterns in the abundances and spatial distribution of the NO_2 enhancements are obtained by perturbing the lightning within the GEOS-Chem 3-dimensional chemical transport model. In most cases, the presence of lightning is confirmed with coincident imagery from LIS (Lightning Imaging Sensor) and the spatial extent of the NO_2 enhancement is mapped using nadir observations of tropospheric NO_2 at high spatial resolution from SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) and OMI (Ozone Monitoring Instrument). The combination of the lightning and chemical sensors allows us to investigate globally the role of lightning to the abundance of NO_2 in the upper troposphere (UT). This is the first application of satellite-based limb scattering to study upper tropospheric NO_2. The spatial and temporal distribution of NO_2 enhancements from lightning (May 2003–May 2005) is investigated. The NO_2 from lightning generally occurs at 12 to 13 km more frequently than at 10 to 11 km. This is consistent with the notion that most of the NO_2 is forming and persisting near the cloud top altitude in the tropical upper troposphere. The latitudinal distribution is mostly as expected. In general, the thunderstorms exhibiting weaker vertical development (e.g. $11 \leq z \leq 13$ km) extend latitudinally as far poleward as 45° but the thunderstorms with stronger vertical development ($z \geq 14$ km) tend to be located within 33° of the equator. There is also the expected hemispheric asymmetry in the frequency of the NO_2 enhancements, as most were observed in the Northern Hemisphere for the period analyzed.

ACPD

7, 5013–5051, 2007

EGU

1 Introduction

Lightning generates most of the NO_x (NO_2 and NO) in the low latitude upper troposphere (Lamarque et al., 1996) and has important consequences for atmospheric chemistry and climate (WMO, 1999; Intergovernmental Panel on Climate Change, 2001). However, considerable uncertainty remains in the magnitude of this natural source of NO_x (e.g. Price et al., 1997; Nesbitt et al., 2000). The vertical distribution of the lightning NO_x emissions also requires further study.

Of the available remote sensing techniques to observe lightning-generated NO_2 in the upper troposphere on a global scale, limb scattering is uniquely suited. Limb scattering has a tremendous advantage over solar occultation in terms of data volume and spatial coverage because the latter technique is limited to measuring only when the sun is on the horizon and in the field of view. Limb scattering provides the opportunity for profile information at high vertical resolution with global coverage. Infrared emission techniques also provide global coverage and can measure at night as well, however MIPAS (Michelson Interferometer for Passive Atmospheric Sounding), the only limb-viewing infrared emission instrument currently available, measures profiles of NO_2 with a vertical resolution 9–12 km below 16 km (Funke et al., 2004). The space borne limb scattering instruments currently capable of providing upper tropospheric NO_2 profile information are SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) (Bovensmann et al., 1999) and OSIRIS (Optical Spectrograph and Infrared Imager System) (Llewellyn et al., 2004). SCIAMACHY can measure NO_2 profiles down to the upper troposphere at low latitudes with an effective vertical resolution of ~3.3 km, dictated by coarse vertical sampling and large instantaneous field of view (2.6 km high by 110 km wide at the tangent point). OSIRIS yields NO_2 profiles with a typical vertical resolution of ~2 km (full width at half-maximum of the averaging kernel for the retrieval technique used below) at the median altitude of the observed NO_2 enhancements of 13 km.

The spectrograph of OSIRIS measures limb scattered radiation in the 280–810 nm

ACPD

7, 5013–5051, 2007

range with ∼1 nm spectral resolution. The instantaneous field of view is 1 km×30 km (vertical, horizontal) at the tangent point, allowing it to see through partly cloudy scenes more effectively and providing better vertical resolution than SCIAMACHY, and is ideal for global studies of vertically-structured phenomena such as NO_2 enhancements from lightning. Odin, the satellite bearing OSIRIS, has a polar sun-synchronous orbit with equator crossing times of 06:00 and 18:00 LT. SCIAMACHY, on the other hand, measures only at ∼10:15 LT. One of the advantages of the orbit of Odin is that OSIRIS can observe approximately the same volume of air in the summer hemisphere within 12 h. This advantage is exploited in this study. The related disadvantage is that OSIRIS gets poor coverage in the winter hemisphere. However, since most of the lightning occurs in the summer hemisphere, the orbit is well suited. Furthermore, with observational local times near twilight, NO_x partitioning is more balanced between NO_2 and NO in the tropical upper troposphere, leading to stronger NO_2 absorption signals compared to midday.

In this paper, we reveal the magnitude and spatial and temporal distribution of the observed enhancements and then highlight some interesting case studies.

2 Method

The retrieval of two years of NO_2 profiles from OSIRIS is a computationally intensive, time-consuming process. We start with version 2.4 NO_2 profiles retrieved operationally with a series of processors. The retrieval method for the operational NO_2 product is described in detail by Haley et al. (2004). The processing of the version 3.0 (v3.0) operational data product has been completed during the writing of this paper. This newest version contains a significant improvement: the retrievals will only extend down to cloud top, if present. We find however, that the operational algorithm for determining cloud tops, occasionally misidentifies the upper end of the stratospheric aerosol layer in the tropics as being a cloud top and thus a significant fraction of tropical upper tropospheric data is lost. Thus we use v2.3/2.4 data (available only to May 2005) and an offline cloud top product described below to filter cloudy cases.

The operationally-retrieved profiles are compared with profiles generated by a stacked photochemical box model (McLinden et al., 2000) for the same local time, month, and latitude. The model extends down to ∼10 km at all latitudes, and thus it covers the retrieval range of operational OSIRIS NO_2 (i.e. 10 to 46 km) with comparable vertical resolution (∼2 km).

When

1. the observed profile exceeds the model profile by 1 order of magnitude at any altitude, or

2. the observed volume mixing ratio (VMR) at a given altitude minus its 1 σ uncertainty is greater than the VMR plus the 1 σ uncertainty for the immediately overlying layer,

the limb scan is selected and the data are reanalyzed as follows.

The first step is to check for the presence of clouds using the ∼810 nm limb radiance profile. Five spectral pixels are co-added to reduce the impact of spikes in the data, which result mostly when the satellite is in the region of the south Atlantic anomaly (Heirtzler, 2002). This wavelength is the longest available with the optical spectrograph. For this wavelength, the atmosphere is optically thin even for upper tropospheric tangent heights and there is no limb radiance maximum at tangent heights (TH) above 10 km even for solar zenith angles approaching 90° for clear-sky conditions. Clouds (or aerosol layers with large optical depths) are identified when the limb radiance profile meets either of the following two conditions:

1) if a limb radiance maximum exists above the lowest tangent height, the corresponding tangent height is the cloud top height.

2) if the ∼810 nm limb radiance scale height is <3.84. A 3.84 km scale height threshold is effective for detecting clouds since radiance scale heights of 4 to 7 km occur in the presence of background aerosols in the upper troposphere and lower stratosphere (UT/LS). The value was determined empirically after extensive examination of cloud

ACPD
7, 5013–5051, 2007

EGU

products generated with different threshold values. At lower threshold scale heights, the algorithm fails to detect thin clouds, whereas at higher threshold scale heights, more false positives are included, e.g. clouds are found when they are not present.

If both cloud identification conditions are met during a scan, the cloud top height is defined to be the higher of the two heights. Both of these conditions are conservative in the sense that minor aerosol layers, including the stratospheric Junge layer, are almost never mistaken for clouds. This point is demonstrated in Fig. 1, which shows the top height of clouds observed in the ~31 000 scans processed offline for this work. There are essentially no cloud detections, for example, at Northern Hemisphere mid-latitudes above 20 km, where the Junge layer would be detected if the cloud identification algorithm were sensitive to it.

Of interest, starting abruptly in mid February 2005 and continuing through April 2005, clouds were observed in the intertropical convergence zone (8° N–4° S) with top heights of >18.3 km. This corresponds to the Walker circulation regaining strength according to El Nino indicators such as the analysis by National Centers for Environmental Prediction (NCEP) of zonally averaged 500 mb temperature anomaly (http://www.cpc.ncep.noaa.gov/data/indices/z500t). The separation between these very high clouds and the usual high clouds observed between 14 and 18 km in the tropics is readily apparent in Fig. 1. Polar stratospheric clouds are also observed annually by OSIRIS in a period near the austral spring equinox (4 September–10 October).

The second step of the offline retrievals is to verify (and correct) the altitude registration. Newer (and more correct) pointing information is used in this study that was not available when v2.3/2.4 of the operational NO_2 product was generated. We use the tangent height of the ~305 nm limb radiance maximum (Sioris et al., 2003), also known as the "knee", to verify or correct the altitude registration. Often, no correction is required as Odin's attitude control system is working remarkably well (e.g. within 500 m) and better than expected (Murtagh et al., 2002; Sioris et al., 2003; Haley et al., 2004). The tangent height offset in any limb scan is corrected if the magnitude of the orbital median TH offset is greater than the standard deviation of the TH offsets during the orbit, based on the approach developed for SCIAMACHY (Sioris et al., 2006).

The ∼305 nm knee indicates an annual variation in the pointing offset, with a departure of ∼500 m in June relative to the rest of the year, for which the mean offset is –297 m (Fig. 2). It is theorized that the June anomaly is caused by the Odin spacecraft twisting slightly as it cools due to the satellite being eclipsed from the sun by the Earth. Even if the apparent seasonal pointing drift were ignored, this error source translates to <15% error on the retrieved NO_2 concentration in the tropical upper troposphere (Haley et al., 2004), and thus this is a secondary source of error considering the magnitude of random errors (illustrated below). Assuming the TH offsets determined by the ∼305 nm knee technique are valid, errors in the NO_2 profile will be even smaller since the TH grid is adjusted prior to the inversion and a June bias will be minimized.

After applying any necessary shift to the altitude registration and limiting the retrieval range based on the cloud top height, each scan is reprocessed using the algorithm described previously (Sioris et al., 2003, 2004), with modifications listed hereafter. The inversion scheme was changed to use Chahine's (1970) relaxation method at each iteration. This modification increases the frequency of successful retrievals but slows down the retrieval because a greater number of iterations are required. The advantage of this retrieval scheme is that the vertical resolution of the profiles is ∼2 km, independent of altitude and the retrievals are almost completely independent of the first guess. These advantages are important for the current study because lightning-produced enhancements are relatively rare according to OSIRIS but consist of very large concentrations of NO_2 often confined in a narrow vertical range (e.g. 2 km), near the bottom of the retrieval range where an optimal estimation approach begins to smooth vertical profiles and rely on a priori information. However, judging from the agreement between the two retrieval schemes (not shown here; see Haley et al., 2004), the reliance on a climatological a priori NO_2 is not as significant an issue as the smoothing of the profiles, and both of these issues are minor.

We have added a surface albedo database (Koelemeijer et al., 2003) into the forward (radiative transfer) model, appropriate for clear-sky conditions. Clouds below the field

ACPD
7, 5013–5051, 2007

of view are still ignored, leading to small errors for large solar zenith angles (Sioris et al., 2003). The retrieval also assumes homogeneous atmospheric composition within an atmospheric layer, ignoring the diurnal gradients that exist between the near and far sides of the limb near twilight (McLinden et al., 2006). In assuming so, we speed up the forward modeling required at each iteration of the inversion at the cost of a minor retrieval error (<10%) in general (McLinden et al., 2006).

The retrieved profiles are examined for NO_2 enhancements. A profile is considered to contain an enhancement if, for the altitude at which an NO_2 enhancement of 1 order of magnitude was found in the operational data relative to the photochemical box model, there is both:

1. an increase relative to the immediately overlying retrieval layer in NO_2 volume mixing ratio (VMR), and
2. an increase in NO_2 slant column density at the closest tangent height underlying the enhanced layer, relative to the immediately overlying tangent height

Observed VMR enhancements are quantified by taking the difference in VMR as compared to the nearest overlying local minimum in the vertical profile of NO_2 VMR. Profiles are converted to VMR from NO_2 number density using air number density profiles from the atmospheric database of McLinden et al. (2002) for the appropriate latitude and month. NO_2 vertical column density (VCD) enhancements are calculated by taking the difference in retrieved NO_2 number density compared to the nearest overlying local minimum in the NO_2 number density profile. These number density enhancements are then integrated over the altitude (z) range exhibiting enhanced values. The VCD enhancements are useful for comparison with nadir viewing instruments (see below).

Some enhancements may be due to aircraft, but it is apparent that aircraft NO_x is only a minor contributor because the spatial distribution of the enhancements (Fig. 3) does not correspond to flight tracks. Another minor source of enhancements is tropopause

ACPD
7, 5013–5051, 2007

NO$_2$ from lightning observed by OSIRIS

C. E. Sioris et al.

EGU

folds. We have filtered out many of these enhancements that occur primarily at northern mid latitudes (40–50° N in spring) by analyzing the orbital cross-sections of retrieved NO_2 (VMR as a function of latitude and altitude) to detect anomalously high NO_2 due to a strong descent of stratospheric air. The stratospheric origin of the NO_2 enhancement was confirmed by making use of NCEP 6-h tropopause data (Kalnay et al., 1996). The tropopause in each of these cases lies at a lower altitude than the climatological tropopause, indicating the intrusion of air from above (where NO_2 VMRs are significantly larger). Further analysis with a high resolution 3-D model is underway to confirm whether the enhanced NO_2 is deposited in the troposphere.

Enhancements also occur in the lower stratosphere during polar spring and are related to renoxification of the polar vortex and descent of mid-latitude air just outside the polar vortex by trapped waves (Tomikawa et al., 2003). These enhancements have been filtered out. The OSIRIS operational NO_2 data showed no large enhancements in the upper stratosphere associated with solar proton events in this time period, except on 12 April 2004 (see Randall et al., 2005).

3 Measurement biases

OSIRIS, like all limb scattering instruments, will have a clear sky sampling bias, because limb scans with towering clouds in the field-of-view are omitted from the reprocessing. However, because of the long lifetime of NO_x in the upper troposphere (∼1 week, Jaegle et al., 1998), the NO_2 generated during thunderstorms can be detected after the clouds move away, tens of hours later, thus limiting the severity of this sampling bias.

There is also a spatial coverage bias as discussed in the introduction. For example, the southernmost sunlit (SZA<90°) latitude observed by OSIRIS on the descending orbital phase (morning) at the start of July, August and September 2004 is 2° S, 5° S, and 16° S, respectively. In October, both hemispheres are covered and then the coverage begins to favour the Southern Hemisphere until February, after which the Northern