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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Vertical profiles of lightning-produced NO_2 enhancements in the upper troposphere observed by OSIRIS" *by* C. E. Sioris et al.

C. E. Sioris et al.

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Anonymous referee 2

We thank referee 2 for his/her effort in critiquing our paper. Plenty of valuable suggestions were given. Below, we have italicized comments and reply immediately below in non-italicized letters.

At some points, the paper is lacking clarity. Especially section 2 (Method) is difficult to read. For instance, on P5017, the bullets 1 and 2 indicate criteria for a first data selection.

We agree and regret that the Method section was written unclearly. To clear this up,

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we now write starting at (P5017, L13): "...the limb scan is selected and the data are reanalyzed using the algorithm described previously [Sioris et al., 2003; Sioris et al., 2004], with modifications detailed below.
The first step of this retrieval algorithm is..."

Then, a 'first step' (P5017, L14) is taken to carry out a cloud presence check, which requires two conditions inconveniently called '1)' and '2)' (P5017, L22 and further).

There are two conditions used for cloud top determination. The numbering has been changed from "1." and "2." to "C1)" and "C2)".

This is followed the authors branching out to interpretation of cloud observation (P5018, L12-20).

We agree that cloud observation interpretation does not belong in the "Method" section. We have moved these statements to the "Measurement biases" section, where biases due to clouds are discussed.

Only on P5018, L21, we come upon the 'second step' -altitude registration- that is treated in depth. Subsequently, on P5020, we find two new bullets labelled '1' and '2' that are brought up to explain how retrieved profiles are examined for NO2 enhancements. In summary, the authors try to clarify a long chain of retrieval steps by repeatedly (4 times) using the numbers 1 and 2, while at the same time trying to twist in some interpretation of observations (clouds), and all this is not improving clarity or readability.

We have changed '1' and '2' to 'a)' and 'b)'.

I am surprised that the authors did not use the differences between the OSIRIS measurements at 06:00 and 18:00 LT. Lightning activity is strongest in the late afternoon over land, and one would expect that the 18:00 LT observations reveal higher NO2 enhancements than at 06:00 LT.

We thought of using this and recorded whether each enhancement was on the AM or

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PM side of the orbit. As mentioned in Section 3, the orbit has gradually drifted since launch in 2001 so that the local time in May 2003 is 6:06/18:06 LT and by May 2005, the local time is 6:30/18:30 LT. Although the local time change does not itself seem too significant, because the equatorial regions of the Earth, where most of the lightning is occurring, are near twilight near 06:00 and 18:00 LT, the range of observable (i.e. sunlit) latitudes has decreased in the PM side of the orbit as a result of the drift from 18:00 to 18:30 LT. This introduces a sampling bias and thus we find that, without accounting for this bias, 200 of 283 enhancements are on the AM side of the orbit. Also, the lifetime of NOx in the upper troposphere is much greater than 12 hours (i.e. days [e.g. Jaegle et al., 1998]), so although the production might be favoured in the PM, the NO2 enhancements in the upper troposphere persist through the following morning. Furthermore, convective (towering) cloud formation is more intense in the afternoon relative to the early morning and this may lead to the omission of many PM lightning cases because OSIRIS cannot see through the clouds. This point is clearly made in Section 3 and there is not much that can be done to remove the bias introduced by clouds on the diurnal signal. Realizing this, we were discouraged from pursuing the diurnal variation further.

Specific comments

P5015, line 12-13: at least in this study there seems to be little justification for the claim that ‘global coverage’ is provided by limb scattering techniques. Are other studies available that pose better examples?

We think that Figure 1 shows the global coverage of OSIRIS profiles quite well. We have added a reference to Haley et al. [2004]. Figure 2 of that paper should be helpful.

P5016, L7-8: what is meant by “OSIRIS can observe approximately the same volume of air in the summer hemisphere within 12 h”? Is this based on OSIRIS taking measurements at 06:00 and 18:00 LT? It is not clear to me.

To clarify we write:

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"One of the advantages of the equator crossing times of the Odin orbit is that OSIRIS can observe approximately the same volume of air in the summer hemisphere within 12 hours."

P5016, L18-22: these lines are not very interesting. Could they be shortened? As a matter of fact the complete first paragraph (until P5017, line 1) of the Method-section holds little useful information.

We have removed the first sentence and shortened the second. We now write:

"We start with operationally-retrieved version 2.4 NO2 profiles. The retrieval method for the operational NO2 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."

P5017, L25: please define 'radiance scale height'.

Radiance scale height is now defined as follows: "C2) if the 810 nm limb radiance scale height (H_I) is <3.84. The radiance scale height is a measure of the rate of change of radiance (I) between spectra at successive tangent heights

$H_I = (TH_n - TH_{n+1})/\ln(I_n/I_{n+1})$ (1)

with $TH_n < TH_{n+1}$."

P5018, L10: please give a reference to support "where the Junge layer would be detected".

We have changed this sentence to read "...above 18 km..." rather than "...above 20 km..." and added a reference to Hofmann and Rosen [1981].

P5018, L28: please define tangent height as TH before using it.

The acronym TH has already been defined on p.5017.

P 5019, L2: please explain what a pointing offset is, and how it is determined.

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We have replaced the vague term ‘pointing offset’ with ‘tangent height offset’. We now write:

We use the tangent height of the simulated 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 metres) and better than expected [Murtagh et al., 2002; Sioris et al., 2003; Haley et al., 2004]. The tangent height offset is defined as the difference between the measured and simulated knee TH. 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 TH offset, with a departure...”

P5020, L11-14: How/how much is this different from P5017, L8-12?

In line 10 of P5020, we have changed “both” to “also” in order to clarify. Those are additional criteria applied to the data reprocessed for this study.

P5020, L13: slant column density should be defined.

We have changed the term “slant column density” to the more simple, yet proportional quantity “absorption optical depth”. The text now reads as follows:

"The retrieved profiles are examined for NO2 enhancements. A profile is considered to contain an enhancement if, for the altitude at which an NO2 enhancement was found in the operational data (see criteria 1-2 above), there is also:
a) (...)
b) an increase in NO2 absorption optical depth at the closest tangent height underlying the enhanced layer, relative to the immediately overlying tangent height."

P5021, L1-8: it would be interesting to mention how often these enhancements occur in Spring.

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The NO2 enhancements related to tropopause folds occur 15-20 times per year, roughly one order of magnitude less frequent than the lightning enhancements. We now write:

"There are approximately 15 such enhancements per year."

What climatology was used? The sentence "Further analysis ... the troposphere" seems redundant.

We have omitted the sentence mentioning "climatological tropopause". We have also omitted the sentence the reviewer found to be redundant.

P5021, section 3: why do the authors spent so much (a complete section) on measurement biases? As pointed out in the introduction, the Odin orbit is suitable for a lightning NO2 detection, which is what the authors state they are after. My feeling is that measurement biases would be important if the authors wanted to create a LNO2 climatology from the OSIRIS observations.

The measurement biases are important to understand why the diurnal variation of LNO2 is difficult to study for example (see above). The spatio-temporal sampling bias described in this section is also important to consider when examining the seasonality of the enhancements versus latitude (Figure 5).

P5022, L11: why is meteo for the year 2000 used? Surely 2003-2005 is available.

We now use meteorological data from the year 2004. See reply to reviewer 1.

P5022, L13: there seems to be a typo (line break) after "includes a

This is now corrected.

P5022, L15-18: please give some more detail on the scaling to reproduce mean flash rates. Is this done regionally? Is the scaling the same for the two years with different lightning?

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We have made the following change:

"The spatial distribution of lightning is scaled to reproduce seasonal mean lightning flash rates from the Lightning Imaging Sensor and Optical Transient Detector satellite instruments to improve the spatial distribution of lightning in the model."

to

"The climatological spatial distribution of lightning is scaled locally following Sauvage et al. [2007] to reproduce the climatological seasonal mean lightning flash rates from the Lightning Imaging Sensor and Optical Transient Detector satellite instruments."

The scaling remains the same for two years with different lightning.

What is meant by 'the timing of emissions of lightning NOx'?

We have changed: "The timing of emissions ..." to "The local temporal variation of lightning NOx emissions ...".

If the timing relates to meteorological variables such as cloud height, how does the scaling affect the fate of the LNO2 (downdrafts, rainout, etc.)?

The mean NO2 mixing ratio with and without rescaling are different by about 0.05 pptv around the globe in the upper troposphere. This corresponds to a relative difference of <1.5%, so the fate of the LNO2 is essentially unchanged by the scaling. We now write in section 4:

"This spatial scaling has a minor effect (<1.5%) on the globally-averaged NO2 VMR in the upper troposphere."

P5023, L10-12: the Choi et al.-reference does not provide an observed range "of tropospheric NO2 enhancements due to lightning", but only a simulation with and without lightning that suggests the <2.5x10^{15} molec/cm^{2} enhancement. This should be removed or rephrased.

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We have rephrased as follows: “Choi et al. [2005] simulated a range of tropospheric NO2 VCD enhancements due to lightning, all <2.5x10^{15} molec/cm^2. Their simulations with lightning were in quantitative agreement with GOME observations and consistent with the upper limit of the magnitude of observed enhancements reported here.”

P5023, L16-17: why does neither problem appear major? Because Figures 4a and 4b look alike?

Yes. To be more explicit, we now write: “However, due to the consistency of the versions as evidenced by the similarity of Figures 4a and b, it appears that neither problem is major.”

P5023, L19-21: Figure 4c represents OSIRIS at 10:30 LT, not GEOS-Chem. Should it be 4d-4e as in line 25?

Yes, thank you. We now write:

“The OSIRIS observations are scaled to 10:30 LT using the McLinden et al. [2000] photochemical box model (see also Brohede et al. [2007]) to match the sampling time of the model simulations. This local time was chosen to clearly exclude from the simulated NO2 field any twilight values (from the winter hemisphere) when NO2 VMRs are much higher and changing rapidly. The OSIRIS observations (Figure 4c) provide a similar spatial distribution of NO2 enhancements to those obtained with the GEOS-Chem 3-dimensional tropospheric chemical transport model (Figures 4d-e).”

P5023, L26-27: can you explain why Figure 4e shows that at 12 km, most of the NO2 is from lightning? Is it because the spatial patterns of GEOS-Chem LNO2 and OSIRIS NO2 are correlated? If so, please provide quantitative information.

The quantitative information is provided by Figures 4d-e. It is true that our statement was misleading. It is not clear that most of the NO2 is from lightning in all tropical regions. So we now write:

“Figure 4d-e show that at 12 km, most of the NO2 is from lightning over Brazil, tropical

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Africa, and northern India, for example.”

P5024, L10-12: if OSIRIS observations are higher than GEOS-Chem at 12 km, this may also imply that the vertical distribution of LNO2 in the model is incorrect. Could the authors comment on this?

This comment is already made later in the paper (P5026, L21-24).

P5026, L5-6: what is meant by “the altitude of 12 km chosen from the model simulations”? This raises the question of how representative the model layer is for the OSIRIS observations shown in Figure 4.

We have reworded:

“...which corresponds very well with the altitude of 12 km for the model simulations.”

What was meant is that all altitudes from the ground to the tropopause are simulated with GEOS-Chem, but the model NO2 field at 12 km was selected. The model layer could not be more representative of the OSIRIS observations shown in Figure 4 since the observations are also at 12 km. We might be failing to understand this comment.

P5026, L8-10: bringing up consistency with Choi et al. for the early spring enhancements isn’t very convincing. Choi et al. found 2-3 days with enhanced simulated LNO2 columns over a small part of the Western Atlantic. Either remove or replace ‘found’ by ‘simulated’.

We have removed the sentence citing Choi et al.

P5026, L25: please define “upper tropospheric column enhancements”. Are these the enhancements in one retrieval layer only? Does Figure 6 consist of both 06:00 and 18:00 observations, or do they represent a 10:30 intermediate?

The “upper tropospheric column enhancements” are not necessarily for one retrieval layer only. We have defined this quantity on P5020, L19-22. Figure 6 consists of both 06:00 and 18:00 observations. We now write in the caption:

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"No effort has been made to scale column enhancements to a single local time."

P5026-5027: the statement that 'a large fraction of lightning NO2 enhancements will be difficult to detect with the current generation of satellite nadir instruments' is too pessimistic and therefore misleading. There is evidence in the peer-reviewed literature that GOME and SCIAMACHY are able to observe LNO2. Especially if used in a statistical sense (GOME, SCIAMACHY and especially OMI and GOME-2) the nadir instruments have a tremendous capacity to overcome single-pixel precision limits mentioned by the authors, and in fact their presenting single-day SCIAMACHY and OMI NO2 data in Figures 7 8 goes against their statement. With respect to the latter, I think earlier work done on lightning NO2 observations from nadir instruments receives too little attention in this paper. Similar numbers as given in Fig. 6 are published in the literature, and the manuscript would benefit if the range presented in Fig. 6 is compared to estimates by Beirle et al. (2004, 2006), Boersma et al. (2005). Furthermore, a comparison to the vertical distribution of LNO2 from the model would be most interesting. There are also aircraft observations of lightning NO2 in the upper troposphere (these are sometimes the same as used in the model). Have the authors looked into these (and whether they extend to above 12 km)?

We agree that there is evidence in the peer-reviewed literature that GOME and SCIAMACHY are able to observe LNO2. In Figure 7 and 8, we present the two highest upper tropospheric NO2 column enhancements (see Table 1) obtained in our two year study, so we are not going against our statement of "a large fraction of lightning NO2 enhancements will be difficult to detect...". However we think the statement is realistic, not pessimistic, and clearly not misleading. We are trying to "lead" the readers to understand that limb scattering has a lower detection limit than nadir because of the long path lengths through the upper troposphere for the former geometry. We agree that earlier work done on lightning NO2 observations from nadir instruments received too little attention. We now write:

"Nevertheless, nadir viewing instruments such as GOME have improved our under-

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standing of the contribution of lightning to the tropospheric NOx budget. Using monthly averaging of GOME NO2 vertical column densities over a 10° (lat) x 20° (lon) region, increases of 2x10^{14} molec/cm^2 (e.g. 5.5x10^{14} versus 3.5x10^{14} molec/cm^2) between winter and summer were linked to lightning [Beirle et al., 2004] in Central Australia where other sources of NOx are minor. Beirle et al. [2006] also found a mean tropospheric NOx VCD over 9 adjacent GOME pixels during an isolated summer storm in the Gulf of Mexico. Using their stated NO2 to NOx ratio, effective for the tropospheric column in the presence of lightning-generated NOx, the mean NOx VCD converts to a NO2 VCD of 2.2x10^{15} molec/cm^2. This amount is larger than the tropospheric column enhancement above 11.0 km observed by OSIRIS (Figure 6), but is comparable to the magnitude of tropospheric NO2 column seen by OMI in Figure 8c. Boersma et al. [2005] observed a strong relationship between cloud top height and NO2 column near and above cloud top in annually-averaged GOME data for clouds with top pressures less than 440 hPa. Over ocean, for example, they were able to clearly observe a NO2 VCD increase from 2.3x10^{14} to 7.5x10^{14} molec/cm^2 as cloud top height increased from 6.5 km to 12 km. This study also showed good correlations between annually-averaged tropospheric NO2 VCDs from GOME and lightning NO2 VCDs from a 3-dimensional chemical transport model (named TM3) in many regions, such as Australia, with GOME observed tropospheric NO2 VCDs increasing from 2x10^{14} to 9x10^{14} molec/cm^2 over the range of the TM3-simulated lightning NO2 VCDs. To summarize these three studies, the detection limit of GOME approaches that of a single limb scattering observation when data are averaged over at least a period of a month or when individual storm studies are limited to cases where lightning enhancements are relatively large."

As indicated, the published analyses of tropospheric NO2 from GOME require one of the following:

1) monthly averaging over a relatively large area, 2) annual averaging (over a small area), or 3) lightning-generated NO2 column enhancements that are near the upper

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limit of enhancements from individual storms.

Furthermore, a comparison to the vertical distribution of LNO2 from the model would be most interesting. There are also aircraft observations of lightning NO2 in the upper troposphere (these are sometimes the same as used in the model). Have the authors looked into these (and whether they extend to above 12 km)?

Yes, we "have looked into these" [Martin et al., 2006] and have compared the model vertical distribution with (in situ) observations from an airborne platform. These observations, more suited for studying profile shape than OSIRIS data in our opinion, suggest a more highly peaked profile in the upper troposphere than is assumed in the model. However, the aircraft data (from ICARTT) rarely extended above 12 km. The highest point during the entire campaign is at 12.6 km. We already reference the Martin et al. publication in this paper with respect to this issue of vertical distribution of NOx. We have begun working on using MAESTRO NO2 profiles to compare with the vertical distribution of LNO2 assumed in the model. The NO2 profiles from this instrument, like the in-situ data, have a higher vertical resolution than profiles from OSIRIS. MAESTRO is able to profile into the stratosphere (i.e. well above 12 km).

P5027, L21-22: what is meant by "This trajectory puts ... closer to the line of sight of OSIRIS"?

We have simplified our statement to:

"This trajectory brings the NO2 plume closer to the line of sight of OSIRIS."

P5027, L7: there seems to be a misplaced ')' after nadir.

Fixed, thanks.

P5027, L10: what is the basis for the factor 4? Has this been simulated with GEOSChem?

This has been simulated with the photochemical box model. We now write: "...but,

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based on photochemical box model calculations [McLinden et al., 2000], a factor of 4 is attributable..."

P5029, 26-27: why aren't the LIS-flashes shown here? I think Figure 9 needs to be cleaned up. The HNO3 profile does not add useful information, since no data below the 13.8 km maximum altitude is available (so how do you know it is maximum?), and I think it should be removed. This would leave just NO and NO2 profiles at sunset, something that should be mentioned in 6.3, not just in the figure caption. The x-axis suggests really small concentration (in 1e-9 pptv).

We will show LIS flashes in what will be named Figure 9b. The HNO3 profile does add some useful information since it shows that HNO3 is increasing below 16 km. The reviewer is correct: perhaps the true local maximum lies below 13.8 km, however given the range of the ACE profile, the HNO3 VMR is a local maximum at 13.8 km. In any case we have removed HNO3 from the figure and from a sentence in section 6.3. The x-axis has been corrected. We now mention in section 6.3 as well that:

"The HALOE NO and OSIRIS NO2 profiles have been converted to local sunset with the photochemical model."

P5031, L2-3: the GEOS-Chem low bias of 6-7 pptv seems to be mentioned here for the first time.

We now mention it in Section 5. Here is the revised sentence from Section 5, which quantifies this difference:

"The OSIRIS observations suggest larger background NO2 concentrations in the upper troposphere (by 6 to 7 pmol/mol), but the difference between the observations and the simulations is within the uncertainty in the lightning source strength derived from other recent global analyses [e.g. Martin et al., in press; Sauvage et al., 2007]."

P5031, L10-19: I did not get this from the text - do the authors refer to a particular figure? If so, 'bands' are hard to distinguish given the scarcity of useful observations.

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We accept that there is a scarcity of enhancements in the tropical and southern Atlantic Ocean, even after analyzing two years of data. We have removed related statements. We have kept the statement about the band in the northern Atlantic Ocean since the enhancements there are not scarce (Figure 3) and the magnitude of the enhancements is significant (Figure 4a-c). This band is discussed in the text in any case (P5026, L2-24).

Even though most lightning occurs over land [Christian et al., 2003], NO2 enhancements frequently occur over the ocean due to transport and the long lifetime of NOx in the upper troposphere. In the northern Atlantic Ocean, a band of enhanced NO2 is observed. It appears to be a result of outflow from the southeast US, starting in spring at approximately 25°N and moving to higher latitudes in boreal summer. In contrast, fewer upper tropospheric NO2 enhancements are found in the Pacific and Indian Oceans and lower baseline concentrations of NO2 are observed there, consistent with the simulations.

P5036, Table 1. How are the first and third entries different?

They are profiles from successive limb scans. The latitude is slightly different for example, as are the NO2 VMRs, naturally.

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