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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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Reply to anonymous referee 1

We thank referee 1 for their effort in reviewing our paper. Many valuable suggestions were given. Below, we have italicized comments and reply immediately below in non-italicized letters. To see the figures, please download the interactive comment from: http://www.cfa.harvard.edu/~csioris/OSIRIS/reply-to-ref1.doc

1. My main concern is the interpretation of the data with limited view to lightning only. I agree that the model supports their view, but the model contains only an approximate representation of surface emissions and convective transport and hence may overestimate the lightning contribution. - I think, the authors should not fully exclude that some *of the NO2 enhancements come for surface sources transported upwards within deep convective events.*

The model may overestimate or underestimate the lightning contribution because of poorly represented surface emissions and convective transport. We have added the disclaimer suggested by the reviewer at the end of section 2:

"We do not fully exclude that some of the upper tropospheric NO2 enhancements come for surface sources transported upwards within deep convective events."

2. For example, the fact, that fewer upper tropospheric NO2 enhancements were found in the Pacific and Indian Oceans indicate that part of the NO2 enhancements are caused by surface emissions. Also, the authors note large enhancements, possibly from lightning, in the outflow into the western north Atlantic in early August (page 5026, L10) which they cannot fully explain. I think, the authors cannot exclude that large parts of these enhancements come for the surface. I ask that this possibility is stated more clearly in the text including the conclusions and the abstract.

The reviewer is concluding that the lack of upper tropospheric NO2 enhancements in the Pacific and Indian Oceans indicates that a large part of the NO2 enhancements are caused by surface emissions. However, there are many other factors that can explain this lack of enhancements. One such factor is that lightning NOx production is greater over the southeastern United States than at similar latitudes in East Asia (China) (e.g. Nesbitt et al. [2000]). Also, the reviewer has not made it clear why, if surface emissions are responsible, the Atlantic would show more enhancements than the Pacific. We note that surface emissions in the 2003-2005 period are comparable between North America and China (e.g. Martin et al. [2006]). We realize that the convective transport could be different between these two regions. Other reasons suggesting that the source of "large parts" of the enhancements is not from the surface are listed here.
1) As found by Rynda Hudman et al. [2007] for the same region during a subset of the time period (ICARTT campaign), the NOx VMR is often higher in the upper troposphere

than at the surface. Thus the isolated NO2 enhancements we are finding are not likely to be due largely to surface sources.

2) The height of the enhancements, especially at mid-latitudes ($|lat|>30^\circ$) is out of the reach of most lightning-free deep convective events. These extend typically to <10 km. Lightning is most intense during the cases of deepest convection, when the cloud tops are reaching the altitude of the observed enhancements ($\sim$ 13 km).

3) Furthermore, the fact that the enhancements are much more frequent at 12-13 km than at 10-11 km severely limits the possibility that "large parts" of the enhancements are coming from below.

4) Finally, Martin et al. [2006], also using ICARTT campaign data, find no obvious bias in upper tropospheric CO in the same region, whereas one would be expected if there was a bias in deep convection. Plate 1 of Levy et al. [1999] shows that, at least for boreal summer, lightning is the largest contributor even at a pressure of 315 mb for latitudes between 45°N and 40°S. Figure 9 of this paper shows that surface sources (including biomass burning and soil biogenic emissions) play a very minor role (<20%) at 190 mb at 30° N in winter and summer. Note that Levy et al. [1999] assumed 4 Tg/year of nitrogen from lightning. Despite our reservations noted above, we have stated the possibility clearly in the text that surface sources are contributing in the upper troposphere as mentioned in our reply to comment1. We have added the following sentence in the concluding section: "Coincident profile measurements of trace gases such as CO through the entire troposphere would be useful to quantify the potential role of surface sources."

3. I expect that the signal is strongly sensitive to the presence of clouds and to the assumed NO2 and O3 profiles. I also wonder how important the diurnal gradients are that exist between the near and far sides of the limb near twilight. In particular, I wonder whether the scaling with the photochemical box model form 6:00 LT to 10:30 LT is really sufficiently insensitive to the cloud albedo.

The retrieval is not "strongly sensitive" to clouds. We have cited Haley et al. [2004] (in

ACPD

7, S1926–S1934, 2007

Interactive Comment

addition to Sioris et al. [2003]), who showed that, for OSIRIS, the error only reaches 18% at a solar zenith angle (SZA) of 60°, assuming the (low) cloud had an albedo of 1 and the retrievals are performed with the albedo in the forward model set to 0.3. Since the average SZA of the retrieved NO2 shown in Figure 3 is ∼83°, this is a minor effect. In spite of the foregoing, we have done a sensitivity test using the retrieval algorithm described in the paper and we have selected a case with a significant NO2 enhancement in the upper troposphere to make the test more relevant to the results reported in this paper. We chose the case with the smallest SZA (=63.8°), since the impact of clouds is greater at small SZA. The "TRUE" profile was actually retrieved from OSIRIS observations near Kirov, Russia (58.6° N 49.6° E) following a summer thunderstorm. We have taken this NO2 vertical profile and forward modeled limb radiances at SZA=63.8° with an albedo of 0.8. Then we retrieve the NO2 profile from these generated radiances with the albedo now set to 0.05 (a clear-sky value over a dark surface). As shown, the retrieval error reaches a maximum of 11% at 15 km.

The retrieval is not sensitive to underlying clouds for two main reasons that are not necessarily intuitive:

1) The mean photon path length below the tangent altitude is small relative to the single scattered path length, mostly because of the long line-of sight path through the tangent point (e.g. at 11 km). 2) High-altitude limb radiance spectra from the same limb scan are used as a reference (i.e. I_0). Thus, the high-altitude reference will also have a very similar path length contribution from the portion of the atmosphere below the tangent height of the interest (11 km). This will essentially cancel out the sensitivity to the mean path from multiple scattering off the underlying cloud. This cancellation of path lengths is not perfect, particularly for a tangent altitude near the cloud top, thus the retrieval bias at the corresponding altitude is not zero. We now write: "Clouds below the field of view are still ignored, leading to <10% underestimates for large solar zenith angles [Sioris et al., 2003] (see also [Haley et al., 2004])."
The importance of the diurnal gradients between the near and far side of the limb has

ACPD
7, S1926–S1934, 2007

already been quantified by McLinden et al. [2006] and is cited in the Method section where details regarding the inversion approach are given.
The contribution of the first guess NO2 was quantified by Sioris et al. [2003] and is very minor.
The sensitivity to the a priori ozone profile had not been evaluated before because O3 absorption is not strong in the NO2 fitting window. A 10% perturbation in ozone leads to <0.1% retrieval error in NO2. We now write:

"Sioris et al. [2003] showed that the sensitivity of the retrieved NO2 to the first guess is <1% below 33 km. The sensitivity to the a priori O3 profile has been quantified for this work and is <0.1% for a uniform bias of +10% in O3 concentration from the surface to the top of the atmosphere."

The reviewer is justified in wondering whether the scaling of profiles from 6 AM to 10:30 AM is sufficiently insensitive to cloud albedo. For the photochemical modeling, we are also using the Koelemeijer et al. [2003] surface albedo database. We have already investigated the sensitivity to cloud albedo during the validation of SCIAMACHY NO2 profiles in tropical regions versus balloons measuring at twilight. Thus, it was easy to calculate the sensitivity for a case with an assumed albedo of 0.1, when the true albedo is 0.8. This results in an overestimate in NO2 of 35-40% (as illustrated) near the equatorial tropopause for January at 10:30 AM. However, if the NO2 comparison with GEOS-Chem at $\sim$6 AM were straightforward, the GEOS-Chem profiles could still be different from the truth due to wrong cloud albedo in those simulations. The NO2 near the tropopause increases by up to 16% at SZA=83°AM as cloud albedo changes from 0.8 to 0.1. So, in reality, the differential impact of scaling from 6:00 AM to 10:30 AM is <22% (=38-16) for this large error in surface albedo. The Koelemeijer et al. [2003] database is biased low in terms of effective albedo because it was compiled using clear-sky observations. Low albedo values tend to make the scaled NO2 concentrations at 10:30 larger than they would be for higher, more realistic albedo values since photolysis of NO2 is reduced for darker scenes, particularly near midday. This

ACPD
7, S1926–S1934, 2007

Interactive Comment

could be partly responsible for the high bias in upper tropospheric NO2 from OSIRIS versus GEOS-Chem. In section 5, we now write: "OSIRIS NO2 shown in Figure 4f may have a slight high bias partly due to the low cloud albedo assumed in the photochemical modeling of the scaling factor between local times."

4. On top of page 5020, the authors discuss the impact of errors in ignoring cloud albedo. For their statements they just cite previous studies. I am not convinced about these statements. It would be better to show explicitly that the results do not depend on cloud albedo. The authors may consider showing a case study with albedo values varying within reasonable limits.

See reply to comment 3.

5. The authors compare observations for the period May 2003-May 2005 with simulations driven with assimilated meteorology for the year 2000. This makes a direct comparison of observed and modelled results less certain. Better would be simulations performed for the time period of the observations. At least this problem should be discussed.

We have redone the simulations for the year 2004 and regenerated Figure 4. The two years are very similar.

6. On page 5020, lines 15, the authors discuss NO2 enhancements found in the Saharan desert region over Libya, Egypt and Chad (Fig. 3). I suggest that the authors use a trajectory analysis to identify from where these enhancements could be from.

We performed 12 back-trajectory analyses, one for each of the enhancements. We find that 7 back trajectories, at some point in the 72 hour period preceding the time of the observed enhancement, lie within 1 hour and $\pm 4^{\circ}$ of latitude and longitude of a lightning flash (observed by LIS) whereas the other 5 enhancements did not. The winds come from the west and/or south in the majority of the 12 back-trajectory cases, with the NOx-generating lightning flashes originating over Algeria in 4 of 7 cases.

ACPD

7, S1926–S1934, 2007

Interactive Comment

“We hypothesize that these enhancements are from advected lightning NOx since 12 of the 14 enhancements observed over Libya, Egypt and Chad are unaccompanied by coincident LIS lightning observations or any meteorological record of a thunderstorm, whereas globally, the majority of the NO2 enhancements lie within $\pm 4^{\circ}$ of latitude (lat) and longitude (lon) of LIS-observed lightning occurring earlier on the same day or the previous day. Meteorological data (http://meteo.infospace.ru/wcarch/html/index.sht) for the given days usually show that cloud fraction is small and the air at the surface is very dry. Back-trajectory analyses were performed to investigate the origin of the enhancements. We find that most of the lightning flashes which coincided with the back-trajectories in space (within $\pm 4^{\circ}$ of lat and lon) and time (within 1 hour) occurred over Algeria in the previous 72 hours or less.”

7. Page 5027, line 13: I suggest inserting “large” before “gaps”.

Done.

8. In discussing Fig. 3, the authors note that “many upper tropospheric (UT) NO2 enhancements lie in tropical Africa”. It would be good to see a plot of occurrence frequency versus latitude to quantify the “many”.

We have included a plot of occurrence frequency versus latitude for the enhancements in Africa (Figure y). There are 55 between the tropics of Cancer and Capricorn. We have quantified “many” in the text.

Figure y. Histogram of OSIRIS UT NO2 enhancements detected over Africa versus latitude over the 2 year period (May 2003-May 2005). The latitude at the midpoint of each bin is labeled on the x-axis.

9. Figure 6: As an interesting check on the model’s capability to simulate lightning induced NOx enhancements, I would be interested to see a similar distribution based on the GEOS-Chem model results? If the model is good in reproducing this distribution it would be a strong support for the model’s accuracy. If not (what I expect), the result

ACPD

7, S1926–S1934, 2007

Interactive Comment

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would show the readers the still existing limitations of such model analysis.

We agree that this would be interesting but this requires major effort and is beyond the scope of this paper. We will try to study this in the near future. Thanks for the idea.

10. Figure 7a. The geographic coordinates are not well readable. I have difficulties to find the coordinates (at 23.936°S, 72.074°E) of the lightning spot mentioned in the text. (By the way, these coordinates are given with too many digits.) At present, the axis notation uses letters which are far too small and difficult to find because of the dark background.

We have rounded the lightning flash coordinates to the nearest hundredth of a degree in the text. We produced a figure showing the location of the flash and the location of the OSIRIS scans, rather than using the product from the LIS website. Our figure, although more legible, did not show the variation in background light levels which is presumably due to clouds, since this data is not available to us (only flash information is available). We have decided to delete the figure since the geography of this case is 'painted' in the text.

11. I would have preferred to have used SI units instead of the non-SI units "pptv" etc. You hardly measure volume ratios (rather molecular density ratios), and the term "billion" has different meanings in the English and American languages. Also the unit "Tg N/year" does not conform with SI standards. This standard recommends to use Tg/a or Tg a-1, and the reference to Nitrogen mass should be part of the text. The term pptv is explained in the text, but ppbv (legend of Fig. 3) is not.

We have changed "pptv" to "pmol/mol" and "ppbv" to "nmol/mol" throughout the paper and in the figure captions. We now use "Tg/year of nitrogen".

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ACPD

7, S1926–S1934, 2007

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