Atmos. Chem. Phys. Discuss., 10, C9592–C9599, 2010
www.atmos-chem-phys-discuss.net/10/C9592/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Direct satellite observation of lightning-produced NO_x" *by* S. Beirle et al.

S. Beirle et al.

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Received and published: 10 November 2010

We thank reviewer 2 for his/her positive feedback and constructive comments. Below we respond to the specific/technical comments point-by-point.

General Comments:

My suspicion is that, despite the radiative transfer modeling that indicates fairly good "visibility" for LNO_x *in the middle portion of a cloud, there is something that we just don't know well enough about radiation behavior in this type of cloud. Therefore, I would recommend that a short sensitivity study be performed to consider the effect of relaxing the 1 hour criterion to perhaps 2 or 3 hours and rerunning the analysis. This amount of time will still be short enough to minimize chemical loss, but will allow at least some of* *the LNO_x to be transported to regions just outside (clear air) or at the edge of the cloud (partly cloudy conditions) where it might be more visible. This would necessitate consideration of flash counts in pixels upwind of the SCIAMACHY observation pixel being considered.*

Reply: According to the reviewer's suggestion, we repeated our analysis for WWLLN flashes within the penultimate hour, i.e. 60-120 minutes before the SCIAMACHY overpass. For these "old flash events", the overall mean CF decreased slightly (from 0.97 to 0.94). But the derived TSCDs and PEs are not fundamentally different, in particular not higher than those for the "fresh" events.

The quantitative relation of NO_2 TSCD to WWLLN flashes *within the SCIAMACHY pixel* is of course more uncertain, and less justified, for the "older" flashes. But counting the *upwind* flashes instead, as suggested by the reviewer, would require cloud resolving model simulations, involving accurate knowledge of a) the vertical placement of the LNO_x and b) the horizontal wind fields for the respective height levels. This information is not available on the required spatial and temporal resolution and accuracy.
Nevertheless, we agree that an additional study, focussing on aged LNO_x under cloud free conditions, would be a valuable complement to our current study. However, the strongest drawback, in our point of view, would be the need to determine the number of flashes which actually contributed to the LNO_x within each satellite ground pixel, which again requires accurate knowledge of the LNO_x altitude and the relevant wind fields. Furthermore, the interference from anthropogenic NO_x, and uncertainties of the stratospheric estimation are probably more critical, since the aged LNO_x is diluted and larger areas have to be averaged.

Presumably a comparison of the FRESCO (derived from O2 A-Band observations by SCIAMACHY) cloud top heights with IR cloud cloud tops might also yield some information concerning why the visibility of LNO_x appears to be poor. If the FRESCO cloud top is not much below the IR top, then the volume of cloud being seen by the instrument will be small, and the resulting NO_2 columns will be small. I recommend doing such a comparison and including it in the paper.

Reply: We agree that additional information of cloud top temperatures (CTTs) from IR measurements could carry complementary information on the cloud structure. We thus tried to investigate CTTs from AATSR on ENVISAT from the GRAPE project (http://badc.nerc.ac.uk/data/grape/) for the sample events in table 2. However, there are two difficulties:

- the swath width of AATSR is smaller ($\approx$ half) than that of SCIAMACHY. Thus, for events at the SCIAMACHY swath edges (like #115 or #225), no matching AATSR data is available.
- the selected events seem to be too extreme to fit in the GRAPE retrieval; over the considered events, Grape output data are flagged as missing (Retrieval quality flag is set to 0, meaning the fit “failed to converge”), and direct AATSR brightness temperatures are unphysical (down to 0 K for 11μm).

Thus, though we can not make a direct quantitative comparison of vis/IR cloud heights, the AATSR data indicates exceptionally high clouds in the UTLS which let the GRAPE retrieval fail. The closest available (valid) brightness temperatures are below 200 K for events #191, #208, #261, and #266. Consequently, in the vis range, a large volume of the cloud has actually been “seen”.

Specific Comments:

p. 18257, line 18: please replace “came up” with “became possible”

Reply: Changed to “have become available” according to reviewer 1.

p. 18263, line 6: please replace “several” with “$\sim$20-30”

Reply: Done.

p. 18263, lines 20-21: should note that the WWLLN data prior to 2007 have now been reprocessed with

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the new algorithm and the DE should now be greater that what is computed here.

Reply: We added this information in the revised manuscript.

p. 18264, Section 2.3: What is not considered here is variability of the WWLLN DE with time of day. I thought that propagation of wavelengths detected by WWLLN was better at night than during the daytime. If so, the DE values used here should be smaller than the diurnally averaged values obtained by comparing WWLLN flash counts with the OTD/LIS climatology. See additional comments regarding the results of the instantaneous DE obtained in Section 4.3.

Reply: We agree that the diurnal cycle of WWLLN DE is potentially important and should also be investigated quantitatively. We therefore compared WWLLN flash counts, grouped in 1-h bins according to local time, to the climatological LIS/OTD diurnal cycle. As statistics of WWLLN are rather poor, particularly over oceans, we do not derive spatially resolved DE maps, but instead divide the total number of WWLLN flashes 2006-2008 for a given time of the day by the respective number of LIS flashes. This was done for oceans and continents separately.

We found a mean DE of 8.2% over oceans and 1.7% over continents. Interestingly, we do not find a clear day/night pattern, but instead only moderate fluctuations (of the order of $\pm$ 10% relative change over oceans).

The finding of generally higher instantaneous DE can thus not be explained by the diurnal cycle of WWLLN DE as derived from comparison to LIS/OTD. Rather, it is probably related to the selection of events with high FRD in our study.

This investigation on the diurnal cycle of the WWLLN DE has been added to Appendix A.

p. 18265, Section 2.4: I think at least some error in the analysis arises from assuming that all of the LNO_x produced during the hour prior to SCIAMACHY overpass remains in the pixel being analyzed. A 30 m/s wind in the upper troposphere yields a transport of 108 km, which is larger than the 30 x 60 km

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pixel. Use of single pixels likely results in missing some portion of the LNO_x that is produced. An analysis was conducted using 10 x 10 pixels, but it was for the event #191 east of Florida which appeared to be contaminated by pollution outflow. I would recommend some more sensitivity analyses of using more than one pixel (maybe much less than 10 x 10), but for pixels not affected by pollution.

Reply:

The neglection of outflow of LNO_x out of the SCIAMACHY pixel indeed introduces some uncertainty in our study (in Sect. 4.5, we consider a possbile underestimation of T up to 50%, which would correspond to an underestimation of P by 100%). We recognize the necessity of a more quantitative discussion of this aspect.

We therefore revised our reasoning concisely:

1. Our determination of PE is based on the assumption that the SCIAMACHY observation can be related to the LNO_x produced over the last 60 minutes within the SCIAMACHY pixel. We justify this approach by claiming that the dimensions of a SCIAMACHY ground pixel (60 km * 30 km) correspond to distances reached within 1 hour for upper tropospheric winds. From the figures 3-8 (3-5 in the revised manuscript), mean wind speeds of the convective systems can be roughly estimated by the distance between the blue (60 minutes back) and red (0 minutes back) flash dots, which actually results in about 30-60 km/h. The 30 m/s (108 km/h) quoted by the reviewer is a rather high value (see, e.g. Huntrieser et al., 2008, ACP 8-921, table 4c). Please note also that the considered flashes, *on average*, happened 30 minutes ago, not 60 minutes.

2. We are aware that some fraction of the LNO_x is "lost" by outflow from the SCIAMACHY ground pixel. However, this is at least partly compensated by some inflow of LNO_x from neighbouring pixels (which is neglected as well). In addition, we generally underestimate the FRD by our restriction to 1 hour.

3. The event maps generally do not indicate a considerable enhancement of the neighbouring TSCDs in wind direction (as indicated by the movement of WWLLN flashes).

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Thus, we see no evidence that outflow to neighbouring pixels would let us seriously overlook the produced LNO_x by focussing on the single event groundpixel.

4. Considering a larger area generally would indeed partly solve the issue of possible NO_x outflow; however, it would also introduce new uncertainties: The larger the considered area, the longer time periods have to be considered for the flashes, and the quantification of the number of flashes that actually contributed to the area under consideration does not become easier.

5. Our focus on a single SCIAMACHY ground pixel was motivated by the fact that, by our selection of high FRD, the resulting TSCDs (for literature values of PE) would be high above background levels. For larger areas, FRD – and thus the expected TSCD enhancement due to lightning – generally becomes lower. Therefore, the impact of a background in NO_2 TSCDs gets stronger, and the potential interference from NO_x sources other than lightning gets worse.

To quantify this reasoning, we also estimated PE for the events involving the 8 direct neighbours, i.e. an area which is 9 times larger. Since the spatial dimensions are increased by a factor of 3, we also count flashes back to 3 hours. The resulting PEs are, on average, higher by a factor of 2. But this is almost solely a consequence of the decreasing denominator (flash densities for 3x3 pixels, i.e. mean FRDs integrated over 3 hours, are, on average, reduced to 47%), while the numerator (TSCD) remains more or less constant (84%). In other words: we again have to conclude that for most events, we do not "see" any LNO_x, and the observed NO_2 TSCDs have another origin! For the category B events, however, PE is higher by a factor of 1.8 for event #261, but even reduced by a factor of 0.7 for event #225. Mean TSCDs are about half for the 9-pixel approach for both events.

Though the virtually higher PE for the 3x3 pixel study is mainly reflecting the impact of background (i.e. non-lightning) NO_x, it can still serve as an upper bound of the uncertainty of our PE estimate due to the neglection of outflow, which is thus less than a

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factor of 2. Sect. 2.5 and our discussion in Sect. 4.5 has been revised accordingly.

p. 18271, line 2: include Ott et al. (2010) P $\sim$ 20 - 42 x 10^{25} molec/flash

Reply: We added Ott et al. (2010) P $\sim$ 22 - 42 x 10^{25} molec/flash (derived from the 360-700 mol/flash given in their conclusions).

p. 18275, line 15: The result of a much larger instantaneous DE at 10 AM is surprising. I would have anticipated a value smaller than the climatological DE due to poorer propagation of the VLF signals in the daytime. But maybe the fact that you are considering only relatively large flash rates (and perhaps relatively large peak currents) in the analysis more than compensates for this. Perhaps add some comments on this subject to this section.

Reply: We revised the respective paragraph in accordance to our analysis of the diurnal cycle of the climatological DE of WWLLN. We suspect that the selection of events with high flash rates results in the high instantaneous DE, and we are particularly sensitive for flashes with high peak currents.

p. 18280, line 22: I'm having trouble figuring out what is being said here. Please clarify.

Reply: We modified the respective paragraph to

"PE estimates in literature might be generally biased high ("publication bias", Scargle et al., 2000): Observations of high PE ("positive" results) have likely been published, while observations of low PE ("negative" results) might have been discarded as non-significant or non-conclusive. But for the estimation of a sound, unbiased mean, information on both tails of the statistical distribution of the NO_x production per flash is needed. We encourage feedback to this hypothesis from scientists having performed in-situ measurements."

p. 18282, Appendix A1: You could compare your DE in 2008 for the Costa Rica region (mostly ocean) with *that (∼22%) of Bucsela et al. (2010) for July/Aug 2007 (after algorithm upgrade). I think the comparison looks pretty good.*

Reply: We decided to omit a comparison to Bucsela et al. (2010), since
(a) a direct quantitative comparison is difficult due to the strong gradient of our DE for this region (about 10% at the coast up to 30% at 700 km afar, and undefined values beyond due to the low climatological LIS/OTD FRD), and
(b) the flash scaling factors derived by Bucsela et al. (2010) are also linked to LIS (via Eqs. 6&7 therein), so it is not surprising to find similar values.

Instead, we added a further comparison to the recent study on WWLLN DE by Abarca et al. (2010), who compared WWLLN to the U.S. NLDN.

p. 18283, lines 15-16: I don't think you know this absolutely for certain. Could this result also suggest that the OTD/LIS climatological flash rates are biased low? There is a tremendous amount of processing that must go into creating these climatologies from very undersampled data. The NASA processing must include consideration of the DE of the OTD and LIS instruments and extrapolate from a very small actual view time. Maybe some comments on the uncertainty of the OTD/LIS climatology might be order somewhere in the paper.

Reply: We agree that it is more adequate to choose a less definitive formulation here. We also added a short discussion of the LIS/OTD uncertainy in Appendix A; in particular, over oceans, the number of actual LIS/OTD counts is rather small. However, while the low number of flash observations over some oceanic regions (by both LIS/OTD and WWLLN) causes some scatter of the derived values for D_{clim}, it can not explain a *systematic* effect of the order of a factor of 3.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 18255, 2010.

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