**Atmospheric
Chemistry
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Discussions**

Interactive comment on "Direct satellite observation of lightning-produced NO$_x$" *by* S. Beirle et al.

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We thank reviewer 1 for his/her positive feedback and constructive comments. Below we respond to the general/specific/technical comments point-by-point.

General Comments:

The events investigated are limited regionally by 1) the detection efficiency of the WWLLN, which seems to be low over oceans, and 2) the presence of air pollution which could contaminate the NO$_2$ signal from lightning. Though the authors have used all available data, because of these limitations it is not truly a global study and I think that could be stated more clearly in the introduction and conclusion.

Reply: We agree that the limitations due to detection efficiency of WWLLN and po-

tential interference from anthropogenic pollution relativize the term "global", and we revised introduction and conclusions accordingly.

It seems to me that a major issue is the use of climatological detection efficiencies applied to specific storms. As the authors note, lightning is a highly variable process and it is possible that DE varies strongly between different events in the same locations. The authors investigate this using LIS overpasses and find large differences, but this is difficult because LIS overpasses are so short in duration and lightning activity can vary strongly over the lifetime of a storm. I wonder if it would be possible to compare with ground-based networks which are available in the US and Europe. I understand that for most of these locations, the levels of pollution are too high to be considered for the PE analysis, but it may give some information into how reliable the climatological DE estimates are.

Reply: The estimation of WWLLN DE is indeed one of the most challenging tasks of this study. We make use of the established LIS/OTD climatology to estimate the spatial dependency of WWLLN DE; to account for the change of WWLLN performance with time, we estimate annual mean DE maps.

In Appendix A1, we compared our estimated DE to literature values: Jacobson et al. (2005) investigated WWLLN DE over Florida by comparisons to a ground based detection system, and found a DE of about 1%, while we estimate a value of 0.89%.

An additional study on WWLLN DE has been published recently by Abarca et al. (2010), using the NLDN as ground truth. In table 2 therein, they number the CG+IC DE over the US for 2007-2008 as 2.9%, matching our value for this period and region (1.8% in 2007 and 3.9% in 2008). This comparison was added in the revised paper.

For the instantaneous DE, our comparison with individual LIS counts reveals high fluctuations (which is partly due to the short time intervals of coincident measurements) and indicates systematically higher instantaneous DE for the detected events, which can not be explained by the diurnal cycle of WWLLN DE. Instead, we suspect this being a consequence of our selection of events with high flash rate densities.

We agree that it would indeed be helpful to repeat these evaluations of instantaneous DE with ground-based networks; but unfortunately, they are a) to our knowledge, not

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freely available, and b) mostly limited to continents, i.e. they could generally not be used to evaluate our DE_{inst}, mostly derived over ocean. A systematic analysis of individual flashes determined by WWLLN and, e.g., NLDN, is beyond the scope of this study.

Specific comments:

p. 18259, L13 – Add a sentence defining what an air mass factor is. I think this would be unclear to readers without a remote sensing background.

Reply: We added a short definition of the AMF.

p. 18261, L10-11 – I think more detail on the determination of the stratospheric fraction is necessary. TSCDs are likely to be strongly dependent on the estimate of the stratospheric component and small errors could have a considerable impact on the calculations. What type of uncertainty does this introduce into TSCDs?

Reply: In the revised manuscript, we added the uncertainty of TSCD due to the stratospheric estimation, which is of the order of typically 0.5 to 1×10^{15} molec/cm^2 for low latitudes (see Fig. 6c in Beirle et al., 2010). Thus, for low tropospheric pollution levels (about 1×10^{15} molec/cm^2), relative errors from the stratospheric estimation might indeed be considerable (up to 100%), and a possible bias would be particularly crucial if low NO$_2$ column densities are integrated over large areas.
However, in our study, we focus on localized events (single SCIAMACHY ground pixels) with high FRD, which should, according to literature values of PE, result in significantly enhanced TSCDs. Thus, for our conclusions, the uncertainty due to the stratosphere is not critical. In particular, it can not explain the observation of events with high FRD, but virtually no NO$_2$. We clarified this in the revised manuscript.

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p. 18261, L24-26 – Values for the sensitivity, E, are calculated based on high resolution model simulations of events during the TOGA COARE/CEPEX period. The authors note that E is insensitive to the cloud optical thickness. Could E vary in different regions or different meteorological conditions? If not, why not?

Reply: Our determination of the sensitivity E is based on the model study presented in Beirle et al., 2009. Sensitivities were modelled for one particular thunderstorm episode, which nevertheless covers all the different stages of cb evolution.

We found E being quite insensitive to COT. The reason for this is that the NO_2/NO_x ratio decreases with altitude towards the cloud top, while the box-AMF increases towards the cloud top, making the "NO_x box-AMFs" (see Beirle et al., 2009) quite independent on altitude. We consider this being a fundamental, characteristic pattern of radiative transfer inside (cb) clouds. Of course, the actual numbers for E might be different for other regions/meteorological conditions (and probably also for the same region, but a different model), but we would not expect changes of orders of magnitude, which would be necessary to explain the missing response of LNO_x in observed NO_2 TSCDs. These aspects are discussed in Section 4.2.

p.18264, L13-15 – I think it would be useful to move some of the material, including plots, from Appendix A to this section to help the continuity and to show the regions where the analysis is possible.

Reply: We pondered about this suggestion and understand the reviewer's concern. However, it is not appropriate to illustrate the WWLLN DE by a single figure, as it changes from year to year. Thus, we still would like to keep the current structure, i.e. having a short summary of our DE definition in Sect. 2.3, and keep all details and figures coherently together in Appendix A (which has now been extended by a discussion of the diurnal cycle of the WWLLN DE).

p. 18265, Line 6 – How is the pollution mask defined? There are already a large number of figures so I hesitate to encourage adding another, but I think it would be helpful to show the mask, possibly overlaid on a global plot of NO_2 column densities, since it ultimately limits what cases can be included in the analysis.

Reply: The effect of the pollution mask should indeed be made more transparent for the reader. Thus, we clarified the definition of the pollution mask in Section 2.4 (i.e., regions where the annual mean SCIAMACHY TSCD is above 3×10^{15} molec/cm^2, plus a band of 500 km around, which basically masks polluted regions in continental and coastal US, Europe, and China). Instead of showing the pollution mask itself, however (to avoid having an additional figure), we decided to include the lightning events over "polluted" regions in figures 1 and 2 (309 cases), but marked with different symbols so that they can still be discriminated. In doing so, the reader can comprehend which events are discarded by the pollution mask.

These "polluted" events generally do not show a different behaviour in PE. In particular, there are also several events with no visible NO$_2$ as well. But for some events (over Southern US, Eastern China, and the Pearl River Delta) extremely high TSCDs (up to 20×10^{15} molec/cm^2 for the latter) are observed that are clearly related to convection of polluted boundary layer air masses.

p.18271, Lines 17-22 – Why is this? Is it because there is more potential for pollution or aged LNO$_x$ contamination when larger regions are considered? Since the authors note that the size of the area considered is important in estimating PE, I think some more explanation could be helpful. What does PE look like if intermediate sized areas of 5x5 SCIAMACHY pixels are considered?

Reply: We recognized that our quantitative estimation of a PE of 300×10^{25} molec/flash for the 10x10 SCIAMACHY pixels in the discussion of category A events was not fully appropriate, as it ignores the record of flashes for this event: Our approach of relating NO$_2$ TSCDs to flashes over the last 1 hour for individual pixels is meaningful, because 1 hour approx. corresponds to the dimensions of the SCIAMACHY ground pixel for upper tropospheric wind speeds. But if a larger area is considered for the estimation of LNO$_x$, also older flashes have to be taken into account. If we consider flashes 10 hours back in time (since the spatial scale has been extended by a factor of 10 as well), we find instead a PE of 62×10^{25} molec/flash, which is still a rather high number.

Thus, the basic warning of the respective paragraph remains still the same:
If a larger area with high NO_2 TSCDs (numerator), but low FRD (demoninator) is considered for the estimation of PE, the latter may be largely overestimated. For event #191, FRD is almost zero in north-east direction, where TSCDs are rather high over hundreds of km. It is thus essential to identify the enhanced NO_2 TSCDs as LNO_x first by demanding matching spatial patterns of lightning. We modified the respective paragraph accordingly.

We also considered an intermediate sized area (of 3x3 pixels, and counting flashes over 3 hours) for determining PE for all events. On average, this results in virtually higher PE (by a factor of about 2), but again for the wrong reason: 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! The consideration of larger areas might spuriously hide this outcome.

p.18273, L15 – The authors mention that NO_2 profiles modified by convection are used. Where do these profiles come from and how strongly do they affect the calculated TSCDs? How variable might these profiles be between storms or at different points during the lifetime of a single storm? I think it would be helpful to add some of these details to Section 2.1.

Reply: In our study, we analyze TSCDs (i.e. total SCDs which are corrected for the stratosphere); the *retrieval* of this quantity does not *involve* profile information. (For the determination of E, LNO_x profiles from the cloud resolving model are involved; but E has been found to be rather insensitive to changes in the profile, see Beirle et al., 2009). But, of course, the *measured* TSCD *depends* on the actual profile; generally, the higher the NO_2 (in terms of altitude), the larger the AMFs, but the lower the NO_2/NO_x ratio. For a quantitative correction of these effects, the modification of background

profiles of NO_2 profiles due to convection would be required. But since this information is generally not available with the required accuracy, and the net effect is small since two different effects partly cancel each other out, we neglect it here. We clarified the respective paragraph.

p.18275, Paragraph 3 – This description is very vague. These differences in DE are quite large and while they may be offset by increases in production per flash, it would be good to try to estimate the potential magnitudes of these effects. Would it be possible to include a table similar to Table 2 for a few cases where some detailed estimates of the changes in PE and FRD are given?

Reply: We revised and specified the respective paragraph. We compared the climatological and instantaneous DE in Appendix A2, Fig. A6c. D_{inst} is, on average, higher than D_{clim} by a factor of 3.4. We now also calculated modified PE and FRD for the events with available LIS information, and find similar effects: PE_{inst} is, on average, higher than PE_{clim} by a factor of 2.8. Flash rate densities using the instantaneous DE are infinite for two events, where WWLLN flash counts within the LIS overpass are zero. If those two are excluded, the remaining FRDs F_{inst} are lower than F_{clim} by a factor of 2.1.

Since the higher instantaneous DE can not be explained by the diurnal cycle of WWLLN DE, we extended the discussion of this finding being probably related to our selection of events with high FRD in the revised manuscript. In addition, we also discuss the effects of a prossible selection of high-current flashes on both WWLLN DE and LNO_x PE.

p. 18280, L28 – Isn't the PE around the US likely influenced by pollution outflow or aged LNO_x? If so, I'm not sure that a difference between subtropical and tropical lightning has been demonstrated.

Reply: We mention both aspects (subtropical versus tropical lightning and antropogenic interference) as possible reasons for the relatively high PE over the U.S. in p. 18281, L1-2.

Technical comments:

p. 18256, L3 – Change 'high' to 'large' or 'strong' to distinguish magnitude from altitude.

Reply: Done.

p. 18257, L18 – Change 'came up' to 'have become available'

Reply: Done.

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