Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-556-RC2, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nitrogen oxides in the global upper troposphere: interpreting cloud-sliced NO_2 observations from the OMI satellite instrument" *by* Eloise A. Marais et al.

Anonymous Referee #2

Received and published: 15 August 2018

In this manuscript, upper tropospheric NO_2 columns from two different cloud slicing approaches applied to one year of OMI data are used to study the impact of lightning on upper tropospheric NO_2. First, data from the two retrievals is compared with each other and with aircraft observations taken over North America in the period March – August of the same year. Data from the NASA algorithm are then compared to GEOS-chem model data for two seasons. Scatter plots of NO_2 columns from both model and satellite retrievals against lighting flashes from the LIS/OTD climatology are compared and the conclusion is drawn, that UT NO_2 from OMI is largely dominated by lightning NOx. Finally, spatially and seasonally resolved maps of NOx production per flash are computed from the ratio of retrieved to modelled UT NO_2, and the dependency of these

production rates on LIS lightning properties is evaluated.

Measurements of NO_2 in the UT are sparse and the use of satellite data for validation of model results in this important atmospheric region is of high scientific interest. The approach taken by the authors is interesting and the manuscript overall well written, although I would have hoped to get more details on what exactly was done in many places.

Nevertheless, the paper leaves me a bit helpless as my impression is, that combining the uncertainties of the individual steps taken in this analysis will make the results basically worthless. More specifically,

- the two retrievals which are based on the same data and on quite similar assumptions lead to very different results on UT NO_2,
- the comparison with airborne measurements shows only broad agreement, and that only if data are averaged over large areas,
- the conclusion that the main driver for the observed UT NO_2 variability is lightning Is probably correct in general but clearly not for individual points in Fig. 6,
- computing NOx emission rates per flash by taking ratios between model and measurement in the scattered distributions shown in Fig. 6 seems really optimistic to me.

I'm also surprised by the briefness of the discussion of the log-linear relationship found between lightning frequency and NO_2. Is this a known fact, and is there an explanation for it? The fact that this relationship is not so clear in GEOS-chem data would not lead me to the conclusion that NOx lifetime is lower at high lightning frequencies (how would that follow from the lightning parametrisation used? Are non-linear effects really expected at the relatively coarse resolution of the model?) and that NO_2 observations are uncertain at low concentrations (there are no observations in the model figure). I would rather suspect that other factors such as transport, vertical mixing, and chemistry are also important drivers of upper tropospheric NO_2 in addition to lightning, which would explain that very large changes in lightning frequency are needed to see moderate changes in UT NO_2.

The variations in NOx production per flash shown in Fig. 7 are large in many places, and would be important input for global modelling studies. However, an error bar is needed for these numbers before they can be used, and maybe this is the reason why the authors don't mention them in abstract and conclusions.

In summary, I cannot recommend this paper for publication in the current form. Before it can be published, the authors need to add more detail on the individual steps of the analysis and the data used, they need to provide uncertainty estimates and explain how they were derived, and also should add more discussion on how the fact that lightning is not the only factor affecting UT NO_2 impacts on their results and conclusions.

Minor comments

- Introduction / beginning of section 2: There is a lot of repetition here, please read again and shorten where possible.
- page 3, line 97: Is aerosol really accounted for in the NO_2 air mass factors, and if so, how?
- page 3, line109: What are near-Lambertian clouds?
- page 4, line 124: Why is the slant column offset affecting the UT NO_2 data – I thought this is cancelled by the stratospheric correction?
- page 4, line 133: How do the authors know which signatures in the figures are real, and which linked to misrepresentation of lower tropospheric signals?

- page 7, line 199: I understand that aircraft measurements are screened for stratospheric air masses in tropospheric applications. However, here data are compared to satellite retrievals, and these will - as far as I understand – include such stratospheric air masses if they are in the right pressure range above a cloud. I therefore wonder if this screening really makes sense here.
- page 7, Fig. 4. It is unfortunate, that here another time period is shown than in Fig. 1. As airborne data is collected over a period of 6 months, seasonal variability in UT NO_2 could play a role in the comparison to satellite retrievals. I'd therefore suggest to show all 4 seasons in Fig. 1 or at least to add this figure to the Appendix / Supplement.
- page 8, Fig. 5. Again, I would suggest to add the other seasons as well.

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