Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-556-RC1, 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 #1

Received and published: 27 July 2018

The manuscript by Marais et al. presents an application of satellite-derived NO2 data for the upper troposphere to diagnose NOx sources with the aid of the GEOS-Chem model. Two such products from the OMI instrument on the Aura satellite are evaluated in relation to aircraft data. The NASA product compared slightly better with the aircraft data than did the KNMI product. Therefore, the NASA product is used in comparison with GEOS-Chem output. This comparison suggests that the lightning NOx production in the model is too large, and the authors have scaled it down to better match the OMI-based data. The authors determined that there was no evidence to suggest larger NOx production efficiency per flash in the mid-latitudes than in the tropics. I have some concern about the level of detail of the lightning NOx emissions that are derived as I

describe below. I consider this a major revision. Otherwise, my comments are minor.

Neither of the OMI-derived UT NO2 products compared well with aircraft data. The better correlation (0.64) was with the NASA 450 - 280 hPa product at very coarse (20 x 32 degree resolution). However, this means R^2 = 0.4 and that the satellite-based data only capture 40% of the variance seen in the aircraft data averaged to this resolution. Is this really good enough to constrain a global chemistry model? If one assumes there is sufficient meaning in these data, the comparison with GEOS-Chem suggests that the lightning NOx emission per flash in the mid-latitudes should be reduced from 500 to 260 moles/flash, leading to an overall lightning source strength reduction from 6.5 to 5.5 TgN/year. However, the authors go on to scale the lightning production per flash upward or downward for each 20 x 30 degree grid cell. Any discrepancy between the OMI UT data and GEOS-Chem is being attributed to differences in NOx production efficiency per flash. Given the relatively poor comparison between OMI and the aircraft data and uncertain model UT NOy chemistry, I think this is taking the analysis too far. It is a real stretch to quantitatively believe the values given in Figure 7 and in lines 259 - 269. I would suggest eliminating Figure 7 and perhaps just comparing the derived NOx production per flash values for mid-latitudes as a whole and tropics as a whole. Figure 8 could stay, as although it contains individual grid cell value of NOx production efficiency, it does not contain specific regional values that someone might quote.

Minor Comments:

Introduction section: The authors need to add some more background material on previous uses of OMI (and earlier satellites) data for diagnosing lightning NOx production. The prior literature is in 2 categories: satellite data and model analyses (Boersma et al., 2005; Martin et al., 2007) and satellite-alone analysis (Beirle et al., 2010; Bucsela et al., 2010; Pickering et al., 2016)

line 48: Add Allen et al., 2010 after Tost et al.

lines 52-53: The 100-500 mol N/flash and 3-7 TgN/yr do not match. If one assumes

the OTD/LIS climatological 46 flashes per second, 100 mol N/flash is about 2 TgN/yr and 500 mol N/flash is about 10 TgN/yr.

line 80: OMI was launched in July 2004.

line 90: For what pressure range was the Choi et al. (2014) product? Are there any other differences between that product and the NASA product used here?

lines 94-95: How well does TM4 do at these estimates?

line 101: I don't understand how this difference yields a column for 330-450 hPa.

line 158: ....lightning NOx emissions and convective transport of boundary layer pollution....

line 212: What percentage is this?

lines 216 - 222: Are these comparisons for the model with 6.5 TgN?

line 221: domain average UT NO2 is 19% lower than aircraft data. The opposite bias is present in comparing the model with OMI. Which should you believe?

line 242: Why would this be the case?

line 259: OMI-derived and GEOS-Chem lightning NOx production

line 289: 5.6 TgN/yr doesn't match the 5.5 TgN/year mentioned in line 214

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