Comment on amt-2021-286
Anonymous Referee #1

Referee comment on "Quantification of lightning-produced NOx over the Pyrenees and the Ebro Valley by using different TROPOMI-NO2 and cloud research products" by Francisco Javier Pérez-Invernón et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-286-RC1, 2022

Review of ‘Quantification of lightning-produced NOx over the Pyrenees and the Ebro Valley by using different TROPOMI-NO2 and cloud research products’ by Perez-Invernion et al.

This paper attempts to estimate the lightning production efficiency of NOx over the Pyrenees and northeastern Spain in Spring 2018 from 3 sources of information: lightning flash counts, TROPOMI satellite above-cloud NO2 column observations, and modelled NOx:NO2 ratios in the upper troposphere. Each of these have considerable uncertainties associated with them. The strength of the work is that the authors address these uncertainties carefully in their approach. Strong about this paper is the use of two different lightning flash networks, two standalone TROPOMI NO2 products, and different approaches to correct for the tropospheric background NO2 not caused by lightning. This is interesting in its own right, as there are considerable uncertainties associated with counting lightning flashes, with the satellite retrieval process, and with our knowledge about tropospheric background NO2. It is also interesting because applying this mini-ensemble provides a robustness check on the quantitative LNOx production, which is at the lower end of previously reported estimates.

The paper has some serious shortcomings which need to be addressed before publication in AMT can be considered in my opinion:

- A weak point of the paper is the reliance on just one CARIBIC flight (22 June 2005) over the Pyrenees-Ebro area. I understand that there may not have been many flights available, but the representativeness of this alternative method to estimate the NO2 background is debatable. A model (e.g. EMAC) analysis of NOx and NOy in the upper troposphere 22 June 2005 compared to April-May 2018 would be helpful to assess this concern.
Another major concern is the usage of the concept of the “LNOx air mass factor”. In the DOAS-community the AMF is strictly defined as the ratio between the slant and vertical column, and since TROPOMI detects NO2, the use of a lightning NO2 AMF, followed by a model-driven NOx:NO2 correction factor, would be the appropriate way to present this aspect of the approach. It is thus misleading here to use a LNOx AMF since TROPOMI measures NO2, and not NOx. The authors should present the AMF aspect of their approach more clearly, specifically in Figure 5 – the simulations are now input to the center-stage box ‘LNOx PE’, but in fact the simulations both influence the NO2 AMF (the upper box) and the subsequent NOx-to-NO2 conversion. Also in Eq. (3) the application of an ‘AMF_LNOx’ is misleading and should be replaced by division by an AMF_LNO2, followed by a correction factor that accounts for the NOx-to-NO2 ratio. To clearly present how the model is required for their ultimate quantification is important given (a) future reproducibility of their results, and (b) the need to prevent leading readers into believing that NOx could somehow be detected from TROPOMI.

Figures 6 and 7 actually give little evidence that “areas of high lightning activity coincide with areas with high SCD-NO2”. This undermines an important claim of the paper, i.e. that enhanced TROPOMI NO2 can be traced back to previous lightning flashes. The authors should provide more evidence that there is a relationship between flashes and enhanced NO2, e.g. via scatter plots suggesting a spatial correlation for lightning circumstances, and the absence of enhancements on cloudy days without recent lightning activity. The same unfortunately holds for Figures 8 and 9, while the relationship between lightning and enhanced NO2 is more evident of the low-wind day shown in Fig. 10 and 11.

Specific comments

Figure 1 is not particularly helpful. One way to provide more useful context is to overplot the mean NO2 columns on the map. That way the reader can appreciate the difficulty of distinguishing the lightning NO2 signal from the nearby anthropogenic hotspots such as Toulouse, Bordeaux, and Barcelona.

L34: ‘estima’ --> estimate

L84: Williams et al. (2017) describes the TM5-MP version which is actually used in the NO2 retrieval. I believe this is a more appropriate reference than the Myriokefalitakis-reference.

Williams, J. E., Boersma, K. F., Le Sager, P., and Verstraeten, W. W.: The high-resolution version of TM5-MP for optimized satellite retrievals: description and validation, Geosci. Model Dev., 10, 721-750, doi:10.5194/gmd-10-721-2017, 2017.

L85: where can ‘version 2.1_test’ be found? Please provide a reference. Are v2.1_test data also available for April and May 2018?
L108-109: it is unclear here if the authors have imposed temporal coincidence of lightning flashes with the observation of TROPOMI cloud fractions > 0.95 and OCP < threshold. Or has a time window been taken, such as lightning flashes within a few hours of TROPOMI overpass and TROPOMI fulfilling the above cloud criteria? Please clarify.

P6, Figure 3: I think the authors should show here the detection efficiency for April-May 2018 rather than March 2018 – December 2018. After all, the paper is about the lightning NO2 production in April-May 2018.

L177: it is unclear how the authors have formulated the SCD here. Which slant column do they mean? The total slant column, which also contains contributions from the stratosphere, or the tropospheric slant column?

L180: what is meant with the “absorption of the atmosphere”? I guess this is about the ratio of the slant to vertical (NO2) optical thickness, but it should be clarified.

P9, Figure 4: what was the pressure of the OCP on this day? Please indicate this in the caption. Also indicate the corresponding AMF LNO2 values.

P10, Eq. (1): I suggest to include the TROPOMI measurand, i.e. the NO2 column, here. This is now missing from the equation, which may give the impression that TROPOMI somehow provides a tropospheric NOx column, which is not the case.

L274: the authors state that the free tropospheric NO2 “may be overestimated” in TM5-MP, but I see no supporting evidence for this. Do the authors have any reason to suspect this, or could the TM5-MP NO2 background also be underestimated?

L323 and Tables 1 and 2: the “lower values” of $V_{\text{tropNOx}}$ for the DLR vs. KNMI product are unclear to me. Has the $V_{\text{tropbck}}$ been subtracted to arrive at $V_{\text{tropNOx}}$?

L359: “background NOx ...activity” is printed in italics. Not clear why. Same on line 371 and 393-394.

P18, Table 3: perhaps useful to also include the overall uncertainty estimate in the table.
