

## Reviewer 1

This paper attempts to estimate the lightning production efficiency of NO<sub>x</sub> over the Pyrenees and northeastern Spain in Spring 2018 from 3 sources of information: lightning flash counts, TROPOMI satellite above-cloud NO<sub>2</sub> column observations, and modelled NO<sub>x</sub>:NO<sub>2</sub> 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 NO<sub>2</sub> products, and different approaches to correct for the tropospheric background NO<sub>2</sub> 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 NO<sub>2</sub>. It is also interesting because applying this mini-ensemble provides a robustness check on the quantitative LNO<sub>x</sub> 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:

We thank the reviewer for these encouraging comments and for the time spent for the revision.

- 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 NO<sub>2</sub> background is debatable. A model (e.g. EMAC) analysis of NO<sub>x</sub> and NO<sub>y</sub> in the upper troposphere 22 June 2005 compared to April-May 2018 would be helpful to assess this concern.  
*As the reviewer points out, we have used only one CARIBIC flight because there are not many flight available. In particular, commercial flights usually avoid areas with high convection. Thus, we have only found one flight over North of Spain with significant convection (22 June 2005). Despite using only one case, we have obtained a good agreement with previous airborne NO measurements over convective systems without lightning in Europe during the EULINOX campaign [Huntrieser et al. (2002)]. We think this comparison with other measurements in Europe is sufficient to rely on the NO mixing ratio provided by CARIBIC.*
- Another major concern is the usage of the concept of the “LNO<sub>x</sub> 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 NO<sub>2</sub>, the use of a lightning NO<sub>2</sub> AMF, followed by a model-driven NO<sub>x</sub>:NO<sub>2</sub> correction factor, would be the appropriate way to present this aspect of the approach. It is thus misleading here to use a LNO<sub>x</sub> AMF since TROPOMI measures NO<sub>2</sub>, and not NO<sub>x</sub>. 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 ‘LNO<sub>x</sub> PE’, but in fact the simulations both influence the NO<sub>2</sub> AMF (the upper box) and the subsequent NO<sub>x</sub>-to-NO<sub>2</sub> conversion. Also in Eq. (3) the application of an ‘AMF\_LNO<sub>x</sub>’ is misleading and should be replaced by division by an AMF\_LNO<sub>2</sub>, followed by a correction factor that accounts for the NO<sub>x</sub>-to-NO<sub>2</sub> 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 NO<sub>x</sub> could somehow be detected from TROPOMI.

As the reviewer points out, we have used the AMF  $\text{NO}_x$  to convert the measured SCD  $\text{NO}_2$  to VCD  $\text{NO}_x$ . However, using an AMF  $\text{NO}_2$  instead of an AMF  $\text{NO}_x$  is not appropriate in this case. The convenience of using AMF  $\text{NO}_x$  instead of AMF  $\text{NO}_2$  was explained by Beirle et al. (2009):

“Please note that a two-step conversion (first from  $\text{NO}_2$  SCDs into  $\text{NO}_2$  VCDs using an overall AMF, and then from  $\text{NO}_2$  VCDs into  $\text{NO}_x$  VCDs using a mean  $\text{NO}_2/\text{NO}_x$  ratio) is not appropriate, since both the box-AMFs and the  $\text{NO}_x$  partitioning are height dependent, and they do not vary independently because both are particularly influenced by clouds.”

We refer to Beirle et al. (2009) for more details. We have added this explanation to the manuscript for the sake of clarity.

The use of AMF  $\text{NO}_x$  instead of AMF  $\text{NO}_2$  to calculate the VCD  $\text{NO}_x$  is common in  $\text{LNO}_x$  PE estimates [e. g.: Bucselá et al. (2013), Pcikering et al. (2016), Bucselá et al. (2019), Allen et al. (2019 & 2021)].

- Figures 6 and 7 actually give little evidence that “areas of high lightning activity coincide with areas with high SCD- $\text{NO}_2$ ”. This undermines an important claim of the paper, i.e. that enhanced TROPOMI  $\text{NO}_2$  can be traced back to previous lightning flashes. The authors should provide more evidence that there is a relationship between flashes and enhanced  $\text{NO}_2$ , 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  $\text{NO}_2$  is more evident of the low-wind day shown in Fig. 10 and 11.

We have calculated the Pearson correlation coefficient ( $r$ ) between the SCD of  $\text{NO}_2$  in convective cells with flashes and the total number of flashes reported by ENGLN in each cell averaged over all the studied cases. We have obtained  $r = 0.18$  for TROP-DLR and  $r = 0.11$  for TROP-KNMI. These values indicate a positive correlation between the SCD of  $\text{NO}_2$  and flashes that is larger for the case of TROP-DLR than for TROP-KNMI. This correlation is larger when we use the tropospheric winds to identify the cells that have been influenced by  $\text{LNO}_x$ . We have copied each flash to the cells that are influenced by the  $\text{LNO}_x$  produced by the flash with the purpose of calculating the upwind correlation coefficient by taking into account the transport of  $\text{LNO}_x$ . With that we obtain  $r = 0.20$  for TROP-DLR and  $r = 0.15$  for TROP-KNMI. The received larger correlation coefficients indicate that accounting for the transport of  $\text{LNO}_x$  can improve the estimation of  $\text{LNO}_x$  PE.

This analysis has been added to the manuscript (Section 3.1).

### **Specific comments**

Figure 1 is not particularly helpful. One way to provide more useful context is to overplot the mean  $\text{NO}_2$  columns on the map. That way the reader can appreciate the difficulty of distinguishing the lightning  $\text{NO}_2$  signal from the nearby anthropogenic hotspots such as Toulouse, Bordeaux, and Barcelona.

We have removed Fig. 1. Figures 5-10 already show the difficulty of distinguishing the LNO<sub>2</sub> signal from the nearby anthropogenic hotspots.

L34: 'estima' --> estimate

Done.

L84: Williams et al. (2017) describes the TM5-MP version which is actually used in the NO<sub>2</sub> 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.

We have added this reference.

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?

The v2.1 research product is not automatically produced for all the TROPOMI orbits. We produced it on a case-based demand to analyze particular thunderstorms. We have added this clarification to the manuscript.

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.

We have used a 5 hours time window. We have included this clarification in the manuscript.

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 NO<sub>2</sub> production in April-May 2018.

We have calculated the DE using ISS-LIS lightning data. ISS-LIS is orbiting the Earth in a low Earth orbit. Thus, there are not many coincidences of thunderstorm occurrence and ISS-LIS passage over the particular region. In addition, each point is observed during only 90 seconds during each passage. Therefore, we have included more than one year of data instead of only two months to estimate the DE of ENGLN. In particular, we have found 30 thunderstorms simultaneously detected by ENGLN and ISS-LIS over the area during March 2017 and December 2018. We have added this to the manuscript.

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?

Here we mean the SCD NO<sub>2</sub> measured by TROPOMI (the total slant column). We have added this to the manuscript.

L180: what is meant with the "absorption of the atmosphere"? I guess this is about the ratio of the slant to vertical (NO<sub>2</sub>) optical thickness, but it should be clarified.

This is clarified in lines 203-206:

“We use the LNO<sub>2</sub> and LNO<sub>x</sub> vertical profiles from the simulations to calculate the AMFLNO<sub>x</sub> following Bucsele et al. (2013). We use the TOMRAD forward vector radiative transfer model (Dave, 1965) to calculate the scattering weights for each of the 8 studied cases using the viewing geometry and the cloud properties for each pixel. We obtained AMFLNO<sub>x</sub> values ranging between 0.28 and 0.71.”

We have added a mention of the scattering in these lines.

P9, Figure 4: what was the pressure of the OCP on this day? Please indicate this in the caption. Also indicate the corresponding AMF LNO<sub>2</sub> values.

Figure 4 corresponds to a simulation on 13 May 2018. This was the day in May 2018 with the highest LNO<sub>x</sub> column density in the simulation. Therefore, we use this case to extract the the mean simulated LNO<sub>2</sub> and LNO<sub>x</sub> profiles. However, it is important to note that the model is global (2.8 x 2.8 degrees horizontal resolution) and that lightning is parameterized using the updraught as a proxy. Thus, there can be a disagreement between the observed and the simulated total number of flashes in specific thunderstorms.

The total number of lightning flashes detected by EUCLID during this day was not particularly high (165 total flashes). In particular, the total number of flashes 5 hours before TROPOMI overpass was only 29. Therefore, we have not included the analysis of TROPOMI data for 13 May 2018. As a consequence, we cannot provide the value of the OCP nor the AMF for this case. In fact, this would not be useful, as the measurements of TROPOMI on 13 May 2018 are not influenced by fresh LNO<sub>x</sub>.

P10, Eq. (1): I suggest to include the TROPOMI measurand, i.e. the NO<sub>2</sub> column, here. This is now missing from the equation, which may give the impression that TROPOMI somehow provides a tropospheric NO<sub>x</sub> column, which is not the case.

We have now mentioned that the  $V_{\text{tropLNO}_x}$  is calculated from TROP-NO<sub>2</sub> product. The method to calculate it is provided in the following paragraphs.

L274: the authors state that the free tropospheric NO<sub>2</sub> “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 NO<sub>2</sub> background also be underestimated?

We have rephrased. Evaluating the TM5-MP NO<sub>2</sub> is out of the scope of this work. We have restricted the discussion to the comparison between the free tropospheric NO<sub>2</sub> into the stratosphere provided by STREAM and by TM5-MP models that can influence the estimation of LNO<sub>x</sub>.

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

No, the  $V_{\text{tropbck}}$  is subtracted from the  $V_{\text{tropNO}_x}$  to yield the  $V_{\text{tropLNO}_x}$ . The  $V_{\text{tropNO}_x}$  is calculated with eq. (3) using the TROPOMI product and based on the AMF LNO<sub>x</sub>.

L359: “background NO<sub>x</sub> ...activity” is printed in italics. Not clear why. Same on line 371 and 393-394.

Changed.

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

Done.