Reviewer 2

The authors have analysed several convective systems around the Pyrenees in order to estimate the production of lightning NOx based on TROPOMI NO2 observations.

The Pyrenees are chosen as they are particularly suited for this kind of analysis. While I disagree here (see below), the paper contributes to the scientific question how much NOx is produced by lightning, and it matches the scope of AMT.

The study is generally well written, but there are some inconsistencies and probable bugs in the presented data.

The authors thus have to cautiously check the presented data, correct the existing bugs, and update the LNOx estimates (and, if needed, discussion and conclusion) accordingly.

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

In addition, they should consider the additional comments listed below.

General comments

1. Study region

As indicated in the title and at several places in the manuscript (e.g. line 70), the Pyrenees are meant to be the focus of this study. In the manuscript, the line of argument is that over the Pyrenees (a) lightning frequency is high, and (b) NOx background is low, and therefore the Pyrenees are an ideal region for this kind of study.

In the presented figures, however, most flashes are observed South or North of the Pyrenees, where also significant boundary layer pollution is present, in particular over large cities like Barcelona, Zaragoza, or Toulouse. This weekens the argumentation considerably, and I am not convinced that the study region is a good choice for studying LNOx from satellite, as the uncertainty of background NOx and the possible uplift of boundary layer pollution severely affect the overall uncertainties.

We agree with the reviewer that there is a significant number of flashes outside the Pyrenees regions. Most of the analyzed thunderstorms extended beyond the Pyrenees, including some areas of the Ebro Valley and Southern France. However, all the studied thunderstorms included a considerable total number of flashes over the Pyrenees. The background- NO_x is a combination of pixels inside and outside the Pyrenees. Including pixels over the Pyrenees contributes to a lower background- NO_x and, as a consequence, to a lower uncertainty in the estimations with respect to studies over polluted areas.

We have clearly shown in the figures and in some points of the manuscript that the studied area is not limited to the Pyrenees. However, we acknowledge that in some lines we suggest that the study is focused on the Pyrenees (as in line 70). This could lead to misunderstandings. We have carefully checked the manuscript and emphasized that adjacent regions are also included in the analysis.

2. Background

As most events are not observed over the Pyrenees, the NOx background might be considerably larger in general. The potential uplift of boundary pollution might bias the observed NO2 over the lightnin pixels.

In addition, also the non-flashing pixels over deep convection might be affected by the advection of LNOx.

As we mentioned above, we agree that the studied cases include a significant number of TROPOMI pixels outside the Pyrenees region. We have now emphasized this in the manuscript.

The inclusion of two different NO2 products is quite illustrative. For instance, the differences in the estimated stratospheric column between the products are far larger than the given uncertainty of the tropospheric background. Please comment on this.

The differences in the estimated stratospheric column between TROP-DLR and TROP-KNMI products and their influence on the LNO_x PE estimates are commented in the manuscript. We have now extended the discussion about these differences.

3. "Lost" NOx

As far as I understood, the study only focusses on cloudy pixels. So in case of LNOx produced within a convective system that disappeared by the time of the TROPOMI overpass, the LNOx would be overlooked in the described procedure, while the respective flashes are included. Consequently, PE is biased low. This seems to actually happen on 29 April. I propose to skip 29 April, as there are only few TROPOMI observations available, and the extension of the study area to large parts of France is also confusing.

The reviewer is right. As we mention in the manuscript and as other authors indicate in previous studies, the employed method provides an estimation on the fresh-produced LNO_x. This is the reason why we only include flashes 5 hours prior to TROPOMI overpass. The averaged age of individual flashes ranges between 0.9 hours for 7 May case and 2.3 hours for 26 May case, while it is 1.9 hours for the 29 April case. We have added this to the mansucript.

We have decided to maintain the 29 April case because the averaged age of individual flashes and the total number of flashes are in the same range as for the other cases. However, we have now commented its particular characteristics in the manuscript.

In addition, this effect has to be included in the discussion and the overall uncertainty estimate.

Effects 2 and 3 go in different directions, are hard to quantify, and will increase the overall PE uncertainty.

We have added to the discussion the possible bias to low LNO_x when using only cloudy pixels. Although indirectly, the effect of the possible overlook of LNO_x is included in the estimation of the uncertainty when using different time windows to count the flashes.

Inconsistencies and possible bugs

- Please be consistent with respect to units. Column densities are given in petamolec per cm2 in the text, but in molec per m2 in the figures which makes direct comparisons difficult.

Done.

- Please check tables 1 and 2:

Done, as explained in the following points.

- Mean Vstrat is quite large (> 8e15 molec/cm2 for DLR), while the maps in Fig. 6ff shows values < 4e15 molec/cm2.

We thank the reviewer for pointing this out. There was an error in the headings of Tables 1 and 2. We do not show the mean VstratNO2 in the tables, but the mean product VstratNO2 x AMFstrat employed in eq. (3) and subtracted to the SCD NO₂. We have corrected this error.

- The 30th percentile of Vtropbck is listed as 9.5e15 molec/cm2 for DLR. This would be a quite considerable tropospheric pollution. Please check.

There is a typo. The Vtropbck is 9.5e14 molec/cm2 instead of 9.5e15 molec/m2.

- Please check Figs. 10 and 11. The number of flashes displayed in the figures is considerably different, while the numbers listed in Tables 1 and 2 are almost the same for 28 May. Please also check for the other days.

We have checked that the numbers are correct. As the maps show small points, the visualization of the total number of flashes is not evident.

Minor comments

Line 8: Over the Pyrenees, the NO2 background is low indeed. But for the investigated events over the Ebro valley, this is not the case, see e.g. <u>http://www.tropomi.eu/data-products/nitrogen-dioxide</u>

We have now commented in the manuscript that pixels over polluted areas can influence the background-NO_x in our study.

Lines 31-34: "Nadir-viewing satellite instruments ... estimate the column densities of NO2 over thunderstorms" - I think this is too fuzzy. The satellite instruments measure spectra. This allows quite accurate quantification of NO2 slant column densities. The conversion into vertical column densities is the main challenge, involving further input data (like cloud fraction and surface albedo, so this might be denoted as an "estimate". But usually, the retrieval focusses on cloud free conditions. Focussing on thunderstorm clouds instead is a quite different and challenging setup which should be pointed out here.

We have rephrased.

Lines 45-46: I propose to flip the order: "The horizontal resolution at nadir is 3.6 km \times 7.2 km before 6 August 2019, while it is 3.6 km \times 5.6 km thereafter."

Done.

Figure 1: I propose to show a map of tropospheric NO2 instead in order to assess the "clean background" issue.

We have removed this figure. The rest of the figures already show the map of tropospheric NO₂.

Line 70: "8 deep convective systems in the Pyrenees": The number of actual flashes over the Pyrenees is quite low. The presented events reveal highest lightning activity South and North of the Pyrenees, with several anthropogenic sources present (e.g. Barcelona, Toulouse).

We have now commented that other regions are also included.

Section 2.3: I think it is quite bold to estimate "the" tropospheric background NOx column (!) from one day of aircraft measurements (in-situ measurements at 12 km!). The authors should discuss the limitations and uncertainties of this approach in more detail and compare with model results.

We have extensively commented on this issue in the answer to the reviewer 1 and introduced some changes in the manuscript. As the discussion is public, we refer to the answers to reviewer 1.

Eq. 1: Please specify which quantities are dependent on space (TROPOMI pixel) and which quantities are just scalar (means, percentiles). For this you might use Copernicus style where vectors and matrices are shown in bold type. Please also explain how the total PE is calculated (spatial mean or summation over which area/pixels?).

We have now used the Copernicus style for the vectors and matrices. The reader can now follow the reasoning and the following equations to better understand how the PE is calculated. The PE is influenced by scalars, spatial mean and spatial medians, as can be seen in eq. (1-3).

Eq. 3: Stratospheric VCD and AMF are provided for each TROPOMI pixel. So the stratospheric correction can be performed for each pixel as well. I see no reason to calculate the average here.

We are following the same approach as Allen et al. (2021), who used the average instead of individual pixels. The reason is explained by Allen et al. (2021) when referring to their Eq. (5) including the average:

"...TROPOMI NO₂ product were often missing values for VtropNO2 and sometimes missing values for VstratNO2 and AMFstrat in regions affected by deep convection and lightning. The use of Equation 5 increased the number of pixels available to estimate VtropLNOx and allowed for more robust estimates of PE".

As one of the goals of the present study was to establish a comparison with previous estimates of LNOx over the US by Allen et al. (2021), we have applied the same method.

We have now included this explanation in the manuscript.

Lines 227-231: Several events are outside the Pyrenees, with considerably higher background NOx. In addition, in case of deep convection, tropospheric pollution might be uplifted and transported over considerable distances. So the local tropospheric background estimate over the clean Pyrenees can at best be considered as a lower limit.

We have added parts of this comment to the manuscript. The background estimate over the clean Pyrenees is calculated for days with convection.

Line 251: Please explain why the DLR product is missing.

Files in the database are missing, but the reason is not known.

Line 509: Please list the existing and the new methods here.

We have rephrased.

- Figs. 6ff: Maps of stratospheric AMF and VCD are not that informative. Listing these values in tables 1&2 would be sufficient, but the numbers listed there need to be checked. Instead, I would like to see maps of AMF_LNOx here.

We have now removed the map of stratospheric AMF and introduced the map for AMF_LNOx. We have maintained the map of stratospheric VCD because the size of the figure does not change when removing only one panel.

- Fig. 12 needs some further explanations:
 - how was the median calculated?

The average values shown for each point are calculated over the values provided by ERA5 on the pressure levels 200 hPa, 250 hPa, 300 hPa, 350 hPa, 400 hPa, 450 hPa and 500 hPa. The median values shown in the title of the figure are the spatial median over the average in each pixel. We have introduced this in the manuscript.

- what is the meaning of color?

The color is related with the wind velocity. We have added a colorbar and changed the color to make it more visual.