

Rebuttal to Reviewer 2

We thank the reviewer for the time spent in reviewing this article and for the provided comments. In this document, we respond to all the points in blue text.

Using a global chemistry-climate model, this paper investigates how various formulations of lightning-generated oxides of nitrogen (LNO_x) influences the chemical composition of the atmosphere. Of particular interest is the formulation in which the production of LNO_x per lightning flash decreases with lightning flash frequency, in contrast to the commonly used assumption that the amount of LNO_x produced per flash is constant. The authors find that this formulation leads to larger NO_x mixing ratios in the lower and middle troposphere and lower NO_x mixing ratios in the upper troposphere, with consequences on atmospheric composition also reported.

Uncertainty in the quantification of LNO_x and its atmospheric and climate ramifications remains rather large, and the present paper is an interesting contribution towards assessing that uncertainty through examining the chemistry-climate model sensitivity to LNO_x. I favour publication of the paper, but it requires a major revision, considering the following points.

We thank the reviewer for these encouraging comments.

1. As a starting point of the study, one would want to know how the flash frequencies predicted by the Price and Rind (1992), Grewe et al. (2001) and Luhar et al. (2021) schemes compare with observations. The authors merely state (lines 65-66) that ‘... we use scaling factors that ensure a global lightning occurrence rate of ~45 flashes per second (Christian et al., 2003; Cecil et al., 2014).’ I would like the authors to compare the global distributions of the predicted flash frequencies with observations such as those from Cecil et al. (2014). Once there is a confidence in the prediction ability of these schemes, one can then move on to LNO_x calculations and impacts on the chemical composition of the atmosphere.

Also, please give what the scaling factor values were for the three schemes, and the predicted and observed global mean values of flash frequency for the ocean and land.

We have added the input variables in Table 1 and the simulated annual flash density in Figure S1. The parameterizations were described, implemented into EMAC and compared with OTD/LIS observations by Pérez-Invernón et al. (2022). For more details, we refer to this publication in order to not lengthen this manuscript.

2. The work presented is built around the relationship between the lightning flash frequency and the LNO_x Production Efficiency (PE) per flash shown in Fig. 11(c) of Bucsela et al. (2019), which shows an almost exponentially decreasing relationship. While such a strong relationship is surprising, it is mainly based on observations from three continental regions in northern midlatitudes. Thus, the validity of this relationship for the ocean is untested. Generally, the flash frequency over the ocean is much less than that over the land, and thus this relationship would predict much larger values of the LNO_x production efficiency per flash over the ocean. Whether that is the case, we do not know as the relationship is based on data for land. The authors need to discuss and clarify this point.

Allen et al. (2019) also reported “evidence for a decrease in PE with increasing flash rate on a regional basis” within the tropics, including the oceans. We have added this to the introduction and discussion.

3. Some more details of the derivation of the relationship shown in Fig. 11(c) of Bucsele et al. (2019) should be presented. How does this relationship depend on the grid resolution? Also, it will be useful to provide the functional form of this relationship that the authors have used (or was it some form of interpolation?).

We have included in the revised manuscript how we derive this relationship.

4. Line 35 and throughout: ‘...lightning as a total number of NO_x molecules per flash...’ To remove any ambiguity, best to say if it is NO or NO₂ molecules per flash (I think it’s the former). Similarly, is it moles per flash of NO or NO₂?

Although the emission of lightning into the model are defined as NO emissions, part of the emitted NO is quickly converted into NO₂. Therefore, lightning emissions are traditionally referred as LNO_x.

5. Line 72: ‘We check that the percentage of boxes that contain a flash frequency lower than a specified value...’ Is this to account for the change from the 1° × 1° data analysis grid to the model 2.8° × 2.8° grid?

Yes, it is. We have included in the revised manuscript how we use the relationship derived for 1x1 degree resolution into our model.

6. Line 80: ‘...to derive the forcings for the subsequent simulations.’ This is not clear to me. What type of forcings? Why are they needed? Later, Line 88 says ‘...but using the radiative forcing fields from the BASE simulations’ What exactly are these radiative forcing fields?

A paragraph before, we explain: “We conduct the simulations using the Quasi Chemistry-Transport Model (QCTM) approach (Deckert et al., 2011). The QCTM mode allows for the separation of dynamics and chemistry in order to operate the model as a chemistry-transport model. This means that minor chemical changes do not introduce noise by affecting the simulated meteorology.”

For the QCTM mode, we take the input for radiative forcing (e.g., monthly averages of greenhouse gases CO₂, N₂O, CH₄, F11 and F12) from a previously performed free-running simulation (BASE). Furthermore, we prescribe the methane oxidation in the stratosphere also from the BASE simulation. By following this approach, we ensure that the dynamics of the atmosphere is binary identical in the CTR and the LNO_x simulations. Therefore, the obtained differences between CTR and LNO_x simulations are solely due to different chemistry (different LNO_x production in the case of this study). We refer to Deckert et al., (2011) for more details.

7. Line 81: ‘In these simulations, we impose a production of 1,112 mol per CG flash and 111.2 mol per IC flash...’ Obviously, this is a critical assumption (i.e. the LNO_x ratio IC/CG = 0.1) following Price et al. (1997), and is by no means a certain one. The authors should give some discussion on the implications of the variability of this ratio for their simulations.

The reviewer correctly highlights the significance of the IC/CG ratio and the assumption of 1,112 mol per CG flash and 111.2 mol per IC flash in the CTR simulations. While alternative approaches exist, this scheme is the most commonly employed in climate-chemistry model simulations. The primary objective of this study is to evaluate the impact of the new parameterization for LNO_x production on atmospheric chemistry, relative to the widely used scheme. In the revised manuscript (Section 2.2), we have added a discussion about the uncertainty related to the similar or different production of LNO_x by CG and IC.

8. Table 1: The “LNOfs” simulations use the same moles NO produced per flash irrespective of CG or IC flash, unlike the other simulations. Is this because the CG-IC distinction is implicitly included in the relationship in Fig. 11(c) of Bucsela et al. (2019) used in the present LNOfs simulations?

Yes, it is. Bucsela et al. (2019) derived the used relationship without distinguishing between CG and IC lightning.

9. Line 100: Why only year 2000? Weren’t the simulations done for 8 years?

We believe the reviewer refers to Figure 1 and/or Figure 9. In these figures, we plot the results based on hourly modeled data. Due to computational limitations, we extracted hourly data only during the first year of the simulations. However, we consider that 1 year is enough to show the obtained LNO_x per flash (Figure 1) and the interactions between NO_x and other chemical species on the hourly scale (Figure 9). The rest of the figures showing the influence of the new parameterization of LNO_x PE are based on monthly averaged modeled data over 8 years, as explained in the manuscript.

10. Section 3.1: Table 2 data should be presented in graphical form for consistency with Fig. 11 of Bucsela et al. (2019).

Done. See new Figure 1.

11. Fig. 1: The colour scales are different in each panel which makes it difficult to make a meaningful visual intercomparison (the same issue with some of the other subsequent plots). I would like to see the same scale in the top two row plots and the same in the bottom row plots. Also, I find it uncomfortable to view the top two rows of plots. Can a better colour scheme be used?

We have intentionally used different scales in Figure 2 (Figure 1 in the previous version of the manuscript) because the peak values between the P-L and G simulations vary significantly. We previously considered using a logarithmic scale, but this made comparison with Bucsela et al., 2019, Fig. 3(c) more difficult. The consensus solution was to display the differences between the CTR and LNO_f simulations in row 3. In addition, we have changed the colorbar, so that it is now similar to the colorbar employed by Bucsela et al., 2019, Fig. 3(c).

12. Line 131: ‘...LNOfs simulations produces a spatial distribution of LNO_x that aligns with space-based measurements more accurately (Bucsela et al., 2019, Fig. 3(c)) than...’ This is not convincing as there is no way of telling that, given the different colour schemes and scales used in the two studies.

As mentioned above, we have changed the colorbar, so that it is now similar to the colorbar employed by Bucsela et al., 2019, Fig. 3(c). In addition, we have extended the discussion on the comparison between simulations and observations. In particular, we have highlighted that the agreement is better over ocean.

13. Section 3.3: Not sure why the LNOfs_L and CTR_L simulations are not discussed here.

We considered that including zonal and seasonal analysis of the differences between the simulations CTR_L and LNOfs_L would significantly lengthen the manuscript without providing extra information. The reason is that the differences are qualitatively similar to the differences between the LNOfs_P and CTR_P simulations, but smaller in absolute numbers. We have explained this in the revised version of the manuscript.

14. Line 206: ‘...where negative values represent a reduced LNO_x injection in the LNOfsL simulation’ Check.

Here we show the difference in the LNO_x injection between the LNO_{fL} and the CTR_L simulations. We have added “...compared to the CTR_L simulation” for clarity purposes.

15. Page 8: Figures 5–8 are discussed before Figure 4?

We do not think so. New figures 4 and 5 (3 and 4 in the previous version of the manuscript) is discussed for the first time at the beginning of Section 3.2. However, it is revisited after discussing the global maps of Figures 6-9.

16. Line 249: ‘During all the seasons, the LNOfs simulations produce more tropospheric ozone than the corresponding CTR simulations in the tropics, causing more disagreement with measurements...’ I am unable to see this in the difference plots, exacerbated by the fact that the scale is different in the plots.

This can be seen in Figures 11-14. The first two columns indicate that, within the tropics, the mixing ratio of ozone is higher in the simulations than in the observations (predominantly red colors). The third column shows that the new scheme results in an even higher mixing ratio of ozone within the tropics.

17. Line 286: ‘Therefore, the results obtained in this study should be regarded as the upper limit...’ Please say this in the abstract too.

Done.

18. Line 267: ‘...resulting in a better agreement with measurements (Jockel et al., 2016, Fig. 29).’ I suggest the authors reproduce Fig. 29 of Jockel et al. to make comparison easier.

We have included this figure in the supplement. We consider this manuscript is already too long and we prefer not adding more figures to the main text if not necessary.

19. The reference Bucsela et al. (2021) is only an AGU conference abstract. I question the usefulness of it.

The reviewer is right, but we consider this reference useful for the discussion of the limitations and uncertainties of our work. It provides new estimations of LNO_x PE that could influence the relationship previously reported by Bucsela et al. (2019), which is a key element of this study.

20. Both the terms ‘climate-chemistry model’ (e.g. in the title) and ‘chemistry-climate model’ (e.g. in the abstract) have been used. Please keep consistency (I think most researchers use the latter).

Done.

21. Lines 354 and 363: The https addresses of these two references seem to have been swapped.

Corrected.