## Reply by the authors to Referee #2's comments on

"Assessing and improving cloud-height based parameterisations of global lightning flash rate, and their impact on lightning-produced NO<sub>x</sub> and tropospheric composition" (#acp-2020-885)

## Anonymous Referee #2 (RC4)

We are grateful to the Referee for taking the time to read our manuscript and making a number of valuable comments. In the following, we provide our responses to these comments (the Referee's comments are shown in blue). The locations of the changes made refer to those in the non-tracked version of the revised manuscript.

Luhar et al. 2020 implement an alternative lightning flash rate parameterization following Boccippio 2002 in the ACCESS-UKCA global chemistry climate model. The new parameterization is evaluated by comparison to satellite observations of lightning and showed that it yields a better agreement than the default lightning parameterization (PR92). This study then assesses the impact of the new parameterization on the model simulation of NOx, O3, OH and CO. Not surprisingly, the results demonstrate that the relatively small amount of NOx emitted by lightning, leads to a disproportionately large impact on middle-to-upper tropospheric chemistry. Accurate representation of lightning and lighting NOx thus is essential to accurate chemistry and climate models.

I have two major comments. First, a model run utilizing the parameterization following Boccippio 2002 (referred BO02 hereafter) is missing. Table 1 lists out the four parameterizations discussed in this study. The new alternative parameterization proposed in this study is quite similar to BO02. As I read it, the only significant difference is changing the linear coefficient by a factor of 2 for lightning parameterization over ocean (Fo). Sec 3.6 shows that switching from PR 92 to this new parameterization improves model's performance on producing lightning flash rates. However, it's unclear that whether this improvement can be achieved by just switching to BO02 or not. This addition of a model run using BO02 is needed to demonstrate that the modification on BO02 suggested in this study is essential.

**Response:** We thank the Referee and agree with the suggestion made. We have now included results from an additional simulation (Run 5) corresponding to Boccippio's (Bo02) flash-rate parameterisations (Eqs. (9) and (10) in our paper). As hinted by the Referee, the Bo02 parameterisations and the new/alternate parameterisations (Eqs. (18) and (20)) used in Run 2 (TS1), which are based on the Bo02 approach, only differ in the values of their linear coefficients.

It is clear from Table 1 that Run 5 (Bo02) leads to lightning flash frequencies over the ocean that are twice as large as the observations.

**Changes in manuscript:** As mentioned above, Run 5 (Bo02) has been added in Section 3.5 and the results discussed in the paper.

Second, in Sec 4.2 and 4.4, the authors incorporated ground-base in-situ observations of O3 and CO and compared the model's simulations against the observations. They concluded that the model using the new parameterization outperforms the one using PR92 and yields better agreements of O3 and CO with in-situ observations. However, the results shown from Figure 12 and Figure 18 are not convincing enough to support the conclusions. The lightning parameterizations only lead to

marginal changes in monthly averaged O3 and CO, and these effects are not obviously responsible for reconciling the difference between model and observation.

**Response:** We agree with the Reviewer. The focus here needs to be more on how the flash-rate modification impacts ground-level  $O_3$  and CO, and less on the model-data comparison. We recognise that lightning  $NO_x$  alone cannot explain the model-data differences and that there would be other factors at play that are responsible for the large differences between the modelled values and observations at some of the sites. The relevant text in the paper has been modified accordingly.

**Changes in manuscript:** In Section 4.2 (P33L13–23), we add "Mauna Loa is located at an elevation of 3397 m on an island which is smaller in size than the grid resolution of the model and therefore it is difficult to correspond the sampling height to a particular vertical model level. We used the modelled concentrations from the bottom model level for all sites. The two model simulations describe the observed monthly variations reasonably well, except at Mauna Loa and Mace Head (the relatively large disagreement at Mauna Loa is likely due to the model resolution issues). There are small, but noticeable, differences in the modelled ozone from the two simulations. The relative change in the modelled yearly averaged O<sub>3</sub> at these ground-based sites with the use of the new lightning parameterisation is small, at 5.9%, 1.3%, -1.9%, 5.9% and 0%, respectively. There is some improvement in the modelled seasonal variation at Ushuaia, Cape Grim and Minamitorishima with the new LNO<sub>x</sub> scheme, but for the other two sites the model-data differences are much larger than those due to the LNO<sub>x</sub> changes. Generally, factors such as model's transport and chemical mechanisms, and input precursor emissions and their distributions are probably more influential in governing ozone model-data differences than LNO<sub>x</sub> near the Earth's surface."

In Section 4.4 (P40L16–21), we say "With the use of the new lightning parameterisation, the relative change in the modelled yearly averaged CO at Ushuaia, Cape Grim, Mauna Loa and Mace Head is -8.1%, -9.8%, -3.8%, and -0.3%, respectively. The modelled ground-level CO is affected only very marginally by the flash-rate modification compared to the magnitude of the model-data differences, except at Ushuaia and at Cape Grim during the austral summer. Clearly, as in the case of ground-level O<sub>3</sub>, the lightning changes alone do not reconcile the differences between the modelled CO and observations."

We now also present a comparison with the SHADOZ ozonesonde measurements following a suggestion by Referee #1 (P34L7–P35L17).

Overall, this paper appears an incremental improvement but it does offer some new insights and should be published after attention to these comments and the ones below.

Response: Thank you for your comment.

Other specific comments:

Page 4 Line 5-6: All cited papers listed above evaluate performances of PR92 over either land or ocean, or both. This statement doesn't hold with respect to all existing studies.

**Response:** The intended emphasis in the statement was on 'properly', that is to say 'fully'. But following the Referee's comment, we have deleted the sentence.

Changes in manuscript: The sentence has been deleted.

Page 5 Line 4: What's the chemical timestep? I understand the model timestep of 60 min as too large to solve chemistry properly. But if I am incorrect, some extra words to explain would be helpful.

**Response:** The model dynamical timestep is 20 minutes, the UKCA chemical solver is called every hour. It is a symbolic backward Euler solver with Newton-Raphson iteration, and runs to convergence, halving the step when required. Further information on the chemical solver used and its performance is given by Esenturk et al. (2018, Geosci. Model Dev., https://doi.org/10.5194/gmd-11-3089-2018).

**Changes in manuscript:** The above text is added and the reference Esenturk et al. (2018) included in the 2<sup>nd</sup> paragraph of Section 2 (P5L11–13).

## Page 6 Line 10: The threshold of 5km for cloud thickness looks arbitrary, authors should discuss this in relation to the estimate of lightning flashes.

**Response:** In the model's lightning scheme, a threshold convective cloud scale needs to be specified for it to constitute a thunderstorm. We use a minimum convective cloud thickness (i.e. the height of cloud top minus the height of cloud base) of 5 km for the lightning NO<sub>x</sub> to be activated. The cloud base and top are diagnosed on a time-step basis from the physical model's convection scheme. The selected threshold of 5 km is consistent with observations of the vertical scale of thunderstorms presented by several researchers, viz. Price and Rind (1992, 1993), Molinié and Pontikis (1995), and Ushio et al. (2001), which have a minimum value of approximately 5 km. Boccippio (2002) considered the Price and Rind (1992) data for cloud tops greater than 6 km.

While prescribing a minimum convective cloud thickness of 5 km for lightning is somewhat arbitrary, having no such threshold value would be unrealistic because then it would be implicitly assumed that a convective cloud always translates to a thunderstorm, and this would lead to unrealistically high flash rates. For example, removing this constraint in our base model (with the PR92 lightning scheme) increased the average global flash rate by 44%.

**Changes in manuscript:** Part of above discussion added to 2<sup>nd</sup> paragraph of Section 2.1 (P6L20–26). Also, please refer to the last paragraph of Section 3.6 (P22L6–17) on applying a scaling factor to modelled flash rate.

## Page 8 Line 12: Based on the discussion of electrical dipole, cloud thickness seems to be a better parameter representing dipole separation and size of charge centers. It is not obvious to me that it links to cloud-top height.

**Response:** In the conceptual picture of a thunderstorm as an electrical dipole used in developing the scaling relationships for the electrical power generated by the thunderstorm, it is assumed that the two cloud charges are spherical (each with radius R) and the dipole separation is 2R. To derive an operationally useful and empirically testable scaling relationship, it is further assumed that the dipole separation varies as cloud-top height. Boccippio (2002) justifies this approximation by observations that in many storms the lower negative charge region remains relatively constant in height and that most upper positive charge is carried on small ice crystals with negligible terminal velocity. Thus, cloud-top height can be taken as a linear approximation of dipole separation.

**Changes in manuscript:** In the paper just above Eq. (5), we add "This assumption is based on observations that in many storms the lower negative charge region remains relatively constant in height and that most upper positive charge is carried on small ice crystals with negligible terminal velocity."

Page 25 Line 11-13: The argument of better agreement over ocean using model run 2 is not convincing in terms of large uncertainty in the calculation. Nv,trop from CAMS is calculated using the average of the two curves as Nv,trop,180, which leads to overestimate compared to Nv,trop calculated using model run1 (PR92) and underestimate compared to Nv,trop calculated using model run 2 (TS1) over the tropical region. Note in Figure 9b, the column NO2 ( $_0.5X1015$ ) within the latitudes  $\pm 30^{\circ}$  is comparable to column NO2 ( $_0.3X1015$ ) over the reference longitude shown in Figure 8. The better agreement between model run2 and CAMS shown in Figure 9b may be predominantly attributed to the uncertainty introduced in Nv,trop,180. To make the result more convincing, two model runs should be compared to two CAMS column NO2 datasets calculated using Nv,trop,180 from model run1 and run2, respectively.

Response: Thank you for this comment. A similar comment was also raised by Referee #3.

With regards to comparing the tropospheric NO<sub>2</sub> columns, since we did not have  $N_{v,trop,180}$  directly from observations, we used the model generated latitudinal variation of  $N_{v,trop,180}$  in the derivation of the 'observed'  $N_{v,trop}$ . The quantity  $N_{v,trop}$  thus obtained was then used to compare with the modelled  $N_{v,trop}$ . But, as the Referee has rightly pointed out, this approach influences the model-data comparison because the data then partially depend on the model results which in turn biases the comparison in favour of a better model performance.

The Referee's suggestion that  $N_{v,trop,180}$  calculated separately for model Run1 and Run 2 should be used (rather than the average of the two) would not alleviate the core issue because  $N_{v,trop,180}$ going into the determination of the observed  $N_{v,trop}$  would still be dependent on the model results.

We have now used a much more justifiable approach whereby we calculate  $N_{v,trop,180}$  directly from the Ozone Monitoring Instrument (OMI) satellite data of tropospheric NO<sub>2</sub> columns (<u>http://www.temis.nl/airpollution/no2.html</u>; Boersma et al., 2017, 2018) and use this in the CAMS reanalysis data to obtain  $N_{v,trop}$ . With this, the model performance does not turn out to be as strong as before (as expected), but there is no change in the overall conclusion from the model-data comparison.

**Changes in manuscript:** The quantity  $N_{v,trop,180}$  is now calculated using the Ozone Monitoring Instrument (OMI) satellite data of tropospheric NO<sub>2</sub> columns (<u>http://www.temis.nl/airpollution/no2.html;</u> Boersma et al., 2017, 2018) and the model-data comparison is revised accordingly.

The pertinent Section 3.7.3 has been fully revised, including Figures 8 and 9 and Table 5, and additional references of Boersma et al. (2017, 2018). We have also changed the section heading from "Tropospheric NO<sub>2</sub> verification" to "Modelled tropospheric total column NO<sub>2</sub> and validation".

References of Boersma et al. (2017, <u>http://temis.nl/qa4ecv/no2col/QA4ECV\_NO2\_PSD\_v1.1.compressed.pdf</u>; 2018, Atmos. Meas. Tech., <u>https://doi.org/10.5194/amt-11-6651-2018</u>) added.

Figure 14: It's very hard to conclude that the new parameterizations lead to modelled ozone closer to the observations from this figure. A better visualization is suggested, for instance, set zero to white color, use relative difference plot etc.

**Response:** Point taken. We have redrawn the plots (now Figure 15) with the range around zero set to white colour. Also, these are now relative difference plots (rather than absolute difference). An

additional Figure 15d is given showing the relative difference between the concentration modelled without any  $LNO_x$  and the observations. The text has been changed to describe the modified plots.

Changes in manuscript: As above.