Reply by the authors to Referee #1's comments on

"Assessing and improving cloud-height based parameterisations of global lightning flash rate, and their impact on lightning-produced NO_x and tropospheric composition" (#acp-2020-885)

Anonymous Referee #1 (RC3)

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.

General Comments: Well written manuscript that is nearly ready for publication.

Thank you.

Scientific Comments:

Abstract reads well. My only quibble is that PR92's deficiencies over water have been well known for years. Perhaps you should go with . . . via the Price and Rind (1992) (PR92) formulation, whose water parameterization is known to greatly underestimate flashes.

Response: We agree with the Reviewer.

Changes in manuscript: The sentence in the abstract has been modified as follows:

"... via the Price and Rind (1992) (PR92) parameterisations for land and ocean, where the oceanic parameterisation is known to greatly underestimate flash rates."

Also, "as expected" inserted in the sentence to read "... the oceanic parameterisation, as expected, underestimates the observed flash-rate density severely,"

P5 L19: Be clear that the k referred to in line 19 is the same as the one referred to in equation (2). This could be done by removing the line 19 bullet and including this sentence in the preceding paragraph.

Response: Point taken.

Changes in manuscript: The suggested change made.

P6L23: What is the justification for parameterizing the thickness of the cold cloud region in terms of latitude when you could use temperature profiles from the driving model? What errors are induced by assuming a simple dependence on latitude for the cold-cloud region?

Response: In the ACCESS-UKCA model, the method of apportioning the total number of flashes into cloud-to-ground (CG) and intracloud (IC) flashes is based on the commonly used empirical parametrisation of Price and Rind (1993) which was developed using cloud height and IC/CG ratio data for 139 individual thunderstorms. In the Price and Rind (1993) parameterisation, the ratio $z_R = IC/CG$ increases as a sole function of the thickness (*dH*) of the cold cloud region in thunderstorms (from 0°C to cloud top), and *dH* is parameterised as a decreasing function of latitude.

ACCESS-UKCA uses the Price and Rind (1993) parameterisation in its entirety, including the thickness of the cold cloud region as a function of latitude. We find that, on average, 24% of the modelled flashes are CG flashes, which is the same amount obtained by Price and Rind (1994) using the same parameterisation.

As stated by the Referee, another possible way to compute the thickness of the cold cloud region is to directly use the temperature profiles from the model. There may be some sensitivity of the ratio $z_R = IC/CG$ to which of the two ways *dH* is calculated and its impact on LNO_x. However, we think that this sensitivity would be relatively small in our study considering the following.

Firstly, in the present model setup, the CG and IC proportions are only used in estimating the vertical LNO_x profile (the amount of LNO_x produced per CG and IC flash is taken to be the same). Therefore, the total LNO_x production is unaffected.

Secondly, Price and Rind (1993) observed that the correlation between observations of the thickness of the cold cloud region and the dH parameterisation based on latitude was quite high (= 0.90).

Thirdly, our estimate of CG flashes being 24% of the total is similar to the value 20% obtained by Barthe and Barth (2008) who directly computed dH as the thickness between the average height of the 0°C isotherm across the model-generated cloud and the average height of where the computed total hydrometeor mixing ratio decreases to 10⁻⁵ kg kg⁻¹ at the top of the cloud.

Changes in manuscript: The last paragraph of Section 2.1 (P7L12–23) is expanded to read:

"We use Eq. (4) along with Eq. (3) to calculate the total flash rate $f_{L,O}$ which is then apportioned into cloud-to-ground (CG) and intracloud (IC) flash rates using an empirical parameterisation for the ratio $z_R = IC/CG$ developed by Price and Rind (1993) (PR93) based on thunderstorm observations in the western United States. In this parameterisation, z_R increases as a function of the thickness (*dH*) of the cold cloud region in thunderstorms (from 0°C to cloud top), and *dH* is parameterised as a decreasing function of latitude. The PR93 parameterisation has been used frequently, with further validation for case studies reported by Pickering et al. (1998) and Fehr et al. (2004). Allen and Pickering (2002) and Grewe et al. (2001) used it in global atmospheric chemistry models, with the former evaluating it for cases in the US. The averaged values of z_R and the CG to total flash ratio obtained from the PR93 parameterisation in the present study are 3.14 and 0.24, respectively. These values are comparable to $z_R \sim 4$ and the CG to total flash ratio ~ 0.2 obtained by Barthe and Barth (2008) using *dH* calculated directly from modelled cloud temperature and total hydrometeor mixing ratio in the PR93 parameterisation. Using IC/CG measurements, Bond et al. (2002) derived a parameterisation for z_R as a linearly decreasing function of latitude and obtained $z_R = 3.76$ and the CG to total flash ratio = 0.21 over the tropics (35° N -35° S)"

P12L25: How different would NO and NL be if you used the same value of k1 for land and ocean points, i.e., would the difference and values be comparable to that shown in Michalon.

Response: This sentence was meant to be qualitative in nature. It is difficult to establish an exact equivalence between the N_0 and N_L values used by Michalon et al. (1999) in deriving their oceanic flash rate (F_0) expression and the k_1 values used in our parameterisations, because the power law dependence on cloud-top height is different in these expressions. But neglecting the differences in this dependence and assuming the same logic as Michalon et al.'s in deriving their oceanic flash rate, the different k_1 values for land (k_{1L}) and ocean (k_{10}) can be linked via $k_{10}/k_{1L} = (N_0/N_L)^{2/3}$. So, if k_{10} is assumed to be the same as k_{1L} (= 1.612 × 10⁻⁵) then $N_0 = N_L$. On the other hand, for $k_{10} = 0.7 \times 10^{-5}$ as calculated for the oceanic component in the new parameterisation, (N_0/N_L)^{2/3} = $k_{10}/k_{1L} = 0.43$. So, for $N_L = 600$ per mg used by Michalon et al.

we get $N_O = 170$ per mg compared with $N_O = 50$ per mg used by Michalon et al. Thus, the differences in k_1 and N_O/N_L do not match.

Changes in manuscript: Considering the above, we simply modify the sentence (P14L22–24) as "... for land and ocean can be interpreted in terms of N_0 and N_L being different with $N_0 < N_L$."

P13 L5: When specifying flash rates, is continental-marine synonymous with land-ocean or is the convection in grid boxes within x km of the coast classified as continental in character? Or to put it another way: Should there be a transition zone over the ocean where flashes are still parameterized as if they are continental in character?

Response: We use 'continental-marine' and 'land-ocean' synonymously. In the model, any grid cell that has a non-zero land surface fraction is considered land for the purposes of lightning NO_x calculation. Conversely, only grid cells with 100% water surface coverage are considered ocean.

Changes in manuscript: This is made clear in the 2nd paragraph of Section 2.1 (P7L9–11).

P16 L10-11 How sensitive is the conclusion (that Run 2 does the best) to the choice of meteorological model? Or to put it another way, how likely is it that the Michalon approach would do better if the cloud top heights were taken from a different model?

Response: We do not have an alternative meteorological model or a convection scheme to test the sensitivity of the calculated lightning flash rates. The atmospheric component of the ACCESS-UKCA model used is the UK Met. Office Unified Model (UM). The convective cloud bottom level and top level used in the flash rate calculation are diagnosed from the UM convection scheme. Evaluation of the distribution of cloud depths simulated by the UM has been reported in studies such as Klein et al. (2013) and Hardiman et al. (2015) (referred to in the paper).

Some sensitivity of flash-rate parameterisations to meteorology can be examined by doing a model run with meteorological nudging. We have done additional model simulations with meteorological nudging, and summarise the results in a new Section 5 titled "Model simulations with meteorological nudging." In short, the average flash rates obtained from Run 2 with nudging were approximately 5% higher than those from the free running Run 2 in Table 1 (so a scaling factor of 0.95 would make the nudged model flash rates approximately match the free running model flash rates).

The Michalon et al. approach may or may not do better if the cloud top heights were taken from a different model. Our oceanic parameterisation is based on flash rate vs. cloud-top height observations as presented in Figure 1, and is, by design, able to represent the flash rate data in Figure 1 better than does the Michalon et al. parameterisation. Also, our parameterisation is consistent with Boccippio's (2002) scaling relationships.

One commonly used approach to get the modelled flash rate right is to first calculate a scaling factor that is the ratio of the observed globally averaged flash rate to the modelled globally averaged flash rate, and then apply this scaling factor to the modelled flash rate for subsequent model runs. This approach is useful as long as the relative global spatial distribution of flash rate is represented realistically. Otherwise, for example with the PR92 approach, the use of global scaling would lead to an overestimation of flash rate (and LNO_x) over land and underestimation over the ocean, although the global average flash rate (and LNO_x) may be correct.

Changes in manuscript: New Section 5 on "Model simulations with meteorological nudging" included. Some discussion on scaling is already given in the last paragraph of Section 3.6 (P22L6–17).

P18 L21: What do you mean by "significant spatial differences"? High-biases? Low-biases?

Response: Point taken.

Changes in manuscript: The sentence is modified (P21L1–5) to read "However, as shown in Figure 5d, ACCESS-UKCA with the new flash-rate parameterisations (Run 2) simulates the oceanic distribution of flash density much better than the PR92 scheme, although it is clear that there are some significant spatial differences (e.g. low bias over western Indian Ocean near southern Africa, and high bias over equatorial Indian Ocean and the Pacific) compared to the corresponding observations and climatology."

P23 L17: What is your rationale for distributing IC and CG emissions as you do? Is it motivated by the results of Pickering, Ott. Other? Wouldn't sub-grid scale mixing lead to some overlap in altitude between where NO from IC and CG flashes is deposited? How did you choose 500 hPa as the dividing line?

Response: In our model, once the amount of LNO_x at a grid point location at a time step has been computed, it is distributed evenly in the vertical in log-pressure coordinate from 500 hPa to the cloud top for intra-cloud (IC) flashes, and from 500 hPa to surface for cloud-to-ground (CG) flashes. The method is motivated by the data analysis of Price and Rind (1993). Their observations from 139 thunderstorms cover cold cloud thickness (i.e. the cloud top height minus the freezing level) values ranging between 5.5–15 km and freezing level values between 2.7–5 km. The ratio $z_R = IC/CG$ increases from 0 to 4.6 with cold cloud thickness from 5.5 to 15 km, but remains relatively constant with freezing level. We take the level below which the CG generated LNO_x is distributed as the observed minimum freezing level plus half of the minimum cold cloud thickness, i.e. $(2.7+5.5/2) \approx 5.5$ km. The selected 500 hPa level is closest to this 5.5 km value.

The use of the log-pressure (rather than linear pressure) coordinate yields a vertical distribution of LNO_x that has more LNO_x released at higher levels.

The non-uniform shape of the averaged modelled vertical distributions of LNO_x is largely caused by the averaging of the LNO_x profile from every time step over spatial and temporal variations in the cloud-top height.

Our averaged model profiles of LNO_x in Figure 7 compare better with Ott et al. (2010). But we believe more observations are required to better understand the nature of LNO_x distribution in the vertical and to constrain it.

Changes in manuscript: A new paragraph added just prior to Section 3 (P9L1–11).

P28 L13: Several of the sites you selected are in regions where the impact of NOx from lightning is minimal. Please add a comparison or two to profiles from locations within the SHADOZ network that may be more affected by lightning-NOx. You could use boundary layer values from the profiles if you wish to retain your focus on the surface.

Response: The selection of the ground-based sites, viz. Ushuaia, Cape Grim, Mauna Loa, Minamitorishima and Mace Head, was based on these sites being either oceanic or coastal and covering a range of latitudes. This emphasis on oceanic/coastal location was given because of the

relatively large difference between the PR92 oceanic flash rate parameterisation and the new one which would directly impact the modelled ozone at such locations.

But it was clear that LNO_x impacts surface ozone very little compared to that at higher altitudes. And in that regard, we thank the Reviewer for the suggestion about the use of the SHADOZ (Southern Hemisphere ADditional OZonesondes) ozone profile data (<u>https://tropo.gsfc.nasa.gov/shadoz/</u>). We have now processed the SHADOZ data and made a comparison with the modelled ozone profiles at eight sites for the year 2006.

Changes in manuscript: In the Figure 12 description, we add "Apart from data availability and covering a range of latitudes, the site selection was based on these sites being either oceanic or coastal so that the relatively large difference between the PR92 oceanic flash rate parameterisation and the new one could be examined against the observations."

We have now included a comparison with the SHADOZ observations in Section 4.2 (P34L7–P35L17) and a new Figure 13. The comparison supports the results from the new oceanic flash-rate parameterisation for all southern hemispheric sites.

P28L15: Is LNOx an important source of NOx at any of these sites? Are differences between the models larger in the winter or summer? I would assume the latter; however, it isn't clear at Ushuaia for e.g..

Response: As mentioned in the response above, only oceanic/coastal sites were selected because the two flash rate parameterisations mostly differ in their oceanic treatment.

There were no NO_x measurements at these sites, and here (in Figure 12) we compared ozone. There are small, but noticeable, differences in the modelled ozone as a result of the differences in the two flash rate parameterisations at these ground-based sites, which suggests that there is an impact of LNO_x , but it is much smaller compared to that the at higher altitudes (e.g. mid troposphere) (which is clear from the SHADOZ comparison in Figure 13 and the zonal difference plot in Figure 14c).

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_x near the Earth's surface.

There is no clear indication from the plots (Figure 12) if the differences in ozone between the two models are larger in the winter or summer, except for Ushuaia where the differences are larger during winter to spring. It is difficult to link the seasonal differences between the two modelled ozone variations linearly to those in LNO_x due to the complexity of ozone chemistry coupled with transport.

Changes in manuscript: In the Figure 12 description, we add (P33L13–25) "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 O₃ 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_x scheme, but for the other two sites the model-data differences are much larger than those due to the LNO_x 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_x near the Earth's surface. There is no clear indication if the differences in ozone between the two models are larger in the winter or summer, except for Ushuaia where the differences are larger during winter to spring."

P33 L3: You should state that the LNOx-induced increase in OH due to the new scheme exacerbates the model high bias in OH burden. – but a softer adjective than "exacerbate" is fine.

Response: Point taken.

Changes in manuscript: Added (P39L15–17) "The LNO_x-induced increase in OH due to the new scheme adds to the model high bias in the OH burden in summer, whereas it reduces the magnitude of the bias in winter with the bias shifting from low to high."

P36 L6: Would it make sense to give the locations of all of the data sets here as opposed to referring back to the manuscript or the acknowledgements section.

Response: Point taken.

Changes in manuscript: The locations/sources of the datasets used are now provided under "Data availability" (P43).

Technical Details:

P2L11 NO and NO2 have already been defined.

Changes in manuscript: Correction made.

P5L18 mb — hPa

Changes in manuscript: Correction made.

P6 L12: as a function of and H — as a function of H

Changes in manuscript: Correction made.

P6 L14: (discusses later) — (discussed later)

Changes in manuscript: Correction made.

P10 L10: when used in global models — when used over water in global models

Changes in manuscript: Correction made.

P28 L8: global ozone — global annual mean ozone Changes in manuscript: Correction made. P31 L1 The three "by"s are not necessary and can be removed.

Changes in manuscript: Correction made.

P32 L8: At mid-troposphere — In the mid-troposphere

Changes in manuscript: Correction made.

P34 L2 The NMSE and r values suggest a mixed result. — Be specific as to what you mean here.

Changes in manuscript: The sentence is deleted, and we add (P40L16–21) "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 O₃, the lightning changes alone do not reconcile the differences between the modelled CO and observations."

P36 L1 "perhaps currently not well constrained" - poorly constrained

Changes in manuscript: Correction made.