

We are very grateful to David Stevenson for his constructive comments. We provide the comments (in bold) and our replies below. Amendments/additions to the text are highlighted in blue. The revised manuscript is provided as an additional supplement.

**Review of ‘Lightning NO<sub>x</sub>, a key chemistry-climate interaction: impacts of future climate change and consequences for tropospheric oxidising capacity’, by Banerjee et al.**

### **General Comments**

**This paper presents results from a series of model experiments with the chemistry-climate model UKCA, investigating the influence of climate change on changes in lightning NO<sub>x</sub> emissions, and the consequences for future levels of tropospheric ozone and hydroxyl radical. The paper is interesting and well presented, but I do have some suggestions for adjustments.**

**My main concerns are about how methane is treated and the consequences for how the simulations are interpreted. Methane concentrations are held fixed (p8757 l16). This is done for reasons of computational efficiency, but this is an artificial model imposition, and it has important influences on model results. Clearly, in the real world, methane concentrations are not held fixed and would be free to respond to any changes in OH.**

**Fixing methane concentrations in the model means that when lightning NO<sub>x</sub> increases, and OH increases, and the methane lifetime reduces, methane concentrations cannot respond. Since methane is an ozone precursor, this mechanism of influencing ozone is not included in the simulations. If methane concentrations were not held fixed, methane would slowly adjust to a new (lower) equilibrium concentration, and the resultant change in ozone would be less than is reported. Similarly, the changes in OH would also differ from those reported. The differences in the changes in ozone and OH between a fixed methane run and a run where it were not held fixed are likely to be substantial, and this must be more clearly acknowledged than it currently is (e.g., this major caveat is not mentioned in the Abstract). It is relatively easy to calculate the approximate impact of holding methane fixed on the results, e.g., following the methods described in Stevenson et al. (2013). If this issue and the minor comments below are addressed, then I recommend this paper is published in ACP.**

With regard to methane feedbacks, please see our response to Point 11 in the Specific Comments.

Please note that Point 5 of the Specific Comments details how we distinguish LNO<sub>x</sub> concentration from its emissions in the text; all of the following responses incorporate these modifications.

### **Specific Comments**

**1. p8759 l8: Reference is made to the interactive calculation of LNO<sub>x</sub>, but this hasn’t been described yet. I think it makes more sense for the last paragraph of Section 2.1 to probably move to the end of Section 2.2.**

We would prefer to keep this paragraph in Sect 2.1, which describes the various simulations, and reserve Sect. 2.2 for a description of the LNO<sub>x</sub> parameterisation. However, as pointed out, the

interactive calculation requires explanation if mentioned and so we have inserted a reference to the next section:

P8759 L8 [‘To do this, we switch off the interactive calculation of LNOx \(see Sect. 2.2\) and instead...’](#)

**2. p8759 l10-11: ‘The Base(fLNOx) and Base runs should be identical; indeed there are negligible differences...’** This sounds like they are definitely not identical. It is not quite clear how different these two base runs are. Are they slightly different because the LNOx emissions are specified differently? In the Base run, are LNOx emissions varying every timestep? (If so, what is the timestep?) Are these high time resolution variations in LNOx emissions also carried over to Base(fLNOx)? At this stage, you haven’t described exactly how the LNOx emissions are specified and even the upcoming Section 2.2 doesn’t describe the temporal variability in the fully interactive run.

In the Base run, LNOx is calculated in every chemical timestep by UKCA (i.e. every hour). The field is output as a monthly mean diagnostic. The monthly mean climatology of this field is then used as input for the Base(fLNOx) and ΔCC8.5(fLNOx) runs and linearly interpolated to 5-day values. Thus, these runs do not have the high time resolution variations in LNOx. However, the differences between the Base and Base(fLNOx) are within the internal variability in the system. We have made the following modifications to the text:

P8759 L8 [‘...and instead, impose a monthly mean climatology of these emissions obtained from the Base run, which is linearly interpolated to 5-day averages.’](#)

P8759 L18 [‘LNOx is calculated every hour in UM-UKCA following the method applied in the p-TOMCAT model.’](#)

**3. p8759 l27: ‘...H is the cloud-top height (km)...’** Is H height above sea-level or above the surface topography? UKCA uses ‘hybrid height’; is H really hybrid height?

H is not hybrid height and it is the height above ground level (Price and Rind, 1994).

**4. p8760 l9: Why are the NOx emissions distributed evenly in log-P from 500 hPa to the cloud top? How sensitive are your results to this (relatively arbitrary?) choice?**

As indicated by Labrador et al. (2005), and as the reviewer rightly suggests, the simulated NOx, ozone, OH etc. are sensitive to the vertical placement of LNOx. However, there are considerable uncertainties and arguments in the actual vertical distribution. For example, Pickering et al. (1998) suggest a C-shaped vertical distribution (hereafter PICK98), which shows maxima in LNOx in the upper troposphere and boundary layer. On the other hand, Ott et al. (2010) suggest a distribution (hereafter OTT10) where emissions are weighted more toward the middle and upper troposphere and without a boundary layer (BL) maximum. By evenly-distributing the concentration of NOx in log-p and distributing between the ground-500hPa (CG) and 500hPa-cloud top (IC), the UKCA scheme places LNOx more evenly by mass in the vertical (for a given source strength) as compared to both the PICK98 and OTT10 distributions.

For example, in comparison to the tropical continental OTT10 profile presented in Fig. 12 in Ott et al. (2010), the distribution in UKCA (not shown) has lower LNOx (as a fraction of total LNOx) from ~7 km upwards, similar amounts between ~4-7 km and greater LNOx below ~4 km. Compared to the PICK98 profile in the same figure, UKCA shows greater LNOx in all but the lowest ~1.5 km where the BL maximum is missing in UKCA.

From this comparison, we expect UKCA to produce lower NO<sub>x</sub>, ozone and OH from LNO<sub>x</sub> in the upper troposphere, where NO<sub>x</sub> lifetimes are long, relative to the PICK98 and OTT10 distributions. For the same reason, we might expect UKCA to underestimate changes in ozone and OH with increases in LNO<sub>x</sub> as a result of future climate change. The differences in the BL and lower troposphere between different distributions should not cause large differences in simulated ozone and OH since NO<sub>x</sub> lifetimes here are short and other sources dominate the total NO<sub>x</sub> production rate (Labrador et al., 2005).

Note that from a comparison of three vertical distributions (including PICK98 and OTT10), Labrador et al. (2005) were unable to select a best fitting distribution due to the low number of observational campaigns and the large scatter in existing observational data. This highlights that there remain significant uncertainties in the vertical distribution of LNO<sub>x</sub>, and consequently we have made the following amendments to the text:

P8760 L11 'Implementation of the PR92 scheme varies in its details from model to model, generally with an aim to generate lightning flash frequencies and distribution for the present-day atmosphere (as within the development of UM-UKCA) or for a particular choice of total, global LNO<sub>x</sub>. In a model study, Labrador et al. (2005) have demonstrated that, in addition to the overall magnitude of LNO<sub>x</sub>, concentrations of tropospheric trace constituents are also particularly sensitive to the vertical distribution of LNO<sub>x</sub>. However, they were unable to select a best fitting distribution due to the low number of observational campaigns and the large scatter in existing observational data. Compared to other vertical LNO<sub>x</sub> distributions, such as those suggested by Pickering et al. (1998) and Ott et al. (2010), UKCA distributes LNO<sub>x</sub> more evenly by mass in the vertical. As a result, UKCA would simulate lower ozone and OH in the mid and upper troposphere for a given magnitude of total LNO<sub>x</sub>, relative to these distributions. In the lower troposphere and the boundary layer, where NO<sub>x</sub> lifetimes are short, trace gas concentrations are far less sensitive to LNO<sub>x</sub> (Labrador et al., 2005).'

P8760 L14 New paragraph at 'Convection itself is also parameterised...'

P8760 L19 New paragraph at 'As in many sensitivity studies...'

P8768 L4 'We have demonstrated that NO<sub>x</sub> production from lightning, following tropical convection, is a key process through which climate can influence the chemistry of the troposphere. Hence, given its importance, we believe it is crucial to strengthen our confidence in model representations of both convection and LNO<sub>x</sub>. Our results are dependent on the LNO<sub>x</sub> and convective parameterisations utilised. In particular, the vertical profile of LNO<sub>x</sub> affects the simulated changes in ozone and OH, particularly in the UT (Labrador et al., 2005). If we were to employ the vertical distributions of Pickering et al. (1998) and Ott et al. (2010), which weight LNO<sub>x</sub> more greatly to the UT than is done in UKCA, we postulate that even larger changes in ozone, OH and subsequent feedbacks would occur for a given change in total LNO<sub>x</sub>.'

**5. Section 3.1/p8762 l8/throughout: In the Introduction, you define LNO<sub>x</sub> as 'NO<sub>x</sub> produced from lightning'. So I think LNO<sub>x</sub> refers to the fraction of NO<sub>x</sub> derived from lightning, or 'lightning NO<sub>x</sub>' for short. In other places you refer to 'LNO<sub>x</sub> emissions'. But in other places, you just use LNO<sub>x</sub> with the implication that you mean LNO<sub>x</sub> emissions. This may sound pedantic, but I think the text should be tightened up to remove any ambiguity; this would certainly be the case for NO<sub>x</sub> vs. NO<sub>x</sub> emissions, so I think the same applies to LNO<sub>x</sub> vs. LNO<sub>x</sub> emissions. For example, the text (and figure caption) describes Figure 2a as showing the correlation of P(O<sub>x</sub>) and LNO<sub>x</sub>, but the figure actually shows LNO<sub>x</sub> emissions. If every reference to LNO<sub>x</sub> really means the emissions, then I suggest change the opening definition.**

Most references are to the emissions, hence we have taken out 'emissions' throughout the text and changed the opening definition as follows:

P8754 L5 '...the impact of climate change on emissions of NO<sub>x</sub> from lightning (LNO<sub>x</sub>)...'

P8755 L3 'Emissions of NO<sub>x</sub> from lightning (LNO<sub>x</sub>) in the mid and upper troposphere (UT), where the NO<sub>x</sub> lifetime is longer than at the surface, exert a disproportionally large influence on tropospheric chemistry.'

Additional minor rewordings:

P8754 L19 '...primarily as a consequence of greater LNO<sub>x</sub>.'

P8755 L11 'NO<sub>x</sub> from lightning induces production of ozone...'

P8755 L19 'In addition, formation of HO<sub>x</sub> can be induced indirectly,...'

P8759 L18 'LNO<sub>x</sub>...'

P8760 L5 '...resulting in 6 Tg(N) yr<sup>-1</sup> of total, global LNO<sub>x</sub> for the year 2000.'

P8761 L15 'With regard to its geographical distribution, LNO<sub>x</sub> occurs predominantly...'

P8762 L3 '...may result in systematic biases in LNO<sub>x</sub> over this region.'

P8765 L26 'We have assessed the impacts of climate change on emissions of NO<sub>x</sub> from lightning (LNO<sub>x</sub>) and the consequences for tropospheric chemistry using UM-UKCA. Using the Price and Rind (1992, 1994a) parameterisation for calculation of LNO<sub>x</sub>, our year-2000 integrations generate 6 Tg(N) yr<sup>-1</sup> of total, global LNO<sub>x</sub>, which lies...'

P8767 L14 'An analysis of reaction fluxes indicates that the dominant reaction pathways for increased OH production through LNO<sub>x</sub> in these regions are HO<sub>2</sub> + NO (directly, following production of NO<sub>x</sub>) and O(<sup>1</sup>D) + H<sub>2</sub>O (indirectly, through photochemical ozone and hence O(<sup>1</sup>D) production).'

**6. p8763 l27-28 '...decrease in ozone is seen throughout the troposphere...primarily due to increased humidity in a warmer climate.'** This is a relatively old and well known result, and probably deserves a reference (e.g., Thompson et al., 1989; Johnson et al., 1999; Doherty et al., 2013).

Inserted a reference to Thompson et al. (1989).

**7. p8764 l4 (l10; also p8766 l14) Figure 4 does not show zonal changes.**

We have amended as follows:

P8761 L17 'Figure 1 shows changes in the tropically averaged (20°S-20°N), annual mean distribution of LNO<sub>x</sub>...'

P8764 L4 'Figure 4 illustrates changes in the tropically averaged (20°S-20°N), annual mean distribution of OH...'

P8764 L10 'An analysis of species concentrations and reaction fluxes...'

P8766 L14 '...we also analysed changes in the distribution of LNO<sub>x</sub> within the tropics.'

**8. p8765 l6-8 Better to use years (rather than months), the 'normal' unit used for methane lifetime.**

Have changed units to years throughout the text.

**9. p8765 l9 'This might have implications...' I think there is little doubt: it will.**

Have changed as suggested.

**10. p8766 l21 ‘P(Ox) contributes to the tropospheric ozone burden...’** This is a slightly odd way of putting it: it is quite possible for P(Ox) to increase whilst the ozone burden decreases (e.g. if Ox destruction increases by more than P(Ox)).

The sentence aimed to highlight that increases in P(Ox) would lead to *direct* increases in the ozone burden. To avoid misleading a reader, we have changed the sentence to read:

P8766 L21 ‘The tropospheric ozone burden increases correspondingly by...’

**11. p8767 l21-26 Indeed – the fact that your experiments used fixed methane concentrations have important caveats for both the reported changes in ozone and OH. These major caveats needs to be reflected in the Abstract: the quantitative results quoted there would change significantly if the experiments were repeated with methane concentrations free to adjust, and in the current Abstract, the reader does not know that methane concentrations are held fixed in the modelling.**

We agree and have detailed the caveats related to this model imposition more carefully in the amended text as follows:

- The caveat has been added to the Abstract:

P8754 L10 ‘The total tropospheric chemical odd oxygen production (P(Ox)) increases linearly with increases in total LNOx and consequently, tropospheric ozone burdens of  $29 \pm 4$  Tg(O<sub>3</sub>) (RCP4.5) and  $46 \pm 4$  Tg(O<sub>3</sub>) (RCP8.5) are calculated here. By prescribing a uniform surface boundary concentration for methane in these simulations, methane driven feedbacks are essentially neglected. A simple estimate of the contribution of the feedback reduces the increase in ozone burden to 24 and 33 Tg(O<sub>3</sub>), respectively.’

- We have added:

P8763 L3: ‘As a result, the decrease in burden of  $34 \pm 4$  Tg(O<sub>3</sub>) due to  $\Delta O_3pre$  is just outweighed by the increase in  $\Delta(CC8.5 + O_3pre)$ , although by using a fixed methane surface concentration in these simulations, the additional feedbacks on ozone and OH are not included (see Sect. 3.4). Nevertheless, it appears that reductions in the ozone burden due to emission policies could be counteracted by future changes in climate.’

- We have removed ‘somewhat’ on P8765 L14 since we have quantified the feedbacks related to this effect (see below).
- We have ended Sect. 3.3 at the end of P8764 and added a subsequent section describing the impacts of changes in OH on methane (including, as suggested, a quantitative description of the methane feedback) and VSLs:

### ‘3.4 Consequences for methane and other trace gases

Since OH is the primary tropospheric oxidant, substantial enhancements in its abundance, such as those shown in Sect. 3.3, can have ramifications for a range of other chemical species. For example, oxidation by OH is the main loss process for atmospheric methane. Hence, there are potentially global consequences through perturbation of the methane lifetime. A measure of this effect can again be deduced from the  $\Delta CC8.5(fLNOx)$  run. Relative to Base, a reduction of 1.79 years in the methane lifetime against loss by OH ( $\tau_{CH_4+OH}$ ) is calculated for  $\Delta CC8.5$ ; in contrast, a smaller reduction of 1.04 years is found for  $\Delta CC8.5(fLNOx)$ . Inclusion of changes in LNOx thus contributes 0.75 years to the reduction in  $\tau_{CH_4+OH}$  due to climate change.

Changes in  $\tau_{\text{CH}_4+\text{OH}}$  will have implications for both chemistry and climate through methane's role as a tropospheric ozone precursor, an OH sink and a greenhouse gas. However, by fixing a uniform lower boundary condition for methane, such feedbacks are essentially neglected within these experiments. If methane concentrations were allowed to respond to decreases in its lifetime with climate change, lower methane concentrations would be simulated at equilibrium in a future climate, with a lower increase in ozone burden and an enhanced increase in OH. The strength of the response is determined by the model dependent methane feedback factor,  $f$  (Fuglestad et al., 1999). Using a further integration in which methane is increased by 20% in the chemistry scheme only (not otherwise discussed here), we derive a value of 1.52 for  $f$  in our model, which lies on the upper end of the large literature range (1.19-1.53) (Prather, 2001; Voulgarakis et al., 2013; Stevenson et al., 2013). From this, we obtain an estimate of equilibrium methane concentrations, following the methodology detailed in Stevenson et al. (2013), and equilibrium ozone burdens, following Wild et al. (2012). We find that accounting for methane adjustments lowers the ozone burden in future climate simulations by, on average, 5 Tg(O<sub>3</sub>) (RCP4.5) and 13 Tg(O<sub>3</sub>) (RCP8.5). The corresponding increases in ozone burden relative to Base are 24 Tg(O<sub>3</sub>) (RCP4.5) and 33 Tg(O<sub>3</sub>) (RCP8.5), which still represent substantial increases with future climate change and greater LNOx. The adjusted increase in burden in  $\Delta(\text{CC8.5}+\text{O3pre})$  (33 Tg(O<sub>3</sub>)) is now more comparable to the adjusted decrease in  $\Delta\text{O3pre}$  (32 Tg(O<sub>3</sub>)).

OH is also important in determining the lifetime of very short-lived substances (VSLS). There is currently considerable interest in the role of VSLS in stratospheric ozone depletion following rapid convective transport into the upper troposphere-lower stratosphere (UTLS) region. Increased oxidation of VSLS by OH in the UT in a future climate could serve to counteract increased stratospheric VSLS loading following enhanced convective lofting into the tropical tropopause layer (TTL) and subsequent transport into the lower stratosphere. The effect could be particularly important over the Maritime Continent, since it is a region characterised by both high deep convective activity and coastal emissions of VSLS (Hosking et al., 2010). These feedbacks add weight to the importance of future changes in LNOx.'

- The caveat has been added to the Summary:

P8767 L21 'The resulting changes in methane concentration and subsequent feedbacks are not simulated by these experiments. Since methane is both a tropospheric ozone precursor and an OH sink, we expect that a shorter  $\tau_{\text{CH}_4+\text{OH}}$  would feedback negatively into LNOx driven increases in ozone but positively into increases in OH. For ozone, we have estimated that accounting for adjustments in methane concentration in a changing climate would lead to increases in the ozone burden of 24 Tg(O<sub>3</sub>) (RCP4.5) and 33 Tg(O<sub>3</sub>) (RCP8.5). Although, as expected, these are smaller than the simulated changes reported above (of 29 and 46 Tg(O<sub>3</sub>), respectively), they still represent substantial increases through future climate change. Since methane is a greenhouse gas, we would also expect a negative feedback onto climate change through its radiative forcing effect.'

## 12. p8777 Figure 3. Maybe it is also worth plotting the difference between Figure 3a and 3b?

We do not think another figure here would add to the current conclusions that tropospheric ozone generally increases with increases in LNOx under future climate change (Fig. 3a), decreases if lightning is held fixed (Fig. 3b) and that these changes influence lower stratospheric ozone in these simulations.

## References (additional to those already in the paper)

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**Doherty, RM, et al. (2013) Impacts of climate change on surface ozone and intercontinental ozone pollution: A multi-model study, *J. Geophys. Res. Atmos.*, 118, doi:10.1002/jgrd.50266**

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