#### **Responses to Reviewer 1**

We would like to thank Reviewer 1 for their helpful suggestions. Below, we show the reviewer comments (in bold) and our responses. Please note that we have grouped some of the suggestions together. Amendments/additions to the text are highlighted in blue.

Please also note that 'LNOx emissions' has been replaced by 'LNOx' throughout the text (see Point 5 in our response to Reviewer 2).

The paper investigates the impact of a future climate on changes in lightning NOx, ozone and OH by means of climate-chemistry model simulations. The paper is well written and certainly suited for publication after revisions.

1a. The lightning changes are not investigated at all. However they are a crucial part of the lightning NOx simulation. I suggest to include some more information why LNOx is increased. Is more convection occurring or is altitude of the convection increasing and hence the parameterisation is increasing the LNOx (5th power)? Note that a different lightning parameterisation is giving a decrease in lightning, because the number in convective events decreased and the increased intensity did not compensate (Grewe, 2009). This result is in agreement with Brinkop (2002) and DelGinio et al. (2007). This should be discussed since it affects significantly the conclusion on the compensation of a reduction of precursor emissions by increased lightning NOx-emissions.

1b. 8761 / Section 3.1 Changes in lightning is a key to this investigation. However, the causes for the changes are not investigated. Figure one suggests that the tropopause altitude increases in a future climate. Is this true for the convective heights? Or is the stratification of the troposphere getting more stable and hence the convective events are getting more intense (higher), but rare? I suggest to include some more analysis on the reasons for the lightning changes in a future climate.

1c. 8754 l9-10 Changes in convection were not investigated!

1d. 8756 l1-4 I think all the mentioned papers parameterise the flash frequency depending on the cloud top heights. It often has been argued that this is a statistical rather than a physical relationship. Other studies using convective mass flow or updraft estimates based on the convective mass flow predict a decrease in lighting NOx production (Grewe, 2009 and Dahlmann et al. 2011). There are studies suggesting that convective activity might decrease in the future in terms of number of events, but the individual events might be stronger. If that is true, what will happen with the lightning NOx? What is the more important parameter, - the decrease in total number of convective events or the increase in intensity of each individual event? It seems that Price and Rind, since the intensity is parameterised with the fifth order is more important and that an updraft parameterisation is less sensitive to the intensity and hence the number of events dominates. This would lead to a decrease in LNOx! See also e.g. Brinkop, 2002 and DelGenio et al., 2007.

Convective cloud-top height (CTH) was diagnosed every hour in our simulations. We have investigated changes in the *intensity* (depth) of convective events by analysing histograms of CTH, which are shown below in Fig. R1 for the Base and  $\Delta$ CC8.5 runs as examples. Over each of the three convectively active regions, the distribution is shifted towards higher CTH in  $\Delta$ CC8.5, relative to Base,

with the mean increasing by 23.6% (Maritime Continent), 9.3% (Africa) and 4.6% (South America). This indicates an increase in the depth of convection with climate change, which is consistent with an increase in tropopause height. As the reviewer points out, since LNOx is proportional to the  $^{5^{th}/2^{nd}}$  (continental/marine) order of CTH in the PR92 parameterisation, LNOx is sensitive to even small changes in the upper tail of the distribution. Hence, the increase in intensity of convective events is a driver of increasing LNOx with future climate change in our simulations. The largest increases in CTH occur over the Maritime Continent, leading to the largest increases in LNOx over this region (Fig. R1).



Fig. R1. Histograms of CTH over the convectively active regions of the tropics for the Base and  $\Delta$ CC8.5 runs. The regions are defined as in Russo et al. (2011). Qualitatively similar changes are seen in histograms of CTH for  $\Delta$ CC4.5 as for  $\Delta$ CC8.5, relative to Base.

We have obtained a measure of the *frequency* of convection by counting the (area-weighted) number of convective events (defined as the number of non-missing values of CTH in the model output) and dividing by the total possible number of convective events over all timesteps and all grid cells within a selected region. By this crude measure, the frequency of convective events increases in  $\Delta$ CC8.5, relative to Base, by 12.4% and 3.6% over the Maritime Continent and Africa respectively, but decreases by 5.2% over South America. The changes in frequency do not scale simply with the radiative forcing between  $\Delta$ CC4.5 and  $\Delta$ CC8.5, although changes in the intensity do. We have not investigated the mechanisms behind these changes, but this is beyond the scope of this study (see e.g. Chadwick et al., 2013).

Thus, in our simulations, increases in the intensity of convective events is the major driver of increases in LNOx with future climate change. The increase in the frequency of convective events also plays a role over the Maritime Continent, and, to a lesser extent, over Africa. Over South America, the frequency decreases in  $\Delta$ CC8.5. Therefore, the effect of increased intensity on LNOx outweighs the decrease in convective frequency.

We agree that a parameterisation based on updraught speed / mass flux could result in different LNOx changes to one based solely on the PR92 method. Analysis of convective updraught mass fluxes (Fig. R2) shows increases in the climatological mass flux with climate change at most altitudes over Africa and the Maritime Continent, and particularly so for the latter. Hence, a parameterisation based on mass flux would also be expected to lead to increases in LNOx over this

region with climate change. In contrast, such a parameterisation would likely result in decreases in LNOx over South America due to the general decrease in mass flux. This may, of course, depend on the particular details of the parameterisation.



Fig. R2. Climatological updraught mass flux profiles over the convectively active regions of the tropics for the Base and  $\Delta$ CC8.5 simulations.

We have made the following modifications to the text to further explain the changes in convection: P8754 L8 'LNOx is simulated to increase in a year-2100 climate by 33 % (RCP4.5) and 78 % (RCP8.5), primarily as a result of increases in the depth of convection.'

P8761 L5 Taken out ', which reflects changes in convection'

P8762 Inserted paragraph at the end of Sect. 3.1 'Changes in LNOx can result from changes in both the intensity (depth) of individual convective events and the overall frequency of convection. Distributions of convective cloud-top height (CTH) (not shown) indicate a shift towards greater CTH under future climate change. For example, in  $\Delta$ CC8.5, mean CTH increases by 23.6% (Maritime Continent), 9.3% (Africa) and 4.6% (South America) relative to Base, where the regions are defined as in Russo et al. (2011). These increases in the depth of convection are consistent with rising tropopause heights (Fig. 1). Using the number of CTH occurrences as a crude measure of the overall frequency of convective events, we find increases of 12.4% and 3.6% over the Maritime Continent and Africa, respectively, but a decrease of 5.2% over South America in  $\Delta$ CC8.5. Since the PR92 parameterisation for LNOx is highly sensitive to the magnitude of CTH, it is the increases in the depth of convection, scaling with the climate forcing, which primarily lead to increases in LNOx in our simulations. The effect of the parameterisation is highlighted over South America in  $\Delta$ CC8.5, where, although convection occurs less often on average, LNOx still increases due to an increase in the depth of convection. The largest increases in LNOx occur over the Maritime Continent because this region is associated with the largest increases in both the frequency and depth of convection.'

P8766 L4 'We simulate greater LNOx at the year 2100 under two scenarios for future climate change: RCP4.5 and RCP8.5, with LNOx increases of 2 Tg(N)  $yr^{-1}$  (33 %) and 4.7 Tg(N)  $yr^{-1}$  (78 %), respectively, primarily in response to increases in the depth of convection.'

P8766 Removed L11-12 'Nonetheless, our simulated increase in LNOx in a future climate is in qualitative agreement with most of the ACCMIP models.'

Inserted 'Note that we have not explored other LNOx parameterisations and some studies using alternate approaches, such as those based on convective mass fluxes, have found different sensitivities for lightning changes under a warmer climate (e.g. Grewe et al., 2009). However, the PR92 method employed here is commonly adopted in state-of-the-art chemistry-climate models, such as most of the ACCMIP models (Lamarque et al., 2013).'

P8766 L17 Inserted at end of paragraph 'The Maritime Continent is associated with the largest increases in both the overall frequency and depth of convection, which explains the largest increases in LNOx found over this region.'

2a. The paper suggests at several text passages and Figures a linear relationship between lightning NOx, the production of ozone, and the ozone burden. Actually the figures (Fig. 2) and the table 1 clearly shows the saturation of the chemical regime. Only changes in LNOx and changes in P(Ox) are linearly correlated.  $\rightarrow$  dLNOx  $\sim$  dP(Ox). The ozone production (LNOx 6 $\sim$  P(Ox)) is not linear. And even the changes in the ozone burden react in a non-linear way. A 40% saturation is found. This part should be revised properly. (See also comments below).

2b. 8754 Might sound picky, but I think it is important. P(Ox) increases linearly with increases in LNOx. Not: with total LNOx. dP(O3)=a\*dLNOx is ok but certainly not other versions like: P(O3) = a \*LNOx or dP(O3)=a\*LNOx.

Section 3.2 / Fig. 2 Two notes on the linearity of the system:

2c. It could be worth mentioning the non-linearity of the system. E.g. doubling the LNOx from 6 to 12 TgN is not doubling the P(Ox), since it increases from  $\sim$ 4700 Tg/y to  $\sim$ 5700 Tg/y, only. Only the changes are linear.

2d. 8766 "A positive and linear relationship between LNOx and P(Ox) is found" No that is not true, see above. Linear would imply doubling of LNOx doubles P(Ox) that's not true. Only the perturbation is linear.

We believe a 'linear' relationship between P(Ox) and LNOx implies P(Ox) = a\*LNOx + b where b is not necessarily 0. We do not believe it implies *direct* proportionality between the two variables (i.e. b=0). Here, of course, b is not 0 since LNOx is not the only factor contributing to P(Ox). However, to relieve ambiguity in the definition, we have made the following modifications to the manuscript:

P8754 L10 'The total tropospheric chemical odd oxygen production (P(Ox)) increases linearly with increases in total LNOx...'

P8762 L11 '...a highly linear fit between the changes in P(Ox) and LNOx is found.'

P8766 L18 'A positive and linear relationship is simulated between the changes in LNOx and global, tropospheric chemical  $O_x$  production...'

P8767 L2 'The linear relationship between the increases in LNOx and P(Ox)...'

We do not believe it is necessary to point out that Fig. 2a does not pass through the origin (i.e. doubling LNOx does not double P(Ox)), since this would trivially explain that there are factors other than LNOx which drive chemical ozone production. P8762 L8 points out that LNOx is only one driver of P(Ox) in the troposphere and the paragraph starting at P8762 L16 discusses some of the other important influences.

3a. (repeat of comment 2a) The paper suggests at several text pasages and Figures a linear relationship between lightning NOx, the production of ozone, and the ozone burden. Actually the figures (Fig. 2) and the table 1 clearly shows the saturation of the chemical regime. Only changes in LNOx and changes in P(Ox) are linearly correlated.  $\rightarrow$  dLNOx  $\sim$  dP(Ox). The ozone production (LNOx  $6 \sim P(Ox)$ ) is not linear. And even the changes in the ozone burden react in a non-linear way. A 40% saturation is found. This part should be revised properly. (See also comments below).

3b. The ozone burden change seems to react pretty non-linear on the LNOx increase. Fg. 2b: blue line base->CC4.5 gives an increase of 30 TgO3 per 2.04 TgN changes. This rate of change would (linearly) give 70 TgO3 for the run CC8.5, but only 43 TgO3 increase is found, which is already a deviation from lienarity by 40%. A remarkable saturation effect! And this is true for all sets of simulation. I propose not to fit the data in Fig. 2b but to draw lines betwen the individual data points and further discuss this non-linearity in the section.

Due to this apparent saturation effect, which is not evident for P(Ox), we have not asserted anywhere in the text that the ozone burden increases linearly with increases in LNOx (only that the burden does increase). LNOx changes are the dominant driver of changes in P(Ox) with climate change in our simulations; in contrast, several factors are important in driving changes in the burden. The deviation from linearity arises due to the increased dominance of humidity driven losses and non-linear increases in STE (with increases in LNOx). We agree that the linear fits in Fig. 2b could be misleading. We will now show lines connecting the data points. The figure and its caption will be amended accordingly:

Fig. 2 Caption 'Linear fits in (a) and connecting lines in (b) are drawn between runs which differ only in their climate states.'

We have also added the following text to describe the effect:

**P8763 L3** 'In contrast to P(Ox), the changes in ozone burden and LNOx are non-linearly related, since several factors, and not just LNOx, contribute significantly to changes in the burden in a warmer climate. From Fig. 2b, it is also evident that the decrease in burden of  $34 \pm 4 \text{ Tg}(O_3) \text{ yr}^{-1}$  due to  $\Delta O3 \text{ pre...'}$ 

4a. The conclusions are to some extend exaggerated. I do not think that ozone from lightning is one of the key parameters for climate simulations. Ocean, sea-ice, carbon-cycle, feedbacks are key parameters. The tropospheric ozone is only a part in climate simulations.

Our focus is on chemistry-climate interactions. Of course, there are many major factors which affect the climate but the title of our paper, and we believe the text, make it clear that we are not discussing those other processes.

## 4b. 8754 I24 Projections of future climate might be too general. Ozone changes are only contributing by a small part to climate change and LNOx is then a part of that. Still it is important. The conclusion should focus more on future ozone projections. And it is not consistent with the argument given on page 8760 I 20 'Our goal ...'

The conclusions, including the concluding paragraph, primarily assert that changes in climate will impact on chemistry, and not vice versa. We focus on ramifications for future tropospheric oxidising capacity (ozone, OH, methane lifetime), which is consistent with P8760 L20-23. The subsequent

feedbacks onto climate are only postulated as *potentially* important and magnitudes are not given. E.g. radiative forcings associated with changes in tropospheric ozone are not discussed; the radiative feedback of changes in methane concentration is mentioned only qualitatively on P8765 L9-10 and P8767 L25-26. We have amended the abstract to make this clear:

P8754 L22 'We emphasise that it is important to improve our understanding of LNOx in order to gain confidence in model projections of composition change under future climate.'

#### References

Brinkop, S., Aspects of convective activity and extreme events in a transient climate change simulation, Meteorol. Z. 11, 323-333, 2002.

Dahlmann, K., Grewe, V., Ponater, M., Matthes, S., Quantifying The Contributions Of Individual NOx Sources To The Trend In Ozone Radiative Forcing, Atmos. Environm. 45 (17), 2860-2868, DOI: 10.1016/j.atmosenv.2011.02.071, 2011.

Del Genio A. D., M.-S. Yao, J. Jonas, Will moist convection be stronger in a warmer climate?, Geophys. Res. Lett., 34, L16703, doi:10.1029/2007GL030525, 2007.

Grewe, V., Impact of Lightning on Air Chemistry and Climate, In: Lightning: Principles, Instruments and Applications Review of Modern Lightning Research, Betz, Hans Dieter; Schumann, Ulrich; Laroche, Pierre (Eds.), 524-551, Springer Verlag, 2009.

References added to text:

Grewe, V., Impact of Lightning on Air Chemistry and Climate, in: Lightning: Principles, Instruments and Applications, Review of Modern Lightning Research, edited by: Betz, H. D., Schumann, U., Laroche, P., Springer Science+Business Media B. V., 524-551, doi:10.1007/978-1-4020-9079-0\_25, 2009.

Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A. and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, Geosci. Model Dev., 6, 179-206, doi:10.5194/gmd-6-179-2013, 2013.

#### **Responses to Reviewer 2**

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.

Review of 'Lightning NOx, 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 NOx 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 116). 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 NOx 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 LNOx concentration from its emissions in the text; all of the following responses incorporate these modifications.

#### **Specific Comments**

## **1.** p8759 I8: Reference is made to the interactive calculation of LNOx, 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 LNOx 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 I10-11: '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  $\Delta$ CC8.5(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 NOx, ozone and OH from LNOx in the upper troposphere, where NOx 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 LNOx 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 NOx lifetimes here are short and other sources dominate the total NOx 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 LNOx, 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 LNOx, concentrations of tropospheric trace constituents are also particularly sensitive to the vertical distribution of LNOx. 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 LNOx distributions, such as those suggested by Pickering et al. (1998) and Ott et al. (2010), UKCA distributes LNOx 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 LNOx, relative to these distributions. In the lower troposphere and the boundary layer, where NOx lifetimes are short, trace gas concentrations are far less sensitive to LNOx (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 NOx 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 LNOx. Our results are dependent on the LNOx and convective parameterisations utilised. In particular, the vertical profile of LNOx 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 LNOx 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 LNOx.'

5. Section 3.1/p8762 l8/throughout: In the Introduction, you define LNOx as 'NOx produced from lightning'. So I think LNOx refers to the fraction of NOx derived from lightning, or 'lightning NOx' for short. In other places you refer to 'LNOx emissions'. But in other places, you just use LNOx with the implication that you mean LNOx 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 NOx vs. NOx emissions, so I think the same applies to LNOx vs. LNOx emissions. For example, the text (and figure caption) describes Figure 2a as showing the correlation of P(Ox) and LNOx, but the figure actually shows LNOx emissions. If every reference to LNOx 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 NOx from lightning (LNOx)...'

P8755 L3 'Emissions of NO<sub>x</sub> from lightning (LNOx) 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 LNOx.'

P8755 L11 'NOx from lightning induces production of ozone...'

P8755 L19 'In addition, formation of HOx can be induced indirectly,...'

P8759 L18 'LNOx...'

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

P8761 L15 'With regard to its geographical distribution, LNOx occurs predominantly...'

P8762 L3 '...may result in systematic biases in LNOx over this region.'

**P8765 L26** 'We have assessed the impacts of climate change on emissions of NOx from lightning (LNOx) and the consequences for tropospheric chemistry using UM-UKCA. Using the Price and Rind (1992, 1994a) parameterisation for calculation of LNOx, our year-2000 integrations generate 6 Tg(N)  $yr^{-1}$  of total, global LNOx, which lies...'

**P8767** L14 'An analysis of reaction fluxes indicates that the dominant reaction pathways for increased OH production through LNOx in these regions are  $HO_2 + NO$  (directly, following production of NOx) and  $O(^{1}D) + H_2O$  (indirectly, through photochemical ozone and hence  $O(^{1}D)$  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 LNOx...'

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 LNOx within the tropics.'

### 8. p8765 I6-8 Better to use years (rather than months), the 'normal' unit used for methane lifetime.

Have changed units to years throughout the text.

#### 9. p8765 I9 '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  $34 \pm 4 \text{ Tg}(O_3)$  due to  $\Delta O3$  pre is just outweighed by the increase in  $\Delta(CC8.5 + O3$  pre), 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 ( $\tau_{CH4+OH}$ ) 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  $\tau_{CH4+OH}$  due to climate change.

Changes in  $\tau_{CH4+OH}$  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 (Fuglestvedt 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( $O_3$ ) (RCP4.5) and 13 Tg( $O_3$ ) (RCP8.5). The corresponding increases in ozone burden relative to Base are 24  $Tg(O_3)$  (RCP4.5) and 33  $Tg(O_3)$ (RCP8.5), which still represent substantial increases with future climate change and greater LNOx. The adjusted increase in burden in  $\Delta$ (CC8.5+O3pre) (33 Tg(O<sub>3</sub>)) is now more comparable to the adjusted decrease in  $\triangle O3$  pre (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  $\tau_{CH4+OH}$  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.

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