We are grateful to Reviewer 2 for their thoughtful comments. We provide our responses below in blue. Line and page numbers refer to the track changed manuscript.

Please note that in the process of reviewing this manuscript, an error was corrected in the radiative forcing calculations. The implications for the results are minor: the differences for whole-atmosphere, stratospheric and tropospheric RFs are less than  $0.02 \text{ W m}^{-2}$  in magnitude. The figures, tables and text (highlighted in yellow) in the revised manuscript have all been updated to reflect the corrected calculations.

## General comments:

I find the paper by Banerjee et al. original, clear and very well-written, and it fits well into the scope of ACP. The paper builds on previous work in Banerjee et al. (2016), but takes it one step further by quantifying radiative forcing. Although the results are based only on a single model, the paper is original in the sense that detailed chemistry is included both for the troposphere and stratosphere, and the fact that several chemical/climatic drivers are studied. I recommend acceptance of the paper, but I also have some comments/concerns that need to be addressed first. Please see specific comments below.

## Specific comments:

Page 1, line 15: Since RCP8.5 is considered rather extreme, it would be interesting, if possible, to have an estimate for O3 RF due to methane also for the RCP4.5 scenario. Do you expect the results from the methane perturbation experiment for RCP8.5 to be relatively linear, so that you can approximate the O3 RF due to RCP4.5 methane by scaling down the results from that experiment?

Previous studies suggest that there is a small non-linearity in the response of tropospheric ozone to changing  $CH_4$  abundance (Wild, 2007) but a fairly linear response of stratospheric ozone (Revell et al., 2012). These studies did not determine the associated linearity or lack thereof in ozone RF; for the relatively small RF values we are considering, we suspect a fairly linear relationship. However, we are unable to perform any further integrations at this stage to test this.

Page 2, line 29: For comparison, it would be useful to mention the forcing in 2000 from Stevenson et al.

We have added the forcing in 2000 (and have removed the rounding of their figures) on P2L32:

...suggests a tropospheric ozone RF of  $-0.033 \pm 0.042$  W\_m<sup>-2</sup> (multi-model mean  $\pm 1\sigma$ ) due to climate change up to 2100 under the RCP8.5 scenario, which is a negligible change from the forcing in <u>the</u> year 2000 of  $-0.024 \pm 0.027$  W m<sup>-2</sup> (both relative to 1850) (Stevenson et al., 2013).

Page 3, line 10-12: It is mentioned that there are previous studies on either tropospheric or stratospheric ozone RF. I would like to see some comparison in the Results section on how the results of those studies compare to the results obtained in this paper.

We have already compared our results to previous studies in the following instances: a qualitative similarity in the stratospheric RF between our  $\Delta$ CC8.5 experiment and 4xCO2 scenarios (P12L10), a quantitative comparison of the future tropospheric RF between our climate change experiments and the multi-model results in Stevenson et al. (2013) (P13L11), the cancellation between the stratospheric SW and LW forcings in scenarios of ODS changes Arblaster et al. (2014) (P14L18). We have now added a comparison of our  $\Delta$ CH4 results to Portmann and Solomon (2007):

P17L13: Around a third <u>A</u> small fraction (~15%) of the whole atmosphere RF is due to the stratospheric ozone RF ( $\frac{0.050.03}{0.050.03}$  W\_m<sup>-2</sup>, Fig. 2Fig. 1), which is the same as the estimate of 0.03 W m<sup>-2</sup> in Portmann and Solomon (2007) for the same CH<sub>4</sub> increase.

P15L27: <u>As in  $\triangle$ ODS</u>, there might also be some contribution of stratospheric ozone changes to tropospheric changes through stratosphere to troposphere transport of air containing higher ozone amounts. Our estimate of the whole-atmosphere CH<sub>4</sub>-driven ozone RF (0.18 W m<sup>-2</sup>) is greater than the previous estimate of 0.13 W m<sup>-2</sup> in Portmann and Solomon (2007) for the same CH<sub>4</sub> increase. The difference is due to the larger tropospheric RF (0.15 versus 0.10 W m<sup>-2</sup>); note that they did not directly diagnose the tropospheric RF due to the simplicitly of their tropospheric chemistry scheme, which could explain the difference.

Other studies of the ozone RF have focused on the *historical* rather than the *future* RF, so it is difficult to make a like-to-like comparison. In P3L13, we have inserted the references of Portmann and Solomon (2007) (who assess drivers of future stratospheric ozone RF) and Stevenson et al. (2013) (who assess future tropospheric ozone RF) to highlight the comparisons we aim to make.

Page 4, line 12: Is 10 years spin-up enough for the ODS simulation, considering that the ODSs are only perturbed at the surface?

In the  $\triangle ODS$  and  $\triangle CH4$  experiments, initial conditions of ODSs and  $CH_4$ , respectively, were also perturbed in order to reduce the required spin up time. Moreover, the mean age of stratospheric air is relatively short in this model (up to 4 years), so a 10-year spin up period is enough for stratospheric concentrations to reach steady-state. This was confirmed by checking the time series of long lived tracers (ODSs,  $CH_4$  and  $N_2O$ ) at various latitudes and altitudes. We have added:

P4L11: The initial atmospheric concentrations of ODSs and  $CH_4$  were also perturbed by the same factor in  $\Delta ODS$  and  $\Delta CH4$ , respectively, in order to reduce spin up time.

P4L24: <u>It was confirmed that this spin up period was long enough for stratospheric concentrations of perturbed gases to reach steady state.</u>

Page 5, line 3-5: I assume the tropopause height is higher in the climate perturbation experiments (especially in the RCP8.5). Perhaps I misunderstand something, but if the tropopause height is the same in all RF calculations, wouldn't that lead to a wrong split between tropospheric and stratospheric contribution to O3 RF?

There are advantages and disadvantages of employing a fixed tropopause height in the RF calculations. The advantage is that it facilitates a like-to-like comparison with previous studies that have made the same choice (Dietmüller et al., 2014; Nowack et al., 2014; Stevenson et al., 2013). A fixed tropopause height also maintains the same mass of air in the troposphere and stratosphere so that attribution of the ozone RF (to its changing concentration/distribution) is not confounded by changing air mass. However, a fixed tropopause does not consider the changing split between stratospheric and tropospheric ozone, as the reviewer points out. We have investigated impacts of a rising tropopause under climate change, and find only small effects, which we highlight in the following instances:

Table 2: added two rows ( $\Delta$ CC4.5(trophgt) and  $\Delta$ CC8.5(trophgt)).

P1L22: <u>Considering the increases in tropopause height under climate change causes only small</u> differences ( $\leq |0.02| \text{ W m}^{-2}$ ) for the stratospheric, tropospheric and whole-atmosphere RFs.

P5L22: In the climate change experiments,  $\Delta$ CC4.5 and  $\Delta$ CC8.5, the tropopause rises; the ramifications for employing a climate-consistent tropopause height for the ozone RF will be shown to be small (see Sect. 3.1).

P14L30: Finally, we note that, in order to maintain consistency with previous studies (Dietmüller et al., 2014; Nowack et al., 2014; Stevenson et al., 2013), the values of the ozone RF discussed thus far do not consider the effect of the increase in tropopause height under climate change. We calculate that employing climate consistent tropopause heights causes only small differences ( $\leq |0.02|$  W m<sup>-2</sup>) for the stratospheric, tropospheric and whole-atmosphere RFs (Table 2).

P19L27: Increases in tropopause height under climate change have a negligible ( $\leq |0.2| \text{ W m}^{-2}$ ) impact on ozone RFs under both the scenarios of climate change considered here.

Page 5, line 29: Figure 1 is not really discussed before page 9, after the discussion of Figs. 2 and 3. I suggest to change the order of the figures to reflect the order in which they are discussed.

We have changed the order of the figures such that Figure 1 shows the total ozone RFs, Figure 2 shows the LW and SW RF components, then Figure 3 shows the vertical ozone profiles.

Page 6, line 7: Not all cases show ozone RFs <0.1 W m-2. The methane case is ~0.2 W m-2.

We thank both reviewers for pointing this out. Even considering the  $\Delta$ CH4 experiment, the wholeatmosphere ozone RFs are small compared to the direct RF from WMGHGs. Hence, we have only modified the sentence on P7L1 slightly:

... the whole-atmosphere ozone RFs are small ( $\leq 0.12$  W\_m<sup>-2</sup>) ...

Figure 3 caption: "d.p." - I assume this means "decimal points". Is that a common abbreviation?

We think the figure is clearer without the rounding so have updated the figure and removed the abbreviation.

Page 9, line 6-7: Could the ozone reduction in the tropical lower stratosphere be related to a higher tropopause in RCP8.5?

Yes, a part of this ozone reduction will be related to a higher tropopause, though the impact is difficult to separate from the effects of strengthening tropical lower stratospheric upwelling. We have included a qualitative note (P12L8):

; this is driven by an increase in the upwelling mass flux by 27%, with an additional contribution from a higher tropopause also being likely.

Page 9, line 17: On page 2, line 28 it states that Stevenson et al. got a value of -0.03 + -0.04 W m-2 due to climate change up to 2100 under RCP8.5. Any idea why the value calculated here is so much higher (0.08 W m-2) and well outside their uncertainty range?

-0.03 W m<sup>-2</sup> is the ozone RF due to climate change between 1850-2100. We discuss in P13L12 that the RF between 2000-2100 (RCP8.5) can be calculated from Table 2 in Stevenson et al. (2013) as ~0.01 W m<sup>-2</sup> with an inter-model range of  $\pm 0.07$  W m<sup>-2</sup>. Our calculated result of 0.07 W m<sup>-2</sup> lies on the upper bound of this inter-model range, and could be due to a larger sensitivity of LNO<sub>x</sub> to surface temperature in our model. We have inserted (P13L16):

<u>Our value of 0.07 W m<sup>-2</sup> is on the upper end of the inter-model range and could reflect a particularly</u> large sensitivity of  $LNO_x$  to climate in our model: 0.96 Tg(N) yr<sup>-1</sup> K<sup>-1</sup> (Banerjee et al., 2014) compared to a multi-model mean of 0.37 ± 0.06 Tg(N) yr<sup>-1</sup> K<sup>-1</sup> for the same 8 CCMs discussed above (calculated using Table S2 of Finney et al. (2016)).

Page 10, line 1: Since the tropopause definition is the same in all RF calculations, wouldn't the tropospheric and stratospheric contributions be incomparable between the RCP8.5 and RCP4.5 experiments (see also earlier comment)?

Please see response to earlier comment.

Page 11, line 3: Table 2 says 0.02 and not 0.03 W m-2 DU-1.

We have updated both instances with the revised and more precise values of  $0.035 \text{ W m}^{-2}$ .

Page 13, line 26-27: The O3 RF from the CH4 experiment is greater in JJA both in the southern and northern hemisphere. In the southern hemisphere, I would expect the photochemical ozone production to be lower during JJA than DJF?

On increasing methane, the pattern of tropospheric column ozone increase in the SH resembles the climatological seasonal cycle. The higher column ozone (and its increases in the  $\Delta$ CH4 experiment) just south of the Equator in JJA is likely due to greater interhemispheric transport from the NH (since the bulk of ozone production occurs in JJA in the NH). We have added this suggestion (P17L17):

As with  $\Delta O3$  pre, the largest RFs are found in JJA<u>in the NH</u> due to greater photochemical ozone production, and an ozone increase, during this season; this likely dominates background ozone concentrations and causes a slightly larger ozone increase (and associated RF) in the SH during JJA than during DJF.

Page 15, line 9-12: On page 6, line 8, RF values for WMGHG are 3 and 6 W m-2 for RCP4.5 and RCP8.5, respectively, and with a reference to Myhre et al. (2013). Here it is given as 2 and 6 W m-2 with a reference to van Vuuren et al. (2011). Would be good to be consistent.

We thank the reviewer for pointing this out. The correct values are 2 and 6 W m-2 as shown by Fig. 10 in van Vuuren et al. (2011). We have amended P7L2:

(roughly 32 and 6 W<sub>m<sup>-2</sup></sub> for RCP4.5 and RCP8.5, respectively<u>, as shown by Fig. 10 in -(Myhre et al., 2013)</u>van Vuuren et al. (2011)).

Page 15, line 16-17: Is it possible to say something about how important future N2O changes may be for O3 RF, based on, if available, any estimates/indications in the literature? Would be good, if possible, to discuss the importance of this effect relative to the effects explored in the paper.

The final line of the manuscript mentions Portmann and Solomon (2007), which, to our knowledge, is the only study that has calculated the indirect RF of  $N_2O$  through ozone. We have expanded this discussion (P21L16):

The contribution of this effect to future ozone RF over the 21<sup>st</sup> century may also be important <u>To our</u> knowledge, only one study to date has investigated the indirect RF of N<sub>2</sub>O through ozone (Portmann and Solomon, 2007). Using a 2D model, this study calculated a stratospheric ozone RF of 0.026 W m<sup>2</sup> and a whole-atmosphere RF of 0.038 W m<sup>-2</sup> associated with a 150 ppbv increase in N<sub>2</sub>O between 2000 and 2100. This whole-atmosphere ozone RF is smaller than found for any of the perturbations in

our study. Nonetheless, the ozone response to increased  $N_2O$  and its associated RF could be better quantified in future studies using 3D chemistry-climate modelsand warrants future investigation.

Technical corrections:

Page 1, line 12: "Wm-2" should be "W m-2". Please correct throughout the manuscript.

Corrected.

Figure 1 caption: Degree signs are missing from e.g., "90S-90N". Also, I cannot see that "SH" and "NH" have been defined.

Corrected.

Page 10, line 25: Please fix parenthesis for the reference.

Corrected.