

We thank Reviewer 1 for their positive judgement of the manuscript and their constructive 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.

I judge this to be well-written and original paper on an important issue, which represents a significant advance in understanding of the future drivers of ozone change in both the troposphere and stratosphere. I recommend acceptance after relatively minor modification. My more important comments are indicated with a M

1:12 “tropospheric ozone precursor” – this is ambiguous, as it needs to be made clear this excludes methane (the ambiguity is emphasised by line 1:26 referring to methane as a tropospheric ozone precursor, and it also being a important result in this paper that methane is a stratospheric ozone precursor)

We agree that this should be clarified. We have changed the phrase on P1L12 to 'non-methane tropospheric ozone precursor'.

1:14, 2:5, 11:1 and elsewhere: The paper would be helped if it could be made clear when (for example) increases due to strat-trop exchange are due to there being more ozone to transport, rather than more advection doing the transport. Perhaps a terminology could be proposed that distinguishes the two?

We have only mentioned stratosphere-troposphere exchange (STE) a few times and so introducing new terminology might cause confusion. Instead, we simply add a clarification in each instance of why STE is changing:

P1L14: ... which is ~~mainly~~ driven by an increase in tropospheric ozone through stratosphere-to-troposphere ~~exchange~~transport of air containing higher ozone amounts.

P13L1: ... by an increase in STE ~~that is caused by a strengthened stratospheric circulation~~, ...

P14L12: The importance of the ~~stratospheric ozone changes for RF~~stratosphere in this experiment is found instead in ~~the enhancing of~~STE by virtue of there being more stratospheric ozone available for transport; ~~which this~~ is the primary driver of changes in tropospheric ozone in the middle and high latitudes (Fig. 1; Banerjee et al. (2016)).

P19L18: ~~;~~ ~~which~~ ~~This RF~~ mainly arises from increases in tropospheric ozone driven by ~~increased~~ stratosphere-to-troposphere ~~exchange~~transport of air containing higher ozone concentrations.

2:7-8 It is unclear (and indeed it may be unclear in Myhre et al.) whether the forcings on line 1:30 assign all the ODS forcing to stratospheric ozone and all the ozone precursor forcing to tropospheric ozone. I feel that one important result in this paper is that there may be a need for some better terminology to capture these effects.

Myhre et al. (2013) do not assign all the ODS forcing to stratospheric ozone and all the ozone precursor forcing to tropospheric ozone, and they do recognize their remote effects. We have clarified on P2L4:

The emission-based estimates of historical ozone RF in Myhre et al. (2013) include the effects of changes in both stratospheric and tropospheric ozone.

We agree that careful terminology is required in all future studies. Indeed the remote effects of ODSs and ozone precursors on ozone RF are not clear in any of the figures in Myhre et al. (2013). We have inserted on P19L31:

We recommend that future studies of ozone RF aim to attribute total (stratospheric + tropospheric) ozone RF to particular emissions and further separate this into stratospheric and tropospheric components, with the use of careful terminology. For example, we recommend the emissions-based view of RF in Fig. 8.17 of Myhre et al. (2013) that shows the total ozone RF for each emission ('O<sub>3</sub>' bars), but with an additional quantification of 'O<sub>3</sub>(strat)' and 'O<sub>3</sub>(trop)' in each case.

2:20 and in addition, the role of NO<sub>x</sub> in forming nitrate aerosols (see e.g. Myhre et al)

We have mentioned this briefly on P2L18:

However, ~~an there are~~ added complications ~~is the potential for of~~ further climate impacts through changes in concentrations of nitrate aerosol and changes in concentrations of the hydroxyl (OH) radical (Myhre et al., 2013); ~~only the latter effect is explored in this study~~. Changes in OH concentration which perturb ...

M3:3 -3:16 I feel there needs to be more of a discussion about what is left out. It seems no aerosol forcing is included in the simulations (at least, it is not mentioned) and a more major issue that emerges later is that the authors have had to make a methodological choice – most notably the methane perturbations calculations are performed at present-day ODS concentrations, which might significantly impact the results. Although this is flagged later in the paper, I feel it is a major restriction that needs raising earlier, and returning to in the conclusions.

We have added the following discussions:

P4L17: There are some forcings and interactions that we do not consider in this study. Firstly, our focus lies on estimating the future ozone RF from emitted gases. We do not simulate any associated aerosol forcing, with aerosol precursor emissions and their oxidant fields being held fixed in all simulations (following the scheme of Bellouin et al. (2011)). Secondly, the 'snapshot' experiments of this study do not consider various transient interactions. For example, the background conditions of NO<sub>x</sub> and ODSs affect CH<sub>4</sub> concentrations, but this coupling is not considered when perturbing NO<sub>x</sub>, ODSs and CH<sub>4</sub> individually in the ΔO<sub>3</sub>pre, ΔODS and ΔCH<sub>4</sub> experiments (potential consequences for the CH<sub>4</sub>-induced ozone RF are, however, discussed in Sect. 3.4).

P19L15: We also note that the ozone response to increasing CH<sub>4</sub> will likely vary over time as the background conditions (e.g. NO<sub>x</sub> and ODS loadings) change: these impacts have not been simulated in the time-slice experiments of this study and warrant future investigation.

M4:1-2 “surface concentrations”. I struggled to understand this. If, in the ODS and CH<sub>4</sub> experiments, it is the surface concentrations that are perturbed, does this mean that the perturbation has then to propagate through the atmosphere by advection? If this is the case, given the age of air in the

stratosphere is several years, a 10-year integration (line 4-13) is hardly long enough for the perturbation to impose itself (especially as the results seem to be averaged over this 10 year period). I feel sure I am misunderstanding here, and some improved clarity should help.

Each integration is 20 years long consisting of a 10-year spin up and 10-year analysis period (P4L24). In the  $\Delta$ ODS and  $\Delta$ CH<sub>4</sub> experiments, initial conditions of ODSs and CH<sub>4</sub>, 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<sub>4</sub> and N<sub>2</sub>O) at various latitudes and altitudes. We have added:

P4L11: The initial atmospheric concentrations of ODSs and CH<sub>4</sub> were also perturbed by the same factor in  $\Delta$ ODS and  $\Delta$ CH<sub>4</sub>, respectively, in order to reduce spin up time.

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

4:20 Stevenson et al. (2013) indicate that the ozone radiative forcing is significantly dependent on the spectral file used in the Edwards and Slingo code. Since this radiative forcing plays such an important role in this paper, it would be good practice, perhaps in the Supplementary, to be specific as to what spectral file is used here. There may be further details of version numbers in the UM-UKCA that could be usefully documented at the same time

The names of the spectral files used in the RTM are for LW: spec3a\_lw\_hadgem1\_wz\_spec and for SW: spec3a\_sw\_hgem1\_in6e\_mean\_spec. We have added this as a footnote on Page 5.

Table 1: Somewhere it may be good to spell out what makes up the WMGHGs (again in the Supplementary?). Some/all of the ODS are part of this? And in deltaO3pre, is the biomass burning assumed to be non-anthropogenic, as that is the implication of the label.

Some (but not all) of the ODSs are radiatively active. The long-lived CFCs (CFC-11 and CFC-12) are WMGHGs and are thus included in this definition. We have added the following sentence to P4L2 and Table 1's caption: "Here, the WMGHGs considered are CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, CFCs, HCFCs and HFCs."

The Supplementary Material only contains Table S1, which pertains to methane feedbacks, so we do not feel a description of WMGHGs here is appropriate.

Despite biomass burning being largely of human-induced origin, it is conventionally considered as separate from anthropogenic emissions (from the combustion of fossil fuels). We follow the IPCC AR5 / ACCMIP definition in Lamarque et al. (2010): "...anthropogenic (defined here as originating from energy use in stationary and mobile sources, industrial processes, domestic and agricultural activities) and open biomass burning emissions.". We have referenced this paper in P4L5.

6-1: Since only adjusted forcings are presented (which is perfectly fine) it may be worth a note that some of the adjusted LW forcing is due to the SW-driven temperature changes – so the separation between SW and LW is not always a completely clean one.

The effect of SW-driven temperature changes is well known to be an important contribution to the adjusted LW forcing for changes in stratospheric ozone. We have mentioned this on P5L18:

The stratospheric temperature adjustment strongly affects the calculated LW (and hence total) RF for stratospheric ozone changes, with the adjustment being largest where the SW-driven temperature changes are largest (Forster and Shine, 1997).

6-7: “all” – this does not seem to be the case for dCH<sub>4</sub> according to the table.

We thank both reviewers for pointing this out. Even considering the ΔCH<sub>4</sub> experiment, the whole-atmosphere 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 ( $\ll 0.12 \text{ W}_m^{-2}$ ) ...

6-7: although not essential, adding the total column ozone change would be useful for this table.

We do not discuss total column ozone changes and so we would prefer to omit these values and avoid unnecessary clutter in the table.

6-14: Without going to the other paper, it is not clear what the equivalence is. Is it forcing equivalence, or stratospheric- temperature-change equivalence?

We have clarified the definition of Carbon Dioxide Equivalent; please see the amended paragraph under the next comment.

9-11: A minor point, but the “which is driven” part of this sentence might be better at the end of the sentence on line 7, where the ozone reduction is first mentioned (it would also shorten this long sentence).

We have updated the paragraph beginning P12L5 to improve coherency:

The difference between the two scenarios arises mainly from the stratospheric ozone RF, which is less negative in ΔCC4.5 ( $-0.04 \text{ W}_m^{-2}$ ) than in ΔCC8.5 ( $-0.15 \text{ W}_m^{-2}$ ) (Fig. 2 Fig. 1, Table 2). Fig. 3 Fig. 2a further shows that this difference stems from the LW, rather than the SW, contribution to RF. As Sect. 4 will discuss, the stratospheric LW contribution to RF in ΔCC8.5 is dominated by the effects of a reduction in ozone in the tropical lower stratosphere (Fig. 4 Fig. 3b); this is driven by an increase in the upwelling mass flux by 27%, with an additional contribution from a higher tropopause also being likely. Qualitatively similar conclusions have been drawn for larger 4xCO<sub>2</sub> perturbation experiments (Dietmüller et al., 2014; Nowack et al., 2014). In contrast, ΔCC4.5 shows a small positive stratospheric LW RF (Fig. 3 2a), ~~which~~ This can partly be explained by more comparable changes in tropical lower stratospheric ozone (driven by an increase in the upwelling mass flux by 10%) and upper stratospheric ozone (Fig. 4 Fig. 3b). Indeed, in a related study focusing on tropical column ozone (Keeble et al., 2017), we find that the change in lower stratospheric ozone, ~~which is driven by increases in the tropical upwelling mass flux (by 10 and 27% in ΔCC4.5 and ΔCC8.5, respectively),~~ scales more strongly with GHG concentration (~~0.03 DU per ppmv of Carbon Dioxide Equivalent (CDE)~~) than the change in upper stratospheric ozone, which is driven by cooling from CO<sub>2</sub> (0.02 DU ppmv(CDE)<sup>-1</sup>): 0.03 versus 0.02 DU per ppmv of CO<sub>2</sub>-equivalent, where CO<sub>2</sub>-equivalent is the concentration of CO<sub>2</sub> that would cause the same RF as the mixture of all GHGs.

10-21 This sentence implies that all halocarbons are ODS’s (as otherwise what is the point of comparing them?). I might guess that a significant fraction of the 2000-2100 halocarbon forcing is from non-ODSs.

This is a good point: the HFCs are greenhouse gases but are not ODSs, so we have modified the comparison (P13L31):

This offsets around half-a quarter of the estimated direct RF of the ozone-depleting halocarbons between 2000-2100 under RCP4.5, which we estimate to be around  $-0.22 \text{ W m}^{-2}$  as the difference between the total halocarbon forcing ( $-0.15 \text{ W m}^{-2}$ ) between 2000-2100 under RCP4.5 (Meinshausen et al., 2011) and the non-ODS halocarbon (HFC) forcing (around  $+0.07 \text{ W m}^{-2}$  from Fig. 1 of Xu et al. (2013)).

11:3 “0.03” – the table says 0.02

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

11:18-20 I was not sure what the logic of adding ODS and dO3Pre (but excluding CH<sub>4</sub>) was. What point was trying to be made?

The effects of  $\Delta\text{O}_3\text{pre}$  and  $\Delta\text{CH}_4$  have often been compared and contrasted within the literature [West et al., 2007; Stevenson et al., 2013]. Thus, it is recognised that the climate penalty from future increases in CH<sub>4</sub> would negate the climate benefits from reductions in non-methane ozone precursor emissions. Here, we wish to highlight the additional competing effect of ODS reductions (albeit a smaller effect than CH<sub>4</sub> increases) that has previously been overlooked. We have clarified this reasoning on P14L30:

The ozone-derived climate effects of changes in non-methane ozone precursor emissions and CH<sub>4</sub> have often been compared (e.g. Stevenson et al., 2013; West et al., 2007). Indeed, we find in the next subsection that future increases in CH<sub>4</sub> abundance would negate the climate benefits of reductions in non-methane ozone precursor emissions. However, we here emphasise that these benefits could also be negated by future reductions in ODSs, which has previously not been noted: In comparison to the results for the  $\Delta\text{ODS}$  experiment discussed in Sect. 3.2, the whole-atmosphere ozone RF in  $\Delta\text{O}_3\text{pre}$   $\Delta\text{ODS}$  is over half the magnitude of the RF in  $\Delta\text{O}_3\text{pre}$  is similar in magnitude but opposite in sign (Fig. 2Fig. 1, Table 2) indicating that the combination of these perturbations would result in a smaller net ozone RF. This is an important point since the ozone derived climate benefits of reductions in non-methane ozone precursor emissions that have been highlighted in previous studies (e.g. Naik et al., 2005) could be negated by future decreases in ODSs. These climate benefits could be further negated under future increases in the abundance of CH<sub>4</sub>; this possibility is now explored.

11:32 (and 1: 16) A minor query about the “a third” – in the table it is (0.05/0.19) nearer a quarter, although the third may be consistent with the fraction prior to rounding.

The revised values show a smaller relative contribution of stratospheric ozone RF. We have amended the following instances:

P1L17: A third small fraction (~15%) of the ozone RF due to the projected increase in methane results from increases in stratospheric ozone.

P15L15: Around a third A small fraction (~15%) of the whole atmosphere RF is due to the stratospheric ozone RF ( $0.050.03 \text{ W m}^{-2}$ , Fig. 2Fig. 1), ...

M12:15-17 As noted above, this is a major caveat which I think requires more flagging earlier in the paper and in the conclusions. It might help the discussion if it could be stated clearly how different the chlorine loading is between 2000 and 2100.

This has been addressed in a previous comment. In addition, we have included the numerical changes in ODS boundary concentrations (and other species) in the caption of Table 1.

14:16-21 It is worth adding that this estimate of the methane effect is without the climate-change induced component of the ozone change resulting from CH<sub>4</sub> increase (which I guess may be more like the dCC4.5 case, as methane won't strongly impact on upper stratospheric temperatures) and so the methane component could be even larger.

The direct impact of increased CH<sub>4</sub> on stratospheric temperatures would likely reduce the total CH<sub>4</sub>-driven ozone RF: a cooling of the upper stratosphere would induce an increase in ozone and a reduction in downwelling SW radiation. We have added a qualitative statement to this effect in P19L12:

Note that the imposed changes in CH<sub>4</sub> are uncoupled from the radiation scheme and so do not, by design, affect atmospheric temperatures. The overall effect of an increase in CH<sub>4</sub> abundance would include a cooling of the upper stratosphere that induces an ozone increase, which we suggest might reduce the SW and total ozone RF. This component of the CH<sub>4</sub>-driven ozone RF is here instead included in the ΔCC8.5 simulation.