

## RESPONSE TO REVIEWER #2

The authors are very grateful to Reviewer 2 for their positive comments and suggestions. The original review (black) and our responses (blue) are provided below. Additions/updates to the text are given in green. Page/line numbers in our responses refer to the track changed manuscript.

The authors explore the roles of future climate change, changes in ozone-depleting substances (ODSs), and reductions in non-methane ozone precursor emissions in both stratospheric and tropospheric ozone changes, using a global chemistry-climate model comprising both stratospheric and tropospheric chemistry. They also carry out an analysis on associated changes in ozone chemical budget terms. The paper is well written and the analysis is quite thorough. The paper is within the scope of ACP. It should be accepted after the authors have addressed the following comments.

Specific comments: Why do you choose not to include methane changes in the RCP8.5 scenario simulation? It would be an interesting perspective to see how much impact such a significant increase of methane would have on both stratospheric and tropospheric ozone. Maybe you could elaborate on this in your introduction.

By the "RCP8.5 simulation", we assume the reviewer means the  $\Delta$ CC8.5 simulation. In this simulation, we aim to isolate *the radiative impacts* of changes in WMGHGs on ozone i.e. a climate change signal. If we were to run a simulation under *full* RCP8.5 forcings, we would have included methane changes within the chemistry scheme.

Moreover, we wished to supplement the relatively few CCM studies that explore ozone changes in the RCP8.5 scenario *without* the assumption of a more than doubling of methane abundance, which carries high uncertainty and can swamp the effects of other drivers of ozone change (e.g. Revell et al., 2015; Young et al., 2013). We have removed part of this reasoning from P3, L4-5 and clarified fully on P4, L14-21: "Note that future methane emissions are highly uncertain and changes in its abundance, particularly at RCP8.5, will likely have large tropospheric and stratospheric impacts (Randeniya et al., 2002; Fleming et al., 2011; Revell et al., 2012, 2015; Young et al., 2013) that are not the focus of this study. Instead, we wish to isolate other drivers of ozone changes, in particular, the role of a change in mean climate state at RCP8.5, without the assumption of a large increase in methane abundance. Hence, the methane boundary condition is kept fixed in all sensitivity tests, although its radiative forcing effect is included in future changes to climate."

To complement the set of process-based experiments described in this study, we do have additional simulations that perturb methane to year 2100 RCP8.5 levels *only within the chemistry scheme*. This perturbation has been applied individually, combined with climate change and with reduced ODSs; the results will be described in a follow-up study.

P30648 line 3: You should state that the purpose of using  $O_x$  is to account for the chemical cycling of the species in this family of  $O_x$ , and  $O_3$  is the most abundant member of this family. The  $O_x$  family should also be defined here. Do you also express ozone dry deposition in the format of  $O_x$ ? How much is  $O_3$  deposition if that's the case (in Table 2)?

We have removed (P3, L9-10): "...or odd oxygen ( $O_x$  = species which rapidly interconvert with ozone)..." and inserted (P3, L11-15): "In practice, many studies calculate the budget of odd oxygen ( $O_x$ ) to account for species that rapidly interconvert with ozone. In this study,  $O_x$  is defined as the sum of ozone,  $O(^3P)$ ,  $O(^1D)$ ,  $NO_2$ ,  $2NO_3$ ,  $3N_2O_5$ ,  $HNO_3$ ,  $HNO_4$ , peroxyacetyl nitrate (PAN), peroxypropionyl nitrate (PPAN) and peroxyethacrylic nitric anhydride (MPAN). Although the exact definition varies between studies, in any case, ozone represents the majority of  $O_x$ ."

We have removed the above definition of  $O_x$  from the caption of Table 2 and reminded the reader that it is defined in the Introduction: "The definition of  $O_x$  employed here is given in the Introduction."

For consistency with  $P(O_x)$  and  $L(O_x)$ , deposition does account for all  $O_x$  species, and thus includes deposition of  $NO_y$  species as well as ozone dry deposition (hence the abbreviation to  $D(O_x)$ ). In the Base run, ozone dry deposition is  $871 \text{ Tg}(O_3) \text{ yr}^{-1}$ , which accounts for 86% of total  $O_x$  deposition; this fraction does not differ substantially between the experiments (84-90%). We thank the reviewer for raising this point since it should be highlighted that ACCENT and ACCMIP reported only ozone, and not  $O_x$ , deposition. P12, L30 - P13, L2 now read: "Chemical production ( $P(O_x)$ ), loss ( $L(O_x)$ ) and deposition are well within  $1\sigma$  of the multi-model means; we compare the dry deposition of ozone here (see Table 2) but consider deposition of all  $O_x$  ( $D(O_x)$ ) hereafter."

We have added values for the dry deposition of ozone in brackets in Table 2 within the  $D(O_x)$  column, and inserted into the caption: "Note that in this study, the  $D(O_x)$  term totals dry deposition of ozone (listed in brackets) plus deposition of those nitrogen compounds that are classed as  $O_x$ , whereas the multi-model mean values report only the former. The same applies in the calculation of  $\tau_{O_3}$ ."

We have updated Table 2 similarly for the ozone lifetime and modified P22, L4-10 to read: "Here,  $\tau_{O_3}$  is calculated as the tropospheric ozone burden divided by total  $O_x$  loss (chemical and deposition).  $\tau_{O_3}$  in the Base experiment closely matches the ACCENT and ACCMIP mean values; note that for this comparison, only the deposition of ozone, and not  $O_x$ , is considered in the  $\tau_{O_3}$  definition (Table 2, bracketed values). Changes about a baseline  $\tau_{O_3}$  of  $22.5 \pm 0.1$  days (Table 2) as a result of each type of perturbation are now considered."

P3069 line 7: should note that methane does not follow either scenario, and is fixed.

Done - see first reply.

P30649 lines 7-11: should remove "However. . .on these topics" and the previous sentence needs to be followed by citations.

The sentence "However...on these topics" highlights that our study does not investigate mechanisms that underlie changes in the global circulation. To clarify, we have changed it to "We do not discuss the detailed mechanisms that underlie changes in the global circulation (e.g. McLandress and Shepherd, 2009; Butchart et al., 2010; Hardiman et al., 2013)."

P30652 lines 4-5: should give definition of the tropopause used here.

Done.

P30652 line12: Does it make sense to assess temperature changes (especially the lower atmosphere) in an atmosphere-only model? Please comment.

We would first like to clarify that irrespective of the origin of the temperature changes, they are shown to facilitate in understanding the ozone changes.

In the climate change cases, it makes sense to also assess tropospheric temperature changes since they are mainly determined by changes in SSTs/sea ice and, hence, reflect the climate sensitivity of the coupled model from which these boundary conditions are obtained for the year 2100 (here, the HadGEM2-CC model).

In the case of the  $\Delta ODS$  and  $\Delta O_3pre$  experiments, SSTs/sea ice are fixed at Base values, which strongly limits any tropospheric temperature response that would otherwise occur e.g. due to ozone-radiative feedbacks onto climate. This has been mentioned in the case of  $\Delta O_3pre$  (P10, L9-10), but is

now also mentioned for  $\Delta$ ODS (P9, L23-27): "Note that the tropospheric temperature response cannot be assessed here since it is strongly limited by the use of fixed, year 2000 SSTs and sea ice. The effect is likely to be small: McLandress et al. (2012) find only small tropospheric warming (Antarctic) and cooling (Arctic) due to ozone recovery between 2001-2050 in their model."

Stratospheric temperature changes are likely to be less sensitive to the details of atmosphere-ocean coupling. Under climate change, the direct radiative effect of WMGHGs is the primary driver of stratospheric temperature changes (see Oberländer et al. (2013), who have separated the direct radiative effect from the indirect effect through SST changes). Similarly, shortwave absorption by ozone in  $\Delta$ ODS dominates the stratospheric temperature change in this experiment.

P30652 line 17-20: These statements are rather vague. Can you describe what specifically will be discussed in the following subsections?

The specifics (zonal/annual mean ozone/temperature and column ozone changes) have been detailed in the preceding lines with reference to the appropriate figures. Perhaps "stratospheric processes" is ambiguous, so it has been replaced with "changes in the large-scale stratospheric state" to tie in with the previous sentences.

P30654 line 9-10: Please note that same prescribed SSTs are used in these perturbation runs as in the Base run so the model cannot realistically capture temperature changes in response to changes in ODS and precursor emissions, especially in the lower atmosphere.

Done - please see comment before last.

P30654 lines 25-26: How much is this as a percentage increase?

3%. Have included % changes for all quoted DU changes.

P30655 lines 8-10: More precisely, changes in ozone precursors have limited impact on stratospheric ozone here.

The sentence has been modified (P10, L9-10): "The changes in ozone precursor emissions in the  $\Delta$ O3pre experiment do not have a significant effect on stratospheric ozone abundances."

P30655 lines 18-21: The finding here seems based on Fig 3 so should be placed after the next sentence.

This is a general finding across the set of experiments. Fig. 3 is mentioned thereafter since it demonstrates the only exception to the statement - that of non-additivity in ozone in the  $\Delta$ (CC8.5+ODS) experiment. To make this clear, the sentence "The extent to which...is shown in Fig. 3" has been removed, and the first sentence in the next paragraph has been modified (P10, 26-28): "The exception is the ozone response in  $\Delta$ (CC8.5+ODS), in which two regions of small, but statistically significant, non-additivities are found (shading, Fig. 3b)."

P30656 lines 1 and 2: "change" – should say it is positive or negative. Same with the following "change".

In specifying the sign of the change, the metric ( $d\ln[O_3]/dT^{-1}$ ) used to characterise the ozone-temperature dependence requires definition, so we have modified P11, L1-5 to read: "The effect is caused by a change in the temperature dependence of catalytic ozone loss (positive if evaluated by  $d\ln[O_3]/dT^{-1}$  as in Haigh and Pyle (1982)) with a reduction in halogen loading. This is essentially the same effect found by Haigh and Pyle (1982) in their experiment combining a doubling in CO<sub>2</sub> with increases in ODS concentrations."

P30656 lines 23-24: Could you give references here, i.e. “from theory and previous model studies (references)”?

Inserted references to Haigh and Pyle (1982); Jonsson et al. (2004); Austin et al. (2010); Eyring et al. (2013); Meul et al. (2014).

P30656 lines 24-25: Could you elaborate on the role of methane changes even though you keep methane fixed in perturbation runs.

Inserted (P11, L30 - P12, L6): "Insight into the impact of methane changes, which are not explored here, can also be garnered from previous literature (Randeniya et al., 2002; Stenke and Grewe, 2005; Portmann and Solomon, 2007; Fleming et al., 2011; Revell et al., 2012). These studies conclude that the stratospheric ozone response to increased methane results from a combination of increased HO<sub>x</sub>-catalysed destruction (upper stratosphere), enhanced production through smog-like chemistry (lower stratosphere), and reduced losses due to water-vapour induced cooling and reductions in [ClO<sub>x</sub>]. Overall, Revell et al. (2012) find positive linear relationships between end of 21<sup>st</sup> century surface methane abundances and stratospheric column ozone across the four RCPs in the NIWA-SOCOL CCM."

P30656 lines 25-26: “However. . . impacts on the troposphere” sounds like a conclusion – you normally do not conclude before the analysis.

These lines have been modified (P12, L8-11): "However, changes in stratospheric composition and dynamics might have important impacts on the troposphere. To determine the extent of these impacts, the next section provides a detailed analysis of the troposphere."

P30657 line 9: should give references regarding “multi-model means”

Inserted references to Stevenson et al. (2006), Naik et al. (2013) and Young et al. (2013).

P30657 lines 25-27: I cannot see the synergy between “The balance between the terms” and “the Base ozone burden is close to the ACCENT and ACCMIP ensemble means . . .”. Can you clarify? Also need to clarify in this section that if O<sub>x</sub> dry deposition includes those non-ozone species, and how much is actual O<sub>3</sub> deposition in the mix if that’s the case.

The first phrase is unclear and has been removed. We simply meant that despite a low STE, the burden compares well to ACCENT/ACCMIP.

The deposition issue has been clarified (see previous comment and reply).

P30658 lines 22-23: “Figure 4a shows that consideration of NCP alone, . . ., would suggest reductions in ozone burden” – I don’t think there is a strict linear relationship between NCP and ozone burden. Ozone burden is determined by the loss rate and its lifetime.

We did not mean to imply such a strict relationship between NCP and the ozone burden. We simply compare changes in NCP to changes in STE (the focus of this study) as they are terms of a similar magnitude in the O<sub>x</sub> budget. Overall, the aim is to highlight STE as a considerable additional source of ozone (as well as its likely impact on the ozone lifetime).

Please also see the response to Reviewer 1’s comment regarding Page 30658, Lines 22-24, the tracked changes in Sect. 4.2, paragraphs 1-4 and the minor re-wording to P1, L28 and P25, L1-3.

P30659 lines 6-7: Sensitivity is usually expressed quantitatively. The sentence is also vague. How about replace "sensitivity" with "response"?

Done. The sentence has been elaborated (see next reply).

P30659 lines 7-17: What do these tell us? What is the useful message? Regarding the statement "the sign of the change in the ozone burden is not agreed upon by models", do you mean different models in one experimental setup or in different experimental setup? The cited model or multi-model studies have different emission and climate scenarios so it should not be directly compared.

The reviewer is correct that care must be taken to distinguish between the dependence of changes in the ozone burden on scenario specific factors (precursor emissions, total radiative forcing) and model specific factors (climate sensitivity, BDC response, LNO<sub>x</sub> changes etc.). The aim of this discussion is to emphasise the latter. Despite the general consistency in the sign of changes in individual O<sub>x</sub> budget terms amongst models (decreases in NCP and ozone lifetime; increases in STE and L+D), the overall change in burden will depend on the precise balance between these, which will differ because of inter-model differences in the above factors amongst others.

We agree that the comparison to the ACCENT study is too direct since we use different climate scenarios to them. We have changed the text to now highlight the ACCENT inter-model range.

The experimental setup of Kawase et al. (2011) is very similar to the ΔCC4.5 and ΔCC8.5 runs of our study so we have made a closer comparison with their results.

As was already stated, the ACCMIP results are not comparable; these are included to give an idea of changes under the RCPs including all forcings e.g. methane changes affecting chemistry that are not included in our experiments.

Please see the modified text (P15, L3-23).

P30659 lines 22-24: A bit of jump here; could you give some context as to why methane adjustments are discussed? Why not move this prelude to the next section?

Sect. 4.2 details changes in the ozone burden; the neglect of methane feedbacks in these experiments is a caveat to these results that requires discussion. The following changes add more context:

The heading of Sect. 4.3 has been changed to "Implications of methane adjustments for the ozone burden"

A new sentence has been inserted at the beginning of Sect. 4.3: "The tropospheric ozone burden is also affected by the method in which the methane boundary condition is applied in the model."

The prelude at the end of Sect. 4.2 has been modified and moved to the end of Sect. 4.3: "Having discussed changes in the ozone burden, the following subsection further explores the tropospheric O<sub>x</sub> budget and investigates the underlying causes of the changes in NCP and STE."

P30661 lines 2-3: Quantifying the individual importance of these processes is not beyond the scope of this study I would say; you could analyse the chemical budget. In the lack of relevant diagnostics, you should note that, e.g., "We do not individually quantify these processes ..."

Indeed, we did not output fluxes through all the relevant reactions, and have changed the sentence to: "We do not quantify the relative importance of these separate drivers."

P30665 lines 29 – 2 next page: It is not surprising that with the reduction of ODS, tropospheric ozone has a substantial increase through STE, which offset chemical loss of ozone through increased water vapour.

The competition between these two effects is not surprising so we have replaced the word "interesting" with "notable" (P21, L13). However, it is the magnitude of the offset that we emphasise here. Even at RCP8.5, for which increases in humidity-driven ozone losses are large, the effect of

STE is dominant throughout most of the troposphere. It is important to highlight this result in the UM-UKCA model since the magnitude of this offset is likely to be model dependent and might not be so large in another model (see also discussion in Stevenson et al. (2006), Sect. 4.1.4).

It is not only the *magnitude* of the total offset associated with increased STE that is notable, but the fact that it affects ozone amounts in different regions of the troposphere depending on the driver (i.e. changes in climate or reduced ODSs), which, to our knowledge, has not been highlighted in any previous study.

P30668 lines 17-19: I think you'd better say "although we cannot verify such assumption here due to lacking relevant diagnostics in this study. . ." rather than saying "it is beyond the scope of this study".

P23, L21-23 now read: "...although we cannot verify such an assumption due to the relevant diagnostics not being available and further sensitivity tests would be required."

P30669 line 10: Could replace "stratosphere" with "changes in stratosphere"?

We would prefer to leave this as is since "changes" appears more than once in the points that follow.

P30670 lines 2-3: "the upper troposphere is a key region for ozone as a radiative forcing agent." - This is not the finding from this study, you might want to say "this should have implications for the climate feedback as UT is a key region for ozone as a radiative forcing agent".

Done.

P30670 line 23: add "and uncertainties" after "differences"

Done.

P30689 Figure 8: Why don't you use the same colour scale for a) and b), and e) and f)?

Done.

Technical corrections:

P30648 Line 21: missing "et al." in Collins and Sudo citations.

Done.

P30651 line 20: missing "et al." in citation.

Done.

P30661 line 16: "MeO2" should be denoted as "CH3O2"

Done in any instance this appears, including Fig. 5a (x-axis label) and Fig. 5b (legend).

P30662 line 8: see above regarding "MeO2"

Done - see above.

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