

RESPONSE TO REVIEWER #1

The authors would like to thank Reviewer 1 for the positive rating of this manuscript and suggested points for improvement. Please see the original review (black) and our responses (blue) below. Additions or updates to the text are in green. Page/line numbers in our responses refer to the track changed manuscript.

In this paper, the authors have explored the sensitivity of stratospheric and tropospheric ozone, and tropospheric ozone budget to future climate change, reductions in ozone depleting substances, and non-methane tropospheric ozone precursor emissions using an interactive stratospheric and tropospheric chemistry-climate model. The results of this study highlight the importance of stratospheric chemistry and dynamics for determining tropospheric ozone burden under different climate change, ODS and precursor emission scenarios. The paper adds to the body of work on the importance of stratosphere-troposphere exchange for tropospheric ozone by performing an in-depth analysis of the tropospheric ozone budget terms. Overall, the analysis is rigorous and the paper is well-written. I recommend the acceptance of this paper by ACP after the following comments have been addressed:

Page 30647, Line 28: A reference to van Vuuren et al., (2011) would be appropriate here.

Thanks - done.

Page 30648, Line 25: Reference to Revell et al. (2015) would be appropriate here.

This line refers only to those studies that *isolate* the impacts of ODS-driven stratospheric ozone recovery on tropospheric ozone. Revell et al. (2015) do not conduct any simulations which allow these to be decoupled from the radiative effects of greenhouse gases (in particular, CO₂ and its indirect impacts through SST changes). We agree however, that this recent study is relevant for the Introduction so we have inserted a reference to it on P2, L19.

Same for Line 20 on page 30649.

This line specifically refers to the ACCMIP study, so we do not think a reference to Revell et al. (2015) is appropriate.

Page 30650: Do lightning NO_x emissions change in any of the perturbation simulations? Since lightning NO_x is conventionally tied to model convection and therefore climate, I would imagine that it is responding to climate change in ΔCC simulations.

That's correct. A previous study (Banerjee et al., 2014) has analysed changes in convection, lightning NO_x emissions and associated impacts on tropospheric ozone under climate change in these simulations. Inserted (P5, L25-27): "Emissions of NO_x from lightning (LNO_x) are parameterised as a function of cloud-top height (Price and Rind, 1992, 1994) and thus, can vary with changes in convection (Banerjee et al., 2014)."

Page 30652: As described by the authors, any ozone molecule above the thermal tropopause is tagged as "stratospheric", however, it is quite possible that ozone produced in the troposphere can potentially land in the stratosphere due to deep convection in the troposphere, particularly in the tropics. It would be helpful if authors could provide insight into how their definition of O₃S and the tropopause might impact the conclusions of this study.

Indeed, there will be quantitative differences in [O₃S] resulting from the definition of "tropospheric" vs "stratospheric" air. Lin et al. (2012) find that employing the thermal tropopause results in higher surface [O₃S] than when using the 'e90' tropopause (Prather et al., 2011). However, as previously stated, we believe the qualitative conclusions regarding *changes* in O₃S will not be affected by the

tropopause definition. The text has been updated (P7, L10-16): "...; Lin et al. (2012) find in their CCM that seasonally averaged surface O₃S abundances are 5-8 ppbv higher when defined by the thermal tropopause compared to the 'e90 tropopause', which essentially differentiates tropospheric from stratospheric air based on mixing time scales (Prather et al., 2011). However, although there are quantitative differences in absolute O₃S abundances between different tropopause definitions, the qualitative conclusions drawn in Sect. 4.5 regarding changes in O₃S are unlikely to depend upon this choice."

Page 30652, Line 16: Reference to Eyring et al. (2013) with results from the latest CMIP5 simulations would be appropriate here.

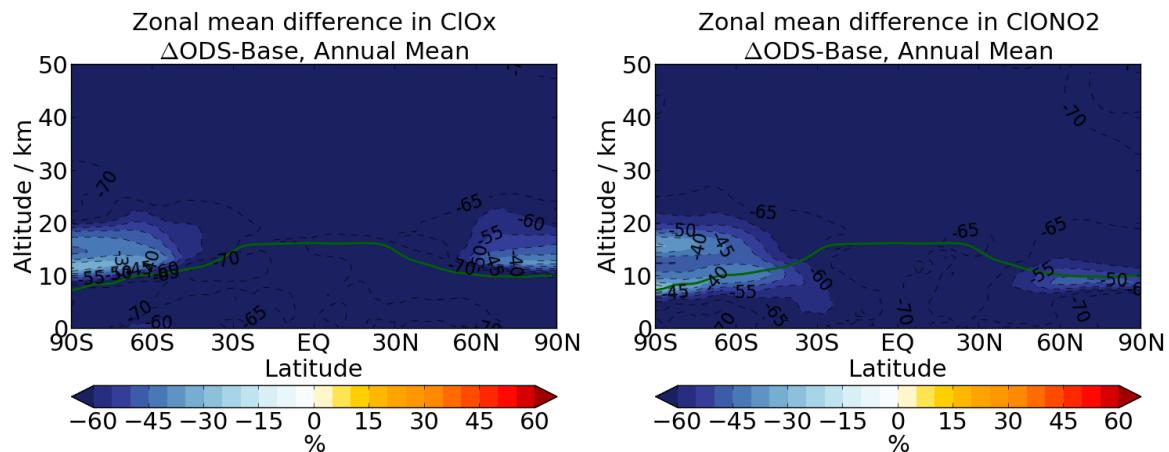
Agreed - the reference has been inserted.

Page 30653, Line 15: The authors mention tropical upper tropospheric increases in ozone are driven by lightning NO_x – are lightning NO_x emissions allowed to respond to climate change?

Yes, this is clarified on P5, L25-27 in response to a previous comment. P8, L20-22 has been modified to: "...; but this is partly mitigated by increases in lightning-derived ozone/NO_x due to deeper convection in a warmer climate (Banerjee et al., 2014)."

Page 30654, Lines 13-16: It is not clear if the authors found the model to produce reductions in the abundance of ClONO₂ reservoir in the ODS experiment. A supplemental plot of modeled changes in ClONO₂ would be helpful here.

Yes, with the large reductions in CFC amounts imposed in the ΔODS experiment, there are large reductions in [ClO_x] and, hence, [ClONO₂] (plots below). This result is expected and inclusion of this plot will probably not add much information. Unless the reviewer objects, we will leave it out of the supplement. We have clarified that it is "not shown" in the text, instead of referencing another study.



Page 30656, Lines 23-24: References are needed after "theory and previous model studies".

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

Page 30657, Lines 13-15: I think it would be useful to provide ACCMIP ozone budget numbers for comparison with the caveat that the budget terms were calculated using 6 models while ozone burden and methane lifetime are calculated using output from ~15 ACCMIP models. Particularly, because many ACCMIP models included interactive stratospheric chemistry.

Please see Sect. 4.1, Sect. 4.6 (paragraph 1) and Table 2, which have been updated to include this comparison.

Page 30658, Lines 22-24: Ozone depositional loss increases for CC4.5 and ODS simulations while it increases for CC8.5. Perhaps the authors could comment on how changes in loss processes (chemical plus dry deposition) affect ozone burden.

Yes, the steady state ozone burden is a product of the ozone lifetime and $L(O_3) + D(O_3)$ (and also $P(O_3) + STE$ if the budget is closed); we refer to the latter quantity as the "turnover flux" for ease. We agree that these quantities are worth discussing in Sect. 4.2. (Note the reasons behind ozone lifetime changes are already detailed in Sect. 4.6.) Please see the tracked changes in Sect. 4.2, paragraphs 1-4.

Page 30659, Line 3: Insert "tropospheric" between higher ozone.

Done.

Page 30659, Lines 4-5: Refer to Table 2 and/or Figure 4 here.

Done.

Page 30659, Lines 7-17: I don't think this is a fair comparison as these models used different assumptions for climate and emissions changes.

Please see response to Reviewer #2, comment regarding P30659 lines 7-17.

Page 30663, Lines 10-11: The reference to Figure 6 without getting into details of the diversity in STE across the perturbation experiments is conspicuously standing out here. I think this sentence could be removed as the figure is discussed in in section 4.5.2.

The reference to Fig. 6 here has been removed. Other than this, the sentence has been retained (to ensure a flow between this subsection and the next) and moved to the end of the previous paragraph (P19, L2-4): "Changes in the residual circulation in the single-forcing experiments will be linked qualitatively to changes in STE in Sect. 4.5.2."

Page 30663, Lines 26-27: A citation is needed here.

A reference (Butchart, 2014) has been inserted and the sentence slightly modified (P19, L20-22): "The latter result is comparable to the CMIP5 multi-model mean increase for the RCP8.5 scenario of 32 % between 2000-2100, extrapolated from the linear rate of change found between 2006-2099 (Butchart, 2014)."

Page 30664, Lines 10-12: Suggest rephrasing to "Figure 8 shows absolute changes in O3S and O3 between Base. . ."

Done.

Page 30671: Lines 5-6: A citation would be helpful here.

Inserted citation to Livesey et al. (2008), which describes ozone measurements made by the MLS instrument. These have previously been assimilated in a CTM in order to improve its representation of STE (Barré et al., 2012) and used to constrain present-day STE variability (Olsen et al., 2013).

Figure 2: Add "Tropical" to y axis title.

Done.

Figure 8: For easy comparison, it would help to use the same color scale for (a) and (b), and (e) and (f). Also, the size of this plot should be increased as the colorbar labels are difficult to read.

Scales - done. These subplots appear to be downsized in the ACPD publication in order to fit the page. We will request that they remain at least the same size as the plots in Fig. 1/3.

References:

Revell, L. E., Tummon, F., Stenke, A., Sukhodolov, T., Coulon, A., Rozanov, E., Garny, H., Grewe, V., and Peter, T.: Drivers of the tropospheric ozone budget throughout the 21st century under the medium-high climate scenario RCP 6.0, *Atmos. Chem. Phys. Discuss.*, 15, 481-519, doi:10.5194/acpd-15-481-2015, 2015.

van Vuuren, D. P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G. C., Kram, T., Krey, V., Lamarque, J.-F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S. J., and Rose, S. K.: The representative concentration pathways: an overview, *Clim. Change*, 109, 5–31, doi:10.1007/s10584-011-0148-z, 2011.

Eyring, V., Cionni, I., Arblaster, J., Sedlacek, J., Perlitz, J., Young, P. J., Bekki, S., Bergmann, D., Cameron-Smith, P., Collins, W., Faluvegi, G., Gottschaldt, K.-D., Horowitz, L., Kinnison, D., Lamarque, J.-F., Marsh, D. R., Saint-Martin, D., Shindell, D., Sudo, K., Szopa, S. and Watanabe, S.: Long-term changes in tropospheric and stratospheric ozone and associated climate impacts in CMIP5 simulations, *JGR*, 10.1002/jgrd.50316, 2013.

References (in addition to those above):

Austin, J., Scinocca, J., Plummer, D., Oman, L., Waugh, D., Akiyoshi, H., Bekki, S., Braesicke, P., Butchart, N., Chipperfield, M., Cugnet, D., Dameris, M., Dhomse, S., Eyring, V., Frith, S., Garcia, R. R., Garny, H., Gettelman, A., Hardiman, S. C., Kinnison, D., Lamarque, J. F., Mancini, E., Marchand, M., Michou, M., Morgenstern, O., Nakamura, T., Pawson, S., Pitari, G., Pyle, J., Rozanov, E., Shepherd, T. G., Shibata, K., Teyssèdre, H., Wilson, R. J. and Yamashita, Y.: Decline and recovery of total column ozone using a multimodel time series analysis, *J. Geophys. Res. Atmos.*, 115, D00M10, doi:10.1029/2010JD013857, 2010.

Banerjee, A., Archibald, A. T., Maycock, A., Telford, P., Abraham, N. L., Yang, X., Braesicke, P. and Pyle, J. A.: Lightning NO_x, a key chemistry–climate interaction: impacts of future climate change and consequences for tropospheric oxidising capacity, *Atmos. Chem. Phys.*, 14, 9871–9881, doi:10.5194/acpd-14-9871-2014, 2014.

Barré, J., Peuch, V.-H., Attié, J.-L., El Amraoui, L., Lahoz, W. A., Josse, B., Claeysman, M. and Nédélec, P.: Stratosphere-troposphere ozone exchange from high resolution MLS ozone analyses, *Atmos. Chem. Phys.*, 12, 6129–6144, doi:10.5194/acp-12-6129-2012, 2012.

Butchart, N.: The Brewer-Dobson circulation, *Rev. Geophys.*, 52, 157–184, doi:10.1002/2013RG000448, 2014.

Haigh, J. D. and Pyle, J. A.: Ozone perturbation experiments in a two-dimensional circulation model, *Q. J. R. Meteorol. Soc.*, 108, 551–574, doi:10.1002/qj.49710845705, 1982.

Jonsson, A. I., de Grandpré, J., Fomichev, V. I., McConnell, J. C. and Beagley, S. R.: Doubled CO₂-induced cooling in the middle atmosphere: Photochemical analysis of the ozone radiative feedback, *J. Geophys. Res.*, 109, D24103, doi:10.1029/2004JD005093, 2004.

Lin, M., Fiore, A. M., Cooper, O. R., Horowitz, L. W., Langford, A. O., Levy, H., Johnson, B. J., Naik, V., Oltmans, S. J. and Senff, C. J.: Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions, *J. Geophys. Res. Atmos.*, 117, D00V22, doi:10.1029/2012JD018151, 2012.

Livesey, N. J., Filipiak, M. J., Froidevaux, L., Read, W. G., Lambert, A., Santee, M. L., Jiang, J. H., Pumphrey, H. C., Waters, J. W., Cofield, R. E., Cuddy, D. T., Daffer, W. H., Drouin, B. J., Fuller, R. A., Jarnot, R. F., Jiang, Y. B., Knosp, B. W., Li, Q. B., Perun, V. S., Schwartz, M. J., Snyder, W. V.,

Stek, P. C., Thurstans, R. P., Wagner, P. A., Avery, M., Browell, E. V., Cammas, J.-P., Christensen, L. E., Diskin, G. S., Gao, R.-S., Jost, H.-J., Loewenstein, M., Lopez, J. D., Nedelec, P., Osterman, G. B., Sachse, G. W. and Webster, C. R.: Validation of Aura Microwave Limb Sounder O₃ and CO observations in the upper troposphere and lower stratosphere, *J. Geophys. Res.*, 113, D15S02, doi:10.1029/2007JD008805, 2008.

Meul, S., Langematz, U., Oberländer, S., Garny, H. and Jöckel, P.: Chemical contribution to future tropical ozone change in the lower stratosphere, *Atmos. Chem. Phys.*, 14, 2959–2971, doi:10.5194/acp-14-2959-2014, 2014.

Olsen, M. A., Douglass, A. R. and Kaplan, T. B.: Variability of extratropical ozone stratosphere-troposphere exchange using microwave limb sounder observations, *J. Geophys. Res. Atmos.*, 118, 1090–1099, doi:10.1029/2012JD018465, 2013.

Prather, M. J., Zhu, X., Tang, Q., Hsu, J. and Neu, J. L.: An atmospheric chemist in search of the tropopause, *J. Geophys. Res.*, 116, D04306, doi:10.1029/2010JD014939, 2011.

Price, C. and Rind, D.: A simple lightning parameterization for calculating global lightning distributions, *J. Geophys. Res.*, 97, 9919–9933, 1992.

Price, C. G. and Rind, D.: Modeling global lightning distributions in a general circulation model, *M. Weather Rev.*, 122, 1930–1939, 1994.

Revell, L. E., Tummon, F., Stenke, a., Sukhodolov, T., Coulon, a., Rozanov, E., Garny, H., Grewe, V. and Peter, T.: Drivers of the tropospheric ozone budget throughout the 21st century under the medium-high climate scenario RCP 6.0, *Atmos. Chem. Phys.*, 15, 5887–5902, doi:10.5194/acp-15-5887-2015, 2015.