

We thank both referees for their positive and constructive comments. Our detailed response is given below (in bold italics). Page and line numbers refer to the updated manuscript.

Response to Anonymous Referee #2

The manuscript presents an investigation of the model-projected evolution of ozone in the tropics (10S to 10N) over the period 1960 to 2100. A transient simulation covering the whole period and following the specified reference scenario for the Chemistry-Climate Model Initiative (CCMI) model intercomparison project, the REF-C2 simulation, is augmented with time-slice simulations for year 2000 and 2100 conditions run under different levels of ozone depleting substances. The influence of changing greenhouse gases (GHGs) and ozone depleting substances (ODSs) on tropical total column ozone are investigated by splitting the total column into upper stratospheric, lower stratospheric and tropospheric components. Linear functions of the change in column ozone due to the effects of GHGs and ODSs are derived and these functions are used to reproduce the evolution of stratospheric ozone column in the full model simulation.

Of significance, the results add to a number of recent papers that underline the importance of ODS-driven changes in ozone on tropical upwelling in the lower stratosphere. The core of the methodology and results presented in the manuscript are, in my opinion, solid. My one significant concern is the way in which the effects of methane and nitrous oxide are treated.

On page 13, Lines 1-4, the authors state:

'In Section 4 we showed that future changes in tropical stratospheric column ozone are driven primarily by changes in: (i) the halogen-catalysed loss; (ii) the strength of tropical upwelling; and (iii) the upper stratospheric cooling induced by GHGs (mainly CO₂).' I would argue that the authors have not, in fact, shown this in general. The inferred causes of changes in partial column ozone are derived from the set of timeslice experiments that only varied GHGs and ODSs. That these are then the only two factors that were found to be responsible for changes in ozone should naturally follow.

On Page 5, Lines 9-11 the authors state ***'In this study we consider the radiative impact of a large number of GHG species (CO₂, CH₄, N₂O, CFCs) and assume that the dominant driver of chemical changes is changes to ODS loadings. In this way, the chemical impact of changing N₂O and CH₄ emissions is not considered here.'*** Since the effects of changing N₂O and CH₄ are not considered it seems difficult to justify the conclusion (Page 16, Lines 21-23) that ***'Results from the simple model indicate stratospheric column ozone changes resulting from future CH₄ and N₂O emissions are of second order on the timescales considered here.'***

While the parameterization of stratospheric column that is derived here is able to reproduce fairly well the evolution of stratospheric column in the transient simulation, the variation of methane is fairly small in RCP6. The parameterized stratospheric column also significantly overestimates the trend from 2020 to 2100, where the full model shows almost no change while the parameterization projects an increase on the order of 5 DU, which could be related to the steadily increasing concentration of nitrous oxide. While the authors have nicely constructed a set of experiments to quantitatively estimate the effects of ODSs and GHGs on tropical ozone, the absence of any methodical investigation of the effects of methane or nitrous oxide would, I believe, rule out making any statements on the importance of these species.

We agree that the role of future CH₄ and N₂O emissions for tropical TCO trends should not be understated. We have added text throughout the manuscript to this effect, and have amended the discussion in section 5 so that it does not appear that we are saying CH₄ and N₂O are not important drivers of future ozone projections.

The reason for claiming, as we do in Section 4, that ‘future changes in tropical stratospheric column ozone are driven primarily by changes in: (i) the halogen-catalysed loss; (ii) the strength of tropical upwelling; and (iii) the upper stratospheric cooling induced by GHGs (mainly CO₂)’ is because the simple model, which includes only terms for chemical ODS and radiative CDE forcings, is able to reproduce with a reasonable degree of accuracy the long-term SCO₃ trend from the fully coupled CCM simulation, which also includes CH₄ and N₂O chemical effects in its detailed chemical scheme. However, we do not know, for example, whether in the UKCA model, N₂O and CH₄ have very large but opposite effects on stratospheric ozone that cancel each other out. There is some indication from the latest WMO assessment that CH₄ and N₂O changes have opposite effects on column ozone, and further work with a number of fully coupled chemistry climate models is required to fully understand the drivers of future ozone concentrations in different regions of the atmosphere. We have expanded where appropriate our discussion on the roles of N₂O and CH₄ and the impacts that not including them in the simple model may have. We have further discussed the role of N₂O and CH₄ for determining differences in future SCO₃ trends between RCP scenarios and the limitations therefore of the simple model in capturing details of the differences between RCPs (e.g. compared to the multi-model results of Eyring et al. (2013)).

Somewhat related to this point, it would be very helpful to the reader if the authors would state what N₂O and CH₄ concentrations were used for the timeslice experiments. I assume all six of the timeslice experiments used the same specifications for N₂O and CH₄ but it would be helpful to know if this were so and what boundary conditions specifically were used.

In all time-slice experiments chemical concentrations of N₂O and CH₄ use prescribed year 2000 concentrations from RCP6.0 as a lower boundary condition. This information has been added to the manuscript (P6L3)

Aside from that my other concerns are minor and are specified below.

Page 4, Lines 22-23: Do the two extra ensemble members that start in 1980 use chemical initial conditions from the original two members that were started in 1960? If not, how are the chemical tracers for these two simulations initialized?

The two extra ensemble members started from 1980 use initial conditions generated by spinning off a 1980 timeslice from one of the ensemble members, which was run for 20 years, New transient simulations were then initialised using different years from this time-slice. This description has been added to the methodology section of the manuscript.

Page 5, Lines 2-3: In Table 1 there seems to be an error in the specifications for TS4.5_ODS as that table says climate for RCP8.5 is used.

The RCP scenarios used for the climate component of each of the experiments has been corrected.

Page 7, Lines 18-20 states ‘These results indicate that over the recent past upper stratospheric ozone depletion resulting from increased Cly concentrations has in part been

offset by radiative cooling resulting from increased GHG concentrations, and that in the future both increased GHG concentrations and reduced stratospheric Cl_y will result in increases in upper stratospheric ozone concentrations.' A very applicable reference to earlier work on this point would be Shepherd and Jonsson, On the attribution of stratospheric ozone and temperature changes to changes in ozone-depleting substances and well-mixed greenhouse gases, *Atmos. Chem. Phys.*, 8, 1435-1444, 2008.

We thank the reviewer for bringing this paper to our attention - the reference has been added to the text and reference list

Page 8, Lines 8-10: 'As was seen for the upper stratosphere, the PCO_3_{LS} response to a given decrease in ODS is dependent on the GHG concentration, (+7 DU for TS2000_ODS - TS2000, +6 DU for TS4.5_ODS - TS4.5 and +4 DU for TS8.5_ODS - TS8.5).' Do you have any explanation for the variations in the response to ODSs across the GHG concentrations?

The variation in the response of ozone to ODS under different GHG loadings is related to the impact of ODS on the speed of the BDC, the temperature dependence of the chlorine catalysed loss cycles and the influence of the upper stratospheric shielding on the lower stratosphere. In all simulations, decreasing ODS concentrations lead to a deceleration of the BDC, decreasing the transport of ozone out of the tropical lower stratosphere and leading to increased lower stratospheric partial column values. The deceleration of the BDC due to ODSs is relatively insensitive to the GHG loading. However, as the stratosphere cools the efficiency of the ClO_x catalysed ozone loss is reduced. Although this plays a minor role in the lower stratosphere, where only a small proportion of the CFCs have been oxidised, it does contribute to the signal seen here. In addition, cooling of the upper stratosphere leads to greatly increased ozone concentrations through both Chapman chemistry and reducing the efficiency of the ClO_x catalysed ozone loss. Increased overhead ozone in turn affects the photolysis rates in the lower stratosphere, slowing ozone production. Decreased production values in the lower stratosphere partially offset the increases from a slower BDC, explaining the variations in the response to ODSs across the GHG concentrations. This discussion has been added to the manuscript.

Page 9, Line 7. Here in reference to Figure 3 the amount of ODSs in the atmosphere is indicated by EESC. Traditionally Equivalent Effective Stratospheric Chlorine has been defined in a very particular way using tropospheric concentrations, age of air and release factors for the decomposition of the ODS compound. Given the way the trace of EESC on Figure 3 looks, I think you would want to refer to Equivalent Stratospheric Chlorine (ESC). Have a look at Eyring et al., Multi-model assessment of stratospheric ozone return dates and ozone recovery in CCMVal-2 models, *Atmos. Chem. Phys.*, 10, 9451-9472, 2010, for an example. You should also quote what value of α , the enhancement factor for bromine, you have used.

Following the definitions of Eyring et al. (2007), we have used ESC rather than EESC. This has been corrected. For α we have used a value of 60. This has been corrected in the text.

Page 12, Lines 12-13. The statement 'The largest rate of change for tropospheric column ozone occurs over the recent past (1960-2000) (Figure 2c), when increases in anthropogenic NO_x emissions (Lamarque et al., 2010) drive increases in ozone production.' A minor point, but I do not think you can rule out the increase in methane over 1960-2000 as contributing. Methane in 1850 was ~800 ppbv, in 1960 it was 1250 and in 2000 it was 1750 ppbv. About one-half of the total increase occurred between 1960 and 2000 and results from

ACCMIP (e.g. Young et al., Pre-industrial to end 21st century projections of tropospheric ozone..., Atmos. Chem. Phys., 13, 2063-2090, 2013) show that the methane increase does account for a good portion of the total increase between 1850 and 2000.

The text of the manuscript has been amended to include the role of CH₄ in historic tropospheric ozone changes alongside the Young et al. reference.

Page 14, Lines 6 and 7: I had trouble reading 'These scenarios include RCP4.5, RCP8.5, RCP6.0 using ODS fixed at 1960 values and RCP6.0 using CDE fixed at 1960 values.' It took a bit of rereading and looking at Figure 7 to understand that not all of RCP4.5, RCP8.5 and RCP6.0 were run using ODS fixed at 1960 values. Is it possible to reword a bit.

The description of the scenarios performed using the simple model has been revised to avoid confusion.

Page 14 Lines 6 and 7: The RCP4.5 and 8.5 results from the parameterization could be compared with Figure 6 of Eyring et al., Long-term ozone changes and associated climate impacts in CMIP5 simulations, J. Geophys. Res., 118, 5029-5060, 2013. They show that going towards 2100, it is actually RCP6 that has the lowest stratospheric column ozone while RCP8.5 is slightly higher. Not to beat on this point too much, but I think the different relative order shown by your parameterization may be due to ignoring the effects of CH₄. Of course, it is a different set of models compared with your parameterization derived from UM-UKCA and that cannot be ignored either.

We have included a comparison of the simple model results with the Eyring et al. multimodel projections to highlight the differences between the scenarios at the end of the century and also include a discussion about how these differences may be due to CH₄ and N₂O.

Page 32 – Figure 6. I may have missed it, but I did not find any discussion of Figure 6 in the text.

Figure 6 should have been referenced in relation to the calculation of the $\frac{\Delta SC03}{\Delta CDE}$ and $\frac{\Delta SC03}{\Delta ESC}$ – this has been added to the manuscript.