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 #1

General comments:

Keeble et al. examine tropical ozone trends between 1960-2100 in an ensemble of chemistry-climate model simulations following RCP 6.0. They examine trends in the upper stratosphere, lower stratosphere and troposphere, and use a set of sensitivity simulations to quantify the chemical effects of CFCs, and the radiative effects of greenhouse gases (CO2 + N2O + CH4 + CFCs). They have laid the foundation for a thorough analysis of projected tropical ozone trends, which will be of interest for the stratospheric ozone community, however I do have a number of issues with the paper in its present form that I think should be addressed before the paper is published in ACP.

The authors do not include the chemical effects of CH4 and N2O in their sensitivity simulations. As the authors themselves note (P3L14-16): "the atmospheric concentration of these species, and by extension future concentrations of HOx and NOx radicals, is therefore highly sensitive to assumptions made about their future emissions." I would have thought this a good reason to include them in the analysis, particularly as CH4 and N2O are not currently regulated, unlike the CFCs. I also do not agree with statements such as (p.13): ". . .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)." You did not look at changes in N2O and CH4 chemistry, so how can you say that they are not important drivers? Or, that "the changes in HOx and NOx chemistry resulting from future changes in CH4 emissions would appear to be of second order on the timescales considered" (P13L27-28). A number of studies show that stratospheric ozone is controlled by CO2, CH4 and N2O in the second half of the 21st century (see e.g. Butler et al. 2016 and references therein), and I think the authors need to address this. N2O is the most important ODS currently emitted (Ravishankara et al., 2009), and while reductions in CFCs and increases in CO2 will have a major effect on ozone this century, I am skeptical that N2O can be considered of secondary importance, especially since its chemical effects were not included in this analysis.

The paper has been amended to make clear that our analysis considers only the radiative effects of GHGs and the chemical effects of halogenated ODS species. Where appropriate we have added to the text discussion on the role of N2O and CH4 in chemical ozone depletion and links to tropical column ozone. Our key rationale for claiming that ODS and the radiative effects of GHGs are the key drivers of tropical stratospheric column ozone values comes from the ability of the simple model to reproduce with a reasonable degree of accuracy the long-term trends in stratospheric column ozone from 1960-2100 as modelled by the transient CCM simulation. While differences between the transient simulation and simple model may result from not including (among other things) CH4 and N2O, the very fact that the simple model is able to reproduce the main features of modelled SCO3 values from the CCM (i.e. rapid ozone loss in the late 20th century, a minimum around year 2000 and a gradual increase throughout the 21st century) highlights the important role ODSs and GHGs play in determining future ozone trends. Nevertheless, as highlighted by the reviewer and as we now discuss in the text, the simple model does not quantitatively reproduce SCO3 changes in all periods, and we now include a more balanced discussion about the limitations of the simple model.

I would also like to see a fuller discussion of how the authors' results compare with existing chemistry-climate model studies. For example, they could be compared with the sensitivity studies of Butler et al., 2016; Eyring et al., 2010; Fleming et al., 2011; Oman et al., 2010 and Revell et al., 2012; full citations are given below. Do the authors' results confirm results from existing studies? Do they show something new?

Fuller discussion of how our results from both the transient CCM simulations and the simple model compare with the existing literature has been added throughout the manuscript where appropriate.

As well as comparing the results with other model studies, I think the authors should compare their RCP 6.0 simulation with observations where available, to show how well their CCM performs in the tropics.

Merged observational data from the Bodeker ozone dataset has been used to add observed total column ozone anomalies relative to the year 2000±5 to Figure 1. A comparison between the model and the observations has been added to the text (P7L7).

I am not convinced that the simple model discussed in Section 5 is reliable. It shows (Fig. 7) that stratospheric ozone abundances at the end of the 21st century are higher in RCP 4.5 than they are in RCP 6.0, which is higher again than in RCP 8.5. This is in direct contrast to results from existing CCMs, which show that ozone is highest in RCP 8.5 > RCP 6.0 > RCP 4.5 (see e.g. Fig. 2-23 from Chapter 2 of the WMO 2014 Ozone Assessment). And why does ozone decrease over time when ODSs are held fixed – surely GHG-induced stratospheric cooling should cause ozone to increase? C.f. e.g. Fig. 6 from Fleming et al. (2011).

As the reviewer states, and as discussed in the reply to major comment 1, the simple model does not include a number of processes, chief amongst them the chemical effects of CH4 and N2O, which are likely to contribute to SCO3 trends in the different RCP scenarios. Differences between the multi-model results of Eyring et al (2013) and our study may also result from differences in the chemical and radiative sensitivity of the UM-UKCA model to halogenated ODS and GHGs (e.g. in the sensitivity of the BDC to changes in CDE), as discussed in the manuscript (P17L13).

Note that Fig. 7 is not directly comparable to Fig. 2-23 from Chapter 2 of the WMO 2014 ozone assessment which shows *total* column ozone. This includes tropospheric ozone changes, which overwhelm the stratospheric ozone response. We instead include a comparison of Fig. 7 to Fig. 6b in Eyring et al., 2013 in the text (P17L4) along with a discussion on the end of century SCO3 values in the simple model and compared with other studies, as the reviewer suggests (P17L8). We have caveated our discussion to highlight that the chemical effects of future CH4 and N2O changes are not included in the simple model and that this may account for some of the discrepancy between the simple model and CCM projections by the end of the 21st century and also in the relative projections from the simple model for different RCPs.

Stratospheric ozone decreases over time when CDE increases but ODSs are held constant (cyan line Figure 7) because the reduction in lower stratospheric ozone concentrations due to transport (due to a strengthening BDC) is able to more than compensate for increases in upper stratospheric ozone (due to CO_2-induced cooling) in our simulations. This point is discussed and quantified in the manuscript (section 4.2).

The figures are generally well presented. I do have some ideas for splitting them up and recombining the various subfigures to improve the flow of the discussion (noted later on). The tables contain a few errors, which I have also noted later on in this review.

These points are discussed below

Specific comments

- The authors repeatedly refer to ozone recovery and “super-recovery.” I understand what they are referring to, however the terminology is not correct. Ozone is projected to increase through the 21st century because (i) CFCs decrease; (ii) GHG-induced stratospheric cooling (mostly by $CO2$) increases. Any ozone increase induced by (ii) is not a "recovery," because it was CFCs that caused late 20th century ozone depletion in the first place. I would prefer that such statements surrounding recoveries and super-recoveries are worded more carefully.

Where appropriate reference to recovery has been replaced with the phase ‘return to 1980s values’ or words to this effect.

- Reactive chlorine is referred to as Cly and ClOx. It would improve readability if one term was used consistently.

We thank the reviewer for pointing this out and have corrected references to Cly (which refers to inorganic chlorine species) and ClOx (=Cl + ClO i.e. active - ozone destroying - chlorine species).

- Be careful when referring to ODS-driven ozone loss. Here you refer to ODSs (in your timeslice simulations) as Cly+Bry containing species, and do not include N2O, which is also an ODS.

Where appropriate we have specifically referred to halogenated ODS in the introduction, and in the methodology section of the paper now highlight that we refer only to the halogenated ODS throughout the discussion of our results (P6L17).

- The discussion of partial column ozone differences (3.2) is difficult to interpret since the drivers of ozone change are given only in the following section. The discussion of drivers of ozone change needs to come sooner. I suggest splitting up figure 2, and combining fig. 2a with figs. 3 and 4; combining fig. 2b with fig. 5; and combining fig. 2c with zonally-resolved plots (discussed later in this review). Then the partial column differences and their drivers in each region of the atmosphere can be discussed sequentially.

We have taken this suggestion under consideration. However, we believe that it is of foremost importance to first identify the trends in total and partial column ozone (particularly with a focus on whether ozone in each region is projected to return to 1960s values), and then to later discuss the mechanisms behind these changes in the subsequent section. We feel that this subdivision into model projections and then mechanisms driving those changes best conveys the key findings of the paper. We have added a line in the introductory paragraph of Section 3 (P7, L3) which states that a detailed description of the mechanisms driving the changes presented in the section will be explored in Section 4.

- In the introduction, you discuss the benefits of the stratospheric ozone layer for human health, however a discussion of the harmful effects on tropospheric ozone (as an air pollutant and GHG, and its negative effects on visibility and crop damage) is missing.

A discussion of tropospheric ozone, its key drivers and its important role in air quality and as a GHG has been added to the introduction.

- P2L1-2: At first this reads like a contradiction. The authors need to explain that in the tropics there is a small stratospheric ozone column with a high ozone concentration, and a large tropospheric ozone column with a low ozone concentration, because of the higher tropopause.

The influence of the altitude of peak ozone mixing ratios and high tropopause height on the low tropical column values has been added to the text (P2L6).

- P2L3-4: You might also want to mention projected changes in tropospheric ozone precursors from developing countries.

The focus of this paper is on the long-lived radiative and chemical drivers of total column ozone, with an emphasis of the contribution from stratospheric ozone to TCO, and for this reason we do not discuss in detail the other drivers of ozone concentrations, such as tropospheric ozone precursors, which have only a small impact on stratospheric ozone concentrations (see e.g. Banerjee et al., 2016).

- P2L10-11: note the time period these studies looked at: they show that tropical TCO3 might not reach pre-1980s values by the end of the 21st century.

Specific reference to the fact these projections go to the end of the 21st century has been added to the text (P2L15).

- P2L18: See e.g. Solomon et al., 2016.

We thank the reviewer for drawing our attention to this paper – it has been cited in the text and added to the reference list (P2L25).

- P3L28: While emissions to date indicate that RCP 8.5 is “business-as-usual” at present, I am uncomfortable referring to RCP 8.5 in this way since the methane concentrations by the end of the 21st century are so extreme.

Reference to business-as-usual has been removed from the text

- P4L16: “WCRP/SPARC” -> “IGAC/SPARC” also the reference Eyring et al. (2013) should be changed to Morgenstern et al., 2017. You could also include a sentence describing what CCMI is.

While the Morgenstern et al. (2017) paper provides an overview of the models participating in the first phase of CCMI, Eyring et al. (2013) provide the original and detailed description of the CCMI Reference simulations and is the correct paper to cite here. Furthermore, CCMI itself is well defined within that reference. As we are not conducting a multimodel comparison we feel this manuscript is not the place to add a detailed description of the CCMI project.

- P4L17: Was the chemistry scheme UKCA or CheS+? How are they different? Please provide more details here.

The UKCA chemistry module is available in several possible configurations. We have used two configurations in this study: 1) the transient simulations are run in a configuration with detailed stratospheric chemistry but simplified tropospheric chemistry and 2) the time-slice simulations are run in a configuration with a coupled

stratospheric-tropospheric chemistry scheme. To avoid confusion, we have removed the abbreviations that describe these two UKCA configurations (CheS+ and CheST) and have instead described them in words. Please see Sect. 2 of the final manuscript for these changes.

- P4L23-24: How were the initial atmospheric conditions perturbed for each ensemble run?

Initial conditions for each simulation were generated from perpetual year simulations, with each ensemble member initialised from different years of this perpetual run.

- Table 1 has some errors. I think TS4.5_ODS is supposed to read: climate = 2100 (RCP 4.5) and for TS8.5 climate = 2100 (RCP 8.5).

We thank the reviewer for highlighting the errors in the table – these have been corrected.

- The ODS scenarios developed for the RCPs are all rather similar and similar to the WMO A1 scenario for halocarbons, is this correct? You may want to include some detail here and thus justify why you use Year 2100 ODSs from RCP 4.5 in you TS8.5_ODS simulation.

As the reviewer states, all RCP scenarios have very similar surface concentrations for ODS, which makes the choice of emissions scenario somewhat arbitrary for the future ODS loading time-slice experiments. We have added text to state this (P5, L30).

Furthermore, it is important when conducting a process-based study, as this paper does, to ensure that each perturbation is consistently applied – i.e. that the ODS emission change is identical for each pair of differences. For this reason, we consistently use the RCP4.5 scenario for 2100 ODS concentrations.

- It would be helpful in Table 1 to note that changes in ODSs (Cly and Bry species but not NOx) are imposed only on the chemistry scheme while changes in GHGs (incl. CFCs) are imposed only on the radiation scheme.

This clarification has been added to the table caption.

- For experiment TS2000 do GHGs (i.e. CH4 and N2O) influence chemistry? I think so as this is your "base" run and the other five timeslice experiments are the perturbation experiments, is that correct?

CH_4 and N_2O affect both chemistry and radiation in all the time-slice simulations. However, only the radiative effects of changes to CH_4 and N_2O (alongside CO_2 and CFCs) are considered in TS4.5 and TS8.5. In effect, all 6 time slice experiments are run with year 2000 concentrations of CH4 and N2O in the chemistry scheme, and either year 2000 or 2100 concentrations in the radiation scheme, depending on which time-slice is considered.

- P5L18: Stating that a full description of the simulations is available in Banerjee et al. (2014) is not very helpful as they use a different nomenclature. Please include all relevant details here.

Additional description has been added to the methodology section of the paper to enhance clarity for the reader (section 2).

Figure 1:

- I would like to see some evaluation and discussion of how your CCM performs compared to observations; maybe by plotting observations for when they are available on Figure 1.

Merged observational data from the Bodeker ozone dataset has been added to Figure 1 and a discussion included in the text (Sect. 3.1).

- I am surprised that tropical total column ozone increases by so much in the mid-21st century (Fig. 1), and would like to see more discussion on this, as it is somewhat at odds with the existing literature (see e.g. Fig.2-23 of the WMO 2014 Ozone Assessment, Chapter 2; Fig. 6 of Eyring et al. (2013)). Is the upper stratospheric cooling in the model excessive? Or is too much ozone produced in the troposphere, for example?

We have added to the discussion of Figure 1 a few sentences highlighting the fact that in our simulations TCO3 values do return to pre-1980s values for some part of the 21st century, although other studies (WMO [2014], Meul et al., 2016) suggest this is far from certain (P7L17). Note that TCO3 at 2100 is only ~2 DU lower than in 1980 (following RCP6.0) in Meul et al., 2016, which is more in line with our study. We do not compare our Fig. 1 showing TCO to Fig. 6b of Eyring et al., 2013, which only shows the stratospheric column.

- I am missing a discussion of why TCO3 in the TS2000_ods experiment is so high (higher than in 1960 in the RCP 6.0 simulation). I think this could be because there is very little Cly-induced ozone loss, together with a strong radiative effect from GHGs, which cools the upper stratosphere and thus increases ozone – please discuss this.

Several effects are neglected by comparing the ozone anomalies in any pair of time-slice simulations (such as TS2000 and TS2000_ODS) to anomalies in the transient runs (such as 1960-2000) e.g. the impact of changing GHGs, particularly the cooling effect of CO2, but also the role of CH4 and N2O changes, the role of the solar cycle and different aerosol loadings and SST configurations. From Figure 2c, it is clear that the smaller TCO3 value in the transient run at 1960 compared to TS2000_ODS can in large part be accounted for by an approximately 5 DU reduction in tropospheric partial column values between 1960-2000, the drivers of which are not included in the TS2000_ODS simulation. In contrast, the values at 1960 and in TS2000_ODS are much more similar for the partial stratospheric columns (particularly PCO3_LS) since most of the change in stratospheric column ozone between 1960-2100 is driven by the effect of ODSs.

- P6L23-24: Please be more explicit here. Ozone-destroying chlorine chemistry is temperature-dependent, therefore slows in a colder stratosphere, therefore ozone increases.

We have removed the explanation of this non-linearity here and explained in further detail in Sect. 4.

- P7L3-4: Was there a particular reason that you chose 30 km to differentiate between the upper and lower stratosphere? Please also state the pressure level.

30 km was chosen as the approximate altitude region where ozone changes from being predominantly under photochemical control in the upper stratosphere and predominantly under dynamical control in the lower stratosphere. The corresponding approximate pressure level has been added to the manuscript (P8L10).

- P7L8-11: State why ozone increases, i.e. the GHG-induced stratospheric cooling effect.

This has been added (P8L19)

- P712-13: But as already stated by the authors, the effect of Cly forcing is non-linear and dependent on the climate scenario. So what does it mean to say that a 5 DU increase in ozone can be attributed to Cly over the 21st century, given that you are looking at a year 2000 climate? I think you're getting at that if ODS concentrations in 2000 were equal to the year 2100 values, we would expect ozone to be 5 DU higher, right?

That is correct; we have amended the sentence to highlight the climate dependence of this value (P8L23).

- P7L15: Please be more explicit here about what the Maycock 2016 paper shows – it looks as though you cite it to back up the statement that stratospheric cooling is GHG scenario dependent, but this has been known for a long time.

The specific reference for the effect of CO2 on stratospheric cooling has been replaced with Manabe and Wetherald (1975) and Shine et al. (2003) (P8L26).

- P7L22-23: Are you referring to the difference between the blue circle and triangle, and the difference between the red circle and triangle? It is hard to read from the figure, but looks like it is ~5 DU for each. That is indeed interesting – it implies that in the upper stratosphere, the climate scenario has little effect on Cly-induced ozone destruction? Why would that be?

We have specifically highlighted the runs we are comparing in the text for clarity (P9L5). While there is some effect of the upper stratospheric climate on the 5DU value, the non-linearity occurs above the level of the stratospheric ozone maximum (see Fig. 3 in Banerjee et al., 2016) and thus causes a small, non linear component effect in the column.

- P8L2: "compare red/blue circles with green circle in Figure 2b" – this sort of statement is useful in interpreting the figures, and I encourage the authors to use more of them.

Where appropriate we have used this terminology to improve the clarity of our discussion.

- P8L6-8: this sentence is confusing; please reword it. Namely, what are you comparing to the upper stratosphere?

This sentence has been reworded to aid clarity. It now reads "While increases in both GHGs and stratospheric Cl_y have acted to decrease $PCO3_{LS}$ in the past, in the future the effects of decreasing stratospheric Cl_y and increasing GHG concentrations will have competing effects on $PCO3_{LS}$. This is in contrast to the upper stratosphere where future decreases in halogenated ODS and increases in GHG concentrations are both projected to lead to higher ozone concentrations."

- P8L12-14: Why? Evolution of ozone precursor emissions in RCP 6.0 due to countries cleaning up their air quality?

It is beyond the scope of this paper to explore the processes driving the tropospheric partial column trends in the transient runs in any detail given that the time-slice experiments do not consider this perturbation and the tropospheric chemistry scheme in the transient experiment being less detailed than in the time-slice experiments. All we can say categorically based on our results is that this decrease is driven by neither ODS nor radiative changes. While ozone precursor emissions are

a possibility, there exist several potential drivers. More work would be required to explore this issue in the future.

- P8L15: How do your results compare with the ACCMIP models? (Young et al., 2013).

This sentence refers to the influence of GHGs (and climate) on tropical tropospheric column ozone only. A comparison to Young et al. [2013] is not possible since that study considered the combined influence of all forcings (including e.g. the chemical impacts of a large increase in CH4 concentration). Moreover, as discussed above, the focus of this paper is on the stratospheric processes and drivers of total column changes given the simplified tropospheric chemistry of the transient simulations. As such, we do not compare the tropospheric trends with other models.

- P816-17: You might want to state that this is expected because ODSs are photolysed in the stratosphere, not the troposphere.

We have modified the sentence to clarify this point (P10L1).

- Table 2: how are the contributions to ozone destruction calculated?

Chemical ozone loss rates are calculated by diagnosing fluxes through each of the ozone destroying cycles included in the model and grouping them by family (e.g. ClOx, HOx, etc.) following the method of Lee et al. (2002) in which the rate of odd oxygen destruction is estimated for different catalytic cycles by determining the rates of their rate-limiting steps. This has been added to the manuscript (P10L21).

- Table 2: You show NOx and HOx-induced ozone destruction, although chemical changes in N2O and CH4 were not included in simulations TS2000_ODS, TS4.5 and TS8.5. . . I think you should state this in the table caption to make it clear that any changes in their rates are radiative effects or buffering by Cly.

This point has been added to the table caption

- P9L17: State how much of a reduction in EESC induces an increase in PCO3_US by 5 DU.

This has been added to the text. The sentence now reads "Comparison of $TS2000_{ODS}$ with TS2000 isolates the effects of future changes in ODSs on $PCO3_{US}$; as discussed in Section 3.2, we find that reductions in ESC from year 2000 values to projected values for year 2100 increase $PCO3_{US}$ abundances by 5 DU (8%)."

- P9L19-20: Ox loss through reactions with Ox? Rather the Chapman cycles?

We use this convention to be consistent with ClOx, HOx and NOx. Furthermore, the Chapman cycle refers to a set of reactions which produce, destroy and inter-convert Ox. We refer only to the reaction of O+O3, the reaction of Ox with Ox which leads to destruction of 2 Ox molecules.

- P9L22: The upper stratosphere warms when GHGs are held constant but Cly is decreased from 2000 to 2100 concentrations. Please clarify this.

This point has been clarified in the manuscript (P11L8). The sentence now reads "The upper stratosphere warms by ~2 K (Figure 4) when GHGs are held constant but ODS concentrations are reduced from year 2000 to year 2100 concentrations, consistent

with the effect of increasing ozone concentrations on upper stratospheric temperatures as discussed by Maycock (2016).”

- P9L24: But as well as temperature effects, HOx and NOx cycles will also be buffered by interactions with Cly. This should also be discussed.

This point has been added to the text, which now reads “Reactions involving HOx and NOx have weaker temperature dependencies and are coupled to Cly concentrations through null cycles and the formation of reservoir species, and thus they show smaller increases.”

- P10L1-18: As mentioned earlier, it would be great if the discussion of ozone drivers came earlier.

As discussed above, we feel the clearest presentation of the results in the paper are to sow the total column projections for the UM-UKCA model, subdivide these changes into the partial column changes to identify key differences between the different altitude regions, and then discuss the mechanisms driving these changes

- Figure 5c is not discussed in the text.

This has been rectified in the text (P12L17, P13L2).

- P11L12-14: How are non-linearities accounted for here?

It was found that a linear fit through the data points used for this calculation had an r^2 value of 0.96, and there was no evidence of any consistent non-linearities, and so they are not considered here.

- P11L16-17: CO is also an important ozone precursor.

CO has been added to the list of ozone precursors (P13L24).

- Figure 2c: In the tropical troposphere, different chemistry regimes are at play, and a lot of information can be lost through zonal averaging. For example, in the tropical Western Pacific region ozone loss via the H2O + O(1D) reaction is very important where solar actinic fluxes and humidity are high. However in other regions, ozone production can dominate due to anthropogenic emissions of ozone precursors (biomass burning etc). I think it would be interesting to somehow resolve figure 2c zonally, and discuss a bit more the chemical changes happening there.

The reviewer makes an important point about the regional effects of short-lived species that affect tropospheric ozone. However, the focus of this study is on the radiative and chlorine drivers of ozone concentrations resulting from changes in long-lived GHGs and halogenated source gases. Assessing zonal asymmetries in the drivers of the tropospheric ozone burden is therefore beyond the scope of this study.

- P12L8-9: State where this is shown (Fig. 2c).

This has been added to the text. The sentence now reads “While reductions in ODS affect tropospheric ozone in the extratropics through STE (e.g. Banerjee et al., 2016), in the tropics, ODS have little impact on tropospheric ozone, with $PCO3_T$ increasing by <1 DU in the $TS2000_{ODS}$ experiment compared to TS2000 (see Figure 2c).”

- P12L11: I would argue that ozone precursors are a major consideration, rather than an additional consideration. . . I think you could look at their effects here too, as from Banerjee et al. (2016) I understand you have simulations available where climate and ozone precursor emissions are perturbed separately and together?

As discussed above, given the simplified chemistry scheme of the transient UM-UKCA simulations and the focus of this paper on the long lived GHGs and halogenated ODS, we feel that further consideration of ozone precursors is beyond the scope of this paper

- How were ozone precursor emissions prescribed in your timeslice simulations? The same as RCP 6.0?

Ozone precursor emissions are prescribed to their year 2000 values in all of the time-slice simulations. Thus, we have not isolated the impact of a perturbation to ozone precursor emissions to year 2100 (RCP6.0) values, since the focus of this study is on the impact of climate and ODSs on column ozone. However, in Sect. 4.3 (final paragraph), we use previous studies [e.g. Revell et al., 2015] which have isolated the impacts of ozone precursors to qualitatively infer their likely role in the evolution of the tropospheric ozone column in our transient simulations.

- P12L17: Also compare with the ACCMIP models in Young et al. (2013).

This reference has been added to the manuscript (P14L22).

- P14L6-7: was CDE fixed or CO2? In the text you say that CDE was fixed, but in the legend on Fig. 7 it says that CO2 was fixed. Please use consistent terminology. I think too that the caption for Fig. 7 should provide a description of the experiments shown.

CDE was fixed as multiple GHGs are considered. The legend in Figure 7 has been corrected to reflect this.

- Figure 7: Why does the simple model overestimate ozone loss between ~1990-2070?

As discussed in the replies to major comments 1 and 4, the simple model, by definition, does not capture all the processes that affect ozone in the transient simulation. While the simple model does overestimate the minimum SCO3 values occurring around year 2000, partly owing to the effects of coincident solar maximum conditions that affect the transient runs but are not included in the simple model, the rate of increase from 2000 to 2040 is similar between the simple model and the transient simulation, highlighting the importance of ODS and CDE changes to projected stratospheric ozone increases in the first half of the 21st century. As we state in the paper, the simple model is not intended to replace fully coupled chemistry climate simulations, but rather to act as a framework for identifying the key drivers of future SCO3 changes. The simple model could be further expanded to include additional terms to represent additional processes which would be likely to improve its quantitative fidelity, and it is hoped that this can be done in the future. However, here we present only an assessment of a simple model constructed using terms we had available given the simulations used in this study and compare it to the fully coupled model to highlight i) that the broad trend of the transient simulation is reproduced in a simple, 2 component model, and ii) there remain differences between the two models highlighting the importance of other compounds and processes.

- Discussion of fig. 7: Non-linearities are not discussed; (Meul et al., 2015) may provide helpful background information here.

The text has been amended to include a discussion of non-linearities

- P15L7: you are talking in terms of the total column, right? Again, I am missing a discussion of the role of tropospheric ozone as an air pollutant – even if lower stratospheric ozone losses are balanced in the total column by tropospheric increases, the result is not great for life in the biosphere because of reduced stratospheric ozone shielding the biosphere from UV-B radiation, and increased tropospheric ozone acting as an air pollutant and GHG.

This has been added to the text (P18L8)

- P15L8-10: Again, I disagree since these were the only factors you looked at, so you cannot discount other factors.

The conclusions of the paper has been amended to highlight that we do not consider here the chemical effects of CH4 and N2O and discuss the role they may have on total column ozone.

- P15L28-30: This was not discussed earlier, please include this discussion in the results section.

This is discussed in the final paragraph of section 4.2 of the manuscript

- Please state where your data are available from.

The transient simulations are available as part of the CCMI initiative through BADC. Any further data are available upon request. We have added a short section on page 29 to state the availability of data.

Technical corrections

- P1L18 “significant differences to” -> “significant differences in”
Corrected

- P2L6: Montreal Protocol and its subsequent amendments -> Montreal Protocol and its subsequent Adjustments and Amendments
Corrected

- P2L21: “over the course of the 21st century perturb” -> “over the course of the 21st century are expected to perturb”
Corrected

- P2L23-24: CFCs are source gases for Cly, N2O is a source gas for NOx and CH4 is a source gas for HOx. Please phrase this more carefully.
Corrected

- P2L26: “increases to the rate constant” -> “increases in the rate constant”
Corrected

- P2L27: “decreases to the rate constant” – as above.
Corrected

- P3L6: define Cly and NOy.
Cly and NOy have been defined when first used

- P3L24 onwards: there is no need to refer to "RCP emissions scenarios" or "RCP scenarios." Calling them RCPs is sufficient.
For clarity we prefer to refer to them as RCP scenarios and so do not feel the text needs to be changed.

- P3L28: "rise" -> "increase"
Corrected

- P5L3: "integration given" -> "integration are given"
Corrected

- P6L16: "discussed in" -> "discussed by"
Corrected

- P7L14: "century is dependent" -> "century are dependent"
Corrected

- P8L5-6: units are in italics.
Corrected

- P9L3: "62 DU" – it says 63 DU in Table 2.
Corrected

- P11L18: NOx: fix subscript.
Corrected

- P11L30: "increase in LNOx at RCP 8.5" -> "increase in LNOx in RCP 8.5"
Corrected

- P12L15: Meinhausen -> Meinshausen
Corrected

- P13L26 "emissions of GHGs" -> "the radiative effects of GHG emissions"
Corrected

- P13L28: dynamic -> dynamical
Corrected

- P15L3: troposphere height -> tropopause height
Corrected

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 N_2O and CH_4 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 Cly 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 PCO3_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 alpha, 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 alpha 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 NOx 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 CH_4 and N_2O.

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 $\frac{\Delta SCO3}{\Delta CDE}$ ***and*** $\frac{\Delta SCO3}{\Delta ESC}$ ***– this has been added to the manuscript.***

Diagnosing the radiative and chemical contributions to future changes in tropical column ozone with the UM-UKCA chemistry-climate model

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Abstract. Chemical and dynamical drivers of trends in tropical total column ozone (TCO3) for the recent past and future periods are explored using the UM-UKCA chemistry-climate model. A transient 1960-2100 simulation is analysed which follows the representative concentration pathway 6.0 (RCP6.0) emissions scenario for the future. Tropical averaged (10°S-10°N) TCO3 values decrease from the 1970s, ~~reaching~~reach a minimum around 2000, and return to their 1980 values around 2040, consistent with the use and emission of halogenated ozone depleting substances (ODS), and their later controls under the Montreal Protocol. However, when the ozone column is subdivided into three partial columns (PCO3) that cover the upper stratosphere (PCO3$_{\mathrm{US}}$), lower stratosphere (PCO3$_{\mathrm{LS}}$) and troposphere (PCO3$_{\mathrm{T}}$), significant differences ~~to~~in the temporal behaviour of the ~~total column are~~partial columns is seen. Modelled PCO3$_{\mathrm{T}}$ values under the RCP6.0 emissions scenario increase from 1960-2000 before remaining ~~steady under this particular emissions scenario~~ approximately constant throughout the 21st century. PCO3$_{\mathrm{LS}}$ values decrease rapidly from 1960-2000, remain ~~steady until around~~ constant from 2000-2050, before gradually decreasing further ~~to~~ from 2050-2100, never ~~recovering~~returning to their 1980s values. In contrast, PCO3$_{\mathrm{US}}$ values decrease from 1960-2000, before ~~rapidly~~ increasing rapidly throughout the 21st century, ~~recovering~~returning to 1980s values by ~2020, and ~~are~~reach significantly higher ~~than 1980s~~ values by 2100. Using a series of idealised UM-UKCA time-slice simulations with ~~varying~~ concentrations of well-mixed greenhouse gases (GHG) and halogenated ODS species set to either year 2000 or 2100 levels, we examine the main processes that drive the PCO3 responses in the three regions, and assess how these processes change under different emission scenarios. Finally, we present a simple, linearised model to describe the future evolution of tropical stratospheric column ozone values based on terms representing time-dependent abundances of GHG and halogenated ODS.

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1 Introduction

Total column ozone (TCO3) ~~values have~~has a direct effect on human health by preventing harmful ultraviolet (UV) radiation from reaching the surface. It is therefore important to gain a quantitative understanding of how TCO3 values may evolve over the 21st century. While ozone mixing ratios are on average highest in the tropical stratosphere, tropical TCO3 values are the lowest of any region outside of the Antarctic ozone hole (World Meteorological Organization (WMO), 2014)~~.~~, due in part to the maximum ozone mixing ratios being found at higher altitudes in the tropics and the tropopause height being higher there than in mid and high latitudes. This fact, combined with the high population of many tropical countries, means it is important to understand the various factors that will affect TCO3 values over the course of the 21st century.

The discovery of the ozone hole by Farman et al. (1985) ultimately led to controls on the emissions of CFCs and other ozone-depleting substances (ODS) through the Montreal Protocol and its subsequent ~~amendments~~Adjustments and Amendments (WMO, 2014). As a result, stratospheric concentrations of inorganic chlorine are expected to decline throughout the 21st century (e.g. Mäder et al., 2010), and stratospheric ozone concentrations in the mid- and high latitudes are projected to ~~recover~~return to their pre-1980s values (Eyring et al., ~~2013~~2013a; WMO, 2014). However, future projections of tropical TCO3 abundances show a large ~~uncertainty~~inter-model range (e.g. Austin et al., 2010; WMO, 2011; 2014), with recent studies indicating that tropical TCO3 ~~values~~ may not ~~recover~~return to pre-1980s values by the end of the 21st century despite reduction in stratospheric halogenated ODS concentrations (e.g. Eyring et al., ~~2013;~~ 2013a; Meul et al., ~~2014).~~ 2016).

In the extra-polar stratosphere, local ozone concentrations are determined by the balance between production and destruction of ozone through gas phase chemical reactions, plus transport into and out of the region of interest (e.g. Brewer and Wilson, 1968; Garny et al., 2011). O_x mixing ratios (where O_x, or odd oxygen, is defined as the sum of ozone (O_3) and atomic oxygen (O)) are determined by sets of photochemical reactions first described by Chapman (1930) plus ozone destroying catalytic cycles involving chlorine, nitrogen, hydrogen and bromine radical species (e.g. Bates and Nicolet, 1950; Crutzen, 1970; Johnston, 1971; Molina and Rowland, 1974; Stolarski and Cicerone, 1974). Unlike in the polar lower stratosphere, heterogeneous processes play only a minor role in determining tropical TCO3 abundances, although this can change after large volcanic eruptions (e.g. Solomon et al., 1996; Telford et al., 2009~~) and could also be affected by~~), in association with aerosol transport within the Asian summer monsoon circulation (Solomon et al., 2016), or as a result of proposed stratospheric aerosol geoengineering schemes (e.g. Weisenstein et al., 2015; Tang et al., 2016).

Changes in anthropogenic emissions ~~over the course of~~during the 21st century are expected to perturb stratospheric ozone chemical cycles involving O_x, ClO_x (Cl+ClO), NO_x ($NO+NO_2$) and HO_x ($OH+HO_2$) in two ways. Firstly, the radiative effects of well-mixed GHGs affect both gas phase kinetics and stratospheric dynamics. Secondly, some GHGs~~, i.e. CFCs,~~

~~N_2O and CH_4, are~~ also act as source gases for reactive ~~ClO_x, NO_x and HO_x~~ species: CFCs are source gases for inorganic chlorine (Cl_y), N_2O is a source gas for reactive nitrogen (NO_y) and CH_4 is a source gas for HO_x.

Cooling of the stratosphere due to increased GHG concentrations, particularly CO_2, increases stratospheric ozone concentrations through both increases ~~to~~in the rate constant for the reaction $O+O_2+M$, leading to an increase of the ratio of O_3 to O, and decreases ~~to~~in the rate constant for the reaction $O+O_3$ (e.g., Barnett et al., 1974; Haigh and Pyle, 1982; Jonsson et al., 2004). In a similar way, the rate constants for the catalytic loss cycles involving NO_x, HO_x and ClO_x radicals are also temperature dependent (e.g. Brasseur and Hitchman 1988; Randeniya et al., 2002; Rosenfield et al., 2002; Stolarski et al., 2015), and so the combined efficiency of these cycles for destroying stratospheric ozone is also affected by GHG-induced stratospheric temperature changes ~~in GHG concentrations~~.

~~The~~Changes to emissions of CFCs, N_2O and CH_4 will alter the concentrations of ClO_x, NO_x, and HO_x radicals, affecting the catalytic cycles that destroy ozone (e.g., Chipperfield and Feng, 2003; Ravishankara et al., 2009). While future stratospheric halogen loadings are expected to decrease throughout the 21st century, emissions of CH_4 and N_2O, which are not regulated in the same way as halogenated ODS, are associated with greater uncertainty. The atmospheric concentration of these species, and by extension future concentrations of HO_x and NO_x radicals, is therefore highly sensitive to assumptions made about their future emissions.

The physical climate response to increases in GHG concentrations is expected to include increasing tropopause height, an acceleration of the Brewer-Dobson Circulation (BDC), and changes in the width of the region of the tropical upwelling ~~region~~in the lower stratosphere (e.g. Butchart et al., 2006, 2010; Garcia et al., 2007; Lorenz and DeWeaver, 2007; Shepherd, 2008; Li and Austin, 2008; Shepherd and McLandress, 2011; Hardiman et al., 2014; Palmeiro et al., 2014). ~~Dynamical changes~~Changes in the strength of the BDC affect ozone concentrations by directly transporting ozone out of the lower stratosphere (e.g. Plumb, 1996; Avallone and Prather, 1996), and by controlling the ~~amount~~abundance of ~~reactive~~ Cl_y, NO_y and HO_x ~~that determine~~, which determines the chemical processing of ozone (e.g. Revell et al., 2012, Meul et al., 2014). In addition to the mean advection of ~~airmasses~~air masses, quasi-horizontal mixing along isentropes is also important for the transport of stratospheric chemical constituents (Hall and Waugh, 1997). However, in the tropics horizontal mixing is relatively weak due to the existence of a sub-tropical transport barrier, the tropical pipe, which acts to some extent to isolate the tropical lower stratosphere from the mid latitudes (Waugh 1996; Neu and Plumb, 1999).

~~Furthermore, changes to emissions of CFCs, N_2O and CH_4 will alter the concentrations of ClO_x, NO_x, and HO_x radicals, affecting the catalytic cycles that destroy ozone (e.g., Chipperfield and Feng, 2003; Ravishankara et al., 2009). While future stratospheric halogen loadings are expected to decrease throughout the 21st century, emissions of CH_4 and N_2O, which are not regulated in the same way as ODS, are associated with greater uncertainty. The atmospheric concentration of these~~

~~species, and by extension future concentrations of HO_x and NO_x radicals, is therefore highly sensitive to assumptions made about their future emissions.~~

Since the photochemical lifetime of ozone is long in the lower stratosphere and short in the upper stratosphere, ~~it is expected that~~ the relative importance of the chemical and dynamical processes described above will vary with altitude, with dynamical changes playing ~~an~~a more important role for ozone in the lower stratosphere and gas phase chemistry ~~controlling ozone concentrations~~a more important role in the upper stratosphere. This makes it challenging to understand the sources of uncertainty and inter-model differences in future tropical ~~total column ozone~~TCO3 trends (e.g. WMO, 2014).

Alongside changes to stratospheric ozone concentrations, tropospheric ozone abundances are projected to change throughout the 21st century due to changes in future emissions of anthropogenic and natural species, particularly ozone precursors (e.g. CO, CH_4, NO_x and VOCs) and changes in climate (e.g. Eyring et al., 2013a; Banerjee et al., 2016; Meul et al., 2016). Changes to emissions of ozone precursors directly affect tropospheric ozone concentrations by affecting chemical production through reactions between NO_x, hydrocarbons and CO, which account for ~90% of local ozone production (Denman et al., 2007). While changes to ozone precursors are not considered in this study, changes in climate can affect tropospheric ozone abundances by changing water vapour, lightning NO_x emissions (LNO_x) and stratosphere-troposphere exchange of ozone (STE) (e.g. Thompson et al., 1989; Eyring et al., 2013a; Young et al., 2013; Revel et al., 2015; Banerjee et al., 2016). These changes are an important consideration when assessing tropical TCO3 trends resulting from changes in GHG and halogenated ODS. It is important to note that while stratospheric column ozone prevents harmful UV radiation reaching the surface, tropospheric ozone is associated with a number of harmful effects on human health, air quality and the environment as it is an air pollutant and GHG (e.g. West et al., 2007; Revell et al., 2015). Therefore, any benefits related to increases in TCO3 resulting from increased tropospheric partial column values could be offset by the negative effects of increased surface ozone concentrations.

To assess the impacts of future anthropogenic emissions on atmospheric chemistry and climate, a number of representative concentration pathway (RCP) scenarios based on different assumptions about future socio-economic development have been developed (van Vuuren et al., 2011). While stratospheric chlorine loadings are predicted to decrease in the future in all RCP emissions scenarios, emissions of CO_2, CH_4 and N_2O are associated with greater uncertainty and hence follow a wider range of pathways between the different RCP scenarios (WMO, 2011, 2014; IPCC, 2013; ~~Meinhausen~~Meinshausen et al., 2011). For example, CH_4 and N_2O emissions are projected to decline during the 21st century in ~~the~~ RCP2.6~~ scenario~~, peak around the year 2040/2080 in ~~the~~ RCP4.5/RCP6.0~~ scenarios,~~ respectively, and ~~rise~~increase monotonically throughout the century in ~~the "business as usual"~~ RCP8.5~~ scenario~~. The multitude of drivers and processes that affect atmospheric ozone abundances motivates the use of chemistry-climate models (CCMs) to explore ~~how column ozone may evolve~~changes in TCO3 over the 21st century under ~~a range of~~the different RCP ~~emissions~~ scenarios (e.g. Eyring et al., ~~2013~~2013a; Iglesias-Suarez et al., 2016).

Here we present results of a modelling study that assesses projected trends in tropical column ozone. The aims of this paper are to: 1) analyse separately the contributions from different altitude regions to future tropical column ozone trends; 2) quantitatively determine the ~~mechanistic~~major chemical and physical drivers of ~~projected~~the modelled partial tropical column ozone trends~~ using a number of idealised CCM simulations~~; and 3) formulate a simple model to estimate future tropical stratospheric column ozone changes and ~~their dependence on~~the contribution from the key drivers ~~discussed~~identified in 2) to these changes. The emphasis here is on the impact of halogenated ODS and the climatic effects of well-mixed GHGs on ozone chemistry and transport. We therefore do not consider the chemical effects of future N_2O and CH_4 emissions, which will also contribute to future tropical column ozone trends (e.g. Butler et al., 2016; Revell et al., 2012) and show differences in their future concentrations across RCP scenarios (Meinshausen et al., 2011). We recognize that the future evolution of tropical ozone will depend, *inter alia*, on the ODS, GHG and tropospheric ozone precursor emissions scenario. Some of these are regulated, some are not and some will respond to climate change. Accordingly, the aim is not to predict the precise evolution of tropical column ozone, but rather explore the contributions from the drivers stated above~~.~~ to future changes over a particular sub-set of scenarios. By breaking down our analysis into different vertical regions within which ozone levels are governed by fundamentally distinct processes, we aim to develop some general understanding of the processes that will affect tropical column ozone throughout the 21st century.

Section 2 describes the CCM simulations used for this study. In section 3, the modelled column ozone trends are discussed and separated into contributions from the upper stratosphere, lower stratosphere and troposphere, before the key drivers of column ozone trends in these separate altitude regions are discussed in section 4. ~~Section 5 presents a discussion on the validity of treating the effects of individual key drivers as additive (i.e. without any co-dependence). In section 6~~In section 5, we produce a simple linear model to describe future tropical stratospheric column ozone changes as a function of GHG and halogenated ODS concentrations. Finally, the results are summarized in section ~~7~~6.

2 Model setup and experimental design

~~We~~For this study, we use version 7.3 of the Met Office's Unified Model HadGEM3-A (Hewitt et al., 2011) coupled with the United Kingdom Chemistry and Aerosol scheme (hereafter referred to as UM-UKCA). The model is run in atmosphere-only mode with a horizontal resolution of 2.5° latitude by 3.75° longitude, 60 vertical levels up to 84 km, and prescribed sea surface temperatures and sea ice extents. For this study, two configurations of UM-UKCA were used which are described below.

~~An~~We use an ensemble of transient simulations following the experimental design of the ~~WCRP~~IGAC/SPARC CCMI REF-C2 experiment, which adopts the RCP6.0 scenario for future GHG and WMO (2011) recommendations for ODS concentrations (Eyring et al., ~~2013) was~~2013b). These simulations were performed using ~~the~~a configuration of UKCA with an extended ~~Chemistry of the Stratosphere (CheS+)~~stratospheric chemistry scheme~~. The REF-C2 experiment adopts the~~

~~RCP6.0 scenario for future GHG and ODS emissions. The CheS+ scheme is an expansion of~~ to that described by Morgenstern et al. (2009~~)~~), in which halogen source gases are considered explicitly, resulting in an additional 9 species, 17 bimolecular and 9 photolytic reactions. ~~CheS+ contains only a simplified~~ The tropospheric chemistry scheme~~.~~ in this configuration of UKCA is relatively simplified. This ~~model~~configuration of UKCA was used for the recent SPARC ~~Assessment of~~ Report on the Lifetimes of Stratospheric Ozone-Depleted Substances, their Replacements and Related Species (SPARC 2013; Chipperfield et al., 2014) and is described in detail in Bednarz et al. ~~(2016). In total, four ensemble members are used in this study: two simulations run from 1960 to 2099 and two simulations run from 1980 to 2080. The four ensemble members have identical time dependent boundary conditions, but differ in their atmospheric initial conditions, thereby providing an estimate of internal atmospheric variability.~~(2016). The model is forced at the lower boundary with sea surface temperatures and sea ice fields taken from a ~~parent coupled atmosphere-ocean HadGEM2-ES integration. All transient simulations~~coupled atmosphere-ocean HadGEM2-ES integration (Jones et al., 2011). In total, four ensemble members are used in this study: two integrations run from 1960-2099, and two integrations run from November 1980 to December 2080. The latter two ensemble members were initialised using different atmospheric initial conditions taken from a supporting perpetual year 1980 integration. The four ensemble members have identical time-dependent boundary conditions, thereby providing an estimate of the contribution from internal atmospheric variability to simulated temporal variability and trends. All transient integrations used in this study include the effects of the 11-year solar cycle in both the radiation and photolysis schemes.

The transient simulations described above include both the radiative and chemical effects of ~~changing~~time-varying anthropogenic ~~emissions.~~source gases, specifically CO_2, CH_4, N_2O and halogenated ODS. In order to separate the relative radiative and chemical contributions to future tropical ozone differences, the transient simulations were supplemented by time-slice ~~simulations~~integrations performed using ~~the Chemistry~~a configuration of ~~the Stratosphere and Troposphere (CheST)~~UKCA with a coupled stratosphere-troposphere chemistry scheme~~.~~ as described by Banerjee et al. (2014). This scheme ~~is~~includes a ~~combination of the UM-UKCA~~more detailed tropospheric chemistry scheme (O'Connor et al., 2014) and the original UM-UKCA stratospheric ~~(~~chemical scheme described by Morgenstern et al~~.,~~. (2009~~) chemical schemes, and is described in detail in Banerjee et al. (2014).~~). Six time-slice experiments were performed ~~using~~ with this configuration of UM-UKCA that include different prescribed SSTs and sea ice, GHG and halogenated ODS concentrations~~. The combinations~~ (Banerjee et al., 2016). These include a set of ~~forcings used for each time-slice integration given~~simulations in ~~Table 1. Each time-slice was run for 20 years, with~~which the ~~first 10 years discarded as spin-up for the model.~~

~~In order to separate the~~physical climate state alone (e.g. SSTs, sea ice, radiative ~~and chemical responses of ozone abundances to changes in anthropogenic emissions, only the radiative impacts of variations in~~ effects of GHG concentrations ~~and only the chemical effects of changes to ODS concentrations are considered in these simulations. I.e. while differences in GHG concentrations directly affect temperature and circulation, they do not affect the concentration of reactive radical species~~

~~(e.g. Cl_y, NO_y and HO_x), while the radiative effects of ODS changes are not included. In this study we consider the radiative impact of a large number of GHG species (CO_2, CH_4, N_2O, CFCs) and assume that the dominant driver of chemical changes is changes to ODS loadings. In this way, the chemical impact of changing N_2O and CH_4 emissions is not considered here. The impact of changing GHG concentrations is expressed in terms of differences in Carbon Dioxide Equivalent (CDE; IPCC, 2007).~~

~~The time-slice experiments include 3 pairs of simulations with a physical climate state (e.g SSTs, sea ice, radiative GHG concentrations) commensurate with either~~) is perturbed from a year 2000 ~~or 2100 conditions, with~~baseline to year 2100 conditions taken from either the RCP4.5 or RCP8.5 ~~scenarios. Each~~ scenario. Note that when perturbing the physical climate state, GHG concentrations are not perturbed in the chemistry scheme i.e. the chemical impacts of changing N_2O, CH_4 and CFCs are not considered. The chemical effects of ODS in particular are considered as a separate perturbation: in the chemistry scheme, each pair of experiments in turn uses ~~chemical~~halogenated ODS loadings for either the year 2000 or 2100. ~~The~~The RCP4.5 scenario is used to determine the year 2100 halogenated ODS levels, although the exact scenario followed is arbitrary since all RCPs show similar projections for future ODS emissions (Meinhausen et al., 2011). The resulting set of time-slice experiments are named accordingly~~, e.g.~~ to reflect the climate condition and chemical ODS loadings, e.g. TS2000$_{ODS}$ includes year 2000 climate conditions and year 2100 chemical ODS loadings, while TS4.5 includes year 2100 climate conditions following the RCP4.5 scenario and year 2000 ODS loadings (see Table 1). ~~A full description of these simulations is provided in Banerjee et al. (2014).~~In all time-slice experiments chemical concentrations of N_2O and CH_4 use prescribed year 2000 concentrations from RCP6.0 as a lower boundary condition, and thus their chemical effects are not considered in this study. In principle, further time-slice experiments could be performed to also explore the chemical impacts of changes in tropospheric ozone precursors, unregulated short-lived halogen compounds and N_2O and CH_4 changes. However, owing to limitations in computational resource we focus our attention on the effects of ODS and GHG-driven changes in climate. Each UM-UKCA time-slice experiment was run for 20 years, with the first 10 years discarded as spin-up for the model.

The ~~use~~design of the time-slice ~~simulations with different combinations~~experiments allows for a quantitative separation of ~~near-present day~~the radiative and ~~end~~chemical effects of some of the known drivers of stratospheric ozone changes over the 21st century~~ forcings enables the processes that drive projected~~, which can then aid in the interpretation of the simulated time-dependent changes in tropical column ozone ~~trends~~ in the transient ~~experiments to be determined.~~ integrations. We purposefully use time-slice experiments with different combinations of forcings to those in the transient simulations (time-slices are run for RCP 4.5 and 8.5 while the transient simulation is run for RCP 6.0), so that we can assess linearities in the ozone response to both ODS and GHG changes.

Throughout the remainder of this study the impact of changing GHG concentrations is expressed in terms of differences in Carbon Dioxide Equivalent (CDE; IPCC, 2007), while ODS will be used to refer only to the halogenated ozone depleting substances, and does not include N_2O, itself an important ozone depleting substance (e.g. WMO, 2014; Ravishankara et al., 2009). ODS concentrations have been calculated using the equivalent stratospheric chlorine (ESC) definition of Eyring et al. (2007), where ESC = $Cl_y + \alpha Br_y$, and $\alpha = 60$.

3 Modelled column ozone trends

The analysis presented in this study focuses on area weighted averages over 10°~~-~~S-10°~~-~~N. While previous studies of tropical ozone trends have used a broader region to define the tropics, typically from 25°~~-~~S-25°~~-~~N (e.g. Austin et al., 2010; Eyring et al., 2010; Meul et al., 2014), Hardiman et al. (2013) show that, in an ensemble of CMIP5 models following the RCP8.5 scenario, as the magnitude of the tropical upwelling mass flux is projected to increase over the 21st century, the width of the region of upwelling narrows at altitudes below 20 hPa. In order to avoid the impacts of changes to the width of the region of tropical upwelling resulting from increases in GHG concentrations, in this study we use a narrower definition of the tropics. However, the results presented in this study ~~are~~were not changed significantly when a broader definition of the tropics (30°~~-~~S-30°~~-~~N) ~~is~~was used.

In this section, we first describe the changes in tropical total column ozone (defined herein as 0-48 km) and then the partial column trends for the upper and lower stratosphere and the troposphere. The processes driving these changes are then explored in Section 4.

3.1 Total column ozone differences

Figure 1 shows tropical averaged TCO3 anomalies relative to a ~~year 2000±5 averaged value~~baseline period of 1995-2005 from 1960 to 2100 for each individual ensemble member (grey lines) and the ensemble mean 11-year running mean (black line). ~~The ensemble mean 11-year running mean TCO3 abundances in the transient~~The baseline period 1995-2005 is chosen so that the transient simulations can be directly compared to the year 2000 time-slice experiments (see Section 2). Also shown in Figure 1 are tropical averaged TCO3 anomalies from version 2.8 of the Bodeker Scientific total column ozone dataset (purple line; Bodeker et al., 2005). There is generally good agreement between the modelled tropical column ozone anomaly values and the Bodeker dataset, specifically with regards to the long-term changes during the period the model and observations overlap, and the magnitude of interannual variability. The ensemble mean 11-year running mean TCO3 abundances in the transient UM-UKCA simulation are generally anti-correlated with the long-term changes in stratospheric chlorine levels, consistent with other studies (e.g. Eyring et al., ~~2013~~2013a). There is a ~~sharp decrease~~decline in tropical TCO3 of ~6 DU from the mid-1970s to 1990, coincident with increases in stratospheric Cl_y concentrations resulting from the emission of halogenated ODSs. TCO3 values remain relatively low from 1990 to 2010, before ~~more~~ gradually ~~recovering~~returning to 1980s values by ~2040 and to 1960s values by ~2050, after which they remain relatively constant from 2050-2090. Beyond 2090 there is evidence for a further ~~decreases~~decrease, bringing column values once again below their 1980s values. This behaviour is broadly consistent with previous studies (e.g. Oman et al., 2010; Eyring et al., ~~2013; Meul et al., 2014).~~2013a; Meul et al., 2014), with the main exception being that while other studies show an increase in tropical TCO3 over the first half of the 21st century, they do not generally indicate a return to 1980s values. Results from the UM-UKCA transient simulations show that tropical TCO3 values may return to pre-1980s values for part of the 21st century, but by the end of the century will begin to decrease again. Superimposed on the TCO3 11-year running mean is the signal of the 11-year solar cycle, which leads to variations of <5 DU between solar maximum and minimum. The large degree of natural variability simulated in the model highlights the difficulties in assessing ozone trends and return dates from relatively short observational records (as discussed ~~in~~by Harris et al., 2015).

The time-slice experiments, plotted in Figure 1 as discrete points (circles and triangles), show the dependence on the RCP scenario of modelled year 2100 tropical TCO3 values. Tropical TCO3 increases by around 12 DU when stratospheric Cl_y loadings are decreased from their year 2000 values to year 2100 values under fixed year 2000 GHG conditions (TS2000 - $TS2000_{ODS}$, shown by the difference ~~in~~between the green symbols in Figure 1). However, the same decrease in stratospheric Cl_y abundances leads to slightly smaller increases in tropical TCO3 when future changes in climate are also included according to the RCP4.5 or RCP8.5 scenario (11 DU for TS4.5 – $TS4.5_{ODS}$ and 10 DU for TS8.5 – $TS8.5_{ODS}$, shown by the differences between the blue and red symbols in Fig. 1, respectively). We explore the processes controlling these changes in Section 4.

The ~~climate dependence of the~~ effect ~~of a reduction in ODS results from the temperature dependence of ClO_x radical chemistry~~ on ~~ozone destruction (e.g. Haigh and Pyle, 1982). The effect~~ tropical TCO3 of future ~~radiative~~climatic changes resulting from increases in GHGs~~, and the associated changes in climate state, on TCO3~~ is ~~explored~~seen by comparing the TS4.5 and TS8.5 time-slice ~~simulations~~integrations with TS2000. Under a more moderate increase in GHG concentrations (TS4.5 - TS2000, compare green ~~to~~and blue circles Figure 1), tropical TCO3 increases by 4.0 DU between year 2000 and 2100, while under a much larger GHG concentration change (TS8.5 - TS2000, compare green ~~to~~and red circles in Figure 1), tropical TCO3 values show no change, indicating a non-linear response to the magnitude of GHG forcing (Banerjee et al., ~~2016).~~2016). The causes of this are discussed further in Section 4.

3.2 Partial column ozone differences

Projected trends in tropical ozone concentrations show a complex vertical structure~~.~~ (WMO, 2014). In this section we assess modelled changes in tropical ozone partial columns~~ (PCO3)~~ for the upper stratosphere (30-48 km; $PCO3_{US}$), lower stratosphere (tropopause to 30 km; $PCO3_{US}$) and troposphere ($PCO3_T$). The 30 km boundary between the lower and upper stratosphere corresponds to an approximate pressure of 15 hPa. This level is chosen as an approximation for the transition region between ozone being predominantly under photochemical control in the upper stratosphere and predominantly under dynamical control in the lower stratosphere. Note that we ~~employ a scenario consistent~~take into account any changes in tropopause height~~ to calculate the partial columns~~, as defined by the lapse rate tropopause (WMO, 1957~~)~~), when calculating the partial columns within each experiment, rather than using a fixed ~~height definition~~altitude. Thus, any changes in tropopause height will affect the lower stratosphere and tropospheric partial columns even if the vertical distribution of ozone concentrations is unchanged.

PCO3$_{US}$ values decrease by around 4 DU from 1960 to the late 1990s (Figure 2a), consistent with the increasing stratospheric Cl$_y$ concentrations over this period. From around 2000 onwards, PCO3$_{US}$ values increase rapidly due to a combination of decreased stratospheric Cl$_y$ concentrations and the GHG-induced stratospheric cooling effect, returning to 1980 values by ~2020, and 1960 values by ~2040. From 2040, PCO3$_{US}$ values continue to increase to around 3-4 DU above their 1960s values by 2100 – the well-known ozone "super recovery" effect (Chipperfield and Feng, 2003).

The time-slice experiments show that an increase in PCO3$_{US}$ values of ~5 DU can be attributed to Cl$_y$ changes over the 21st century (calculated as the difference between the green symbols in Figure 2a~~).~~), although the exact magnitude of this increase is dependent on the background climate, as discussed above. As well as responding to changes in stratospheric Cl$_y$, PCO3$_{US}$ values in the late 21st century ~~is~~are dependent on the ~~RCP~~GHG emissions scenario~~ followed, which determines~~, since CO$_2$ is the ~~magnitude~~main driver of stratospheric cooling (e.g. Manabe and Wetherald, 1975; Shine et al., 2003). The~~Maycock, 2016).~~ ~~PCO3$_{US}$ for~~ TS4.5 and TS8.5 experiments, which consider only the ~~radiative effects of changes~~differences in ~~GHGs~~physical climate resulting from GHG increases, both show higher TCO3 values than TS2000 (+5 DU for TS4.5, comparing green and blue circles in Fig. 2a, and +12 DU for TS8.5~~).~~, comparing green and red circles). These results can be used to calculate an approximate change in tropical PCO3$_{US}$ ~~abundance~~ per unit change in CDE ~~and EESC concentration, which gives values~~ of $\frac{\Delta PCO3_{US}}{\Delta \mathrm{CDE}}$ ~~≈ 0.02 DU ppmv^{-1} and~~ ≈ 0.02 DU ppmv^{-1} and per unit change in ESC of ~~$\frac{\Delta PCO3_{US}}{\Delta EESC} \approx -1.72$ DU ppbv^{-1}.~~ $\frac{\Delta PCO3_{US}}{\Delta \mathrm{ESC}}$ ≈ -1.72 DU ppbv^{-1}. These ~~results~~relationships indicate that over the recent past upper stratospheric ozone depletion resulting from increased Cl$_y$ concentrations has in part been offset by radiative cooling resulting from increased GHG concentrations~~,~~ (consistent with Shepherd and Jonsson, 2008), and that in the future both increased GHG concentrations and reduced stratospheric Cl$_y$ will result in increases in upper stratospheric ozone concentrations. However, as discussed for TCO3, the impact of ODS changes on upper stratospheric partial column abundance is dependent on GHG concentrations (compare blue/red circles with blue/red triangles in Figure 2a).

As was found in the upper stratosphere, the modelled historical trend in PCO3$_{LS}$ is strongly negative, with a decrease of ~6 DU from 1960 to the late 1990s (Figure 2b). However, the projected future ~~trends~~trend in PCO3$_{LS}$ ~~differ~~differs greatly from ~~those~~that in the upper stratosphere. From 2000 to ~2050, modelled PCO3$_{LS}$ abundances remain approximately steady, before~~ again~~ decreasing during the latter half of the 21st century.

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The time~~-~~-slice experiments demonstrate the competing effects of decreasing stratospheric Cl_y and ~~the radiative~~ changes ~~following~~in physical climate from increasing GHG concentrations on ~~ozone in the tropical lower stratosphere~~$PCO3_{LS}$ over the ~~course of the~~ 21st century. As in the upper stratosphere, projected decreases in stratospheric Cl_y result in ~~increases to~~ an increase in $PCO3_{LS}$ ~~values~~between year 2000 and 2100 (compare green triangle and circle in Figure 2b). However, ~~radiative~~ changes ~~following increases in~~to the physical climate from increased GHG concentrations lead to decreases in $PCO3_{LS}$ ~~values~~ (compare blue/red circles with green circle in Figure 2b). ~~-~~For changes in GHGs alone, the magnitude of the ~~lower stratospheric partial column ozone~~$PCO3_{LS}$ response increases with the magnitude of the CDE perturbation (-4 DU for TS4.5-TS2000, -16 DU for TS8.5-TS2000). As for upper stratospheric partial column values, changes to tropical $PCO3_{LS}$ values per unit change in CDE and ~~EESC~~ESC concentrations can be calculated, ~~-~~giving ~~$\frac{\Delta PCO3_{LS}}{\Delta CDE} \approx -0.03\ DU\ ppmv^{-1}$~~ $\frac{\Delta PCO3_{LS}}{\Delta \mathrm{CDE}}$ ≈ -0.03 DU $ppmv^{-1}$ and ~~$\frac{\Delta PCO3_{LS}}{\Delta EESC} \approx -1.92\ DU\ ppbv^{-1}$. In comparison to the upper stratosphere, while past~~ $\frac{\Delta PCO3_{LS}}{\Delta \mathrm{ESC}}$ ≈ -1.92 DU $ppbv^{-1}$. While increases in both GHGs and stratospheric Cl_y ~~and GHG concentrations~~ have ~~led to decreased lower stratospheric ozone concentrations~~acted to decrease $PCO3_{LS}$ in the past, in the future ~~ozone recovery resulting from decreased~~the effects of decreasing stratospheric Cl_y and increasing GHG concentrations will ~~compete with ozone~~have competing effects on $PCO3_{LS}$. This is in contrast to the upper stratosphere where future decreases ~~resulting from increased~~in halogenated ODS and increases in GHG concentrations~~.~~ are both projected to lead to higher ozone concentrations. As was seen for the upper stratosphere, the $PCO3_{LS}$ response to a given ~~decrease~~change in ODS is also 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~~).~~, see Figure 2b).

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From 1960 to 2000, the tropical tropospheric partial ozone column ($PCO3_T$) increases by approximately 5 DU (Figure 2c), then remains constant until 2040, before increasing again by ~2 DU by 2060. There is some ~~evidence~~suggestion from the transient simulations that for the RCP6.0 emissions scenario tropical ~~tropospheric partial ozone column~~$PCO3_T$ values decrease to year 2000 values during the final decade of the 21st century. The time-slice experiments indicate that tropical ~~tropospheric column ozone is~~$PCO3_T$ values are relatively insensitive to ~~differences~~changes in ~~GHGs~~the physical climate state alone for the two RCP scenarios considered here, with values in TS4.5 and TS8.5 both increasing by ~5 DU. As expected, tropical ~~tropospheric column ozone~~$PCO3_T$ shows no significant response to changes in ODS concentrations irrespective of the GHG loading~~. Note~~, as the long-lived halogenated ODS species are not oxidised until they reach the stratosphere. However, we remind the reader that by design the time-slice simulations do not explore the ~~role~~chemical roles of tropospheric CH_4, NO_x and volatile organic compounds (VOCs~~),~~) emissions, which are likely to be important drivers of tropospheric ozone changes in the transient simulations; ~~their likely roles are~~this is discussed further in Section 4.3.

4 Drivers of column ozone changes

~~As discussed above, both ODS and GHG concentrations drive ozone~~ The results in Section 3 show that changes in ~~the upper stratosphere, lower stratosphere and troposphere. However, while decreases in ODS result in increases in ozone throughout the stratosphere, increases in GHGs drive increases in upper~~ stratospheric Cl$_y$ and the physical climate effects of GHGs have distinct impacts on partial column ozone~~, but decreases in lower stratospheric partial column ozone. The contrasting impacts of these forcings as a function of~~ in different altitude ranges. These behaviours reflect the various chemical and transport processes that form the dominant control on tropical ozone abundances in different regions. In the following sections the major mechanisms operating in each of the three partial column regimes are explored.

4.1 Upper Stratosphere

As discussed in Section 3.2, PCO3$_{US}$ values are projected to ~~recover~~return quickly to pre-1980 values over the next few decades and ~~to~~ continue to increase throughout the 21st century, leading to super-recovery of ~~the partial column~~PCO3$_{US}$ by 2100 (Figure 2a). This can be seen further in Figure 3, which shows annual mean PCO3$_{US}$ values plotted as a function of stratospheric ESC at 45 km, for both the time-slice and transient simulations. Data for the transient integrations covering 1960-2100 are shown as crosses, with different colours denoting different 20 year periods. From 1960 to 2000, as ESC concentrations rapidly increase by ~3 ppbv, PCO3$_{US}$ abundances decrease by ~3 DU. From 2000-2100, as ESC concentrations decrease, PCO3$_{US}$ abundances are projected to increase, but the trend from 2000 to 2100 does not retrace the trend from 1960 to 2000. Instead, the transient integrations indicate a larger change in PCO3$_{US}$ per unit change in ESC in the future compared to over the past owing to the higher background GHG concentrations.

The time-slice experiments can be used to quantify the separate and combined effects of GHG-induced changes in climate and the chemical effects of ODS on PCO3$_{US}$ changes. The upper rows in Table 2 give values for the TS2000, TS2000$_{ODS}$, TS4.5 and TS8.5 simulations of PCO3$_{US}$ abundances, chemical loss of O$_x$ through reactions with each of the key chemical families (halogens, HO$_x$, NO$_x$, and Ox), chemical production and O$_x$ lifetime. ~~The simulated upper stratospheric partial ozone column under near present day conditions (TS2000) is 62 DU.~~Chemical loss of O$_x$ is calculated following Lee et al. (2002) with the total rate of O$_x$ destruction calculated as the sum of the rates of each chemical ozone loss cycle included in the model chemical scheme. As discussed above, N$_2$O and CH$_4$ concentrations are kept constant in the chemical scheme in all time-slice simulations, and thus any change in NO$_x$ and HO$_x$-induced ozone destruction result only from chemical feedbacks through coupling to temperature or to Cl$_y$ reactions. The simulated PCO3$_{US}$ under near present day conditions (TS2000) is 63 DU. Net chemical loss is 48 DU day^{-1}, with the major loss being due to catalytic cycles involving NO$_x$ (39%), with smaller contributions from HO$_x$ (22%), halogens (20%) and O$_x$ (19%). The average chemical lifetime of ozone in the tropical upper stratosphere (calculated as the burden divided by net chemical loss) is 1.3 days. These results are consistent with previous studies (e.g. WMO, 1998; Grooß et al., 1999; Meul et al., 2014).

~~PCO3$_{US}$, as a function of stratospheric EESC at 45km, for both the time-slice and transient simulations is shown in Figure 3. Annual mean values for the transient simulations covering 1960-2100 are shown as crosses, with different colours denoting different 20 year sections of the simulations. From 1960 to 2000, as EESC concentrations rapidly increase by ~3 ppbv, PCO3$_{US}$ abundances decrease by ~3 DU. From 2000-2100, as EESC concentrations decrease, PCO3$_{US}$ abundances are projected to increase, although the trend from 2000 to 2100 does not retrace the trend from 1960 to 2000, as highlighted by the shaded points for the transient simulations. Instead, the transient simulations indicate a larger change in PCO3$_{US}$ per unit change in ODS in the future compared to the past owing to the increasing GHG concentrations (i.e. the "super-recovery" of ozone).~~

~~The time-slice experiments can be used to quantify the radiative effects of GHG increases and the chemical effects of ODS on PCO3$_{US}$ changes.~~ Comparison of TS2000$_{ODS}$ with TS2000 isolates the effects of future changes in ODSs on PCO3$_{US}$; as discussed in Section 3.2, we find that reductions in ~~EESC~~ESC from year 2000 values to projected values for year 2100 increase PCO3$_{US}$ abundances by 5 DU (8%). Table 2 shows that net chemical O_x loss in TS2000$_{ODS}$ is reduced by 5% compared to TS2000, driven predominantly by large decreases in O_x loss through catalytic cycles involving halogens, which are reduced by 63%. O_x loss through reactions with HO_x, NO_x and O_x all increase, predominantly due to the increase in ozone concentrations, but also due to temperature changes, which are themselves a response to increases in ozone (e.g. Maycock, 2016).~~Shine et al., 2003).~~ The upper stratosphere warms by ~2 K (Figure 4) when GHGs are held constant but ODS concentrations are reduced from year 2000 to year 2100 concentrations, consistent with the ~~multi-model results presented~~effect of increasing ozone concentrations on upper stratospheric temperatures as discussed by Maycock (2016). The reaction $O+O_3$ has a strong temperature dependence and becomes faster at higher temperatures, thereby further increasing O_x loss in TS2000$_{ODS}$ relative to TS2000. Reactions involving HO_x and NO_x have weaker temperature dependencies~~,~~ and are coupled to Cl_y concentrations through null cycles and the formation of reservoir species, and thus they show smaller increases.

In addition to the ~~reductions~~projected reduction in ~~EESC~~halogenated ODSs, the cooling of the stratosphere induced by increased GHG concentrations (mainly CO_2) will be a major driver of future PCO3$_{US}$ changes. Comparison of TS8.5 with TS2000 quantifies the impact of GHG changes alone on PCO3$_{US}$. As the chemical lifetime of O_x is short in the upper stratosphere, transport changes are expected to have a relatively minimal effect on projected ozone trends. Instead, PCO3$_{US}$ changes between TS2000 and TS8.5 are driven by the response of reaction rates to the simulated temperature changes. The tropical upper stratosphere in TS8.5 is ~11 K cooler than in TS2000 (see Banerjee et al., 2016). This leads to a PCO3$_{US}$ increase of 12 DU (21%), which is driven predominantly by a decrease in the reaction $O+O_3$, but also by a change in partitioning of O_x due to the acceleration of the reaction $O+O_2+M \rightarrow O_3+M$ (Jonsson et al., 2004; Banerjee et al., 2016).

The relationship between PCO3$_{US}$ and upper stratospheric temperature for the transient and time-slice experiments is shown in Figure 4. From 1960 to 2000, temperatures and PCO3$_{US}$ both decrease. ~~The ozone decreases are~~During this period the decrease in $PCO3_{US}$ is driven predominantly by increasing ODSs, ~~which deplete ozone chemically~~as described above. Decreased ozone concentrations in turn reduce upper stratospheric heating, thereby reducing temperatures (e.g. Forster and Shine, 1997; Shine et al., 2003). From 2000 to 2100, as temperatures decrease further, mainly due to cooling from ~~increased GHG~~increasing CO_2 abundances, ozone concentrations increase, driven predominantly, as discussed above, by a reduced rate for the reaction of $O+O_3$ and decreased ODS concentrations. These increases in ozone offset part of the stratospheric cooling due to rising ~~GHG~~CO_2 concentrations (Maycock, 2016). The impact of temperature on $PCO3_{US}$ can be isolated by fitting lines through the sets of time-slice experiments with the same ODS loadings (i.e. TS2000, TS4.5 and TS8.5). We find that the relationship between $PCO3_{US}$ and upper stratospheric ~~ozone column increases by ~~~temperature is approximately $\frac{\Delta PCO_{US}}{\Delta T_{US}} = 1$ DU K^{-1}, which, when combined with decreasing ODS, drives the super~~-~~-recovery of $PCO3_{US}$.

4.2 Lower Stratosphere

In comparison to the upper stratosphere, the chemical lifetime of O_x in the tropical lower stratosphere is long (>1 month, see lower rows in Table 2), so dynamical processes play a much more important role in determining ozone abundances there. A strengthening of the BDC, which is projected to occur in the future in response to ~~increasing~~increases in GHGs (e.g. Shepherd and McLandress, 2011; Hardiman et al., 2014; Palmeiro et al., 2014), would therefore have a significant effect on tropical lower stratospheric ozone. We use the Transformed Eulerian Mean residual vertical velocity ($\overline{w}^*$; Andrews et al., 1987) at 70 hPa as a measure of the strength of the advective part of the BDC~~.~~ in the lower stratosphere. In the transient ~~REF-C2~~ simulations, the annual and tropical (10ºS-10ºN) mean $\overline{w}^*$ at 70 hPa increases by around ~~20~~40% from ~0.20 mm s^{-1} in 1960 to ~0.~~24~~28 mm s^{-1} in 2100.

Consistent with the important role of the BDC in determining tropical lower stratospheric ozone abundances, there is a strong negative correlation (~~r = -~~R = -0.76) between annual mean $PCO3_{LS}$ and $\overline{w}^*$ values at 70hPa (Figure 5a). By plotting ~~$\overline{w}^*$~~$\overline{w_{70}^*}$ vs. CDE concentration as a function of time for the transient experiment (Figure 5b~~)~~), and by comparing across the time-slice ~~runs~~experiments with constant ODS ~~loadings~~loading (i.e. TS2000, TS4.5 and TS8.5~~)~~), an approximate value for the acceleration of the BDC per unit increase in CDE can be calculated. From these experiments, a value of $\frac{\Delta \overline{w_{70}^*}}{\Delta \mathrm{CDE}} \approx 2\mathrm{x}10^{-4}$ mm s^{-1} $ppmv^{-1}$ is calculated. ~~By comparison,~~The strong negative relationship between $PCO3_{LS}$ and $\overline{w_{70}^*}$, and in concert the positive relationship between $\overline{w_{70}^*}$ and CDE concentration, combine to give a negative relationship between $PCO3_{LS}$ and CDE concentration, as shown in Figure 5c and quantified in Section 3.2.

The chemical effects of ~~changes in ODS concentrations~~changing ODSs also impact on the modelled BDC strength~~ of the BDC by~~. The TS2000 and $TS2000_{ODS}$ experiments are used to quantify this relationship as $\frac{\Delta \overline{w_{70}^*}}{\Delta ESC} \approx 5.4\mathrm{x}10^{-3}$ mm s^{-1} $ppbv^{-1}$.

This indicates that, per molecule, ODS increases have a greater effect on the BDC than GHGs. Previous work using the UM-UKCA model has indicated that an acceleration in stratospheric circulation, particularly the lowermost branch of the BDC, is to be expected from increased springtime polar lower stratospheric ozone depletion and the resulting increase in meridional temperature gradients (Keeble et al., 2014; Braesicke et al., 2014). Our results also corroborate the findings of Polvani ~~and Wang~~et al. (2017) who highlight the dominant impact of ODS on tropical lower stratospheric temperature and ozone through changes in tropical upwelling between 1960-2000. Results from this study suggest that reduced future ~~recovery of~~ polar lower stratospheric ozone depletion following reduction in ODS concentrations will act to slow the BDC, partly offsetting the acceleration expected due to increased GHG concentrations.

The impact of ODS changes on the speed of the BDC, along with the temperature dependence of the ozone depleting chemistry and the influence of upper stratospheric ozone shielding on the lower stratosphere, result in a non-linear dependence of the PCO3$_{LS}$ response to ODS on GHG loading, as was found in the upper stratosphere. In the time-slice experiments, the effect of the year 2000 to 2100 decrease in ODS on PCO3$_{LS}$ is: +7 DU for TS2000$_{ODS}$ - TS2000, +6 DU for TS4.5$_{ODS}$ - TS4.5 and +4 DU for TS8.5$_{ODS}$ – TS8.5 (compare circles and triangles of the same colour in Figures 5b and 5c). As described above, decreasing ODS concentrations lead to a deceleration of the BDC and an increase in PCO3$_{LS}$. However, as the stratosphere cools the increase in overhead ozone column reduces photolysis rates in the lower stratosphere, slowing ozone production and acting to decrease PCO3$_{LS}$, as discussed above. Together these opposing mechanisms explain the difference in the PCO3$_{LS}$ response to ODSs changes under different GHG concentrations.

The combined influence of GHGs and ODS on the strength of tropical upwelling can largely explain the three distinct periods of behaviour in tropical PCO3$_{LS}$ seen in Figure 2b. Firstly, between 1960-2000, the partial column shows the largest rate of change as the effect of GHGs and ODS on tropical upwelling reinforce one another, both strengthening the tropical upwelling and reducing PCO3$_{LS}$~~.~~, while increasing stratospheric Cl_y concentrations also enhance chemical ozone depletion. Secondly, between 2000-2040~~,~~ increasing GHG concentrations lead to an acceleration of the BDC acting to reduce PCO3$_{LS}$ values while decreasing ODS ~~reductions (~~concentrations slow the BDC and ~~the associated polar ozone recovery)~~decrease chemical O_x loss (Figure 5), and ~~increasing GHGs compete in their effects on tropical upwelling and the partial column, which~~as such PCO3$_{LS}$ remains relatively constant during this time. Finally, between 2040-2100, by which time ~~the ODS influence on~~further changes in ozone ~~has already been~~and the BDC due to ODSs are reduced significantly, the effect of increasing GHGs on tropical upwelling dominates and PCO3$_{LS}$ values ~~again~~ show a ~~decreasing~~negative trend.

~~-~~Finally, we note that in addition to changing to the strength of the BDC, increasing GHG concentrations also affect PCO3$_{LS}$ values by decreasing chemical production as a result of increased overhead column ozone (see Section 4.1). Table 2 shows how O_x production in the lower stratosphere responds to changes in ODS and CDE concentrations. Compared to TS2000, lower stratospheric O_x production in TS2000$_{ODS}$ and TS8.5 has decreased, consistent with the increased partial column abundances in the upper stratosphere in these simulations. Using this information we can calculate the response of lower stratospheric O_x production to changes in upper stratospheric partial column abundance; we estimate that tropical lower stratospheric O_x production will decrease by 0.1 DU day^{-1} for each additional DU of ozone in the upper stratosphere.

4.3 Troposphere

The primary factors affecting future tropospheric ozone are likely to be changes in the emission of ozone precursors (CO, CH_4, NO_x and VOCs) and changes in climate. Changes in climate can affect tropospheric ozone abundances in several ways, including changes in water vapour amounts, lightning NO_x emissions (LNO_x) and stratosphere-troposphere exchange of ozone (STE) (e.g. Thompson et al., 1989; Young et al., 2013; Banerjee et al., 2016). Future ODS-driven stratospheric ozone recovery is also projected to increase tropospheric ozone abundances through STE (e.g. Zeng and Pyle, 2003; Banerjee et al., 2016). Here, we first use the time-slice simulations to deduce the role of climate change and ozone recovery on future tropospheric column ozone changes. Then, we discuss the likely drivers of the partial column evolution between 1960-2000 in the transient simulations, where changes in ozone precursors must also be considered.

~~Climate change resulting~~Changes in the physical climate from ~~changing~~increased concentrations of ~~CO_2~~GHGs in the TS4.5 and TS8.5 experiments ~~enhances~~enhance tropical tropospheric column ozone by around 4 DU relative to TS2000. The increases are driven primarily by LNO_x, which increases by 2 and 4.7 Tg(N) yr^{-1} under the RCP4.5 and RCP8.5 scenarios, respectively (Banerjee et al., 2014). In fact, a further sensitivity experiment in which the climate is allowed to change according to ~~the RCP8~~TS8.5~~ forcings~~, but LNO_x ~~is~~values are kept fixed at TS2000 values (not otherwise discussed; see Banerjee et al., 2014) shows a 3 DU decrease in tropospheric column ozone. ~~The~~This reduction ~~arises~~results from increases in tropospheric humidity under a ~~warming~~warmer climate (e.g. Thompson et al., 1989). Thus, the increase in LNO_x ~~at RCP8~~in TS8.5 contributes 7 DU to the increase in ~~tropospheric column ozone~~tropical $PCO3_T$.

A further increase in ~~the tropospheric partial column~~tropical $PCO3_T$ arises from the increase in the height of the tropopause under a warmer climate. ~~In the ensemble mean of~~In the transient simulations (which follow the RCP6.0 scenario), the ensemble mean annual mean tropopause height increases by ~~800m~~800 m from ~16.1 km ~~in the year 2000~~ to ~16.9 km ~~in the~~between year 2000 and 2100. The impacts of increasing tropopause height on ~~tropospheric column ozone~~tropical $PCO3_T$ are calculated as the difference between the full ~~tropospheric column from the column~~$PCO3_T$ values calculated using ~~the~~a consistent tropopause height from the values calculated using a fixed year 2000 tropopause height of 16.1 km. This ~~analysis~~calculation indicates that ~~tropopause height~~ the increase in tropopause height between 2000-2100 accounts for ~1.5 DU of the increase in ~~the total tropospheric column~~tropical $PCO3_T$ in the transient experiment.

~~In contrast to the effects of climate change,~~While reductions in ODS ~~primarily~~ affect tropospheric ozone in the extratropics through STE (e.g. Banerjee et al., 2016~~). Conversely,~~), in the tropics, ODS have little impact on tropospheric ozone, with ~~the partial column~~$PCO3_T$ increasing by <1 DU in the $TS2000_{ODS}$ experiment compared to TS2000~~.~~ (see Figure 2c).

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The time-slice sensitivity experiments ~~suggest~~indicate that the net effect of changes in the climate ~~change acts~~will be to increase ~~tropospheric column ozone~~tropical PCO3$_T$ in the transient simulations. ~~As discussed above, an additional consideration~~However, in the transient simulations (~~that are~~ run under all-forcings at RCP6.0) ~~are changes in~~ tropospheric ozone levels are also determined by the chemical effects of ozone precursors, ~~which here include~~including CH_4, CO and NO_x. The largest rate of change for ~~tropospheric column ozone~~tropical PCO3$_T$ occurs over the recent past (1960-2000) (see crosses in Figure 2c), ~~when~~during which time increases in anthropogenic NO_x and CH_4 emissions ~~(~~have driven~~Lamarque et al., 2010) drive~~ increases in tropospheric ozone production (e.g. Lamarque et al., ~~.~~2010; Young et al., 2013). After 2000, all the RCP scenarios project strong reductions in anthropogenic NO_x and NMVOC emissions (~~Meinhausen~~Meinshausen et al., 2011), which would in turn drive tropospheric ozone reductions. However, in the transient experiment tropospheric column ozone remains steady up to ~2040, partly due to the compensating effects of climate change, as suggested by the time-slice simulations, but also due to increasing tropospheric CH_4 concentrations (Young et al., 2013; Revell et al., 2015). The ~~rise~~increase in ~~tropospheric column ozone until~~tropical PCO3$_T$ up to 2060-2080 and its subsequent decline is consistent with the evolution of CH_4, which maximises around 2080 ~~at~~in the RCP6.0 scenario.

5 Developing a simple model for predicting stratospheric column ozone change in the tropics

Future projections of ~~total column ozone~~tropical TCO3 are strongly dependent on the assumed pathway for anthropogenic emissions, for which there is a great deal of uncertainty, particularly in relation to emissions of CO_2, CH_4 and N_2O. CCMs are commonly used to assess possible future changes in ozone under a small number of well-defined scenarios, e.g. the ~~Representative Concentration Pathways (~~RCP~~; van Vuuren et al., 2011)~~ scenarios used in the IPCC Fifth Assessment Report~~.~~ (IPCC, 2013). These emissions scenarios are neither forecasts nor policy recommendations, but instead are chosen to represent a range of possible global socio-economic and technological pathways for the future. In order to comprehensively quantify the response of the chemistry-climate system to such emissions scenarios, long, computationally expensive model simulations are required. However, simpler models can also be used to identify which processes dominate future trends and to explore the composition response to a wider range of emissions scenarios.

In Section 4, we ~~showed that future changes in tropical stratospheric column ozone are driven primarily by changes in: (i)~~quantified the impacts of halogen-catalysed ozone loss~~; (ii)~~, changes in the strength of tropical upwelling~~;~~ and ~~(iii) the~~ upper stratospheric cooling induced by ~~GHGs (mainly~~GHG changes (predominantly CO_2~~).~~) on the tropical stratospheric ozone. Furthermore, the partial column ozone trends in the upper and lower stratosphere were found to be, to first order, linearly dependent on ~~ODS~~ESC and CDE concentrations~~.~~ (see Figures 3 and 5). This conclusion was derived from the transient ~~runs based on~~experiments adopting a single emissions scenario and multiple time-slice ~~runs~~experiments based on 3 additional scenarios, and so is valid for a range of possible CDE and ~~ODS~~ESC concentrations. In this section, we describe a simple, computationally inexpensive linearised model that can be used to ~~estimate~~explore how tropical stratospheric column

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ozone ~~changes~~may change under a much wider range of future ~~emission scenarios~~ESC and CDE concentration pathways than are typically explored by comprehensive CCMs~~ (e.g. Morgenstern et al., 2017).~~. We emphasise that the experiments described in this study do not allow us to distinguish the effects of other chemical species, such as N_2O and CH_4, on stratospheric ozone, and thus the simple model does not attempt to include the effects of these species, which also vary substantially amongst RCP scenarios.

The simplest version of such a model has a linear dependence of tropical stratospheric column ozone (SCO3) on GHG concentrations (expressed in CDE) and ~~ODS. We justify this simple approach based on the approximately linear dependencies of ozone found in UM-UKCA and shown in Figure 3 and Figure 5. Such a model has~~ESC of the form:

$$SCO3_t = SCO3_{t_0} + \left(\frac{\Delta SCO3}{\Delta CDE} . \left(CDE_t - CDE_{t_0}\right)\right) + \cancel{\left(\frac{\Delta SCO3}{\Delta ODS} . \left(ODS_t - ODS_{t_0}\right)\right),} \qquad \cancel{(1)} \left(\frac{\Delta SCO3}{\Delta ESC} . \left(ESC_t - ESC_{t_0}\right)\right) \cancel{,} .$$

where the subscripts t_0 and t signify the reference year and the year the model is solving for, respectively. The constants $\frac{\Delta SCO3}{\Delta CDE}$ and ~~$\frac{\Delta SCO3}{\Delta ODS}$~~$\frac{\Delta SCO3}{\Delta ESC}$, which represent the SCO3 change due to surface CDE and ~~ODS~~ESC perturbations, respectively, ~~can be~~are calculated using the time-slice simulations~~, as these simulations~~ which perturb ~~ODS~~ESC and GHGs separately. ~~$\frac{\Delta SCO3}{\Delta ODS}$ was~~The parameter $\frac{\Delta SCO3}{\Delta ESC}$ is calculated by averaging the values obtained from the three pairs of simulations with different ODS loadings, but the same ~~CO_2 mixing ratios. The term~~GHG concentrations, i.e. from the SCO3 differences between the green triangle and green circle in Figure 6 divided by the difference in surface ESC concentration between these runs. Similarly, the parameter $\frac{\Delta SCO3}{\Delta CDE}$ was calculated ~~by averaging~~as the average of the linear fits through the ~~two sets of~~ three pairs of time-slice simulations with the same ODS loading, but different GHG ~~mixing ratios.~~concentrations, i.e. the green, blue and red circles in Figure 6. Using this method, values of $\frac{\Delta SCO3}{\Delta CDE}$ = -0.005 DU ppmv^{-1} and ~~$\frac{\Delta SCO3}{\Delta ODS}$~~$\frac{\Delta SCO3}{\Delta ESC}$ = -3.64 DU ppbv^{-1} were obtained. The parameters for the simple model ~~is~~are therefore ~~trained~~derived using the time-slice simulations and it can then be compared against the transient simulations in order to determine its ability to reproduce output from the same comprehensive CCM under a different scenario. These comparisons are shown in Figure 7 for the RCP4.5, 6.0 and 8.5 scenarios alongside annual mean stratospheric ozone column values from the transient UM-UKCA RCP6.0 simulations.

Projections of SCO3 made using the simple model following the RCP6.0 scenario for GHG and ODS (~~purple~~magenta line, Figure 7) can be compared with the fully-coupled ~~RCP6.0~~ transient simulations (grey lines, Figure 7). Overall the agreement between the simple model and the fully-coupled CCM is reasonable ~~and confirms that~~ over the ~~major drivers of future tropical SCO3 change are the emissions of GHGs and ODS. However, it is also surprising as the atmospheric processes~~

~~involved are complex and, in many cases, non-linear. For example, the changes in HO_x and NO_x chemistry resulting from future changes in CH_4 and N_2O emissions would appear to be of second order on the timescales~~140 year period considered. ~~Similarly, the dynamic feedbacks resulting from increasing vertical velocities on radical production from N_2O, CFCs and CH_4 are limited in magnitude. One of the reasons for this is most likely the limited latitude range of the region studied.~~ The ~~effects of increasing N_2O and CH_4 concentrations on ozone destruction are expected to~~ simple model does capture the main features of modelled SCO3 values from the CCM, with rapid ozone loss in the late 20th century, a minimum around year 2000 and a gradual increase ~~with latitude (e.g. Revell et al., 2012), while the effects of dynamical feedbacks are more likely to appear at the edge of the tropical pipe or at low altitudes due to branching in the BDC (e.g. Garny et al., 2011).~~throughout the 21st century. Year 2100 SCO3 values are in good agreement between the simple model and CCM, with both indicating that tropical averaged SCO3 values will not return to their 1960 values despite reductions in halogenated ODS concentrations following the implementation of the Montreal Protocol. However, there are important quantitative differences between the simple model and fully coupled CCM results, which likely result from the neglect of additional important chemical controls on stratospheric ozone in the simple model (e.g. N_2O, CH_4). For example, the simple model overestimates the maximum extent of tropical SCO3 depletion occurring around year 2000, partly a result of the solar maximum in that year, and remain below the CCM values for the first half of the 21st century. Furthermore, the rate of increase in the later half of the 21st century is overestimated in the simple model compared to the CCM. This is likely due to the increased importance of HO_x and NO_x catalysed ozone destruction in the later part of the century associated with increases in CH_4 and N_2O (e.g. Ravishankara et al., 2009; Fleming et al., 2011; WMO, 2014; Butler et al., 2016), which are neglected in the simple model. In general though, there is good qualitative agreement between the simple model and CCM, which highlights the importance of GHG and ODS as major drivers of tropical SCO3 in the future.

As well as using the simple model to calculate SCO3 projections under the RCP6.0 scenario, additional emissions scenarios have also been investigated~~. ~~, and are also shown in Figure 7. These scenarios include the RCP4.5~~,~~ and RCP8.5~~, RCP6.0 using ODS fixed at 1960 values and RCP6.0 using CDE fixed at 1960 values. Projected SCO3 values for each of these~~ scenarios ~~are shown in Figure 7. It should be noted that the SCO3 projections calculated are sensitive to the point at which they are initialised. Figure 7 shows a number of~~ (green and yellow lines respectively), a scenario using time varying CDE concentrations following RCP6.0 with fixed 1960 values for ESC (light blue line) and a scenario using time-varying ESC concentrations with fixed 1960 values for CDE (dark blue line). All scenarios were initialised from 1960s SCO3 values ~~taken from~~in the transient ~~simulations.~~

~~Caveats exist for developing and~~ experiment. The scenario from the simple model using ~~such simple models. The constants $\frac{\Delta SCO3}{\Delta CDE}$ and $\frac{\Delta SCO3}{\Delta ODS}$ likely vary with latitude, and so those calculated for this study could not be used for projections of exratropical stratospheric column values. Furthermore, they are likely to vary between~~ fixed 1960 ESC values (light blue) highlights the projected decreases in tropical SCO3 resulting from GHG induced increases in the speed of the BDC, which is markedly different ~~CCMs.~~to the rest of the stratosphere where GHG induced cooling leads to increased ozone mixing ratios. The results from the simple model indicate that by 2100 tropical SCO3 is lower following the RCP8.5 scenario, and higher following the RCP4.5 scenario, than the RCP6.0 scenario. This is because the reductions in lower stratospheric ozone from the acceleration of the BDC, which approximately scales with CDE (see Figure 5b), overwhelm any ozone increases in the upper stratosphere resulting from decreasing ESC concentrations and cooling of the upper stratosphere, as discussed in Section 4.2. These results from the simple model are in contrast to Eyring et al. (2013a) who used output from CCMs that participated in CMIP5 and found that by 2100 SCO3 values are expected to be lowest under RCP6.0 and slightly higher under RCP8.5 (see their Figure 6b). This difference is partly due to not including the chemical effects of N_2O and CH_4 in the simple model, as in the RCP8.5 scenario CH_4 levels at 2100 are more than double those in RCP4.5/6.0 and N_2O values in 2100 are around 7% higher in RCP8.5 compared to RCP6.0 (Meinshausen et al., 2011). The differences between the end of 21st century SCO3 values in the simple model and the results of Eyring et al. (2013a) may also reflect different sensitivities of UM-UKCA to radiative and chemical drivers compared to the CMIP5 multi-model ensemble. For example, the parameters $\frac{\Delta SCO3}{\Delta CDE}$ and $\frac{\Delta SCO3}{\Delta ESC}$ likely vary between different CCMs. Indeed, differences between these ~~values~~parameters in different CCMs would indicate varying ~~sensitivities~~sensitivites to ~~CO_2~~GHG and ODS changes and may help in the identification of which processes have high uncertainty and should be explored in more detail.

Lastly, the parameters used in the simple model have been derived for a tropical band (10°N-10°S) but are likely vary substantially with latitude, so those calculated for this study could not be used to examine projections of extratropical SCO3 values. The aim of such a model is not to replace fully-coupled CCMs, but to provide a simple and computationally inexpensive way of exploring possible future SCO3 changes in the tropics. In this capacity, it appears to offer considerable promise and could act as a valuable complementary approach to the 2D model studies which are currently used to investigate multiple scenarios (e.g. Fleming et al., 2011; WMO, 2014).

6 Conclusions

We have investigated the drivers of past and future changes in tropical averaged total column ozone using a number of model runs performed with two ~~versions~~configurations of the UM-UKCA model. Four transient simulations following an RCP6.0 future GHG emissions scenario and WMO (2011) ODS recommendations were performed, with the longest of these simulations spanning the period 1960-2100. ~~These~~The transient runs were supplemented with 6 time-slice experiments run under a range of prescribed GHG and halogenated ODS loadings commensurate with either year 2000 or 2100 levels. Note that in the time-slice experiments only the chemical impacts of changes to ODS loadings and only the radiative impacts of GHG perturbations are considered, and so we ~~are able to separate~~focus on separating the contribution of these chemical and radiative drivers ~~of~~to future tropical ozone column changes. ~~Changes~~ We do not consider explicitly in this study the chemical contributions to tropical ~~total~~ column ozone ~~were split into~~trends of future CH_4 and N_2O emissions. To aid in understanding the effects of the explored drivers on tropical column ozone changes, we analyse temporal trends in three partial ozone columns based on the following altitude ranges: the troposphere, the lower stratosphere (tropopause to 30km) and the upper stratosphere (30-48km). ~~Future stratospheric ozone projections for~~Ozone concentrations in each of these regions are governed by different processes and thus show distinct behaviours that combine to determine the overall evolution of total column ozone.

Future tropospheric ozone changes are driven by a number of processes, including changes to surface emissions of ozone precursors such as CH_4 and NO_x, increased NO_x emissions from lightning associated with ~~increasing GHG concentrations~~changes in convection, and changes to ~~troposphere~~tropopause height. There is a high level of uncertainty associated with future emissions of ozone precursors, linked to uncertainties in anthropogenic emissions, biomass burning and land use changes. While the various RCP scenarios follow a range of future emissions scenarios for many key tropospheric ozone precursors, particularly CH_4, further work is required to explore the impact of changes to tropospheric ozone on TCO3 trends during the 21st century in order to understand to what extent changes in tropospheric ozone column offset decreases in the lower stratosphere. Of course the environmental benefits from reductions in tropospheric ozone as an air pollutant and GHG may considerably outweigh any gains increases in tropospheric ozone could have by balancing the effects of a decreased stratospheric ozone column on surface UV radiation.

~~In the simulations described in this study, it has been shown that projected changes to stratospheric ozone throughout the 21st century are predominantly driven by~~ The chemical ~~changes resulting from differences in ODS loadings and radiative changes resulting from differences in GHG mixing ratios. The transient UM-UKCA simulations run under the RCP6.0 emissions scenario show that by the year 2100 stratospheric column ozone values are increased by 5 DU from the minimum values around the year 2000. However, modelled stratospheric column values in the simulations never recover to 1960s values despite declining stratospheric ODS loadings, due to the competing~~ effects of changes in ~~partial column ozone values in the lower and upper stratosphere.~~

~~Chemical and radiative perturbations~~ODSs and climatic changes due to GHGs drive changes to ~~stratospheric ozone abundances in~~ both the upper and lower stratospheric partial ozone columns. ~~However, while in~~ In the upper stratosphere ~~both reducing~~, where the chemical lifetime of ozone is short (~ 1 day), projected future reductions in ODS concentrations and ~~increasing~~ stratospheric cooling from increased GHG concentrations both lead to increased ~~ozone abundances, in the lower stratosphere decreasing ODS concentrations, which lead to increased ozone abundances, are counteracted by the effects of increased GHG concentrations, which lead to decreased ozone abundances. Furthermore, while in the upper stratosphere GHG and ODS increases lead to chemical changes (by changing gas phase kinetics and the concentration~~upper stratospheric partial column ozone by reducing halogen-catalysed destruction of ~~EESC~~ozone and slowing of the temperature dependent ozone loss cycles, particularly those of the Chapman cycle, respectively. The combination of these two effects is expected to lead to super-recovery of upper stratospheric partial column values above their 1960s values.

~~) which dominate the projection of ozone abundances, in the lower stratosphere ozone differences are the result of changes to the strength of the BDC, which accelerates in response to future GHG increases and decelerates in response to future ODS decreases.~~

~~Changes to lower stratospheric ozone~~Conversely in the lower stratosphere, where the chemical lifetime of ozone is typically >1 month, ~~are~~ the partial column ozone values are predominantly controlled ~~predominantly~~ by changes to transport. Projected increases in GHGs lead to an acceleration of the BDC, which is associated with increased transport of relatively ozone poor air masses into the tropical lower stratosphere, thereby decreasing ozone mixing ratios and the partial lower stratospheric column ozone. The magnitude of acceleration of the BDC is highly correlated with increasing GHG mixing ratios, and so the total effect of transport changes on tropical lower stratospheric ozone depends strongly on the future GHG emissions scenario. Future reductions in lower stratospheric ozone partial column values also result from decreased production of O_x from photolysis of O_2 in the lower stratosphere due to increased overhead ozone concentrations in the upper stratosphere. Analysis of the simulations presented here suggests lower stratospheric O_x production will decrease by 0.1 DU day^{-1} for each additional DU of ozone in the upper stratosphere.

The above points highlight that future projections of tropical stratospheric column ozone are the result of a complex interplay between drivers of ozone trends in the lower and upper stratosphere. The transient UM-UKCA simulations run under the RCP6.0 emissions scenario show that by the year 2100 stratospheric column ozone values are increased by 5 DU from the minimum values around the year 2000. ~~Future concentrations of ozone in the upper stratosphere are driven by changes in both ODS loading and temperature. In contrast to the lower stratosphere, both CO_2 increases and ODS decreases lead to increased ozone concentrations. ODS concentrations are expected to decrease throughout the 21st century due to the restrictions on ODS emissions imposed by the Montreal Protocol and its amendments. At the same time, stratospheric GHG concentrations, particularly CO_2, are projected to increase, leading to stronger radiative cooling of the stratosphere and slowing of the temperature dependent ozone loss cycles, particularly those of the Chapman cycle.~~ However, modelled stratospheric column values in the simulations never return to 1960s values despite declining stratospheric ODS loadings, due to the competing effects of changes in partial column ozone values in the lower and upper stratosphere.

~~The combination of these two effects is expected to lead to super-recovery of upper stratospheric partial column values above their 1960s values.~~

~~Projections of stratospheric column ozone values in the tropics are therefore the result of a complex interplay between ozone trends in the lower and upper stratosphere.~~ Understanding the extent to which dynamically induced decreases in lower stratospheric partial column values counteract upper stratospheric super-recovery is key to making accurate projections of stratospheric column ozone, and requires detailed modelling of both photochemical and dynamical processes under a range of future emissions scenarios. However, output produced by complex, fully-coupled CCMs can be used to create simple linear models which can be used to explore the stratospheric ozone column response to changing surface GHG and ODS concentrations. Simple linear models are computationally inexpensive and can be used to investigate a wide range of emission scenarios much more quickly than ensembles of fully-coupled CCMs. In this work we present a simple linearised model ~~which has been~~ developed from the UM-UKCA experiments to help investigate projections of stratospheric column ozone ~~following different~~ for a range of future emissions scenarios. The model includes ~~terms~~parameters for the dependence of stratospheric column ozone on ~~ODS~~ESC and ~~CDE~~GHGs (expressed as Carbon Dioxide Equivalent) mixing ratios. The simple model was ~~trained~~built using data from the ~~single and combined forcing~~ time-slice UM-UKCA experiments and then its performance compared against the transient ~~model simulations. Good~~integrations. There is reasonable quantitative agreement ~~was found~~ between the simple model and the long-term behaviour of tropical stratospheric column ozone in the fully-coupled RCP6.0 CCM simulations, confirming emissions of GHG and ODS to be ~~the major~~key drivers of long-term future tropical stratospheric column ozone changes. ~~Results from~~However, there are quantitative differences between the ~~simple~~ results of the simple model for other RCP scenarios and previous multi-model ~~indicate stratospheric column ozone changes resulting~~results from ~~future~~ CMIP5 (Eyring et al., 2013a). This is likely to be due to differences in N_2O and CH_4 ~~and N_2O emissions are of second order on the timescales considered here.~~concentrations amongst RCP scenarios, which are neglected in the simple model, and may also be due to different models possessing different sensitivity parameters for stratospheric column ozone to changing ODS and GHG concentrations.

~~While~~In summary, while fully-coupled CCM simulations are required to precisely quantify changes in, and identify the processes responsible for, future atmospheric composition changes, simple models can provide a complementary approach for investigating a broad range of potential emissions scenarios. Furthermore, it is hoped that the model presented here can be further developed to include more parameters (e.g. N_2O, CH_4) by performing more ~~simulations~~integrations, and also to more accurately constrain the terms of the simple model by using ~~simulations~~integrations from more CCMs. This would also allow for a better assessment of the uncertainty of each of the terms used in the simple model.

Availability of data

Data from the transient simulations are available as part of the CCMI initiative through BADC: https://blogs.reading.ac.uk/ccmi/badc-data-access/. All further data are available upon request.

Acknowledgements

The research leading to these results has received funding from the European Community's Seventh Framework Programme (FP7/2007 - 2013) under grant agreement n° 603557 (StratoClim), the European Research Council through the ACCI project (project number: 267760~~),~~) and the Natural Environment Research Council through the CAST project (NE/ I030054/1) and ACM’s Independent Research Fellowship (NE/M018199/1). We thank NCAS-CMS for modelling support. Model

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~~simulations~~integrations have been performed using the ARCHER UK National Supercomputing Service and MONSooN system, a collaborative facility supplied under the Joint Weather and Climate Research Programme, which is a strategic partnership between the UK Met Office and the NERC.

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Simulation name	Climate (SST, sea ice, GHG)	ODS (Cl_y, Br_y)
TS2000	2000	2000
TS2000$_{\mathrm{ODS}}$	2000	2100 (RCP4.5)
TS4.5	2100 (RCP4.5)	2000
TS4.5$_{\mathrm{ODS}}$	2100 (~~RCP8~~RCP4.5)	2100 (RCP4.5)
TS8.5	2100 (~~RCP4~~RCP8.5)	2000
TS8.5$_{\mathrm{ODS}}$	2100 (RCP8.5)	2100 (RCP4.5)

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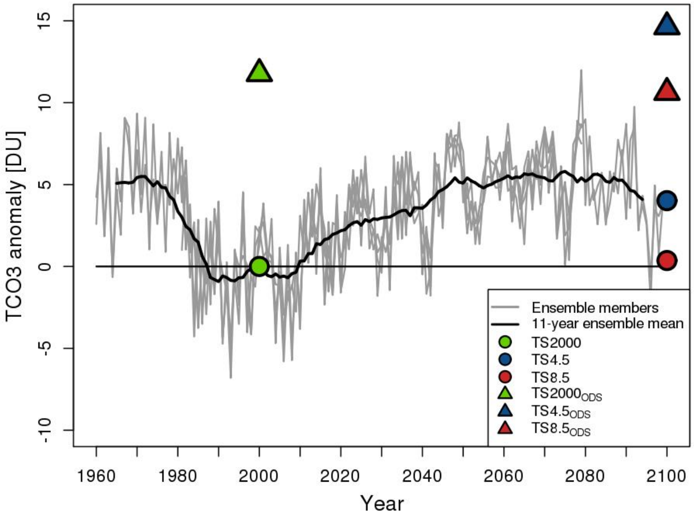
Table 1. Simulation names and corresponding climate (including radiative impacts of GHGs, SSTs and sea ice) and ODS loadings. Note that changes in halogenated ODSs are imposed only on the chemistry scheme while changes in GHGs (CO_2, CH_4, N_2O and CFCs) are imposed only on the radiation scheme. RCP scenario used for future GHG and ODS concentrations given in parentheses.

	Integration	PCO3	Lifetime	Production	Loss	Halogens	HO_x	NO_x	O_x
$PCO3_{US}$	TS2000	63 DU	1 day	48 DU day^{-1}	48 DU day^{-1}	9 DU day^{-1}	11 DU day^{-1}	19 DU day^{-1}	9 DU day^{-1}
	$TS2000_{ODS}$	+8%	+13%	-5%	-5%	-63%	+9%	+4.5%	+19%
	TS4.5	+5%	+8%	-3%	-3%	+2%	+2%	-5%	-7%
	TS8.5	+19%	+27%	-6%	-6%	+4%	+11%	-13%	-21%
$PCO3_{LS}$	TS2000	179 DU	34 days	7 DU day^{-1}	5 DU day^{-1}	1 DU day^{-1}	2 DU day^{-1}	2 DU day^{-1}	1 DU day^{-1}
	$TS2000_{ODS}$	+4%	+21%	-10%	-14%	-65%	-6%	+3%	+7%
	TS4.5	-3%	+8%	-8%	-10%	-5%	-8%	-10%	-8%
	TS8.5	-7%	+23%	-16%	-25%	-17%	-15%	-33%	-37%

Table 2. Partial column ozone values (DU), average ozone lifetime (days), net chemical production and loss and absolute contribution of halogen, HO_x, NO_x and O_x ozone destroying cycles (DU day^{-1}) in the upper and lower ~~stratospheric~~stratopsheric for the TS2000 integration. Percentage ~~differences are given~~change for the $TS2000_{ODS}$, TS4.5 and TS8.5 simulations relative the to TS2000 ~~integration~~simulation.


TCO3 anomaly [DU]
15
10
5
0
-5
-10
1960
1980
2000
2020
2040
2060
2080
2100
Year
Ensemble members
11-year ensemble mean
TS2000
TS4.5
TS8.5
TS2000ODS
TS4.5ODS
TS8.5ODS

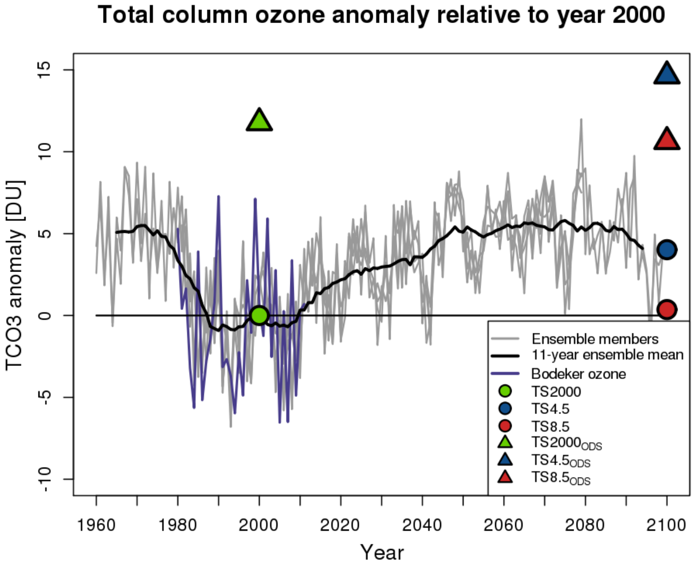
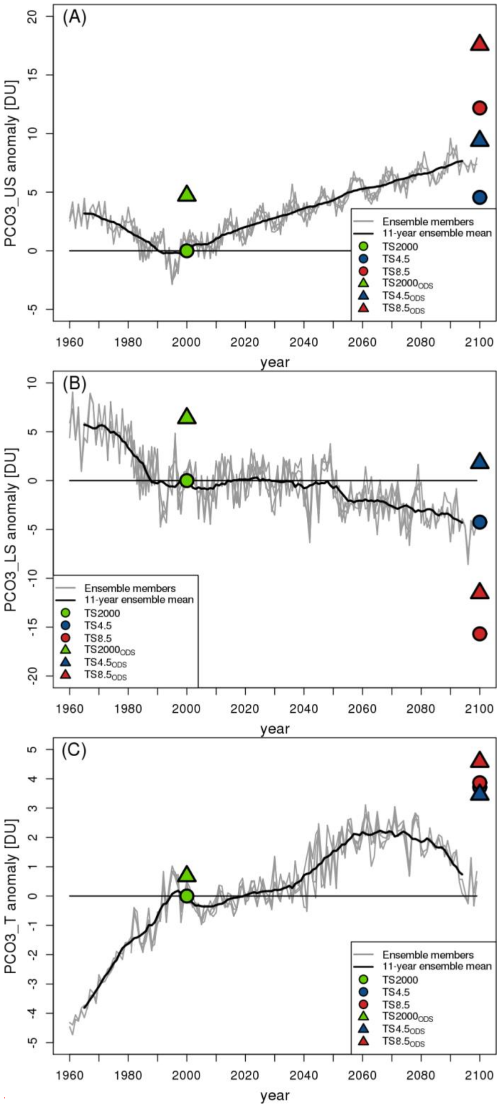


Figure 1. Total column ozone anomalies (in DU) relative to the year 2000±5 mean, averaged over 10°S-10°N for the four transient UM-UKCA experiments following the RCP6.0 future emissions scenario (grey lines), and the ensemble mean 11-year running mean (black line). Coloured circles and triangles represent tropical total column ozone in the time-slice experiments, as given in the figure legend. The purple line shows tropical averaged total column ozone values from v2.8 of the Bodeker dataset (Bodeker et al., 2005).


(A)
PCO3_US anomaly [DU]
20
15
10
5
0
-5
1960
1980
2000
2020
2040
2060
2080
2100
year
Ensemble members
11-year ensemble mean
TS2000
TS4.5
TS8.5
TS2000ODS
TS4.5ODS
TS8.5ODS
(B)
PCO3_LS anomaly [DU]
10
5
0
-5
-10
-15
-20
1960
1980
2000
2020
2040
2060
2080
2100
year
Ensemble members
11-year ensemble mean
TS2000
TS4.5
TS8.5
TS2000ODS
TS4.5ODS
TS8.5ODS
(C)
PCO3_T anomaly [DU]
5
4
3
2
1
0
-1
-2
-3
-4
-5
1960
1980
2000
2020
2040
2060
2080
2100
year
Ensemble members
11-year ensemble mean
TS2000
TS4.5
TS8.5
TS2000ODS
TS4.5ODS
TS8.5ODS

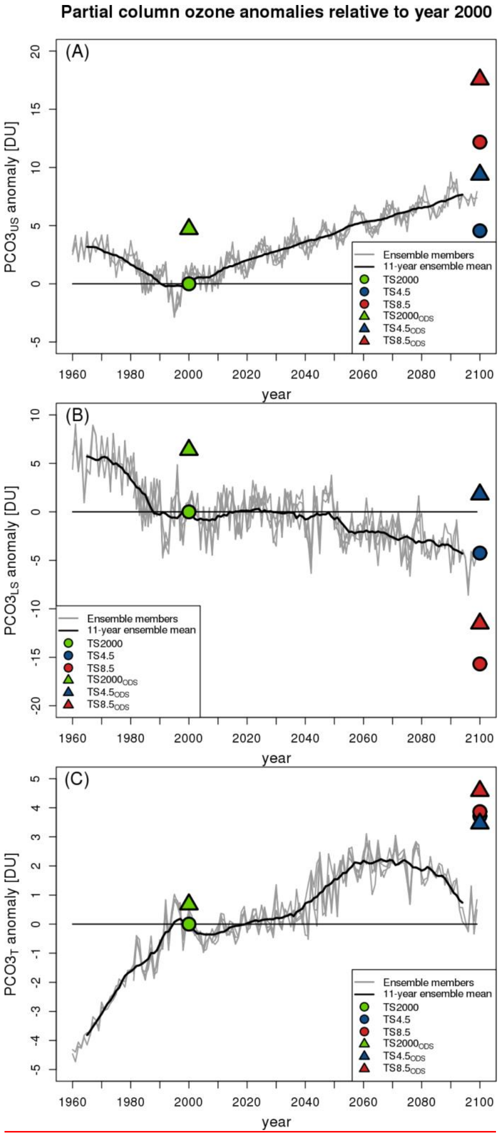
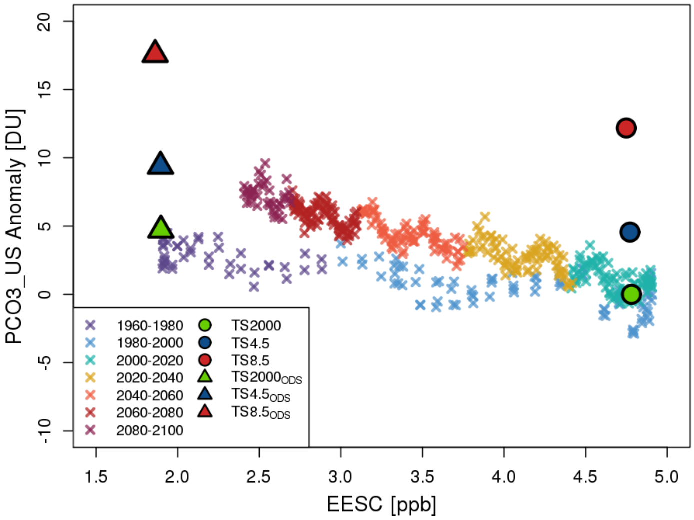


Figure 2. As for Figure 1, but for partial columns for (a) the upper stratosphere (30-48 km), (b) lower stratosphere (tropopause-30 km) and (c) troposphere.


PCO3_US Anomaly [DU]
20
15
10
5
0
-5
-10
EESC [ppb]
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0
1960-1980
1980-2000
2000-2020
2020-2040
2040-2060
2060-2080
2080-2100
TS2000
TS4.5
TS8.5
TS2000$_{ODS}$
TS4.5$_{ODS}$
TS8.5$_{ODS}$

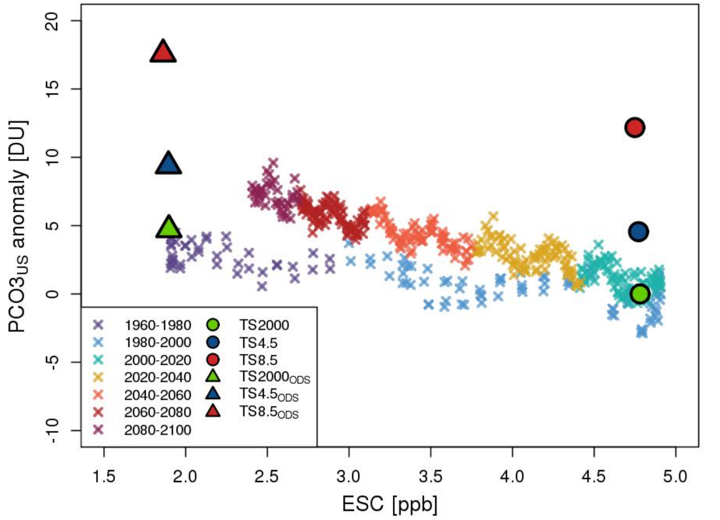
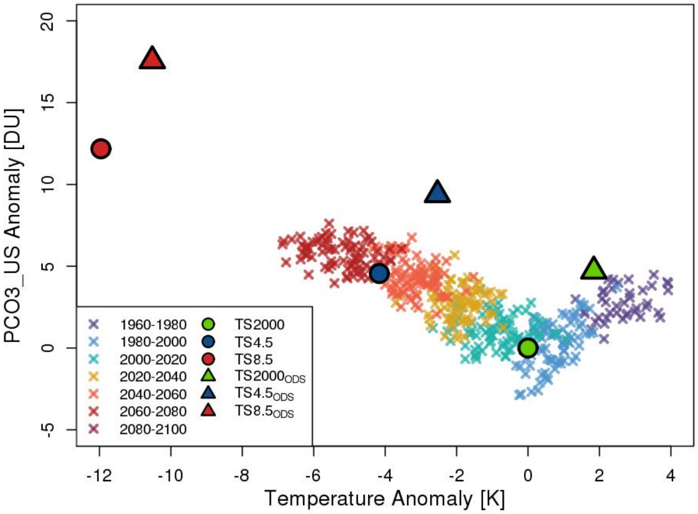


Figure 3. Scatterplot of annual mean upper stratospheric partial column ozone anomalies relative to the year 2000±5 mean (in DU) vs. 45km ~~EESC~~ESC (in ppb) for the transient simulation (crosses) and time~~-~~ slice experiments (circles and triangles). Results from the transient simulations have been coloured in 20 year sections.


PCO3_US Anomaly [DU]
Temperature Anomaly [K]
20
15
10
5
0
-5
-12
-10
-8
-6
-4
-2
0
2
4
1960-1980
1980-2000
2000-2020
2020-2040
2040-2060
2060-2080
2080-2100
TS2000
TS4.5
TS8.5
TS2000$_{ODS}$
TS4.5$_{ODS}$
TS8.5$_{ODS}$

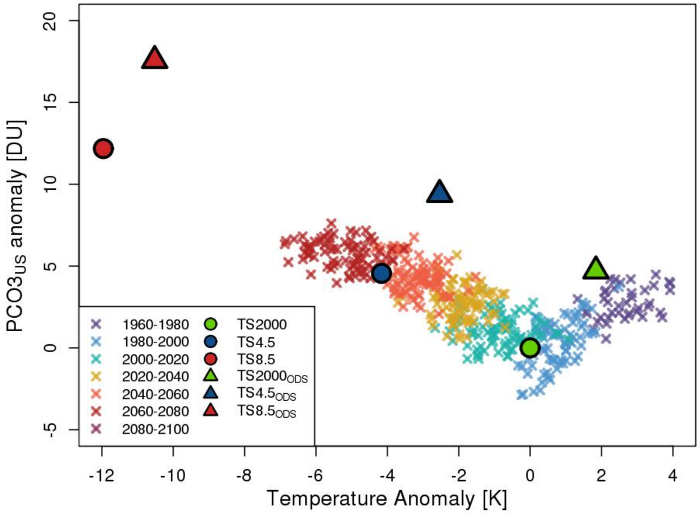
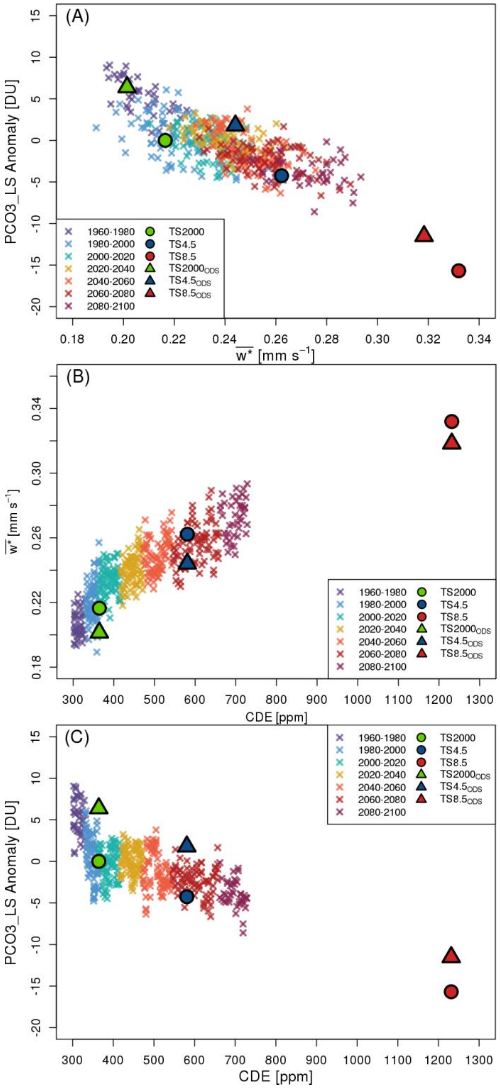


Figure 4. Scatterplot of annual mean upper stratospheric partial column ozone anomalies relative to the year 2000±5 mean (in DU) vs. 45km temperature (in K) for the transient simulation (crosses) and time-slice experiments (circles and triangles). Results from the transient simulations have been coloured in 20 year sections.


(A)
PCO3_LS Anomaly [DU]
15
10
5
0
-5
-10
-15
-20
0.18
0.20
0.22
0.24
0.26
0.28
0.30
0.32
0.34
$\overline{w^*}$ [mm s^{-1}]
1960-1980
1980-2000
2000-2020
2020-2040
2040-2060
2060-2080
2080-2100
TS2000
TS4.5
TS8.5
TS2000$_{ODS}$
TS4.5$_{ODS}$
TS8.5$_{ODS}$
(B)
$\overline{w^*}$ [mm s^{-1}]
0.34
0.30
0.26
0.22
0.18
300
400
500
600
700
800
900
1000
1100
1200
1300
CDE [ppm]
1960-1980
1980-2000
2000-2020
2020-2040
2040-2060
2060-2080
2080-2100
TS2000
TS4.5
TS8.5
TS2000$_{ODS}$
TS4.5$_{ODS}$
TS8.5$_{ODS}$
(C)
PCO3_LS Anomaly [DU]
15
10
5
0
-5
-10
-15
-20
300
400
500
600
700
800
900
1000
1100
1200
1300
CDE [ppm]
1960-1980
1980-2000
2000-2020
2020-2040
2040-2060
2060-2080
2080-2100
TS2000
TS4.5
TS8.5
TS2000$_{ODS}$
TS4.5$_{ODS}$
TS8.5$_{ODS}$

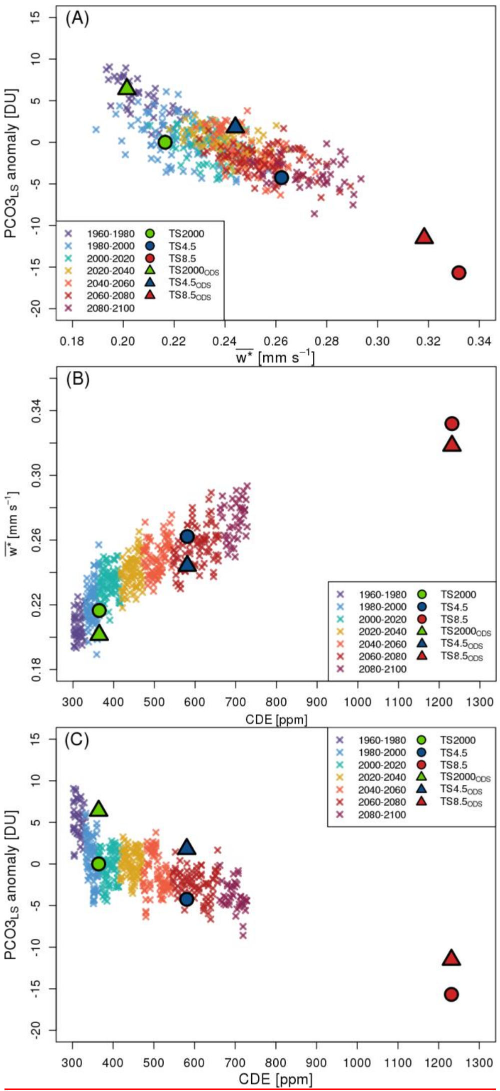
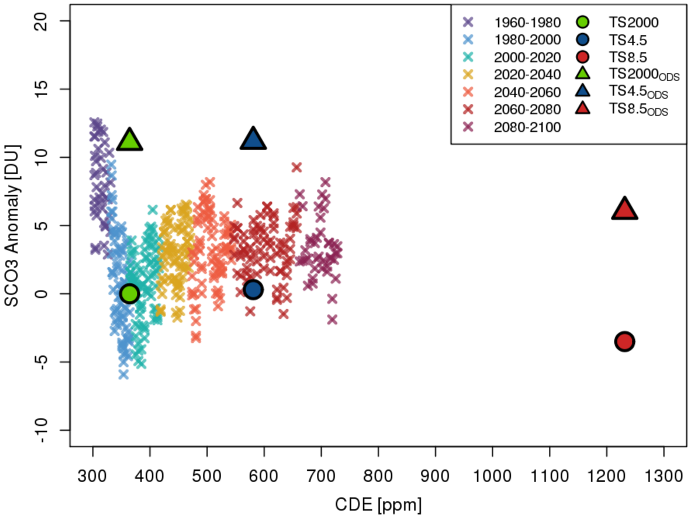


Figure 5. ~~Scatterplot~~Scatter plot of (a) lower stratospheric partial column ozone anomalies relative to the year 2000±5 mean (in DU) vs. 70hPa ~~ẃ*~~$\overline{w}^*$ (in mm/s), (b) 70hPa ~~ẃ*~~$\overline{w}^*$ vs. CDE mixing ratio (in ppmv) and (c) lower stratospheric partial

column ozone anomalies vs. CDE mixing ratio for the transient simulations (crosses) and time--slice experiments (circles and triangles). Results from the transient simulations have been coloured in 20 year sections.


SCO3 Anomaly [DU]
-10
-5
0
5
10
15
20
CDE [ppm]
300
400
500
600
700
800
900
1000
1100
1200
1300
1960-1980
1980-2000
2000-2020
2020-2040
2040-2060
2060-2080
2080-2100
TS2000
TS4.5
TS8.5
TS2000ODS
TS4.5ODS
TS8.5ODS

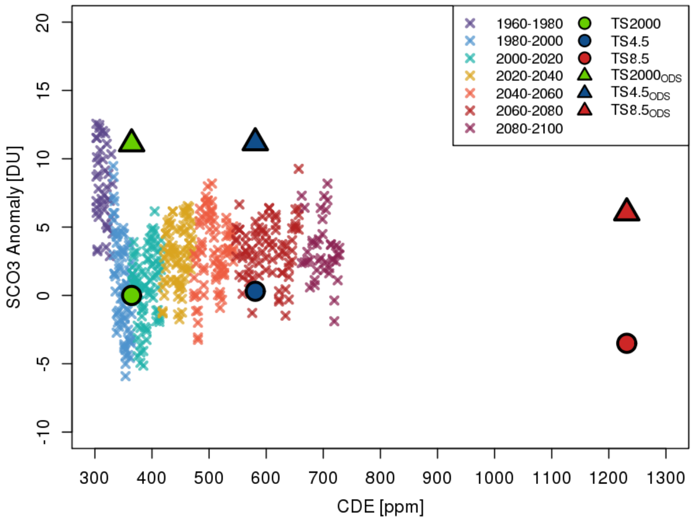
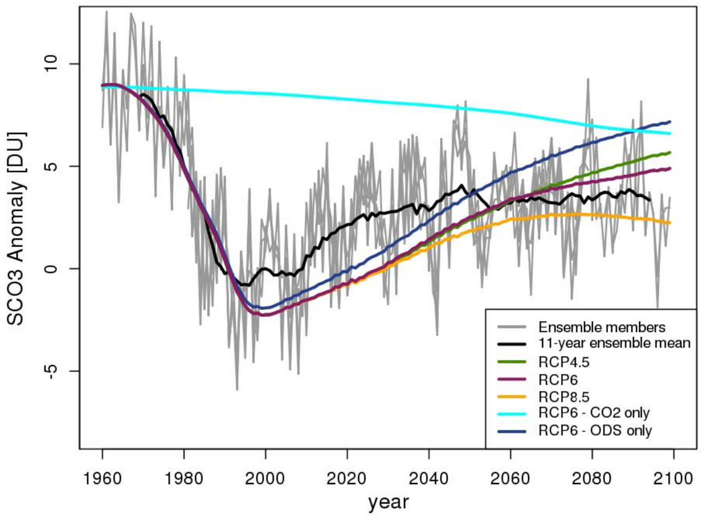


Figure 6. ~~Scatterplot~~Scatter plot of stratospheric column ozone anomalies relative to the year 2000±5 mean (in DU) vs ~~CO_2~~CDE mixing ratio (in ppmv) for the transient simulation (crosses) and time~~-~~-slice experiments (circles and triangles). Results from the transient simulations have been coloured in 20 year sections.


SCO3 Anomaly [DU]
10
5
0
-5
1960
1980
2000
2020
2040
2060
2080
2100
year
Ensemble members
11-year ensemble mean
RCP4.5
RCP6
RCP8.5
RCP6 - CO2 only
RCP6 - ODS only

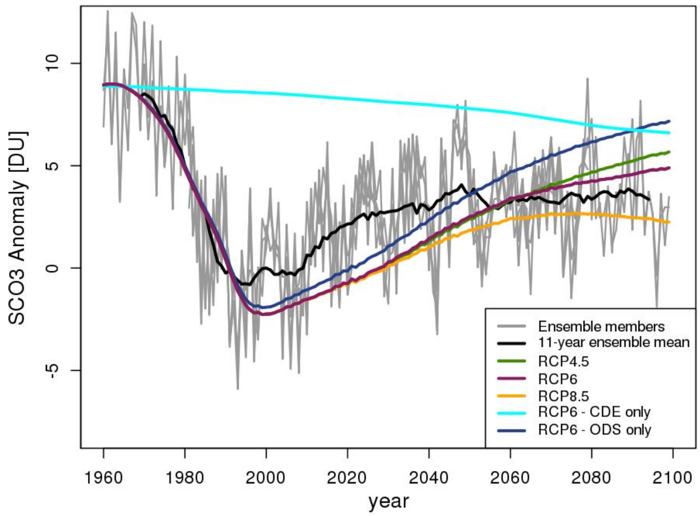

SCO3 Anomaly [DU]
10
5
0
-5
1960
1980
2000
2020
2040
2060
2080
2100
year
Ensemble members
11-year ensemble mean
RCP4.5
RCP6
RCP8.5
RCP6 - CDE only
RCP6 - ODS only

Figure 7. Annual mean stratospheric column ozone anomalies relative to the year 2000±5 mean (in DU) as modelled by the transient simulations (grey lines), with the ensemble mean 11-year running mean also plotted (black line). Results obtained using the simple model are shown for a range of emissions scenarios, initialised to 1960 values taken from the transient run.