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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Diagnosing the radiative and chemical contributions to future changes in tropical column ozone with the UM-UKCA chemistry-climate model" *by* James Keeble et al.

Anonymous Referee #1

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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.

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Discussion paper

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.

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?

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.

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).

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.

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.

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

- 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.

- 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.

- 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.

- 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.

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

- 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.

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

- 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.

- 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.

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

- P4L23-24: How were the initial atmospheric conditions perturbed for each ensemble 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).

- 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.

- 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.

- 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?

- 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.

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.

- 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?

- 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.

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

- 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.

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

- 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?

- 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.

- 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 $\sim$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?

- 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.

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

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

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

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

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

- 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.

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

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

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

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

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

- Figure 5c is not discussed in the text.

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

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

- 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.

- P12L8-9: State where this is shown (Fig. 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?

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

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

- 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.

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

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

- 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.

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

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

- Please state where your data are available from.

Technical corrections

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

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

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

- 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.

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

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

- P3L6: define Cly and NOy.

- P3L24 onwards: there is no need to refer to “RCP emissions scenarios” or “RCP scenarios.” Calling them RCPs is sufficient.

- P3L28: “rise” -> “increase”

- P5L3: “integration given” -> “integration are given”

- P6L16: “discussed in” -> “discussed by”

- P7L14: “century is dependent” -> “century are dependent”

- P8L5-6: units are in italics.

- P9L3: “62 DU” – it says 63 DU in Table 2.

- P11L18: NOx: fix subscript.

- P11L30: “increase in LNOx at RCP 8.5” -> “increase in LNOx in RCP 8.5”

- P12L15: Meinhausen -> Meinshausen

- P13L26 “emissions of GHGs” -> “the radiative effects of GHG emissions”

- P13L28: dynamic -> dynamical

- P15L3: troposphere height -> tropopause height

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