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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Drivers of the tropospheric ozone budget throughout the 21st century under the medium-high climate scenario RCP 6.0" *by* L. E. Revell et al.

Anonymous Referee #3

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GENERAL COMMENTS:

The manuscript by Revell et al. investigates the behaviour of the SOCOL CCM when it comes to capturing tropospheric ozone and related species from the mid-20th century to the late 21st century. Following a brief evaluation for present-day conditions, analysis of a control and two sensitivity simulations provides insight into the role of emissions and climate change in driving ozone changes. The role of NOx is found to be dominant, with methane being the second most important factor and climate changes only having a minor contribution.

Given that the study is based on a single model and a single scenario, its value as a prediction of future ozone is relatively limited. However, through the sensitivity experiments and the diagnostic analysis performed, it manages to provide some mechanistic understanding of the behaviour of this model, and to therefore add to the body of literature that aims to understand the contribution of different processes to past and future tropospheric ozone changes. Since the latter is quite an uncertain area of study, contributions of this kind are useful. Also, the manuscript is well written, and suitable for publication in ACP. However, there are several - mostly minor - amendments that I believe would need to be made before publication (see below).

GENERAL COMMENTS:

- It would have been useful to discuss the results in the context of the performance of the model, using what was found in Sect. 3.1, but also in previous evaluation efforts of SOCOL. The reader is not getting an idea of what are the main implications of those biases for the main features of the evolution of ozone, NOx, and CO+VOCs. Also, it is not clearly discussed what are the implications of model structural deficiencies, e.g. the crude handling of wet removal of HNO3. Maybe all those can be discussed in a "Discussion" session before the conclusions.

- There is very little discussion on how well the model represents OH, which is key for several processes discussed here. It would be useful to give a sense to the reader of how well the model does in capturing present-day OH and/or methane lifetime, and how OH evolves with time in the simulations.

- There is almost no mention of aerosols. I know this is not a central aspect, but there are some things for which the reader is left in the dark: Does the model include any aerosol tracers? I presume not, but Table 1 mentions "Ozone and aerosol precursor emissions", which is confusing. There is also a mention of "16 heterogeneous reactions", and it is not clear whether that refers to the troposphere or the stratosphere, and what fields are used to drive those reactions (e.g. tropospheric aerosol fields). It

would be good to clarify those aspects in the model description section, and to discuss any implications of the lack (?) of aerosols for the ozone/chemistry results (via e.g. photolysis, or heterogeneous processes).

SPECIFIC COMMENTS:

Page 482, Line 9: Later on it appears that climate change includes methane affecting chemistry. You may want to rephrase to "...climate change (including methane effects)", as otherwise the reader gets the first impression that methane changes are ignored.

Page 482, Lines 10-11: In this part of the abstract, it would be useful to make a brief statement on the nature of the RCP6.0 scenario (i.e. optimistic, pessimistic, moderate), and on why it was chosen. I know this is discussed later on, but it would be nice to clarify it to the reader up front (i.e. that it is a "medium-high" scenario, and therefore fairly realistic).

Page 482, Line 13: Please rephrase to "... at 23% compared to 1960", for clarity.

Page 482, Lines 18-19: Not clear what is meant in this context by "...together with the longevity of ozone in the troposphere" - please rephrase.

Page 482, Lines 19-20: "A simulation..." – by when?

Page 482, Line 27: I would suggest rephrasing to "Overall, the results show that, in this scenario, ozone in the future...".

Page 483, Line 17: Better to use the most recent IPCC forcing chapter citation (Myhre, Shindell et al., 2013).

Page 483, Line 21: Worth mentioning CO as well.

Page 484, Line 12: Worth also citing Kawase et al. (2011), who examined this effect.

Page 484, Lines 13-15: Clarify that this effect is due to photolysis.

Page 484, Line 28: It would be better to replace "projections" with "changes", as much of the ACCMIP work that you mention looks at historical changes too, not just future projections.

Page 485, Line 22: Suggest rephrasing "obtaining grades in the midrange" to "obtaining performance grades in the midrange".

Page 485, Line 26: Please delete space between "." and "2".

Page 486, Lines 1-2: Here it is worth summarising the major features of performance in a couple of sentences. They will be helpful later on in the discussion.

Page 486, Lines 18-20: Where do those conversion factors come from? Are they totally arbitrary (I presume not), have they been shown to lead to better performance, do they come from the literature? Worth stating in the text.

Page 486, Lines 28-29: Would the scaling prevent future convective activity changes from modifying the magnitude of lightning NOx production (as well as the areas of occurrence, which is already discussed)? Please discuss.

Table 1: For the ozone and aerosol precursors, pre-2000 emissions are not exactly "observations" (second column) – especially for those short-lived species there has not been that detailed observational information with global coverage. Perhaps it could be rephrased to "Historical emissions until 2000...". Also, for ODSs, until when are observations used?

Page 487, Line 24: I would suggest adding a word so that it reads "These transient simulations...", to emphasise on the fact that the simulations are not time-slice/equilibrium.

Figure 1: Please mention in caption that emissions shown are global total. Also, for NMVOCs, mention that they are non-biogenic.

Page 488, Lines 18-20: I presume the fixing of methane will directly impact both the

radiation and the chemistry? Please clarify in the text.

Sect. 3.1, beginning: Some further details are needed here for the model-observation comparison. E.g. why was 2005-09 chosen (I presume because it is representative of the present-day and because of good data availability). Also, was the model output processed with satellite operators (averaging kernels and a prioris) for TES, to account for observational sensitivity? If not, implications need to be discussed. And with what tropopause definition was the model tropospheric NO2 column calculated?

Figure 2: The title TES/OMI in the second column is a bit misleading, as nowadays there are combined satellite products (e.g. MLS/TES) and the reader may think that is the case here (without reading the text). I suggest labelling every panel with the species being looked at and the instrument/model. There is no need to increase the size of the figure, as there is already enough space between the panels. Also: The panels are not labelled with the letters mentioned in the caption.

Page 489, Line 14: Please change “large burden” to “large ozone burden” for clarity.

Page 490, Line 3: Perhaps use “low” instead of “depleted”, as the latter may imply that there was ozone there that was depleted.

Page 490, Lines 8-10: It would be useful to give the reader a sense of how high OH is in the model. Either the global mass-weighted mean OH concentration, or the methane lifetime would give some good general picture. And it would also be useful to compare against recent multi-model (Voulgarakis et al., 2013) and observational (Prather et al., 2012) estimates.

Page 490, Lines 18-19: Please change “and also to the general high ozone bias” to “and also leads to the general high ozone bias”.

Page 490, Lines 25-28: How does this performance compare to the earlier evaluation by Stenke et al. (2013)? Discussing this here will strengthen the conclusions of the evaluation.

Page 492, Line 23: “are located in different places” – I would add the word “slightly” (different places), as the broader region is the same. Note that in Voulgarakis et al. (2013), multi-model mean surface OH was also lowest over Indonesia and not over the Western Pacific (Fig. 6a).

Page 492, Line 27: Not clear how R5 could be a result of low NOx and high CO+VOCs.

Figure 3: Again, letters that indicate the order of panels (a, b, c. . .) are missing.

Page 493, Lines 1-3: Wouldn’t R1 also be partly responsible for this feature (less NOx so less OH recycling).

Page 493, Line 27: Please add “later” after “as shown”.

Page 494, Lines 1-2: Please explain why (I presume you mean that at this level the satellite instruments, e.g. TES, have a better sensitivity).

Page 494, Line 14: Please add “as seen here”.

Page 495, Line 17: It seems completely offset rather than “partially”, given the flat trends in Fig. 5c.

Page 496, Lines 21-28: Please clarify that this is an effect driven by OH differences. Here, it would actually be useful to show OH evolution plots. Also, relating to this subsection: The authors do not comment on the NOx response to fixing methane. It looks like NOx abundances are entirely driven by chemistry (short-lived emissions and methane) as 1960 and 2100 fCH4 levels look the same, so the meteorological changes (precipitation, lightning) do not play any role. Worth commenting here on how the model set-up (fixed wet removal rates, scaled lightning) could have affected this result.

Page 497, Lines 1-2: Also, it is interesting that for the tropics the fCH4 simulation shows drastically different results for 2100 compared to REF-C2 and fEmis. Any ideas on why that is?

Figure 8: What does the grey colour indicate in panel (b)?

Page 499, Line 10: You could add ", which is a scenario of intermediate severity compared to RCP4.5 and RCP8.5," after "RCP6.0".

Page 500, Lines 15-16: ". . .although their effects are relatively small." – this has not been demonstrated in the analysis.

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