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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric ozone in CCMI models and Gaussian emulation to understand biases in the SOCOLv3 chemistry-climate model" *by* Laura E. Revell et al.

Anonymous Referee #2

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This manuscript quantifies tropospheric ozone biases in two versions of the SOCOL chemistry-climate model, as well as the CCMI models. The SOCOL bias is further investigated using an emulator. I find the methodology novel, and the Discussions and Conclusions is particularly well reasoned and should be of considerable interest to the chemistry-climate modeling community. I do believe the paper could be greatly improved if some choices and details of the methodology are better explained (and perhaps if the paper is slightly restructured) as I explain in my two major criticisms below.

General comments

1) A stronger rationalization of the input parameter choices for the emulator is needed in Section 2.4. An important reason for testing the ozone precursors [variables (1-3)] is that they are a primary candidate for the cause of the systematic high bias in tropospheric ozone among model intercomparisons that use harmonized emissions, such as CCMI and ACCMIP. An important reason for testing (3) should be that SOCOL is very simplistic in its representation of NMVOC chemistry compared to other CCMI models (as an aside: why not vary the yield of CO from NMVOC oxidation separately to the magnitude of NMVOC emissions?). It also seems that variables (4-9) are chosen to reflect developments between SOCOLv3.0 and v3.1...is that correct? If so, I am not sure why, besides (8), they are investigated at all since the authors have already performed a sensitivity test in which they find that inclusion of heterogeneous hydrolysis of N2O5 is the main development that reduces the model's ozone bias between the two versions (P9L31).

2) It seems that the most detailed portion of the paper is focused on quantifying and understanding SOCOL's ozone biases, in part with the emulator, rather than an exploration of biases in the CCMI models (which could be a paper by itself!). With this in mind, the authors might consider first discussing SOCOL biases and then placing the results of the single model study within the wider context of the CCMI models e.g. combining Section 3.1 with the first paragraph of the Discussions and Conclusions. However, I leave this up to the authors. Secondly, and more importantly, please elaborate upon the basics of the emulation technique. Although I appreciate that the authors are probably trying to avoid jargon, as a non-statistician, I find the beginning of Section 2.4 a little confusing. Finally, the emulator experiments are a novel contribution to this field, which should be emphasized in the Introduction and Conclusions to increase the significance of the paper. Perhaps the authors could also speak to the broader goals such as extending the emulation methodology to explore tropospheric ozone variability due to meteorological parameters (e.g. convective parameters) not investigated here, or variability in other metrics such as ozone extremes etc...

Specific comments

P2L21: these fractions were deduced using data over individual sites in the Southern Hemisphere and are not necessarily representative of the whole troposphere.

P2L23: specify that this is the "global tropospheric lifetime" since the ozone lifetime can vary considerably by region.

P2L27: please cite Young et al. (2018) alongside Young et al. (2013) and Parrish et al. (2014).

P3L5: please cite Stevenson et al. (2006) for ACCENT and Young et al. (2013) for ACCMIP.

P3L26 (and P6L21): Do you mean non-additive instead of non-linear?

P4L3: For clarity, specify that SOCOL is a chemistry-climate model.

P4: Provide some information about the stratospheric boundary conditions.

P4L16: A look-up table is an offline, not online, photolysis scheme (in agreement with the last sentence of the paragraph).

P5L14: This is inconsistent with P4L29, which states that methane is prescribed as a "surface mixing ratio", which implies the lowermost model level.

P5L16: Naively, I would not expect methane-induced ozone production to be reduced upon prescribing methane on one level versus multiple levels since it is well mixed in the troposphere.

P6 paragraph 1 and Section 3.1: I wonder how much of the inter-model differences in the tropospheric ozone burden arise from inter-model differences in tropopause height. Could this be quantified by imposing the same tropopause height across all the models and noting the difference in ozone burden?

P6L20: Please see General Comment #2. This sentence is packed with information

and is confusing to a non-statistician.

P6 points 1 and 3: Which type of emissions? Anthropogenic/biomass burning/natural?

P6 point 4: I am unclear as to why this is tested. Emissions are included as surface fluxes (i.e. lowest model level) in both SOCOL versions, and to my knowledge, across most models.

P7 point 5: I would have thought a priori that the number of levels that methane is prescribed on would not matter for tropospheric ozone amounts, and this is confirmed later in the paper.

P7L24: I am not sure why you would test ranges that are not feasible. E.g. the maximum range for methane (4xCH4) is much larger than even RCP8.5 year 2100 amounts relative to present day. Are we then sure the results of the emulator remain meaningful?

P7: The final paragraph explains that physical/meteorological parameters are, by design, not investigated in the emulator experiments. Indeed there could be multiple reasons, besides chemistry, for SOCOL's particularly high ozone bias. This is explained well in the Discussion, but should also be made clear in the Introduction: the methodology used here does not explain (nor is it intended to explain) the entirety of the "remaining ozone bias in SOCOLv3.1" as stated on P3L20.

P8L2 and Section 3.2: Why not also show results for the global mean tropospheric ozone burden, given its discussion in the Abstract and elsewhere.

P8L12: Reference Morgenstern et al. (2017) who discuss familial relationships between the CCMI models.

P8L22: I do not think you can say ECAM-L90 simulates a "better" representation here since there is no comparison to the observations yet.

P9L16: Please provide the ACCMIP MMM global mean tropospheric ozone burden in DU for comparison with CCMI and CMIP5. Also state which, or at least how many,

models were considered in the ACCMIP and CMIP mean.

P9: The CCMI/ACCMIP/CMIP5 comparison is brief. This is fine for the present study, but perhaps the authors could highlight the potential for more detailed future investigation (see also General Comment #2). It would be interesting to see the extent of agreement - or lack thereof - between the different model intercomparisons' simulation of tropospheric ozone, given their different aims and formulations (e.g. a focus on stratosphere-troposphere interactions in the CCMI models vs atmosphere-ocean coupling in CMIP5).

Figures 2 and parts of Figure 4, 5: The continuous scale in these figures makes it difficult to distinguish numerical differences between the sub-plots. I recommend a discrete scale as in Figures 3 and 4c, 4f, 5c, 5f.

P9L30: Do you mean regionally not globally?

P9L33: From Figure 3, it looks like several of the CCMI models also show this bias over the Southern Ocean. Do they share the Wesely deposition scheme?

P10L6: State where this maximum bias occurs.

P10L9, Figure 6: Am I right in thinking that two conditions need to be satisfied in order for the emulator to perform well: having a high R squared value and having the points falling on a 1:1 line? Please clarify.

P10L10: See earlier comment about using inputs outside feasible ranges, which is acknowledged on P10L30. Do these extremes need to be tested?

P10L20: Can we explain this? Does it reflect a NOx titration effect?

P10L17, Figure 7: I am a little confused on what to take from this figure: is the "sensitivity" of tropospheric ozone to each parameter determined by the slopes of the sub-plots? If so, why compare the different sensitivities? To determine which parameters are more "important" for tropospheric ozone variability, it makes more sense to compare the vari-

ance explained by each parameter (Figure 8). Finally, what does the uncertainty in Figure 7 signify? I may be missing the obvious! Please explain Figure 7 clearly or consider removing.

P10L17: "Figure 7 displays the sensitivity of global-mean tropospheric ozone..." but the figure caption suggests the mean is over the Asian region only.

Figure 8: Remove "9 variables" from the figure caption since all 9 variables are not shown.

Figure 8: Could you also show a panel for the global mean burden?

Figure 8: Could you explain why the relative importance of CH4 and CO is smaller over Asia than Europe or the US? It would be better to use the same scale on all the panels.

P11L6: "up to 8 DU regionally"

P11L12: "up to $\sim$30 DU regionally"

Discussions and Conclusions: I very much like this section! I would only conclude with some remarks on the novelty of the emulation technique within this field and its potential future value in the study of ozone biases (see General Comment #2).

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