

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2023-9

Anonymous Referee #1

Referee comment on "Exploring the drivers of tropospheric hydroxyl radical trends in the Geophysical Fluid Dynamics Laboratory AM4.1 atmospheric chemistry–climate model" by Glen Chua et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2023-9-RC1>, 2023

This manuscript presents an examination of hydroxyl radical trends, variability, and sensitivity from the GFDL model AM4.1 for 1980-2014. In addition to a "Base" run and a "Met" run in which all emissions are fixed to 1980 levels, sensitivity simulations are also performed in which emissions for individual species (NO_x , CH_4 , CO, NMVOCs, and ODSs) are fixed to 1980 to isolate spatial and temporal effects on OH abundance. Results suggest that global mean OH concentration has increased by ~5%, mainly due to the competing effects of increasing NO_x and CH_4 . Model validation against OMI NO_2 and MOPITT CO is performed, revealing that NO_2 compares reasonably well while modeled CO trends compare poorly against observations (which reflects more on the emissions inventory than on the model).

Overall, I consider this to be a nice analysis that makes a solid contribution to the literature surrounding OH concentrations at the global scale. Sensitivity simulations like the ones performed here are valuable for gleaning information about the drivers of OH variability, with interesting, if perhaps somewhat expected, conclusions found in the spatial and temporal details of the various analyses of the simulations. I consider the comparison to observations to be sufficient for this study – there are always additional datasets that can be compared against, but for the species examined and the motivation of this work, the two included make sense. Prior literature is well cited, and the present work is well contextualized with comparisons to the results of other studies. The article is well within the scope of ACP, and, after addressing a number of comments included below, I would consider it a good candidate for publication.

Specific comments:

Table 1: Curious that a wavelength cutoff of 310 nm is used for $\text{O}_3 \rightarrow \text{O}^1\text{D}$ photolysis; most other models (e.g., Lelieveld et al., 2016, which you compare to throughout this manuscript) use 330 nm due to the small contribution from the quantum yield tail – see, e.g., Armerdling et al., 1995: <https://pubs.acs.org/doi/pdf/10.1021/j100010a025>. Any idea, or ability to quantify, how much this might affect total primary production in your results?

L225 / Fig. 2b: While the increase in [OH] in the lower troposphere is largest in absolute terms, [OH] values drop in the UT just as a result of pressure. Would be informative to also see this plot in units of pptv.

L235: I'm curious if the authors see any issue with treating CH₄ as a surface boundary condition and making conclusions like "CH₄ caused a negative trend in [OH]". Especially since it's a problem of "the chicken and the egg" and feedbacks between OH and CH₄ are notably missing at the surface, isn't causation particularly difficult to attribute in this case? Since models are generally not set up to do CH₄ fluxes, the model configuration here is understandable; perhaps just worth a note of caution in the text.

Figures 11 and 12: For both panels (b), does this indicate a non-zero emissions trend over the oceans? I don't see why a trend in emissions for either CO or NO₂ should occur, besides for shipping lanes perhaps, but I would expect from the color bar that a zero trend should be depicted as white.

Technical corrections:

L34: "tropospheric" misspelled

L86: "increasing" should be "increase"

L167: Check punctuation; period should be comma.

L203: should be "as well as"

L227: sensitivity misspelled

L243: "the" or "this" should be removed

Figure 2: in panel d), I think the purple bar lost part of its label (should be CH₄+NO_x? I only see "+NO_x")

L296: should be "out of"

Figure 6: Text on panels c and d should be increased in size

L340: "increases" misspelled

L362: "flux" here is a bit hard to decipher, please clarify

L386: "trends" at beginning of line should be removed; is "Iand" a typo

Figure 11 caption: same as above. Also, I'd suggest avoiding repetition between figure captions and text.

L445: "agrees" should be "agree"

L469: Should this be "SLCF" instead? Defined?

L479: should be "(Horowitz et al., 2020)" all in parentheses?