

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, CH₄, 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 CH₄. Model validation against OMI NO₂ and MOPITT CO is performed, revealing that NO₂ 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.

We thank the reviewer for their overall positive feedback as well as specific comments. We address each of the specific comments below:

Specific comments:

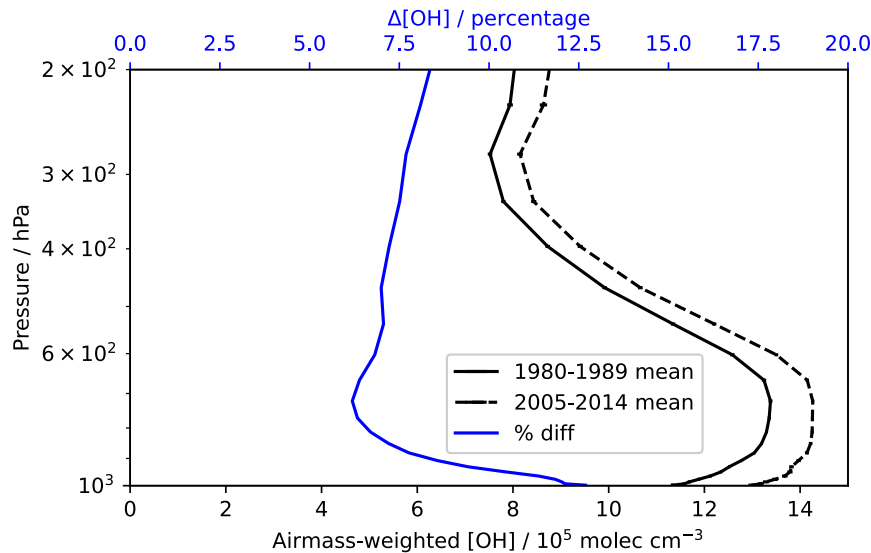
Table 1: Curious that a wavelength cutoff of 310 nm is used for O₃->O₁D photolysis; most other models (e.g., Lelieveld et al., 2016, which you compare to throughout this manuscript) use 330 nm due to the small contribution from the quantum yield tail – see, e.g., Armerding et al., 1995:

<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?

We have changed it to say ‘<330 nm’. In our model, we use the Fast-JX version 7.1 scheme (Wild et al., 2000; Bian and Prather, 2002) to calculate photolysis rates which includes this larger wavelength tail, and does not have a fixed cutoff for wavelength.

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.

The altitudinal profile of [OH] is reported in airmass-weighted concentrations of molecules per volume following other papers reporting [OH] such as Zhao et al. (2019). However, we have now also plotted the percentage change of 2005-2014 mean compared to 1980-1989 mean at each altitude in Fig. 2(b), instead of just presenting the absolute values (this figure is the same whether we use volume mixing ratios or airmass-weighted concentrations of molecules per volume).



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.

We acknowledge the comment and are also aware of the drawbacks of the current modelling setup. However, it is clear from our simulations that the imposed changing methane concentrations are causing significant OH trends. While the feedbacks are missing on the surface, methane and OH are still allowed to interact throughout the rest of the troposphere. Forcing the model with surface methane emissions instead of surface concentrations will likely further amplify these significant OH trends due to the methane self-feedback. In our paper, we have taken more caution to emphasize that we are not using methane fluxes. We have added a paragraph in the 'Implications' section to say 'We acknowledge that CH₄ concentrations are prescribed on the surface in the current model set-up, so this can lead to an underestimation of the surface chemical feedbacks. Including the surface feedbacks would likely amplify the modelled effects of CH₄ on [OH]. This should be further investigated in an emissions-driven run.'

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.

The non-zero emissions trend over the oceans for CO comes from shipping. The non-zero-emissions trend over the oceans for NO_x comes not only from shipping but also from aircraft as well.

Technical corrections:

L34: "tropospheric" misspelled

Done.

L86: "increasing" should be "increase"

Done.

L167: Check punctuation; period should be comma.

Done.

L203: should be "as well as"

Done.

L227: sensitivity misspelled

Done.

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

Done.

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")

Done.

L296: should be "out of"

Done.

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

Done.

L340: "increases" misspelled

Done.

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

Done.

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

Done.

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

Thank you for your suggestion. We have revised it.

L445: "agrees" should be "agree"

Done.

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

Thank you for pointing this out. We have revised it.

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

Thank you for pointing this out. We have revised it.

Thank you for the technical corrections, they have all been addressed and/or amended.

References

Bian, H. and Prather, M. J.: Fast-J2: Accurate Simulation of Stratospheric Photolysis in Global Chemical Models, *Journal of Atmospheric Chemistry*, 41, 281–296, <https://doi.org/10.1023/a:1014980619462>, 2002.

Wild, O., Zhu, X., and Prather, M. J.: Fast-J: Accurate Simulation of In- and Below-Cloud Photolysis in Tropospheric Chemical Models, *Journal of Atmospheric Chemistry*, 37, 245–282, <https://doi.org/10.1023/a:1006415919030>, 2000.

Zhao, Y., Saunio, M., Bousquet, P., Lin, X., Berchet, A., Hegglin, M. I., Canadell, J. G., Jackson, R. B., Hauglustaine, D. A., Szopa, S., Stavert, A. R., Abraham, N. L., Archibald, A. T., Bekki, S., Deushi, M., Jöckel, P., Josse, B., Kinnison, D., Kirner, O., Marchal, V., O'Connor, F. M., Plummer, D. A., Revell, L. E., Rozanov, E., Stenke, A., Strode, S., Tilmes, S., Dlugokencky, E. J., and Zheng, B.: Intermodel comparison of global hydroxyl radical (OH) distributions and their impact on atmospheric methane over the 2000–2016 period, *Atmospheric Chemistry and Physics*, 19, 13 701–13 723, <https://doi.org/10.5194/acp-19-13701-2019>, 2019.