Review of “Inter-model comparison of global hydroxyl radical (OH) distributions and their impact on atmospheric methane over the 2000-2016 period” by Zhao et al for ACP

This study presents a multi-model comparison of the distribution and trends in hydroxyl (OH) radical and an analysis of how the different OH fields influence atmospheric methane trends over the 2000-2016 period. This paper is very relevant to the current debate in the scientific literature about the drivers of recent methane increase in general and specifically the role of OH in this renewed methane growth since 2007. Generally, the paper is well-organized and addresses scientific questions within the scope of ACP.

The authors have analyzed the free-running coupled chemistry-climate model simulations derived from the CCMI project as well as from three additional models not participating in CCMI. Like the ACCMIP models, the models considered here also produce a wide range of OH distributions and agree on the sign of change in OH over the 1980 to the present (2010) time period. The authors largely attribute the reasons for intermodel diversity in the simulated mean OH distributions to differences in the representation of nitrogen oxide, and natural emissions of NMVOCs and their chemistry. I felt that the analysis could be a bit deeper, especially the role of differences in lightning NOx and stratospheric ozone could be investigated. The authors also do not evaluate the models against proxy observational constraints (such as ozone, water vapor, CO, NOx column, methane lifetime) to assess their skill in simulating OH levels. How do we know which model is closer to reality? I acknowledge that this is a difficult question to answer but feel that some effort is needed to evaluate the models. Finally, the authors attribute the increasing trend in tropical OH over 2000 to 2010 to increasing NOx emissions, again without an in-depth quantitative analysis. I have highlighted these and other issues in my specific comments below. I recommend the publication of this paper after it has been revised to include additional evaluation and analysis.

Specific Comments:

L70: Levy (1972) is the wrong reference here. The correct reference should be Levy (1971).

L71: Atomic excited oxygen is denoted as $O(^1D)$. Please revise $O^{1D}$ throughout the manuscript.

L99-103: The tropospheric methane chemical lifetime against OH loss from ACCMIP models was calculated as the global annual mean **atmospheric** methane burden divided by annual mean methane tropospheric loss by OH. Please revise.

L112-114: “The precise scenario…” sounds odd. Please revise to “The precise reasons for stagnation and renewed growth of methane still remain unclear…”

L127-130: With their chemistry model, Dalsoren et al (2016) actually simulate an ~8% increase in OH over the 1970 to 2012 time period. Not only is this trend as large as that inferred by Turner et al and Rigby et al, but is also in a completely opposite direction and is also contrary to the no OH trend deduced by Nicely et al (2018). I think the point that there is a mismatch in the OH trends inferred from observations and simulated by global models should be explicitly highlighted.

L141-144: The configuration of ACCMIP simulations was not ideal for assessing interannual variability in OH due primarily to the fact that these simulations were not performed continuously from year to year but were timeslices with emissions as well meteorology characteristic of the decade being simulated. So in that sense, the ACCMIP simulations did allow one to assess decadal (or multi decadal) but not year to year variability. I think this sentence should be revised to something like this for clarity: “Year-to-year integrations of CCMI and INCA models driven by time-varying emissions and meteorology facilitate the investigation of interannual variability in OH which was not possible using the ACCMIP time-slice simulations”

L158-159: From the perspective of understanding the influence of OH changes on methane, I think the analysis of REF-C1SD is more relevant because it will likely reproduce (or be close to) observed changes in various climate related factors that influence OH (e.g., humidity, temperature) as well as chemical composition since it is nudged to observed meteorology. I would highly encourage the analysis of REF-C1SD simulations if results from at least 5 of the models analyzed here are available.

L168-169: Please clarify what is tropospheric ozone chemistry coupled to - stratospheric chemistry, physical climate?

L172-173: Define the chemical names before using them HCHO (formaldehdye) and C5H8 (isoprene).

L177-178: I think it would be helpful to clarify that after year 2000, the MACCity emissions (therefore REF-C1 simulation) follow the RCP8.5 scenario. It would also be helpful to clarify whether biomass burning emissions are also from MACCity and if they vary from year to year.

L190: According to the description in Morgenstern et al (2017), methane concentrations prescribed in the CCMI models vary in time following the RCP6.0 contrary to the specification (global mean of ~1750ppbv averaged from over 2000-2010) described here. Please clarify.

Section 2.1: It would be helpful if the authors could clarify how tropopause is defined (in order to calculate tropospheric OH from the models) somewhere in this section.

Section 2.2.1: The motivation for using this model and simulations became clear to me only after I read section 2.2.2. I think the motivation for using this model with methane emissions should be clarified up front in this section.

L232: access should be replaced with assess.

L238-239: It is mentioned above that the offline LMDz5B is run with OH field from CCMI models. Does it also use O(1D) fields from the CCMI models?

L245-248: Please clarify which chemistry module is used in the setup here.

L301, L552: Please confirm if this is indeed 1‰ yr-1 or 1% yr-1.

L313-314: Revise "...which overestimation of [OH]..."

L317: IPCC (2011) is missing from the reference list. Also suggest citing the IPCC chapter (though I do not think there is a 2011 IPCC report relevant here) rather than the whole report.

Tables 3, 4, 5, and 7: It would be helpful to provide a quantitative measure of model spread (e.g., coefficient of variation or range) at the end of the columns which would make it easier for the reader to quickly get an estimate of intermodel diversity.

L336-347: In addition to individual model maps, it would be helpful to have a map of the standard deviation of OH concentrations across the models considered here to clearly see the regions of high/low spread. Also, could variability in lightning NOx emissions across models be a cause of the model spread in OH? What about differences in simulated stratospheric ozone across the models; relevant for southern hemisphere OH differences? I think a deeper analysis of the factors that influence OH is needed to assess the reasons for the spread in OH fields.

L356: Is it GEOSGCM or GEOSCCM?

L390-410: Murray et al. (2014) show that lightning NOx plays a key role in controlling OH as also mentioned on lines 414 to 417. How different/similar are the models in their representation of the vertical distribution of lightning NOx emissions?

Figure 3 would also benefit from plot of standard deviation across models for each region.

L430-431: What is the mean year to year variation over this period in units of percent?

L432-434: Are the numbers in parentheses the changes in OH concentrations from 1960 to 1980? If so, it would be helpful to provide percent changes as well.

L447-450: How do the year-to-year variations in OH from CCMI and INCA models compare with the results of Turner et al. (2017), Rigby et al. (2017) and Nicely et al. (2018) using different approaches?

L455-458: During this time period, there have been increases in water vapor as well (e.g., Dessler and Davis 2010) that could potentially influence OH trends in the tropical regions. Admittedly, the increase in NOx emissions has been significant, but I am not sure if the analysis presented here can be used to say, with confidence, that the strong positive trend in OH is being solely driven by NOx emissions. It would be helpful to perform some regression analysis to build confidence in the conclusions here.

L459-460: Changes in stratospheric ozone also dominate in the southern hemisphere and could potentially add to inter-model differences.

L478-485: The sensitivity of CH4 oxidation due to OH to lower tropical tropospheric temperature has been established by prior studies (see John et al., 2012 and references therein), some of which should be cited here. Additionally, the discussion here will benefit from a table of CH4 loss flux simulated by LMDz using the different OH fields for tropics (30S-30N), northern mid to high lat (30N-90N), and southern mid to high (30S-90S) and for three vertical levels. Or these numbers could be plotted in the form of Lawrence plots as in Lawrence et al. (2001) for each OH field. This would clearly show the diversity in the spatial distribution of methane loss resulting from the different OH fields.

L497-499: Is the scaling applied to every year over the 2000-2010 period or just for year 2000?

L510-513: I am not sure if it is surprising that there was a spread in the simulated methane distributions across models, particularly, because the scaling was performed on a global scale (matching the global methane loss flux) rather than at the grid-cell level. While the global OH may match with INCA NMHC OH field after scaling but the spatial distribution may still be different producing differences in the simulated global mean methane distributions. .

L534-535: Is this true for all the models shown in figure 7? From the figure, methane for Run_fix_Oh and run_standard seem to overlap for INCA NMHC and GEOSCCM models.

L545: It should be EMAC-L90MA.

L544-547: Add “relative to the Run_fix_OH” after “....further reducing CH4 mixing ratios by up to 20-30 ppbv in 2016…”

L565: Have the measurements been combined in a specific way to create global mean? Are the model CH4 values sampled at the location of these stations?

L569-571: Reference Figure 8 here.

L574-576: From Figure 8, it looks like that the three lines (black obs, blue Run_standard and grey Run_fix_OH) are overlapping until about 2003. y -axis of Figure 8 also needs concentration units (ppb?)

L576-578: I am not sure if I understand this sentence (especially “fill the gap between model simulations and observations by up to 50%”). Could the authors please clarify and also how these percentages have been calculated.

L609-610: This assertion needs to be substantiated or toned down in the absence of more detailed analysis (due to lack of diagnostics such as OH prod and loss). I believe the Riahi et al 2011 reference is not appropriate here as it documents RCP8.5 emissions but does not comment on chemistry-composition impacts from changes in these emissions.

References:

Dessler, A. E., and S. M. Davis (2010), Trends in tropospheric humidity from reanalysis systems,J. Geophys. Res.,115, D19127, doi:10.1029/2010JD014192.

John, J. G., Fiore, A. M., Naik, V., Horowitz, L. W., and Dunne, J. P.: Climate versus emission drivers of methane lifetime against loss by tropospheric OH from 1860–2100, Atmos. Chem. Phys., 12, 12021-12036, https://doi.org/10.5194/acp-12-12021-2012, 2012.

Levy, H.II (1971), Normal atmosphere: Large radical and formaldehyde concentrations predicted, Science, 173, 141–143.

Nicely, J. M., Canty, T. P., Manyin, M., Oman, L. D., Salawitch, R. J., Steenrod, S. D., et al. (2018). Changes in global tropospheric OH expected as a result of climate change over the last several decades. Journal of Geophysical Research: Atmospheres, 123, 10,774– 10,795. https://doi.org/10.1029/2018JD028388

Rigby M, et al. Role of atmospheric oxidation in recent methane growth. Proc Natl Acad Sci USA. 2017;114:5373–5377

Turner AJ, Frankenberg C, Wennberg PO, Jacob DJ. Ambiguity in the causes for decadal trends in atmospheric methane and hydroxyl. Proc Natl Acad Sci USA. 2017;114:5367–5372.