Reply to RC1: 'Review of "On the role of trend and variability of hydroxyl radical (OH) in the global methane budget"'

Comment: The manuscript first discusses the OH variability and trends in details in relation with precursor emissions and chemistry as used in the chemistry-climate models. Then the modelled OH fields, adjusted to the global mean OH in 2000, are used for CH4 modelling in a box model and a sophisticated 3D model. They show good agreement (?) between the two models for global total CH4 budgets. The manuscript is well written and would be alright for publication in ACP. I would like to draw attention of the authors to a few points as detailed below. Hope these are useful.

Response: We thank the reviewer for his/her helpful comments. All of them have been addressed in the revised manuscript. Please see our itemized responses below.

Comment: line 49: Do you need an update here? Is the present day understanding is unambiguous too?

Response: We change the sentence to (L47-49): "The tropospheric CH_4 mixing ratio has more than doubled between pre-industrial and the present day, mainly attributed to increasing anthropogenic CH_4 emissions (Etheridge et al., 1998; Turner et al. 2019)."
We add the reference: "Turner, A. J., Frankenberg, C., and Kort, E. A.: Interpreting contemporary trends in atmospheric methane, Proceedings of the National Academy of Sciences, 116, 2805-2813, 10.1073/pnas.1814297116, 2019."

Comment: line 56: The citations look to be very restricted in general in these two para of the introduction. May consider expansion. The TransCom-CH4 project was launched to underdstand the sources and sinks budget in comparison with the model transport, for example.

Response: We change the citation to "TransCom-CH4", thank you very much for pointing out this.

Comment: line 72: I am not aware of any proven issues with weakening MCF gradient. Could you pleas expand what exactly are you talking about; meridional, zonal or vertical gradients?

Response: We clarify in the text (L73-74):" ... and the weakening of inter-hemispheric MCF gradients after the 1990s (Krol et al., 2003, Bousquet et al., 2005; Montzka et al., 2011; Prather and Holmes, 2017)."

Comment: line 92: are CH4 budgets from 1980 or 1986

Response: We change the sentence to "... on decadal scale since the 1980s". and we

add in L97-98:" We finally estimate the impact of OH year to year variations and trends on the top-down estimation of global CH_4 emissions over 1986-2010". Since we analyzed the OH variation for 1980-2010 and analyzed the inversion results for 1986-2010.

Comment: line 114: There are 4 issues with OH from CCMI models; you seems to ignore the biases in meridional gradient in OH, and account for the other three (global totals, trends and IAV)

Response: In this work, we are focusing on the temporal variation of OH and the impact on CH_4. The impacts of OH inter-hemispheric gradient have been estimated in Zhao et al. (2020) and therefore less discussed in this study.

Reference:" Zhao, Y., Saunois, M., Bousquet, P., Lin, X., Berchet, A., Hegglin, M. I., Canadell, J. G., Jackson, R. B., Dlugokencky, E. J., Langenfelds, R. L., Ramonet, M., Worthy, D., and Zheng, B.: Influences of hydroxyl radicals (OH) on top-down estimates of the global and regional methane budgets, Atmos. Chem. Phys. Discuss., 2020, 1-45, 10.5194/acp-2019-1208, 2020."

Comment: line 120: Could this also mean that the variabilities you show are not from OH concentrations but due to t-dependent loss rates & dry airmass. Is it possible to tell the readers what would you expect if you scale OH concentrations themselves not weighted by k? including showing it in a 2nd column?

Response: For the scaling, we apply the single global scaling factor estimated for 2000 to every year of the OH field. Hence the scaling will not influence the interannual variation of OH. We have clarified this in the text (L116-117):" The inferred global mean scaling factors are calculated for the year 2000 and each OH field and then applied to the whole period (1980-2010)"

Comment: line 157: I fail to understand why continuous inversion were not done for the period 1994-2010, using the two OH cases. This does not seem to be for reducing computing time, given that 2 years are gone for spin-up and spin-down. Please explain. Also why you need two years of spin-up/down for the box model but only 1-year for the 3D model.

Response: We do the inversion separately for each time period so that the inversions can be run in parallel at the same time. The 3D model inversions are much more computationally expensive than the box model inversions. Hence we only take one-year spin-up and spin-down for the 3D model. We clarify in the text (L159-L160):" We only spin-up/spin-down the 4D variational inversions for one year to save computing time."

Comment: line 185: What do you mean? I see many other negative anomalies are apparently consistent among the models.

Response: We clarify this issue in the text by adding (L187-188):" Only the negative OH anomaly during 2006-2007 (2±1%) is simulated by all models during the four weak El Niño events."

Comment: line 195ff: I agree that the CH4 growth rates are more positive during the El Nino years (discussed in the TransCom-CH4 analysis too). We have to better understand the lower growth rates in CH4 during the La Nina periods - this is quite new concept. (some people talk about Mt Pinatubo for 1993 growth rate anomaly and others do not see a negative anomaly during the La Nina).

Response: Here we want to show that the CH_4 growth rate is smaller during the La Niña years comparing with their adjacent El Niño years. We make the expression more precise in the text (L96-98):" The positive anomalies of $[OH]_{GM-CH4}$ during La Niña events correspond to a much smaller CH_4 growth (e.g. 3.8±0.6ppbv yr^{-1} in 1993 and 2.3±0.8ppbv yr^{-1} in 1999) compared with that during the adjacent El Niño years (Fig. S1)."

Comment: line 200: do you need "processes" here?

Response: We remove the "processes".

Comment: line 204: Is there a reason for different unit (Tg/yr) here?

Response: We correct the typo by changing"Tg/yr" to "Tmol yr^{-1}"

Comment: line 212: should this be "different"? Do you mean the NMVOCs are not included in some of the model or the number of species differ from model to models?

Response: Here we mean the model outputs of OH loss due to reaction with NMVOCS, we clarify this in the text(L212-213):" Besides, there are 12% of OH production and 33% of OH loss not analyzed here due to lack of data in the CCMI model outputs (e.g. output of OH loss due to reaction with NMVOCs included in different models)"

Comment: line 214: My personal choice, but I would have loved to see the actual values presented in this plot. It is fine to adjust different multi-model values to a common 1980 level.

Response: We agree that it will be more straightforward to show the actual values. However, we here adjust the model values to a common 1980 to focus more on the temporal variations from 1980.

Comment: For here and elsewhere, this is specialised journal publication, there is no need for so much space restriction; I mean this can be 1-column figure is the trends are less prominent by the increase of y-axis range. Also for the x-axis tick labels, please consider reducing number of labels or elongate the x-axis or introduce minor ticks. Presently looks a bit clumsy.

Response: We plot the trends of different chemical processes in the same panel to better compare the relative contribution from each process and find out which process is most important for determining OH trend. We replot figure 1-4 to reduce x-axis tick labels as suggested.

Comment: line 220: This is a nice discussion, but I cannot assess the novelty of it given that ACCMIP and CCMI paper have discussed the OH variabilities and budgets in similar fashion, and there are papers by MPI Mainz group on the details of OH budgets. Could you please consider showing the net (P-L) OH trends in a separate panel. When you say OH loss, is the '-ve' sign in the y-tick labels appropriate and consistent with the number in the text?

Response: ACCMIP provides the model outputs for time slices (Naik et al., 2013), which limit the analysis of OH interannual variations. For the studies based on CCMI model simulations, both Zhao et al. (2019) and Nicely et al. (2020) analyzed the OH interannual variations but not the OH budget. The OH budget has already been analyzed in previous single model studies such as Murray et al. (2013; 2014) (as we cite in the manuscript) and Lelieveld et al. (2016) (paper from MPI Mainz group). But to our knowledge, few studies are analyzing the interannual variation of OH budget based on multi-model outputs.

We cite Leliveld et al. (2016) in the text (L201-203): "Here we assess the drivers of OH year-to-year variations and trend by calculating the OH production and loss processes listed in Table 2 following Murray et al. (2013; 2014) and Lelieveld et al. (2016)."

Reference:" Lelieveld, J., Gromov, S., Pozzer, A., and Taraborrelli, D.: Global tropospheric hydroxyl distribution, budget and reactivity, Atmos. Chem. Phys., 16, 12477-12493, 10.5194/acp-16-12477-2016, 2016."

Showing the net (P-L) will certainly help for better understanding the OH variations. However, only 2 of 5 models provide both total OH production and OH loss data, so we cannot give the temporal variations of net OH production and loss based on current model outputs.

For the "-ve" sign (I suppose the reviewer means "negative"), we now clarify this issue in the figure caption.

"Figure 2. Annual total OH tendency (Tmole yr^{-1}) from chemical reactions with respect to the year 1980 with year-to-year variations removed. The positive and negative tendencies represent OH production (left) and loss processes (right), respectively."

"Figure 3. Anomaly of the detrended annual global total OH tendency from reactions $O(^1D)+H_2O$, $NO+HO_2$, O_3+HO_2, and CO+OH. The positive and negative tendencies represent OH production and loss processes, respectively."

Comment: line 236: I was probably asking to present something similar in my earlier comment for OH anomaly in Fig. 1. May be it is good to show the Net OH (production - loss) variabilities as well in a separate panel here (in % change).

Response: As stated in response to the last comments, we cannot assess the net OH production and loss for most of the models due to a lack of corresponding output.

Comment: line 266: How good are the CO emission estimations and also the satellite data? I have heard some issues with the MOPITT data retrievals. Is this model comply with surface CO observations?

Response: The trend and variations of the tropospheric CO column simulated by CCMI models have been evaluated by comparison with MOPITT data retrievals (Strode et al., 2016). Here we are focusing on the OH loss by CO over the whole troposphere (instead of the surface). The consistency of CCMI simulated tropospheric CO column with MOPITT observations can support the model simulated decreasing tropospheric OH loss by reaction with CO during 2000-2010.

We add in the text (L220-222): "The negative trend of CO simulated by CCMI models during 2000-2010 is consistent with MOPITT observations over most of the regions (Strode et al., 2016)."

Despite the finding that atmospheric chemistry models generally capture the CO trend, they usually underestimate the atmospheric CO burden compared to surface and satellite observations. We add in the text (381-383): "For example, underestimation of CO, especially over the northern hemisphere, compared with the surface and satellite observations (Naik et al., 2013; Strode et al., 2016) and bias in atmospheric O_3 column (Zhao et al. 2019)."

We add the reference:" Strode, S. A., Worden, H. M., Damon, M., Douglass, A. R., Duncan, B. N., Emmons, L. K., Lamarque, J. F., Manyin, M., Oman, L. D., Rodriguez, J. M., Strahan, S. E., and Tilmes, S.: Interpreting space-based trends

in carbon monoxide with multiple models, Atmos. Chem. Phys., 16, 7285-7294, 10.5194/acp-16-7285-2016, 2016."

Comment: line 274: How good are these for accounting for the effect of the meteorology. I suppose the temperature effect is taken in to account by doing the k_oh+ch4 anomalyin Fig. 1, but there are likely to have some non-linear interaction between the transport (inter-hemispheric & stratosphere-troposphere exchange) and loss by OH. This effect may be of 2nd order but nevertheless important. Any assessment would be helpful to the readers. This is where a long-term 3-D model based inversion would have helped.

Response: The long-term 3D model inversions can certainly help better assess the impact of OH variations. However, the 3D model inversions are computationally expensive, which limits conducting long-term 3D model inversions using all of the 9 OH fields present in this study. That's why we use the two-box model to do long-term inversions and do 3D model inversions with a focus on four time periods.

We have demonstrated the advantage of both box model and 3D model inversions in the text (L130-134): "The two-box model inversions allow us to easily conduct multiple long-term global scale inversions (1984-2012) with each of the nine OH fields to estimate the global CH_4 emission variations caused by various OH fields. The 4D variational inversions allow us to better represent atmospheric transport, account for the variation of meteorological conditions, and address regional CH_4 emission distributions."

And we have compared the emission changes estimated by two-box model inversions and 3D model inversion in Figure 5, which show that (L302-L306) "Despite the limitations inherent to two-box model inversions, such as treatment of inter-hemispheric transport, stratospheric loss, and the impact of spatial variability (Naus et al., 2019), the two-box model inversion estimates similar temporal changes of CH_4 emissions and losses compare to the variational approach for the four periods, as well as their response to OH changes (Fig.5), on a global scale. Such comparisons reinforce the reliability of the conclusions made from the two-box model inversions regarding changes in the global total CH_4 budget.".

Line280 : It is obvious from this analysis that introducing OH IAV as modelled by the CCMI models will reduce the CH4 emission anomaly. What are the new questions/implications? 1) increase the wetland emission anomaly, 2) decrease biomass burning emission anomaly, 3) some missing process in the OH chemistry (recycling efficiency).

Response: We have discussed the implication on top-down estimated wetland

emissions in the "Conclusion and discussion" (L393-396): "The variational inversions using OH with temporal variations attribute the observed rising CH_4 growth during El Niño to the reduction of CH_4 loss instead of enhanced emissions over the tropics, which are consistent with process-based wetland models that estimated wetland CH_4 emission reductions at beginning of El Niño event (Hodson et al., 2011; Zhang et al., 2018)."

We also discuss the impact on top-down estimated biomass burning emissions (L400-L401): "Also, the negative OH anomaly can reduce the top-down estimated biomass burning CH_4 emission spike during El Niño events, similar to that presented in Bousquet et al. (2006)."

We discuss the implications of including full-chemistry estimated OH by comparing with previous results which only consider OH loss by CO (L362-365): "We estimated that the negative OH anomaly in 1998 reduces the high top-down estimated CH_4 emissions by 10±3Tg yr^{-1}, ~40% smaller than the reduction estimated by Butler et al. (2005) (16Tg yr^{-1}), which only include the OH reduction response to enhanced biomass burning CO emissions. The smaller CH_4 emission reductions (OH anomalies) estimated with CCMI OH fields shows the significance of considering multi chemical processes as included in the 3D atmospheric chemistry model in capturing OH variations and inverting for CH_4 emissions"

Reference: "Butler, T. M., Rayner, P. J., Simmonds, I., and Lawrence, M. G.: Simultaneous mass balance inverse modeling of methane and carbon monoxide, Journal of Geophysical Research: Atmospheres, 110, 10.1029/2005jd006071, 2005."

Comment: line 285: The most important question for the authors is then to convince the readers how you propose to increase CH4 emissions by more than 25 Tg/yr in just 3 years and keep maintaining at that level for the later years.

Response: The large emission increase after 2005 have been reported in previous studies (e.g. Kirschke et al. (2013), Saunois et al. (2017)). We compare the emission increase over the same period with previous study in the text (L279-281):" The CH_4 emissions averaged over 2006-2010 is 20Tg yr^{-1} higher than over 2000-2005, within the range of 17–22Tg yr^{-1} estimated by an ensemble of inversions in Kirschke et al. (2013)."

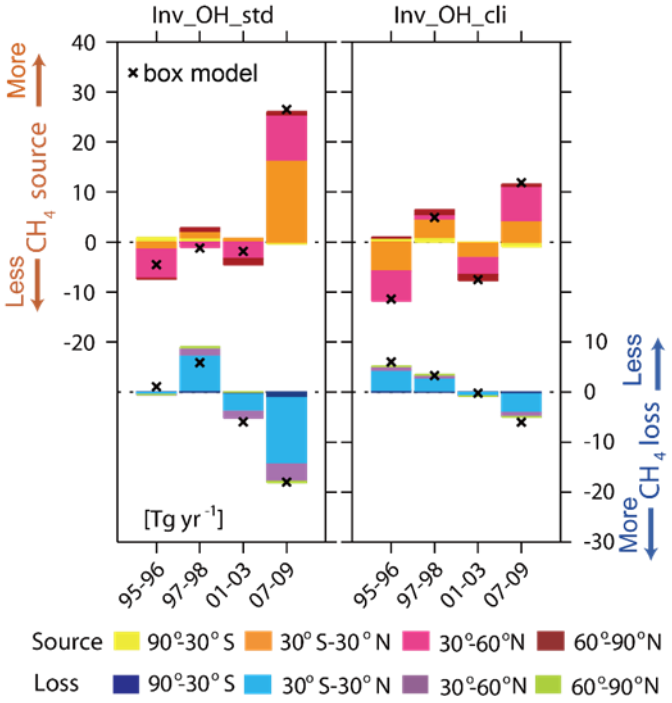
We add the reference: " Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J. G., Dlugokencky, E. J., Bergamaschi, P., Bergmann, D., Blake, D. R., Bruhwiler, L., Cameron-Smith, P., Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E. L., Houweling, S., Josse, B., Fraser, P. J., Krummel, P. B., Lamarque, J.-F., Langenfelds, R. L., Le Quéré, C., Naik, V., O'Doherty, S., Palmer, P. I., Pison, I., Plummer, D., Poulter, B., Prinn, R. G., Rigby, M., Ringeval, B., Santini, M., Schmidt, M., Shindell, D. T., Simpson, I. J., Spahni, R., Steele, L. P.,

Strode, S. A., Sudo, K., Szopa, S., van der Werf, G. R., Voulgarakis, A., van Weele, M., Weiss, R. F., Williams, J. E., and Zeng, G.: Three decades of global methane sources and sinks, Nature Geoscience, 6, 813-823, https://doi.org/10.1038/ngeo1955, 2013."

Comment: line 301: "assess"
Response: Changes as suggested

line 303ff: Consider adding header to each panels - again panel size could be increased for clarity. It is very hard to see the semi-hemispheric emissions in the bars. The right panel adds to the confusion how to read this plot; for me it is much easy to see what you want say from the left and middle panels.

Response: We change the figure as suggested.



Comment: line 366: Did Prather and Holmes estimated OH variability or trends?
Response: Prather and Holmes (2017) showed uncertainties exist in the MCF-constrained OH using two-box models.

Comment: line 372: I do not know true or not true? The authors, I think, understand the trends and IAV in OH simulated by the CCMs still require much testing. Firstly the global mean OH values, which you have adjusted at the very first; the amplitude and phase of the modelled IAV may not be perfect; the longer term trends are still anyone's guess (needs at least one more line of evidence); finally there is unspoken bias in meridional gradients in the modelled OH. If a true variability and trend in OH become available there would be no issues with the top-down modellers to adopt it. There are several inversions which included the OH trends and variability like you discuss here

(e.g., McNorton et al., ACP, 2018)

Response: We discuss the uncertainties in OH simulated by atmospheric chemistry models in the text now in more detail: "However, the CCMI models still show biases that are related to OH production and loss. For example, these include an underestimation of CO especially over the northern hemisphere compared with the surface and satellite observations (Naik et al., 2013; Strode et al., 2016) and bias in atmospheric total O_3 column (Zhao et al. 2019). In addition, the changes in aerosols (Tang et al., 2003) and atmospheric circulation such as the Hadley cell expansion (Nicely et al., 2018) are not discussed in this study. Given the large discrepancy between MCF–based and CCMI model simulated OH trends, and the uncertainties in both model simulated (Naik et al., 2013; Zhao et al., 2019) and MCF-constrained (Bousquet et al., 2005; Prather and Holmes, 2017; Naus et al. et al., 2019) OH, the OH trend after the mid-2000s remains an open problem and more effort is required in both method to explore the close the gap. "

We change L372 to: "The temporal variations of OH, which are generally not well constrained in current top-down estimates of CH_4 emissions, imply potential additional uncertainties in the global CH_4 budget (Saunois et al., 2017; Zhao et al., 2020)."

Comment: line 382: Is there a better reference for El Nino in future climate? I am sure there are

Response: We change the reference to "Berner, J., Christensen, H. M., and Sardeshmukh, P. D.: Does ENSO Regularity Increase in a Warming Climate?, Journal of Climate, 33, 1247-1259, 10.1175/jcli-d-19-0545.1, 2020."

Comment: line 384: I think this is well known since the ACCMIP at the least!

Response: We change the sentence to "Our research emphasizes the importance of considering climate changes and chemical feedbacks related to OH in future CH_4 budget research."