Reply to RC2: 'Referee comments'

This study investigates the impact of CCM calculated OH fields on the long-term trend and interannual variability in global methane emissions as inferred from atmospheric inversions. It is concluded that accounting for the CCM simulated trend in OH implies a significantly larger increase in methane emissions in the past decades than previous estimates that did not consider changes in OH. In addition, correlations between OH variations and El Nino are found that reduce the variability in inferred emission, notably in the Tropics. The manuscript, that is well written, provides a useful contribution to the scientific discussion on the drivers of the global increase of methane, contrasting the view on the role of OH that follows from the more common use of MCF. The study also mentions important limitations of using MCF, suggesting that the CCM-derived OH scenario is more consistent. However, in my opinion the evidence in support of this suggestion is very limited. Besides this point, there are a few other points that need further attention as explained below. Overall, only minor revisions will be needed to accept this paper for publication.

Response: We thank the reviewer for his/her helpful comments. We have now discussed the limitations of the CCMI-derived OH in more detail in the text. All of the other comments have been addressed in the revised manuscript. Please see the itemized responses below.

Comment: The study provides an analysis of the main drivers of variability and trends in OH using output from the CCMs. It is mentioned that both chemical and climatological influences are considered. However, the budget that is provided in Table 2 only lists chemical drivers. A study by Dentener et al (ACP, 1993) (which would be worth referencing in this context) identified an important contribution of meteorology on OH variability. Apart from changes in water vapour, those influences, including temperate and changes in circulating, are not discussed here.

Response: Dentener et al. showed that water vapor is the main meteorological driver of OH trend during 1979-1993. Our analysis also shows that the increase of water vapor can contribute to a positive OH trend by enhancing OH primary production (L217-L219): "The increase in OH primary production is due to an increase in both tropospheric O_3 burden (producing $O(^1D)$) and water vapor (Dentener et al 2003; Zhao et al., 2019; Nicely et al., 2020)."

The impact of water vapor on OH variability is also discussed in the manuscript (L246-L248): "The increase in OH primary production is mainly due to an increase in tropospheric water vapor and O_3 burden during El Niño events (Fig.S3 and S12 in Nicely et al. (2020)), while the increase in OH secondary production is caused by enhanced NO_x emissions (Fig.S3) and O_3 formation (Nicely et al. (2020) related to biomass burning as well as more HO_2 formation by CO+OH."

The temperature mainly influences the reaction rates of OH with other chemical species (CO, CH4, NMVOC, etc.), which is included in our estimation of OH production and loss (Table S1). Nicely et al. (2018; 2020) have proven that temperature has a neglectable impact on OH trend.

Due to a short lifetime, the OH is controlled by local production and loss, thus less directly impacted by atmospheric transport. Nicely et al. (2018) shown that the Hadley cell expansion can have a small impact on tropospheric mean [OH] through changing tropopause height. We add this point in the text (L380-383): "In addition, the changes in aerosols (Tang et al., 2003) and atmospheric circulation such as Hadley cell expansion (Nicely et al., 2018) are not discussed in this study."

We also add the following references:
"Dentener, F., Peters, W., Krol, M., van Weele, M., Bergamaschi, P., and Lelieveld, J.: Interannual variability and trend of CH4 lifetime as a measure for OH changes in the 1979–1993 time period, Journal of Geophysical Research: Atmospheres, 108, 4442, 10.1029/2002jd002916, 2003."
" Nicely, J. M., Canty, T. P., Manyin, M., Oman, L. D., Salawitch, R. J., Steenrod, S. D., Strahan, S. E., and Strode, S. A.: Changes in Global Tropospheric OH Expected as a Result of Climate Change Over the Last Several Decades, Journal of Geophysical Research: Atmospheres, 123, 10,774-710,795, doi:10.1029/2018JD028388, 2018."

Comment: The discussion of the results rightly mentions the different outcomes obtained using MCF or CCM derived OH, and their significance for global methane. From the evidence that is presented it is not possible in my opinion to conclude which of the views is right. Nevertheless, the conclusion section mentions that 'the evidence for increasing OH given by CCMI models and other literature, the accuracy of MCF-based OH inversions after the mid-2000s remains an open problem'. So, in short, the accuracy of MCF is the "open problem" that could explain the disagreement in the opinion of the authors. However, the consistency between CCM's itself cannot be considered as evidence, since these models are not independent and could therefore all be wrong for the same reason. To give an example that might introduce important uncertainty in CCM derived variations, the role of changes in aerosols on OH is not discussed at all. Either appropriate evidence should be presented of CCM's being more accurate than MCF or a more objective position should be taken regarding this question.

Response: We try to discuss the two methods in the text now in a more balanced way:

"However, the CCMI models still show biases that related to OH production and loss. 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 total O$_3$ column (Zhao et al. 2019). In addition,

the changes in aerosols (Tang et al., 2003) and atmospheric circulation such as Hadley cell expansion (Nicely et al., 2018) are not discussed in this study. Given the uncertainties in both atmospheric chemistry model simulated (Naik et al., 2013; Zhao et al., 2019) and MCF-constrained OH (Bousquet et al., 2005; Prather and Holmes, 2017; Naus et al. et al., 2019), and the large discrepancy between two methods, the OH trend after the mid-2000s remains an open problem and more effort is required in both method to close the gap. "

We add the references:
"Strode, S. A., Duncan, B. N., Yegorova, E. A., Kouatchou, J., Ziemke, J. R., and Douglass, A. R.: Implications of carbon monoxide bias for methane lifetime and atmospheric composition in chemistry climate models, Atmos. Chem. Phys., 15, 11789-11805, 10.5194/acp-15-11789-2015, 2015."
" Tang, Y., Carmichael, G. R., Uno, I., Woo, J.-H., Kurata, G., Lefer, B., Shetter, R. E., Huang, H., Anderson, B. E., Avery, M. A., Clarke, A. D., and Blake, D. R.: Impacts of aerosols and clouds on photolysis frequencies and photochemistry during TRACE-P: 2. Three-dimensional study using a regional chemical transport model, Journal of Geophysical Research: Atmospheres, 108, 8822, 10.1029/2002jd003100, 2003."

SPECIFIC COMMENTS

Comment: line 38, 40: the numbers that are mentioned in these sentences lack an uncertainty estimate.

Response: We change "up to 10Tg yr^{-1}" to "up to 10±3Tg yr^{-1}" and "increase by 23Tg yr^{-1}" to "increase by 23±9Tg yr^{-1}".

Comment: line 148: How is Inv_OH_var detrended?

Response: We clarify in the text "Inv_OH_var stands for the inversion using the detrended OH".

Comment: line 249: What is meant by 'response of OH to CO.'?

Response: We clarify by changing the "response of OH to CO" to "response of OH to enhanced CO emissions during the El Niño events".

Comment: line 253: Banda et al 2016 is a good reference to add here for the effect of Mt. Pinatubo.

Response: Thank you very much for providing this valuable reference. We revise the sentence to:" The positive anomaly OH primary production (0.2±0.5Tmol yr^{-1}) is not significant during 1991-1992 El Niño event, maybe due to absorption of ultraviolet (UV) by volcanic SO$_2$ and scattering of UV by sulfate aerosols as well

as reduction of tropospheric water vapor after the eruption of Mount Pinatubo (Bândă et al., 2016; Soden et al., 2020).”

We add reference:” Bândă, N., Krol, M., van Weele, M., van Noije, T., Le Sager, P., and Röckmann, T.: Can we explain the observed methane variability after the Mount Pinatubo eruption?, Atmos. Chem. Phys., 16, 195-214, 10.5194/acp-16-195-2016, 2016.”

Comments: line 280-283: Similar conclusions on emission anomalies during El Nino were drawn by Butler et al, JGR, 2005, which would be useful references and comparing here.

Response: we add in the text (L358-L363): ” 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 reduction (OH anomaly) estimated with CCMI OH fields may reflect the significance of considering multi chemical processes as included in the 3D atmospheric chemistry model in capturing OH variations and inversing CH_4 emissions.”

Comment: Figure 3: what do the lines represent in this figure represent? Figure 5: Emission anomalies are shown compared to what? Initially I assumed that the mean was subtracted. However, the bars for the different time slices don’t add up to zero.

Response: we add the legend in Figure 3 as shown below. The anomalies in Figure 5 are calculated by comparing to the mean CH_4 emissions of Inv_OH_cli over the four time period as demonstrated in the figure caption, we add the value “494Tg” in the figure caption to clarify.
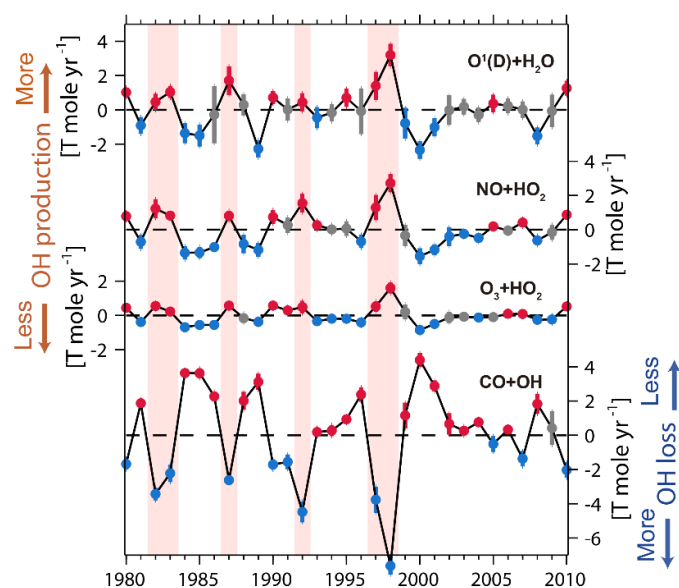
TECHNICAL CORRECTIONS
Comment: line 119: 'For each OH field' io ' .. fields'

Response: Changed as suggested

Comment: line 122: The parenthesis in this sentence should be fixed.

Response: We change the parenthesis to "($[OH]_{GM-CH4}$, weighting factor = reaction rate of OH with CH_4 × dry air mass, Lawrence et al., 2001) as well as of its production and loss rates."

Comments:
line 170: 'This continuous increase in' io 'This continuously increases in', and 'based on MCF inversions' io 'based in the MCF inversions'
line 205: 'NO+HO2' io 'NO+NO2'
line 249: 'anomaly in OH primary' io 'anomaly OH primary'
line 250: 'during the 1991-1992..' io 'during 1991-1992..'
line 322: 'the Tropics' io 'the tropics'
line 324: 'twice that of the inversion' io 'twice of the inversion'
line 334: 'and their impact' io 'and its impact'

Response: The above technical corrections are changes as suggested. Thank you very much for pointing out these.