Reply to RC2: ' Review'

1 Overview:

Review of “Influences of hydroxyl radicals (OH) on top-down estimates of the global and regional methane budgets” by Zhao et al. This review slipped through the cracks as the COVID-19 situation evolved here. My sincere apologies for any hold ups. Zhao et al. present an analysis of a set of methane inversions using a set of 10 different OH fields from the CCMI experiment. They find the magnitude of methane emissions differs by roughly 30% (518-757 Tg/yr) depending on what OH fields they use. Over all, the study is useful in quantifying some of the uncertainties in methane emission estimates due to uncertain OH concentrations. The main shortcoming is the lack of discussion of what actually causes some of the differences (or really any discussion of OH). The figures are high quality but the text is very hard to follow because it’s filled with many acronyms and parenthetical expressions. I would recommend major revisions.

Response: We thank the reviewer for his/her helpful comments. This manuscript is the second step of our previous study, in which we have made a detailed description of the CCMI, INCA and TransCom OH fields and where we analyzed the factors controlling the inter-model differences in OH burden and spatial distribution and the increasing trend of OH simulated by CCMI models (Zhao et al., 2019). This manuscript mainly focuses on the impact of the inter-model differences of different OH fields on the top-down estimates of CH_4 emissions. Thus, regarding discussions on OH, we directly refer to the conclusion from Zhao et al. (2019) (see the response for the comments 2.1) without re-detailing all results. However, we now better explain the link with our first paper in the revised version and include summaries of results from Zhao et al. (2019) in this second paper.

Also, we have rephrased large parts of the original manuscript in this revised version, and especially

Section 3.2, which includes most of the acronyms and parenthetical expressions. All of the other comments have been addressed in the revised manuscript. Please see out itemized responses below.

2 Comments:

2.1 What processes actually drive these differences?

The main issue I feel is totally missing from the manuscript is any discussion of what processes are actually driving some of these differences. I believe there was only a single paragraph (Lines 70-79) even mentioning anything about what might affect OH. For example, do some of the CCMI models or inversions show consistent patterns with known climate oscillations? This was surprising given that this is a paper focused on how OH impacts methane. The obvious question is what leads to these differences/similarities in OH. The authors seem to treat the CCMI models as a black box which makes it hard to gain any understanding of what's happening.

Response:

As mentioned in the overview, this paper follows Zhao et al. (2019) where we analyzed in detail OH fields from CCMI models. Including the main conclusions of this first paper in the updated section 2.1 of this paper provides the required elements on what causes OH differences in CCMI models.

In the introduction, to clarify the link with our previous paper, we added (L112-121):

"This paper follows the work of Zhao et al. (2019), where we analyzed in details 10 OH fields derived from atmospheric chemistry models considering different chemistry, emissions, and dynamics (Patra et al., 2011; Szopa et al., 2013; Hegglin and Lamarque, 2015; Morgenstern et al., 2017; Zhao et al., 2019; Terrenoire et al., 2019). We now aim to build on this previous paper to estimate the impact of these OH fields on methane emissions as inferred by an atmospheric 4D variational inversion system. To do so, we use each of the OH fields in the 4D variational inversion

system PYVAR-LMDz based on LMDZ-SACS (Laboratoire de Météorologie Dynamique model with Zooming capability-Simplified Atmospheric Chemistry System) 3D chemical transport model to evaluate the influence of OH distributions and variations on the top-down estimated global and regional CH_4 budget. Section 2 briefly describes the OH fields and their characteristics and underlying processes (see also Zhao et al., 2019 for more details)"

In section 2.1, we added (L146-L154):

"The inter-model differences of OH burden and vertical distributions are mainly attributed to differences in chemical mechanisms related to NO production and loss. The differences in [OH] spatial distributions are due to applying different natural emissions: for example, primary biogenic VOC emissions and NO emissions from soil and lightning (Zhao et al., 2019). As a result, the regions dominated by natural emissions (e.g. South America, central Africa) show the largest inter-model differences in [OH] (Fig.S1). The CCMI models consistently simulated positive OH trend during 2000-2010, mainly due to more OH production by NO than loss by CO over the East and Southeast Asia and positive trend of water vapor over the tropical regions (Zhao et al., 2019; Nicely et al., 2020). More details can be found in Zhao et al. (2019) and the herein cited literature."

In section 3.1.1 (L310-L314), we added:

"The high $[OH]_{GM\text{-}CH4}$ simulated by SOCOL3 and MOCAGE are mainly due to high surface and mid-tropospheric NO mixing ratio simulated by these two models (Zhao et al., 2019). As analyzed in Zhao et al. (2019), the lack of N_2O_5 heterogeneous hydrolysis (by both SOCOL3 and MOCAGE) and the overestimation of tropospheric NO production by NO_2 photolysis (by SOCOL3) are the major factors behind the overestimation of NO and OH."

Given this, the only major take-away I had from the paper is that "OH can lead to big differences in

methane estimates", but this was already demonstrated by the box modeling papers (and others) that Zhao et al. are highly critical of. For example, the Rigby paper had error bars on their OH fields that bounded zero and the Turner paper had a case where OH didn't change. Both of these led to radically different methane emissions.

Response: Two-box models are an effective tool to assess changes in global CH_4 budget (Rigby et al., 2017, Turner et al., 2017), but we think that 3D analysis is still needed : (i) to check if box-model results are not biased by the oversimplification of atmospheric transport, (ii) to infer the regional CH_4 budgets, and (iii) to estimate methane decadal budgets as box-models are less relevant for estimating budgets than budget changes (Saunois et al., 2019). The study aims to quantify the uncertainties lead by using the prescribed OH fields in 3D model inversions.
Also, we have been more precise in the abstract and conclusions for the reader to get a more complete take away of our work (regions that are sensitive/important, quantitative estimates, comparison with other causes of uncertainties).

We added in the abstract (L25-L27):
"Current top-down estimates of the global and regional CH_4 budget using 3D models usually apply prescribed OH fields and attribute model-observation mismatches almost exclusively to CH_4 emissions, leaving the uncertainties due to prescribed OH field less quantified. "
And we clarified in the text that we conducted inversions using an ensemble of OH fields not only to show that OH can lead to differences in CH_4 emissions but also to quantify the influences at global and regional scales.

In "conclusions and discussion", we compared the uncertainties lead by OH with other causes of uncertainty.

For global total CH_4 emissions (L640-L644):

" Based on the ensemble of 10 original OH fields ($[OH]_{GM\text{-}CH4}$:10.3-16.3 $\times 10^5$ molec cm^{-3}), the global total CH_4 emissions inverted by our system vary from 518 to 757Tg yr^{-1} during the early 2000s, similar to the CH_4 emission range estimated by previous bottom-up syntheses and larger than the range reported by the top-down studies (Kirschke et al., 2013; Saunois et al, 2016). "

For the regional emissions (L647-L654):

" At regional scale (excluding the two highest OH fields), CH_4 emission uncertainties due to different OH global burdens and distributions are largest over South America (37% of multi-inversion mean), South Asia (24%), and China (39%), resulting in significant uncertainties in optimized emissions from the wetland and agriculture and waste sectors. These uncertainties are comparable in these regions with those due to model transport errors and inversion system set-up (Locatelli et al., 2013; Saunois et al., 2016). For these regions, the uncertainty due to OH is critical for understanding their methane budget. In other regions, OH leads to smaller uncertainties compared to that given by Locatelli et al. (2013) and Saunois et al. (2016)."

And we emphasized the importance of the OH spatial distributions on the top-down estimation of regional CH4 budget (L705-713):

" Our results indicate that OH spatial distributions, which are difficult to obtain from proxy observations (e.g. MCF), are equally important as the global OH burden for constraining CH_4 emissions over mid- and high-latitude regions. Constraining global annual mean OH based on proxy observations (e.g. Zhang et al., 2018; Maasakkers et al., 2019) provides a constraint on global total methane emissions, through the necessity of balancing the global budget (sum of source – sum of sinks = atmospheric growth rate). It also largely reduces uncertainties in optimized CH_4 emissions due to OH over most of the tropical regions but not over South America and overall mid-

high latitude regions. Also, the spatial and seasonal distributions of OH is found critical to properly infer temporal changes of regional and sectoral CH_4 emissions."

Back to my point, I would find this manuscript much more useful and compelling if the authors actually highlighted processes and phenomena that lead to similar methane inversion responses. From Holmes et al., ACP (2013; https://doi.org/10.5194/acp-13- 285-2013) we know some of the major processes that influence OH and Turner et al., PNAS (2018; https://doi.org/10.1073/pnas.1807532115) showed how this can with things like ENSO, do the authors see ENSO signals in the methane inversions? A recent paper from Nguyen et al., GRL (2020) tried to look at these feedbacks in a simple model. The authors should at least touch on the processes that influence OH, particularly those that could also influence methane.

Response: We acknowledge that it is important to analyze processes and phenomena that lead to similar methane inversion responses. During the time period of this manuscript (the 2000s), the CCMI model simulated OH show a consistent positive trend (Zhao et al., 2019; Nicely 2020). As stated in our response to comments 2.1, the positive OH trend is mainly due to more OH production by NO than loss by CO over the East and Southeast Asia and positive trend of water vapor over the tropical regions (Zhao et al., 2019; Nicely et al., 2020). We have analyzed the impact of positive [OH] trend during the 2000s on top-down estimates of CH_4 emissions in section 3.2 and section 3.3. The ENSO signal during the early 2000s is very weak (with small year-to-year variations of [OH]) and the time period of this paper very short. Therefore, analyzing the impact of ENSO seems beyond the scope of this paper. However, please note that we have another manuscript submitted to ACP using CCMI models that analyzes the impact of trend and interannual variability of OH on the CH_4 budget on the decadal time scale (1980-2010) with a focus on the ENSO (https://doi.org/10.5194/acp-2020-308).

Yet, we discussed shortly the impact of ENSO in Section 4 (L692-L701):

"The trend and interannual variations of tropospheric OH burden are determined by both precursor emissions from anthropogenic and natural sources and climate change (Holmes et al., 2013; Murray et al., 2014). Based on satellite observations, Gauber et al. (2017) estimated that ~20% decrease in atmospheric CO concentrations during 2002-2013 led to an ~8% increase in atmospheric [OH]. The El Niño-Southern Oscillation (ENSO) has been proven to impact the tropospheric OH burden and CH_4 lifetime mainly through changes in biomass burning from CO (Nicely et al., 2020; Nguyen et al, 2020) and in NO emission from lightning (Murrary et al., 2013; Turner et al., 2018). The ENSO signal is weak during the early 2000s, resulting in small interannual variations of tropospheric OH burden (Zhao et al., 2019). The mechanisms of OH variations related to ENSO and their impacts on the CH_4 budget need to be explored by inversions, but over a longer time period than this study (e.g. 1980-2010, Zhao et al., 2020)."

2.2 Oversight of previous work and faith in the CCMI models

The authors seem to have quite a bit of faith in the CCMI models, more than this reviewer finds to be justified. There are quite a few known shortcomings of the models. For example, the models don't even get the ratio of the N/S gradient in OH correct. Yet the authors are quick to criticize MCF-constrained [OH] fields with seemingly no validation of their own OH fields (e.g., Lines 595 600). Is their analysis consistent with MCF? The authors seem to be arguing that these model derived forward simulations of OH are more reliable than reconstructions. The strongest claims made in this paper seem to be those that are critical of previous work estimating OH (e.g., Rigby and Turner). For example, Lines 595-600, the abstract is dismissive of box modeling results: 'previous research mostly relied on box modeling inversions with a very simplified atmospheric transport". The latter line in the abstract isn't even correct as there has been quite a bit of non-box model work the authors seem to discount or miss: McNorton et al., ACP (2016; https://doi.org/10.5194/acp-16- 7943-2016), Gaubert et al., GRL (2017;

https://doi.org/10.1002/2017GL074987), Rigby et al., PNAS (2017; https://doi.org/10.1073/pnas.1616426114), Turner et al., PNAS (2017; https://doi.org/10.1073/pnas.1616020114), McNorton et al., ACP (2018; https://doi.org/10.5194/acp-18-18149-2018), Maasakkers et al., ACP (2019; https://doi.org/10.5194/acp-19-7859-2019), Naus et al., ACP (2018; https://doi.org/10.5194/acp 19-407-2019), Nguyen et al., (2020; https://doi.org/10.1029/2019GL085706), and He et al., ACP (2020; https://doi.org/10.5194/acp-20-805-2020). About half of these papers use 3-D atmospheric transport models and some even include fully-coupled chemistry (e.g., He et al., 2020), which is more comprehensive than the models used by the authors. The authors should do a more complete reading of the literature as they don't cite Holmes et al., ACP (2013), Murray et al., ACP (2014), or any of Michael Prather's papers.

Response:

About CCMI models and proxy methods.

To date, we have mostly two approaches to estimate regional and global [OH], one using direct atmospheric chemistry modeling and one using proxy tracers, the main one being MCF. We do not have a specific faith in CCMI models (neither we deny the interest of MCF) but here, we choose to investigate the first approach using chemistry models, taking benefit of an important collaborative effort of this scientific community (CCMI experiments) to compare and, in fine, improve their models. Each method has its caveats and, following the comment of the reviewer, we try to balance more things between the two approaches in the updated version of the manuscript.

We added in the introduction(L104-106): "However, the OH fields simulated by atmospheric chemistry models show some uncertainties in both global burden and spatial-temporal variations (Naik et al., 2013; Zhao et al., 2019)"

For the N/S ratio>1 simulated by CCMI models, we reported their ranges, with a clarification in the text (L176-179):

" The inter-hemispheric OH ratios range from 1.0 to 1.5, larger than ones derived from MCF inversions (e.g. Bousquet et al., 2005; Patra et al., 2014), partly explained by the underestimation of CO in the northern hemisphere by atmospheric chemistry models (Naik et al., 2013)."

For lines 595-600, we just wanted to mention that the OH trend simulated by CCMI models (positive) is different than that from MCF inversions, which mainly show a decrease of [OH] after the early 2000s. It is hard to say which one is correct since both of the methods have their caveats regarding trends. For the increasing trend simulated by CCMI models, we have discussed the impact on CH_4 budget in the manuscript by writing with caution:" if the CCMI models represent the OH trend properly, a higher increasing trend of CH_4 emissions is needed to match the CH4 observations (compared to the CH_4 emission trend derived using constant OH).". We do not argue that CCMI models simulate a more realistic OH trend than two-box model inversions and/or proxy-based methods.

For the abstract, we removed "previous research mostly relied on box modeling inversions with a very simplified atmospheric transport".

About missing literature

For previous studies that quantify the impact of OH variations on the top-down estimate of CH4 by 3D models, we acknowledge the missing references and thank the reviewer to have provided them. We added in the introduction (L105-L112):

"The role of OH variations on the top-down estimates of CH_4 emissions has been evaluated using

two box-model inversions with surface observations (e.g. Rigby et al., 2017; Turner et al., 2017, Naus et al., 2019) and 3D models that optimize CH_4 emissions together with [OH] by assimilating surface observations (Bousquet et al., 2006) or satellite data (Cressot et al., 2014, McNorton et al., 2018; Zhang et al., 2018; Maasakkers et al., 2019). The proxy-based constraints usually optimize [OH] on a global or latitudinal scale, the impact of OH vertical and horizontal distributions being less quantified to date. Also, proxy methods do not allow to access underlying processes as direct chemistry modeling (Zhao et al., 2019). "

He et al. (2020) estimated the global CH_4 budget by forward-model simulations and mass balance method and estimated that a 1 % change in OH levels could lead to an annual mean difference of ~ 4 Tg yr^{-1} in the optimized emissions, consistent with our top-down estimates. We cited in Section 3.1.1 (L334-336):

"Where a 1×10^5 molec cm^{-3} (1%) increase in $[OH]_{GM\text{-}CH4}$ will increase the top-down estimated CH_4 emissions ($EMIS_{CH4}$) by 40.4 Tg yr^{-1}, consistent with that given by He et al. (2020) using full-chemistry modeling and a mass balance approach."

Holmes et al. ACP (2013), Murray et al. ACP (2014), Gaubert et al. (2017), Nguyen et al. (2020) analyzed the factors controlling OH variability, we cited them in the conclusions and discussion (L692-L698):

"The trend and interannual variations of tropospheric OH burden are determined by both precursor emissions from anthropogenic and natural sources and climate change (Murray et al., 2014; Holmes et al., 2013). Based on satellite observations, Gauber et al. (2017) estimated that ~20% decrease in atmospheric CO concentrations during 2002-2013 led to an ~8% increase in atmospheric [OH]. The El Niño-Southern Oscillation (ENSO) has been proven to impact the tropospheric OH burden and CH_4 lifetime mainly through changes in biomass burning from CO

(Nicely et al., 2020; Nguyen et al, 2020) and in NO emission from lightning (Murrary et al., 2013; Turner et al., 2018).”

We also cited Prather et al. (2012) and Prather et al. (2017):

L85: “resulting in a chemical lifetime of ~9 years for tropospheric CH_4 (Prather et al., 2012; Naik et al., 2013).”

L86-L88: “However, accurate estimation of [OH] magnitude, distributions, and year-to-year variations needed for CH_4 emission optimizations are still pending (Prather et al., 2017; Turner et al., 2019).”

And in the conclusions and discussion (L729-L731):

“In addition, as suggested by Prather et al. (2017), the OH inversion would benefit from including in their prior data the responses of [OH] to variations of the precursor emissions (e.g. biomass burning and lighting) using the uncertainties estimated by 3D models.”

We added the following references:

McNorton, J., Wilson, C., Gloor, M., Parker, R. J., Boesch, H., Feng, W., Hossaini, R., and Chipperfield, M. P.: Attribution of recent increases in atmospheric methane through 3-D inverse modelling, Atmos. Chem. Phys., 18, 18149-18168, 10.5194/acp-18-18149-2018, 2018.

Holmes, C. D., Prather, M. J., Søvde, O. A., and Myhre, G.: Future methane, hydroxyl, and their uncertainties: key climate and emission parameters for future predictions, Atmospheric Chemistry and Physics, 13, 285-302, 10.5194/acp-13-285-2013, 2013.

Gaubert, B., Worden, H. M., Arellano, A. F. J., Emmons, L. K., Tilmes, S., Barré, J., Martinez Alonso, S., Vitt, F., Anderson, J. L., Alkemade, F., Houweling, S., and Edwards, D. P.: Chemical

Feedback From Decreasing Carbon Monoxide Emissions, Geophysical Research Letters, 44, 9985-9995, 10.1002/2017gl074987, 2017.

Nguyen, N. H., Turner, A. J., Yin, Y., Prather, M. J., and Frankenberg, C.: Effects of Chemical Feedbacks on Decadal Methane Emissions Estimates, Geophysical Research Letters, 47, e2019GL085706, 10.1029/2019gl085706, 2020.

He, J., Naik, V., Horowitz, L. W., Dlugokencky, E., and Thoning, K.: Investigation of the global methane budget over 1980–2017 using GFDL-AM4.1, Atmos. Chem. Phys., 20, 805-827, 10.5194/acp-20-805-2020, 2020.

Murray, L. T., Logan, J. A., and Jacob, D. J.: Interannual variability in tropical tropospheric ozone and OH: The role of lightning, Journal of Geophysical Research: Atmospheres, 118, 11,468-411,480, 10.1002/jgrd.50857, 2013.

Murray, L. T., Mickley, L. J., Kaplan, J. O., Sofen, E. D., Pfeiffer, M., and Alexander, B.: Factors controlling variability in the oxidative capacity of the troposphere since the Last Glacial Maximum, Atmospheric Chemistry and Physics, 14, 3589-3622, 10.5194/acp-14-3589-2014, 2014.

Prather, M. J., Holmes, C. D., and Hsu, J.: Reactive greenhouse gas scenarios: Systematic exploration of uncertainties and the role of atmospheric chemistry, Geophysical Research Letters, 39, L09803, doi:10.1029/2012GL051440, 2012.

Prather, M. J., and Holmes, C. D.: Overexplaining or underexplaining methane's role in climate change, Proceedings of the National Academy of Sciences, 114, 5324-5326, 10.1073/pnas.1704884114, 2017.

Nicely, J. M., Duncan, B. N., Hanisco, T. F., Wolfe, G. M., Salawitch, R. J., Deushi, M., Haslerud, A. S., Jöckel, P., Josse, B., Kinnison, D. E., Klekociuk, A., Manyin, M. E., Marécal, V., Morgenstern, O., Murray, L. T., Myhre, G., Oman, L. D., Pitari, G., Pozzer, A., Quaglia, I., Revell, L. E., Rozanov, E., Stenke, A., Stone, K., Strahan, S., Tilmes, S., Tost, H., Westervelt, D. M., and Zeng, G.: A machine learning examination of hydroxyl radical differences among model simulations for CCMI-

1, Atmos. Chem. Phys., 20, 1341-1361, 10.5194/acp-20-1341-2020, 2020.

Zhao, Y., Saunois, M., Bousquet, P., Lin, X., Berchet, A., Hegglin, M. I., Canadell, J. G., Jackson, R. B., Deushi, M., J öckel, P., Kinnison, D., Kirner, O., Strode, S., Tilmes, S., Dlugokencky, E. J., and Zheng, B.: On the role of trend and variability of hydroxyl radical (OH) in the global methane budget, Atmos. Chem. Phys. Discuss., 2020, 1-28, 10.5194/acp-2020-308, 2020.

Other references were already cited in the previous version of the paper.

2.3 Very difficult to follow The paper is filled with jargon and abbreviations. For example, nearly half of the text in Lines 440-452 are acronyms or parenthetical expressions interjecting things. This was very hard to follow as a reader.

Response: the jargon and abbreviations are mainly used in the 3.2, we reduced the jargon and abbreviations used in this section and organize sentences in the text.