Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-1208-RC1, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influences of hydroxyl radicals (OH) on top-down estimates of the global and regional methane budgets" *by* Yuanhong Zhao et al.

Anonymous Referee #1

Received and published: 16 March 2020

Zhao et al. assess systematically how uncertainties of OH concentrations affects our inference of the global and regional methane emissions and their decadal changes from the existing surface measurement network. The authors performed a series of inversion experiments using varied OH fields and used the standard deviations of an inversion ensemble to represent the uncertainty due to OH fields. The work is very important, as the uncertainty source of prescribed OH fields have not been quantitatively assessed in previous syntheses (e.g., Saunois et al. 2017, 2019). However, the manuscript can be improved with better presentation and in-depth discussion. I'd recommend the publication of this manuscript if the following issues are addressed.

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Discussion paper

1. The manuscript lacks quantitative comparisons of the results with other uncertainty sources (as assessed in literature) of methane emission estimations. The comparison could provide readers both the context and the insight. For example, I am looking for answers to the following questions: how large is the uncertainty due to OH compared to other uncertainty sources (e.g., transport)? Is the uncertainty due to OH the bottleneck for understanding the global and regional methane budget? In which regions, the uncertainty due to OH dominates; and in which regions, they are not that important? Is it adequate to reduce the uncertainty of global mean OH for the purpose of improving estimates for global and regional methane emissions? Or reducing uncertainty in OH spatial distribution is equally important? These questions are interesting to readers and can be addressed by putting the results of this paper in the context of literature (such as Saunois et al. 2017 from the authors' group).

2. The regional results are specific to the observing system (i.e., NOAA surface network). Surface observations are relatively dense in North America and West Europe, but very sparse near South America, Tropical Africa, and Tropical Asia. Therefore, the inversion tends to adjust emissions from regions less constrained by observations, if any global mismatch exists, leading to large spread of estimates in these regions. Inclusion of more observations may lead to different spatial patterns in Fig. 3. It is important to acknowledge that the conclusion about regional emissions applies to only this specific observing system. The authors mentioned site locations when explaining the difference between Inv1 and Inv2; however results from other experiments may also be explained by this factor, at least partly. In addition, OH concentrations are highest over tropics, therefore, it is expected that the difference in OH from varied fields is largest over tropics. This could explain the larger posterior flux range in tropics for Inv1 (Fig. 3).

Other comments

38-40: The sentence reads awkward. Physically, increases in OH burden cannot contribute to increases in emissions. Clarify or rephrase to avoid any confusion.

53-54: The word “additional” is confusing here.

71-72: Unclear what “catalytic chemistry” in this sentence is referred to. Also, the statement “a small perturbation of OH can result in significant change in atmospheric CH4” is inaccurate or ambiguous. The author may want to say “. . . significant change in the budget (or budget imbalance) of atmospheric CH4”.

72-75: There are other OH sources such as O3+HO2, H2O2 photolysis, and OVOCs photolysis that become important depending on the chemical environment, for example, see Lelieveld et al. (2016).

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

78: ÂăDirect measurement of OH is challenging but possible. But estimates of global mean from sparse direct measurements is nearly impossible because the large variation of OH as a result of its short lifetime.

101-103: Optimizations of CH4 emissions together with OH concentrations have been done using 3-D model inversions (e.g., Cressot et al., 2014, Zhang et al., 2018 and Maasakkers et al., 2019), in addition to two-box model analysis. These studies all used satellite data though.

Cressot, C. et al. On the consistency between global and regional methane emissions inferred from SCIAMACHY, TANSO-FTS, IASI and surface measurements, Atmos. Chem. Phys., 14, 577–592, https://doi.org/10.5194/acp-14-577-2014, 2014.

Zhang, Y. et al. Monitoring global tropospheric OH concentrations using satellite observations of atmospheric methane, Atmos. Chem. Phys., 18, 15959–15973, https://doi.org/10.5194/acp-18-15959-2018, 2018.

Maasakkers et al. Global distribution of methane emissions, emission trends, and OH concentrations and trends inferred from an inversion of GOSAT satellite data for

2010–2015, Atmos. Chem. Phys., 19, 7859–7881, https://doi.org/10.5194/acp-19-7859-2019, 2019

Section 2.1: from the description, it is not clear if these OH fields vary from month to month, or are annual mean 3-D fields.

Line 155: What temperature field do you use to compute [OH]GM-CH4 for different models? And how “troposphere” is defined in this calculation? Line 158: Is latitudinal distribution of OH also a factor (and maybe even more important factor) that results in [OH]GM-CH4 > [OH]GM-M ?

Eq. 1. The (x-xb) term is repeated twice.

Line 172: Since only emissions are optimized in the inversion, it’s a bit misleading to say H(x) represents sensitivity to sinks.

Line 205: What about Cl?

Line 231-232: “To separate the influence of OH spatial distributions from that of global mean [OH]. . .”. As commented above, it is unclear whether the OH fields vary monthly or annual mean. If the former, then in addition to influence of spatial distribution, the influence of seasonal variation is also embedded. If the latter, then the study design has a major flaw because the latitudinal distribution of OH has a pronounced seasonal cycle.

Line 239-240: Please denote inv3 and inv4 explicitly after 2007-2009 and 2000-2002 to make it easier to follow.

Line 241-243: I don’t think Inv4-Inv2 represents the impact of OH spatial distribution.

Line 250: Which one has the largest trend, which may be more relevant in this setting?

Line 269-273: This sentence does not flow smoothly within the context (results from Inv1). Remove it or move it somewhere else.

Line 278: Not clear to me how this helps decreasing discrepancies with bottom-up estimates? Fig. 2 does not show the discrepancies are reduced to me. Please clarify. Also please provide the values (and ranges) of bottom-up estimates in the text for a clear comparison.

Line 284-290: Is it possible that the difference is due to the fact that [OH]GM-CH4 is used here instead of [OH]GM-M (which I assume was used in these studies)? It does not convince me that the difference is due to the inter-hemispheric transport and stratospheric loss in 3-D model vs. 2-box model. Choices of hemispheric mean reaction rate of OH+CH4 can also introduce biases in 2-box model.

Line 316: Does the seasonality of OH fields also play a role here?

Line 338: Please explicitly state which uncertainty sources Saunois et al. (2016) considered. The comparison may be misleading otherwise.

Line 343-345: Likely because these regions have high prior emissions, but are not well constrained by surface measurements. So, it should be stated that these regional features are not intrinsic of the atmosphere, but specific to the observing system of interest.

Line 355: what is the “total differences”? How large are they?

Line 364-367: I don’t understand the logic here. I think it is probably related to OH concentration being much higher in tropics than extra-tropics.Âă

Line 379: The range of global total CH4 emissions by Inv2 (551+-2 Tg a-1) should be reported and discussed in 3.3.1, in comparison with Inv1.

Line 418: Be clearer what “global scale increase” in this sentence is referred to. It is ambiguous in the current form.

Line 485-489: The assessment of the uncertainty due to OH fields relative to other uncertainty sources are too qualitative throughout the manuscript. More insight can

be gained by quantitatively comparing to uncertainty estimates in literature such as Saunois et al.

Table 2 Quite confusing. Why global and hemispheric emissions are only shown for Inv1, but the inter-hemispheric differences are shown for both Inv1 and Inv2? Also, unit should be denoted in the caption.

Table 5 With fixed OH field, you still expect an increasing OH sink (and therefore increasing emissions) because of increasing CH4 concentration and temperature. This should be clarified somewhere in the text.

Fig. 2 The R2=0.99 line in the right panel: it should be acknowledged that other sinks of methane (such as soil absorption, Cl, and stratospheric loss) are not optimized and are specified with the same field in these inversions. Uncertainty in these sinks, if considered, will certainly create some spread in the data.

Fig. 3 To interpret this figure, the author should consider the uneven sampling of the surface network. The ranges of inferred regional emissions are large where observations are sparse, because it "costs" the least for the inversion to adjust in these regions. The inference for regional emissions is specific to the particular observing system. Having more surface stations in the southern hemisphere, or including satellite observations, would change the spatial pattern shown in this figure.

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