Reply to RC1: ' Review of "Influences of hydroxyl radicals (OH) on top-down estimates of the global and regional methane budgets" '

Comment: 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.

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

Comments: 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:
1 how large is the uncertainty due to OH compared to other uncertainty sources (e.g., transport)?

Response:

For the uncertainties in global total CH_4 emissions lead by OH, we added the values in section 3.1.1 (L316-326):

" The minimum-maximum range of the CH_4 emissions estimated by the 10 OH fields is almost similar to the range estimated by previous bottom-up studies (542-852Tg yr^{-1} given by Kirschke et al., 2013 and 583-861Tg yr^{-1} given by Saunois et al, 2016) from GCP syntheses and much larger than that reported by an ensemble of top-down studies for 2000-2009 in Kirschke et al. (2013) (526-569Tg yr^{-1}), Saunois et al. (2016) (535-566Tg yr^{-1}) or the recent Saunois et al. (2019) (522-559 Tg yr^{-1}). (Table 2 and Fig. 2). In the three top-down model ensembles, most of the inversion systems use TransCom OH fields, and the reported differences are mainly from different model transport and set-up of the inversion systems (e.g. the observations used in the inversions). Excluding the two

outliers (MOCAGE and SOCOL-3) in Inv1, we find an uncertainty of about 17% in global methane emissions (518 to 611Tg yr^{-1}) due to OH global burden and distributions, while transport model errors lead to only 5% of the uncertainty of the global methane budget (Table 3, Locatelli et al. (2013)). "

For the regional emissions, we now better compare the uncertainties lead by OH with that lead by model transport errors and set-up of the inversion systems given in Saunois et al. (2016) and of Locatelli et al. (2015). We have inserted a new Table 3 summarizes the results.

Table 3. Global, latitudinal, and regional CH_4 emission in Tg yr^{-1} (mean±SD and the [min-max] range of the inversions) calculated by Inv1 and Inv2 during the early 2000s (2000/07/01-2002/06/01) in Tg yr^{-1} (excluding MOCAGE and SOCOL-3). The uncertainties (Unc. = (max－min)/multi-inversions mean) lead by using different OH fields are compared with the uncertainties in CH_4 emissions given by Saunois et al. (2016）and Locatelli et al. (2013).

Study	This study (Impact of OH)				Saunois et al. (2016）	Locatelli et al. (2013)
Period	2000/07/01-2002/06/01				2000-2009	2005
Experiment	Inv1 (Original OH)		Inv2 (Scaled OH)		TD ensemble	Transport model errors
Region	Mean±SD[range[1]]	Unc.	Mean±SD [range]	Unc.	Unc.	Unc.
global	567±34[518-611]	17%	551±2[548-555]	1%	6%	5%
60°-90°N	29±1[27-30]	12%	29±1[27-30]	12%	50%	10%(NH)
30°N-60°N	174±8[158-183]	14%	172±6[159-178]	11%	20%	
0°-30°N	199±14[178-217]	20%	192±1[191-194]	1%	13% (<30°N)	
0°-30°S	147±14[121-167]	30%	140±6[133-153]	14%		24%(SH)
30°S-90°S	19±1[17-20]	18%	18±1[18-19]	9%		
America	45±2[42-48]	11%	45±1 [42-46]	8%	25%	37% (North America)
Canada	27±1[24-28]	17%	27±1 [24-28]	13%	70%	
Europe	27±1 [25-28]	12%	27±1 [25-28]	11%	43%	23%
Russia	33±1 [30-35]	13%	33±1 [30-34]	12%	31%	38%
China	42±5 [33-50]	39%	40±3 [35-43]	20%	11%	25% (Asia)
Southeast Asia	38±3 [34-41]	20%	37±0.3 [36-37]	3%	42%	
South Asia	59±6 [51-66]	24%	57±0.8 [56-58]	4%	44%	
Northern South America	73±9[58-85]	37%	69±4 [65-77]	17%	44%	48% (South America)
Southern South America	33±4[27-39]	37%	31±2[29-36]	20%	94%	
Africa	76±4 [68-82]	18%	74±1 [73-77]	6%	42%-45%	30%

And we added in section 3.2.2 (L387-391 and L400-405):

"The uncertainties in global OH burden and distributions lead to larger uncertainty (maximum－minimum) in top-down estimated CH_4 emissions over the tropics (>20% of multi-inversion mean) and smaller uncertainty over the northern mid-latitude regions (14%) compare with that lead by transport model errors and different observations given by Saunois et al. (2016) (13% over tropics and 20% over northern mid-latitude regions) (Table 3)."

" As shown in Table 3, at regional scales, the uncertainty (maximum－minimum) in top-down estimated CH_4 emissions due to different OH global burden and distributions over Asia and South America (~37% of multi-inversion mean) are of the same order than those lead by transport errors (25% and 48%) or given by Saunois et al. (2016) (~40%). Over other regions, using different OH fields lead to smaller uncertainties (11%-18%) compared to other causes of errors (23%-70%) (Table 3)."

For emissions changes during the 2000s, we added in section 3.2.2 (L555-566):

" We now compare the uncertainty of top-down estimated CH_4 emission changes from the early to the late 2000s due to different OH spatial-temporal variations with that ensemble of top-down studies given by Saunois et al. (2017). For the sectoral emissions, the emission changes from agriculture and waste and from wetland show the largest uncertainties (more than 50% of multi-inversions mean, Inv3－Inv2 in Table 6) induced by OH spatial-temporal variations, comparable to that given by Saunois et al. (2017). On the contrary, the uncertainty of fossil fuel emission changes (24% of multi-inversions mean) is much smaller than that given by Saunois et al. (2017). For regional CH_4 emission changes, the uncertainty induced by OH spatial-temporal variations is usually larger than the multi-inversion mean emission changes (except South Asia) and similar to that given by Saunois et al. (2017). The large differences existing in different top-down estimated regional and sectoral emission changes are mainly attributed to model transport errors in Saunois et al. (2017). Here, our results show that uncertainties due to OH spatio-temporal variations can lead to similar biases in top-down estimated CH_4 emission changes."

2 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?

Response:

For the global CH_4 budget, in the conclusions and discussion, we have demonstrated (L640-L645): " Based on the ensemble of 10 original OH fields ($[OH]_{GM\text{-}CH4}$:10.3-16.3 ×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). The top-down estimated global total CH_4 emission varies linearly with $[OH]_{GM\text{-}CH4}$, which indicates that at the global scale, a small uncertainty of 1 ×10^5 molec cm^{-3} (10%) $[OH]_{GM\text{-}CH4}$ can result in 40.4Tg yr^{-1} uncertainties in optimized CH_4 emissions."

For the regional CH_4 budget, we added in "Conclusions and discussion" (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). "

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?

Response: We added in Section 4 "Conclusions and discussion" (L705-L713):

" 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."

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).

Comments: 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.

Response: We added in the 3.1.2 (L407-415):

" The uncertainties in the top-down estimated regional emissions are not only due to inter-model differences of the regional OH fields but also rely on the distribution of the surface observations used in the inversions. Over the regions with large prior emissions but less constrained by observations (e.g. South America, South Asia, and China), our OH analysis leads to larger uncertainties than regions that are well constrained by observations (e.g. the North America and Canada) (Fig. S3). The results may indicate that on the regional scale, the top-down estimated CH_4 emissions and the uncertainties lead by OH are specific to the observation system retained. If more surface observations (e.g. in the southern hemisphere) or satellite columns with a more even global coverage were included in our inversions, spatial patterns of the top-down estimated CH_4 emissions and their uncertainties (as shown by Fig.3) could be different."

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.

Response: We changed the sentence to (L37-L38):

"From the early to the late 2000s, the optimized CH_4 emissions increased by 21.9±5.7Tg yr^{-1} (16.6-30.0Tg yr^{-1}), of which ~25% (on average) offsets the 0.7% (on average) increase in OH burden "

Comments: 53-54: The word "additional" is confusing here.

Response: We removed "additional"

Comments: 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".

Response: We rephrased the sentence as suggested:" A small perturbation of OH can result in significant changes in the budget of atmospheric CH_4 (Turner et al., 2019)."

Comments: 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).

Response: We changed in the text (L70-L75):

" At the global scale, tropospheric OH is mainly produced by the reaction of excited oxygen atoms ($O(^1D)$) with water vapor (primary production) but also by the reaction of nitrogen oxide (NO) and ozone (O_3) with hydroperoxyl radicals (HO_2) and organic peroxy radicals (RO_2) (secondary production). At regional scales, photolysis of hydrogen peroxide and oxidized VOC photolysis can be important depending on the chemical environment (Lelieveld et al. 2016)."

And we added in the reference list:

Lelieveld, J., Gromov, S., Pozzer, A., and Taraborrelli, D.: Global tropospheric hydroxyl distribution, budget and reactivity, Atmospheric Chemistry and Physics, 16, 12477-12493, 10.5194/acp-16-12477-2016, 2016.

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

Response: We rephrased the sentence to (L77-L89) "Tropospheric OH has a very short lifetime of a few seconds (Logan et al., 1981; Lelieveld et al., 2004), hindering estimates of global OH concentrations ([OH]) through direct measurements and limiting our ability to estimate the global CH_4 sink."

Comments: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.

Response: We added in the text (L105-L111):

"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). "

Comments: 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 ?

Response: We clarified in the text (L167-L169):

"The tropopause height is assumed at 200hPa following Naik et al. (2013) and the 3D temperature field used to compute $[OH]_{GM\text{-}CH4}$ is from ERA Interim re-analysis meteorology data (Dee et al, 2011). "

As we can see in Table 1, if MOCAGE and SOCOL3 OH fields are excluded, differences between $[OH]_{GM\text{-}M}$ and $[OH]_{GM\text{-}CH4}$ are largely reduced. We clarified in the text (L175-L177):

" This is mainly because MOCAGE and SOCOL3 OH fields show much higher [OH] near the surface than in the upper troposphere (Zhao et al., 2019).", and we removed: " as some of the OH fields show distinct vertical distributions".

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

Response: Thank you very much for pointing out this, we removed the (x-xb).

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

Response: We removed the "sinks" as suggested.

Comments: Line 205: What about Cl?

Response: We added in the text (L225-226) "The CH_4 sink by reaction with chlorine is not considered in our LMDz model simulations."

Comments: 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.

Response: OH fields vary monthly in our inversions, the seasonal variations of OH fields can impact inversion results. Thank you for mentioning the role of the OH seasonal cycle, which is not detailed in our analysis. We clarified in the text (in section 2.2 - L186):

" We conduct an ensemble of variational inversions … but different prescribed <u>monthly mean</u> OH fields as described in Sect. 2.1."

In section 2.3, to emphasize the impact of OH seasonal variation, although not analyzed separately in this work. we added:

L254-255: "To separate the influence of OH spatial distributions (including their seasonal variations) from that of the global annual mean [OH]."

L257-258: "As such, Inv2 provides the uncertainty range of CH_4 emissions induced by OH spatial distribution in both horizontal and vertical directions as well as seasonal variations…"

Comments: Line 239-240: Please denote inv3 and inv4 explicitly after 2007-2009 and 2000-2002 to

make it easier to follow.

Response: This has been changed as suggested.

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

Response: Here we mean the difference in Inv4－Inv2 estimated by different OH fields represents the uncertainties lead by the different OH spatial and seasonal distributions since they are all using OH fields scaled to the same value globally for 2000-2002.

We clarified in the text (L268-272):

" Therefore, the difference Inv3－Inv2 reveal the impact of OH on CH_4 emission changes between the early and late 2000s (the yellow box with solid lines of Fig. 1), Inv3－Inv4 separates the impact of OH interannual variations, and the difference Inv4－Inv2 allows assessing the uncertainties of optimized CH_4 emission changes due to different OH spatial and seasonal distributions (the yellow boxes with dashed lines in Fig. 1). "

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

We added in the text (L279):

"…shows the largest year-to-year OH variations and a positive trend of 0.35% yr^{-1} …"

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

Response: We removed this sentence as suggested

Comments: 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.

Response: We removed "help decreasing discrepancies with bottom-up estimations", and we added the number in the text (L316-L320):

"The minimum-maximum range of the CH_4 emissions estimated by the 10 OH fields is almost similar to the range estimated by previous bottom-up studies (542-852Tg yr^{-1} given by Kirschke et al., 2013 and 583-861Tg yr^{-1} given by Saunois et al, 2016) from GCP syntheses and much larger than that reported by an ensemble of top-down studies for 2000-2009 in Kirschke et al. (2013) (526-

569Tg yr^{-1}), Saunois et al. (2016) (535-566Tg yr^{-1}) or the recent Saunois et al. (2019) (522-559 Tg yr^{-1}). (Table 2 and Fig. 2). "

Comments: 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.

Response: For the two-box model inversion, the $[OH]_{GM\text{-}CH4}$ is the same as $[OH]_{GM\text{-}M}$ since the air mass and temperature are homogeneously distributed over space. For 3D model inversion, the optimized CH_4 emissions do not show a linear relationship with $[OH]_{GM\text{-}M}$. One can see that the $[OH]_{GM\text{-}M}$ of CMAM OH field (11.3×10^5 molec cm^{-3}) is a bit lower than that EMAC-L90MA (11.5×10^5 molec cm^{-3}) and CESM1-WACCM (11.4×10^5 molec cm^{-3}), but the top-down estimated CH4 emissions using CMAM OH field (599Tg yr^{-1}) is higher than that estimated using CESM1-WACCM (578Tg yr^{-1}) and EMAC-L90MA (589Tg yr^{-1}).

For the explanation of the difference between two-box model and 3-D model inversions, we agree that the choice of hemispheric mean rate is a more important factor. We added in the text (L342-L345):

"This difference probably results from the different hemispheric mean reaction rates of $OH+CH_4$ applied in box models, but could also be due to different treatments of inter-hemispheric transport and stratospheric CH_4 loss in global 3D transport models compared to simplified box-models (Naus et al., 2019)."

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

Response: Yes, the seasonality can also contribute to the differences in Inv2. As we cannot separate the contribution from seasonal variations and spatial distribution, we emphasized this in Section 2.3 (L245-L255):

"To separate the influence of OH spatial and seasonal distributions from that of the global mean [OH]."

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

Response: We clarify in the text (L387-L391):

"The uncertainties in global OH burden and distributions lead to larger uncertainty (maximum－minimum) in top-down estimated CH_4 emissions over the tropics (>20% of multi-inversion mean) and smaller uncertainty over the northern mid-latitude regions (14%) compare with that lead by transport model errors and different observations given by Saunois et al. (2016) (13% over tropics and 20% over northern mid-latitude regions) (Table 3)."

Comments: 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.

Response: As already mentioned in the first comments, we added in the text (L407-415):

"The uncertainties in the top-down estimated regional emissions are not only due to inter-model differences of the regional OH fields but also rely on the distribution of the surface observations used in the inversions. Over the regions with large prior emissions but less constrained by observations (e.g. South America, South Asia, and China), our OH analysis leads to larger uncertainties than regions that are well constrained by observations (e.g. the North America and Canada) (Fig. S3). The results may indicate that on the regional scale, the top-down estimated CH_4 emissions and the uncertainties lead by OH are specific to the observation system retained. If more surface observations (e.g. in the southern hemisphere) or satellite columns with a more even global coverage were included in our inversions, spatial patterns of the top-down estimated CH_4 emissions and their uncertainties (as shown by Fig.3) could be different."

Comments: Line 355: what is the "total differences"? How large are they?

Response: We clarified in the text (L427-L429):

"… account for 50% of the differences due to both OH burden and spatial distributions… "

Comments: 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.

Response: When scaling all OH fields to the same total global loss, the inter-model difference of OH is reduced by 33% over northern mid and high latitudes and uncertainties in top-down estimated CH_4 emissions are reduced by only 22%. Over northern tropical regions, the inter-model difference in OH is reduced by 67% but the uncertainties in CH_4 emissions are reduced by 93%, as we show in the text. The explanations here, we think, are similar to the comments for Line 343-345, which related that OH over tropical regions is more sensitive to global OH burdens as less constrained by local/direct observations. We clarified this point in the text (L434-L440):

"Over tropical regions, CH_4 emissions are less constrained (with few to none observation sites near source regions) than in the northern extra-tropics, where several monitoring sites located at or near the regions with high CH_4 emission rates and high OH uncertainties (e.g. North America, Europe, and downwind of East Asia). Thus, CH_4 emissions over the tropical regions mainly contribute to match the global total CH_4 sinks (instead of the sinks over the tropical regions only) estimated by inversion systems. When all OH fields are scaled to the same CH_4 losses (Inv2), differences of emissions over the tropical regions are therefore largely reduced. "

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

Response: We added in section 3.1.1 (L347-L350):

" With the OH fields scaled to the same $[OH]_{GM\text{-}CH4}$ (11.1×10^5molec cm^{-3}), the Inv2 simulations (assuming a global total OH burden well constrained) estimated global CH_4 emissions of 551±2Tg yr^{-1} (Table 3), as expected by the scaling. Differences in OH spatial distributions only lead to negligible uncertainty in global total CH_4 emissions estimated by top-down inversions."

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

Response: We add in the text (L491):" the increase in global mean [OH]"

Comments: 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.

Response: We added in section 3.2.2(L555-L566):

" We now compare the uncertainty of top-down estimated CH_4 emission changes from the early to the late 2000s due to different OH spatial-temporal variations with that ensemble of top-down studies given by Saunois et al. (2017). For the sectoral emissions, the emission changes from agriculture and waste and from wetland show the largest uncertainties (more than 50% of multi-inversions mean, Inv3－Inv2 in Table 6) induced by OH spatial-temporal variations, comparable to that given by Saunois et al. (2017). On the contrary, the uncertainty of fossil fuel emission changes (24% of multi-inversions mean) is much smaller than that given by Saunois et al. (2017). For regional CH_4 emission changes, the uncertainty induced by OH spatial-temporal variations is usually larger than the multi-inversion mean emission changes (except South Asia) and similar to that given by Saunois et al. (2017). The large differences existing in different top-down estimated regional and sectoral emission changes are mainly attributed to model transport errors in Saunois et al. (2017). Here, our results show that uncertainties due to OH spatio-temporal variations can lead to similar biases in top-down estimated CH_4 emission changes."

Comments: 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.

Response: We added the global and hemispheric CH_4 emissions estimated by Inv2 to Table2 as suggested. And we included the unit.

Table 2. The global total, hemispheric CH_4 emissions, and inter-hemispheric difference of CH_4 emissions calculated by Inv1 and Inv2 during the early 2000s (2000/07/01-2002/06/01) in Tg yr^{-1}.

Unit: Tg yr^{-1}	Inv1 original OH				Inv2 scaled OH			
	Global	**0-90 °N**	**90 °S-0**	**N-S$_{Inv1}$**	**Global**	**0-90 °N**	**90 °S-0**	**N-S$_{Inv2}$**
Prior	522	384	138	246	522	384	138	246
TransCom	530	368	162	206	549	377	172	205
INCA NMHC-AER-S	518	380	138	242	553	399	154	245
INCA NMHC	552	392	160	232	552	392	160	232
CESM1-WACCM	587	420	166	254	551	400	151	249
CMAM	599	419	180	239	553	399	154	245
EMAC-L90MA	589	414	175	239	555	396	159	237
GEOSCCM	611	424	187	237	550	392	159	233
MOCAGE	716	/[a]	/	/	/	/	/	/
MRI-ESM1r1	553	396	156	240	548	396	152	244
SOCOL3	757	/	/	/	/	/	/	/

Mean±SD	601±78	401±21	166±15	236±14	551±2	393±7	158±7	236±14

[a] We do not analyze the hemispheric CH_4 emission estimated with MOCAGE and SOCOL3 OH field since inversions using the two OH fields calculate much higher CH_4 emissions than using other OH fields.

Comments: 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.

Response: We clarified in the 3.2.1 (L522-L525):

"Keeping OH fields from 2000-2002, top-down estimated CH_4 emissions increase by 16.9±1.9Tg yr^{-1} (14.3-19.3Tg yr^{-1}, Table 5) between the early 2000s (Inv2) to the late 2000s (Inv4) in response to increasing atmospheric CH_4 mixing ratios and temperature. This represents 75% of total optimized emission changes (Inv3－inv2) between the early and late 2000s (21.9±5.7Tg yr^{-1}, Table 5)."

Comments: 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.

Response: We added in the Section 3.1.1 (L334-338): " 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. Other CH_4 sinks including soil uptake and oxidation by $O^1(D)$, which are prescribed in this study, remove 66.7Tg yr^{-1} CH_4. If uncertainties in all the CH_4 sinks were also considered, the correlation between optimized CH_4 emissions and the $[OH]_{GM\text{-}CH4}$ would be reduced. "

Comments: 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.

Response: As stated in previous comments, we added in section 3.1.2 (L407-L415):

"The uncertainties in the top-down estimated regional emissions are not only due to inter-model differences of the regional OH fields but also rely on the distribution of the surface observations

used in the inversions. Over the regions with large prior emissions but less constrained by observations (e.g. South America, South Asia, and China), our OH analysis leads to larger uncertainties than regions that are well constrained by observations (e.g. the North America and Canada) (Fig. S3). The results may indicate that on the regional scale, the top-down estimated CH_4 emissions and the uncertainties lead by OH are specific to the observation system retained. If more surface observations (e.g. in the southern hemisphere) or satellite columns with a more even global coverage were included in our inversions, spatial patterns of the top-down estimated CH_4 emissions and their uncertainties (as shown by Fig.3) could be different. ”