Reply to SC1: ' 'Important evidence to importance of OH but it can have more impact' '

Comments: This study provides important evidence to the importance of improving our estimates of the tropospheric OH sink in other to accurate quantify the CH4 budget. However, I believe that there are three main aspects this study that could make this study have a much stronger impact:

Response: We thank Tonatiuh Guillermo Nuñez Ramirez for the helpful comments. Please see out itemized responses below.

Comments: 1. The study found the largest absolute OH induced differences for Inv1 over northern South America, South Asia and China and at gridcell level over South America, Central Africa, East and South Asia, and mainly for wetlands, and agriculture and waste. While, it is already explained that the distribution of sampling stations is one of the reasons for this. This and further reasons for the larger uncertainty in the Tropics were discussed in detail in Bousquet et al., 2011 (another paper from this groups which I think should be reference at that this point in the paper). Furthermore, there are not only less sensitivity to observations in the Tropics, but also larger uncertainty in the fluxes. As a consequence, the inversion fits in everything in the Tropics which is too costly to accommodate elsewhere. Unfortunately, the Tropics is also the region where most of the OH reaction occurs. Therefore, it is very difficult to make conclusions on how the estimation of Tropical fluxes is affected by the OH assumptions on a regional level. I believe the study is missing either one more scenario where the uncertainties for each source are uniform globally, e.g. 5 nmol m-2 s-1 for wetlands (if there are wetland emissions in the gridcell), and/or include the analysis of the uncertainty reduction and posterior correlations, to determine how well resolved are these regions.

Response: We discussed the impact of the distribution of the sampling stations 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."

We acknowledge the fact that more scenarios could provide additional conclusions but this would necessitate extensive additional work and the paper is already long. Here this study aims to quantify the uncertainties in the current top-down due to uncertainties in OH. Analysis of how the top-down inversion can resolve the regional emissions by testing the uncertainty reduction and posterior correlations can be a separate study in our further study. However, we thank you for the suggestion and keep the idea for future works.

Comments: 2. The main goal of using an inversion is to find the fluxes that best explain the observations. However, we do not get to see how well the observations are fitted by the inversions with the different OH fields. Therefore, we cannot evaluate which features of the different OH distributions are realistic. By knowing for example the spatial distribution of the residuals, or of the correlations between posterior mixing ratio and observations, we can evaluate if certain spatial patterns are realistic. Also the use of aircraft profiles for validation, e.g. over the Amazon (Miller et al., 2007, Beck et al., 2012, Gatti et al., 2015, Basso et al., 2016), Asia (Brenninkmeijer et al., 2007, Baker et al., 2012, Schuck et al., 2010) or across latitudinal transects (e.g. Wofsy, 2011 and Schuck et al., 2012) could provide information on the

realism of the vertical distribution. During the period of the simulation, there were two satellites sensors available SCIAMACHY and IASI with distinctly different sensitivities. SCIAMACHY is more sensitive to the surface, while IASI to the upper troposphere. Using this, it may be possible to say something about how realistic is both the horizontal and vertical distribution.

Response:

In the updated version, we evaluate the inversions using aircraft observations. Usually, one can use the surface observations to evaluate the inversions using satellite data but we do not use satellite data to evaluate the inversions using surface observations. Comparison can still be made but (i) the observations from IASI do not provide the averaging kernel, thus they cannot directly compare with model simulations, and (ii) SCIAMACHY CH4 data experience significant to large systematic errors, limiting strongly the interest of comparison.

We added in the text (L284-L296):

"We evaluate the optimized CH_4 emissions by comparing the simulated CH_4 mixing ratios using prior and posterior CH_4 emissions with independent measurements from the NOAA/ESRL Aircraft Project. The location of the observation site (Table S1) and the vertical profile of the model bias in CH_4 mixing ratios compared with the aircraft observations (model minus observations) are shown in the supplement (Fig. S4a for Inv1 and Fig. S4b for Inv2). The comparisons with independent aircraft observations confirm the improvement of model-simulated CH_4 mixing ratios when using posterior emissions. All of the inversions in Inv1 and Inv2 reach small biases when compared with aircraft observations (right panel of Fig.S4a and Fig.S4b), which means that it is hard to distinguish which OH spatial and vertical distributions are more realistic in terms of quality of fit to these aircraft CH_4 observations. For Inv1, the root mean square errors (RMSE $= \frac{\sqrt{\sum(model-observation)^2}}{n\_obs}$, n_obs is the number of observations) are reduced from up to more than 100ppbv (prior) emissions

to ~10ppbv (posterior). For Inv2, although the CH_4 mixing ratios simulated using prior emissions already match well with aircraft observations (MSE=8-17ppbv), the posterior emissions still reduce the RMSE by up to 10ppbv. ”

We added Table S1 and Figure S4a and Figure 4b in the supplement:

Table S1. Location of the NOAA ESRL aircraft sites.

STATION ID	SITE LOCATION	BOTTOM ALT(m)	TOP ALT(m)	latitude	longitude
CAR	Briggsdale, Co	1658	11879	40° 22’ N	104° 17’ W
HAA	Molokai Island, HI	305	8104	21° 14’ N	158° 57’ W
HFM	Harvard Forest, Ma	582	8063	42° 32’ N	72° 10’ W
PFA	Poker Flat, AK	131	7604	65° 04’ N	147° 17’ W

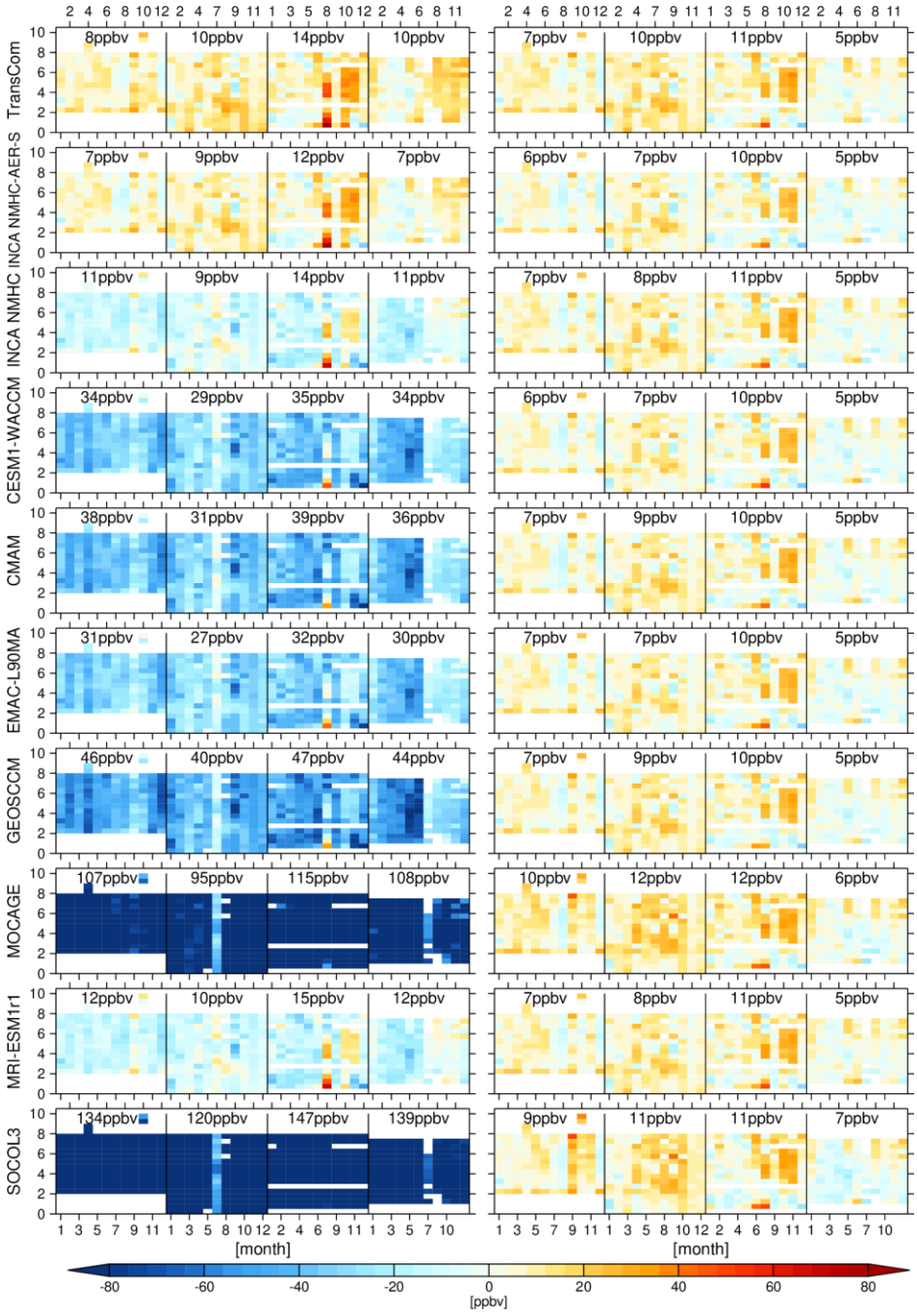

Figure S4a. The vertical profiles of the bias in LMDz simulated monthly CH_4 mixing ratios compare with measurements from the NOAA/ESRL Aircraft Project (model－observations) during 2000/7-2002/6. The left panels show the bias simulated by prior emissions with 10 original OH fields and the right panels show the bias simulated by corresponding posterior emissions from Inv1.The root mean square errors

(RMSE = $\frac{\sqrt{\sum(model-observation)^2}}{n\_obs}$) are shown inset.

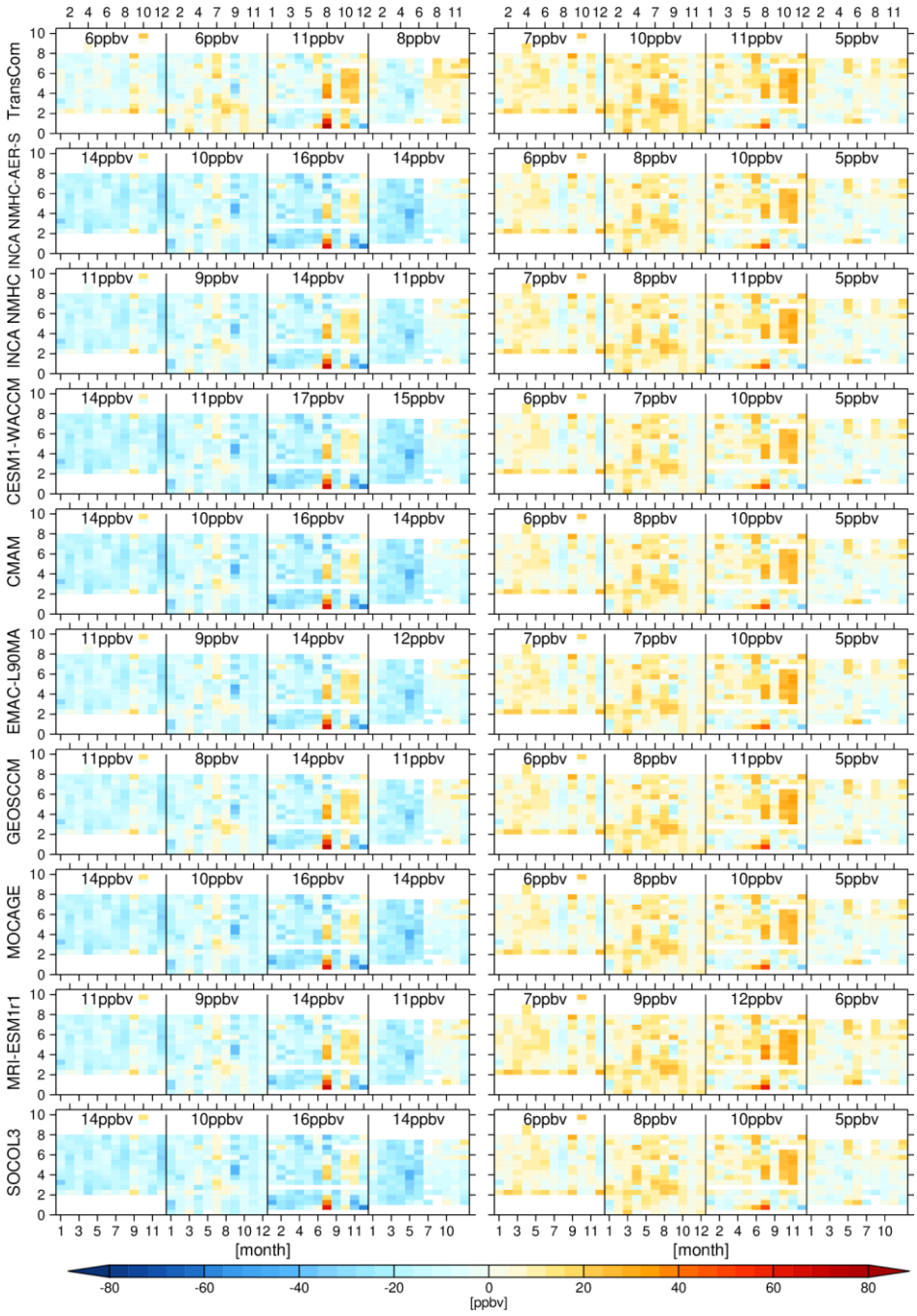

Figure S4b. The same as figure S3a but for Inv2.

Comments: 3. Link to the validation, there is little discussion on the features of the OH fields provided by the models. For example, Patra et al., (2015) determine that observations of CH3CCl3 support a N/S gradient of 1, so more should be done to explain how probable bias in the modeled OH distributions affects the CH4 estimations. Also many of the different features in the spatial distributions OH are caused by known biases in the climate chemistry models, e.g. the NMVOC levels, the CO burden, CO biases, O3 biases (e.g. Naik et al. 2013, Shindell et al., 2006). Here, it would be very interesting to see, for example, if there is a relationship between the N/S ratio of the OH distributions and the N/S ratio of the posterior fluxes (similar to figure 2). Also why are SOCOL3 and MOCAGE such outliers?

Response: We have evaluated the impact of the uncertainties in OH spatial distribution by conducting Inv2. For the relationship between the N/S ratio of the OH and N/S ratio of the posterior fluxes, we have stated in the text (L369-371):" The TransCom OH field, for which OH N/S ratio is 1.0, leads to an inter-hemispheric CH_4 emission difference of 205Tg yr^{-1}, which is 35Tg yr^{-1} (27Tg yr^{-1}) smaller than the mean (minimum) inter-hemispheric difference calculated using other OH fields with OH N/S ratio of 1.2-1.3." We don't think it will be helpful if we further estimate the correlation between the N/S ratio of OH and fluxes because, among the 8 OH fields analyzed here (exclude MOCAGE and SOCOL3), only one has N/S ratio of 1, five having N/S ratio of 1.2, and two of 1.3.

The explanation of SOCOL3 and MOCAGE simulating high [OH] can be found in Zhao et al. (2019) and we added in section 3.1.1 (L310-L314):

" 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."

Comments: We are shown inversions with and without interannual variability in the OH fields. However, due to the increase of tropospheric temperatures, even in the simulations with fixed OH or the fields distributed in the TRANSCOM-CH4 experiment, the lifetime of CH4 will decrease. This effect is not quantified in the paper unless I missed it.

Response: Indeed, temperature changes will impact the CH4 lifetime but we do not test the impact of this effect here.

Comments: As stated in the study, the transport model uncertainty is very large. This means that the distribution of CH4 is model dependent. Therefore, there could be a large uncertainty in the global OH means weighted by the CH4 reaction. I believe an airmass or volume weighted OH means should be at least provided in the supplement and that the comparison with box models or with other models should be done with air mass or volume weighted means, including the relationship in figure 2.

Response: The air mass-weighted [OH] is already given in table 1 and volume-weighted OH was given by Zhao et al. (2019).

We added in section 2.1 (L170):" The volume-weighted [OH] was given by Zhao et al. (2019)."

For the relationship in Figure 2, the air mass-weighted and volume-weighted [OH] do not show a linear relationship with optimized CH_4 emissions since some OH field shows distinct vertical variations. For example, the air mass and volume-weighted [OH] simulated by CMAM model (11.3×10^5 molec cm^{-3} and 10.4×10^5 molec cm^{-3}) is smaller than simulated by EMAC-L90MA model (11.5×10^5 molec cm^{-3} and 11.1×10^5 molec cm^{-3}), but the top-down inversions using CMAM OH field estimated larger CH4 emissions (599Tg yr^{-1}) than that using EMAC-L90MA OH fields (589Tg yr^{-1}).

Comments: On which basis did you choose only 7 of the 20 CCMI simulations?

Response: The same reasons explained in Zhao et al. (2019) as we explained it in section 2.1.

Comments: You mentioned the TRANSCOM-2011 project. However, this was actually known as the TRANSCOM-CH4 (Patra et al., 2011), since there have been several TRANSCOM projects mainly with CO2 . It might be useful to mention, that in Patra et al., (2011), the OH fields from Spivakovsky (2000).

Response: We corrected TRANSCOM-2011 to TRANSCOM-CH4.

We have already mentioned this in Sect 2.1.1 (L161-L163):

"We also include the climatological OH field used in the TransCom simulations (Patra et al., 2011), which uses the semi-empirical, observation-based OH field computed by Spivakovsky et al. (2000) in the troposphere"

Comments: Table 1 and Table 2 are missing the units

Response: We added the units, thank you very much.

Comments: Could you specify which convection parameterization is used? In Locatelli et al., (2015) three parameterizations are used.

Response: We added in the text(L226):

"The deep convection is parametrized based on the Tiedtke (1989) scheme."

We added in the reference: "Tiedtke, M.: A Comprehensive Mass Flux Scheme for Cumulus Parameterization in Large-Scale Models, Monthly Weather Review, 117, 1779-1800, 10.1175/1520-0493(1989)117<1779:acmfsf>2.0.co;2, 1989."

Comments: I think that at least in the supplements you should include the maps of the mean differences

between the scenarios, e.g. Echangeall;Echangef ixoh;Echangevaroh:

Response: Thank you very much for the suggestion. Here we mainly analyzed the regional emission changes for the large emitting regions follow Saunois et al. (2016; 2017) and Locatelli et al. (2013). The differences in emission changes on the grid-scale cannot be well resolved by the global inversions with limited surface observations so it is not analyzed here and we prefer to only provide results aggregated at the regional scale. Analysis of the impact of OH top-down emission changes on grid-scale belongs to future studies doing regional inversions using more dense observation networks (e.g. satellite).

Comments: In figures 4 and 5, would it be possible to show the a priori uncertainties as error bars? In general I find the double axes confusing and maybe a single axis with absolute emissions would be better.

Response: Thank you very much for the suggestion. The prior uncertainties are 100% of the prior emissions (see Section 2.2). So it is the same as the prior emissions which limit the interest to show them in the figure.

We agree that showing the absolute value will be easier to understand. But the aim of Fig. 4 and Fig.5 is to show the uncertainties due to OH. Showing absolute emissions (0-100Tg yr^{-1}) will make it difficult to recognize the spread among different inversions (<20Tg yr^{-1} or even <5Tg yr^{-1}), which is the purpose of this figure.