Anonymous Referee #2

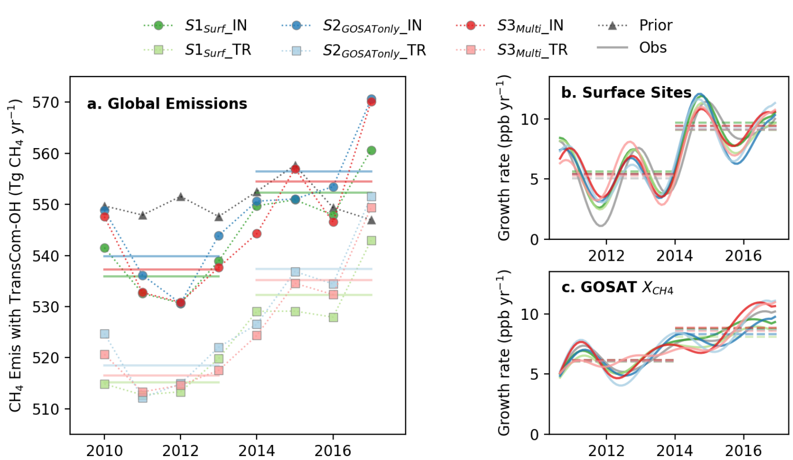
This is a very interesting study about a recent increase in the global growth rate of methane and the use of inverse modelling to disentangle the underlying causes. 6 different inversion set ups are used that lead to very consistent results, which is encouraging. The setup of those inversions addresses uncertainties in the treatment of OH, although the results seems to show very little sensitivity to it. Because of this, the added value of CO and CH2O measurements that are used remains unclear. Besides the treatment of OH, some other factors require further attention as will be explained below.

We thank the reviewer for the interest in our study and for the constructive and insightful comments that help us to improve our manuscript. Please see our point-to-point responses below.

GENERAL COMMENTS

I was surprised to see that the posterior scaling factors for OH remain so close to 1. It is mentioned that low variability is in line with some earlier studies. However, what I am more surprised about is that offsets aren't larger, given the much larger uncertainty in global OH. Looking at figure S6, I see quite a substantial difference in the prior simulation using the two OH fields. It suggests a sizeable difference in the methane lifetime between TRANSCOM and INCA-OH. Surprisingly, this difference does not lead to an OH correction in the inversion, for one are both fields. This suggests, that the updated emissions account for the difference. However, looking at Figure 2, I don't really see a systematic difference between the emissions using the two OH fields either. This must be explained.

First, we would like to clarify some confusion about Figure 2a. In the original plot, there were two y-axises, with a shift of 20 Tg CH_4 emissions per year. Posterior emissions estimated using INCA-OH (shown in circles) are associated with the left y-axis (ranging from 525-575 Tg/yr), while estimates using TransCom-OH are (shown in squares) associated with the right y-axis (ranging from 505-555 Tg/yr). The choice was made to highlight the interannual variations of the posterior CH_4 emissions using both OH fields while setting aside the systematic differences. We have updated the plot as shown below to avoid such confusion.



The validation presented in the supplement concentrates on CH4, which is fine. However, I was surprised not to see anything about CH2O and CO, and how well inversions 1 and 3 fit those data. This makes it very difficult to judge the performance of these inversion components, and how much we can expect them to influence the estimates for CH4.

We did not include evaluation data for CO and CH_2O as they were documented in previous papers using the same inverse system (e.g. Yin et al., 2015 and Zheng et al., 2019), and we wanted to keep the focus on CH_4 in this study. We agree with the reviewer that such evaluation information would be helpful for the interpretation of the CH_4 results, hence we will add evaluation results in the supplementary of the revised manuscript.

It is concluded that the largest contribution to the growth rate increase comes from East Asia and the Tropics. I wonder whether this conclusion may be influenced by the fact that these are also very large fluxes, with large uncertainties. Therefore, you expect the largest adjustments to those fluxes. Suppose the inversion wouldn't know where to put an emission correction. Then the cheapest solution is to distribute it evenly across the globe in terms of fractional deviation from the a prior uncertainty. If that were the case, I suspect that East Asia and the Tropics would stand out also. If East Asia and the Tropics are singled out as main causes explaining the increase, shouldn't that be measured in comparison to this "none-informative" reference rather than absolute emission deviations from the prior?

We agree with the reviewer's insights that the posterior results are regularized by the prior fluxes and associated uncertainties. The ensemble of the 6 inversions included in this study tested uncertainties that stem from observational constraints and OH prior fields, but not from the prior methane emissions. We have added discussion regarding this aspect in the revised manuscript in section 4.3: "We note that the posterior fluxes are regularized by the prior information with climatological fluxes (except for fire) and error statistics that are proportional to the prior fluxes. Nevertheless, our analysis of observational information content using an analytical inverse framework demonstrates that there is information in GOSAT X_{CH4} observations to isolate emissions at continental scales in both mid-latitude and tropics (Fig. 6; Fig. S8), so that the IAV of regionally aggregated fluxes could be well constrained. Furthermore, current bottom-up estimates of both fossil fuel and wetland emissions have large uncertainties (Poulter et al., 2017, Miller et al., 2019, Saunois et al., 2020), introducing uncertain IAV and trend in the prior emissions may degrade the posterior estimates. Therefore, we consider having no prior IAV or trend the best option given that we do not have high confidence in the prior information regarding this aspect. Future follow-up studies that explore uncertainties due to prior information would be very valuable."

We did not compare absolute emission deviations from the prior much in this paper (except for Fig. 4b). As suggested by the reviewer, our paper focused mostly on the temporal changes in the posterior emissions from 2010 to 2017 with each inversion, which avoids direct comparisons of the absolute emission deviations from the prior.

The supplement provides some evaluation of the inversions against surface and total column data. However, I am missing statistical information on the fits, necessary to judge if the a priori and observational uncertainties are chosen in a realistic and statistically consistent manner. This information (e.g. chi2) should be provided.

We have added statistical information on the fits to the revised manuscript as Supplementary Table 4.

SPECIFIC COMMENTS

Page 1, line 14: I do not think that the '[xx]' notation is correction for representing mixing ratios. In chemistry, the notation is used for concentrations, which is obviously something very different. In my opinion, there should be no confusion between concentration and mixing ratio.

We adopted this notation for its brevity, but we agree with the reviewer's comments. We have revised the manuscript accordingly.

Page 6, line 123: which "meteorological reanalysis"?

ERA-Interim reanalysis. Added in the revised text.

Page 6, line 133: Although I understand the rational for using a climatological prior, I nevertheless think it is a problem when investigating the magnitude of trends. Depending on the weight of the prior, the solution will underestimate the trend. As figure S11 confirms, the trend in the climatological prior is biased. Looking at Figure 2b, I get the impression that the trend in the observations is indeed underestimated by the inversion optimized fit. Surprisingly enough the climatological a prior does not affect the a posteriori estimated trend in OH, which I had expected would have accounted for at least part of the missing trend in the posterior solution.

In the original paper, we stated in section 2.2.2, "This choice is made to avoid prior assumptions about the interannual variations (IAV) or trends in the surface emissions so that IAV in the posterior fluxes are primarily driven by assimilated observations." We have further elaborated on our choice of climatological prior emissions (except for fire) as stated above in our response. As Figure S11 shows, different inventories show diverging trends in many cases, and there are various possible driving factors of the recent methane growth rate as mentioned in the introduction of the manuscript. As such, introducing uncertainty trends or IAV into the prior emission does not seem to be a better option.

For Figure 2b and c, we have added Supplementary Table 5 that summarizes the statistics of the estimated growth rates in the posterior model states against observed ones. Compared to the surface observations, the growth rates of the posterior models are not underestimated. Compared to GOSAT X_{CH4}, there are small biases of (-0.33 ppb yr-1) in the surface inversion (S1_TR) that is independent of the GOSAT data.

Table S5. Summary statistics of monthly growth rates comparison between posterior model states and collocated observations as shown in Figure 2b and c. Shaded area indicates that the compared observations are assimilated in the corresponding versions.

	Compared to Surface Obs (ppb)		Compared to GOSAT XCH4 (ppb)	
	Mean Bias	RMS	Mean bias	RMS
$S1_{Surf}\_IN$	0.44	0.96	0.03	0.45
$S1_{Surf}\_TR$	0.11	0.89	-0.33	0.65
$S2_{GOSATonly}\_IN$	0.05	1.44	-0.2	0.48
$S2_{GOSATonly}\_TR$	0.13	1.55	-0.01	0.68
$S3_{Multi}\_IN$	0.21	1.4	0.07	0.62
$S3_{Multi}\_TR$	0.01	1.48	-0.09	0.7

Both the prior emissions and OH are climatology fields. Given the configuration of the inversion and observed changes in the mixing ratio of the three species, the resultant change in OH is very small and the increases in methane mixing ratio are mostly attributed to changes in surface emissions by the inverse system.

Page 6, line 139: Which information supports the 20% uncertainty in weekly OH per latitude band? I wonder what happens if you integrated the a priori uncertainty in OH globally and per year. The number would probably become very small. Maybe that explains why global mean OH is almost not adjusted in the inversion?

We acknowledge the limitation of such error statistics based on empirical choices. This comment is also related to the previous point that the posterior results are regulated by the prior error statistics. We have added more discussion regarding the underlying caveats in section 2.2.2.

Page 8, line 202: By 'loss rate' you mean 'sink' or 'life time'? I guess 'sink' although 'loss rate' suggest rather 'life time'.

We have corrected it to "the total methane sink".

Page 9, line 206: One way to judge how well the inversion is capable to independently estimating the sources and sinks of methane is to look at the posterior correlation between global OH and the global emission. To be able to judge this, it is necessary to provide information on that correlation.

We agree with the reviewer that it is ideal to have the error covariances of posterior fluxes. However, the computational cost is very expensive with a variational inverse system to estimate the posterior error covariances using a Monte Carlo approach. We have added more discussion regarding this point referencing recent methane inverse studies that use an analytical inversion scheme to optimize anthropogenic methane emissions and their trends on a 4º×5º grid, along with monthly regional wetland emissions and annual hemispheric concentrations of tropospheric OH for individual years (Maasakkers et al., 2019; Zhang et al., 2020). The full error covariances could be computed in such an analytical setting, while the nature of the problem is similar.

Page 12, line 245: It would be good to refer to Monteil at al (2013), who were the first to report the difficult to jointly fit surface measurements and GOSAT column retrievals.

We thank the reviewer for pointing out this important reference. It has been added to the discussion.

Page 13, line 274: Looking at figure S9, I find it hard to be convinced by the argument raised here. For China, the p-value is quite high – so the significance of the positive trend is only low. For the Amazon it looks better. However, I still doubt that it is a good idea to only take the seasonal maximum. It makes the analysis sensitive to extreme events and outliers. Looking at the seasonal coverage a longer common period of data coverage could have been defined. At least some other points should be tried to confirm the robustness of these trends.

We noted in the original manuscript that there are considerable sources of uncertainty for such a gradient analysis. Many factors such as varying sampling in space and time, as well as changes in transport, could result in changes in the latitudinal gradient. Nevertheless, we find this piece of information very interesting. Following the reviewer's comment, we have further highlighted the caveats and the need for more observations.

Page 13, line 264: The description of regional emission changes is rather silent about the USA. Numerous papers have discussion the increase in fossil fuel related emissions in the past years, potentially explaining a large fraction of the observed global increase in methane. However, I do not see that back in figure 7, which would be worth mentioning.

We have added more discussion regarding the lack of trend in the US. "Relatively small increase is found after 2014 with flat emissions before, which is consistent with previous studies finding no trend over US before 2012 (Saunois et al., 2017, Bruhwiler et al., 2017)".

Page 14, line 275: The difference between OH and emissions that is mentioned here happens by design, since OH is only allowed to be changed in a zonally uniform manner. There is no reason fundamental reason why the sink couldn't change in similar patterns as the source.

We would like to clarify that only in S1, where surface CH_4 and CO observations are assimilated, the OH fields are optimized in a zonally uniform manner. In Inversions S2 and S3 that assimilate GOSAT XCH_4 observations, OH are optimized per each model grid cell. The different choice for surface inversion was made by the limited spatial coverage of surface stations. We have made this point more clear in the revised manuscript.

Page 17, line 342: Here a connection is made between d13C measurements, and a model analysis that does not account for d13C. Then, how do you know that your results are consistent with d13C? I wonder about the validity of the qualitative arguing in this paragraph. Looking at figure S12, the lags between emission anomalies and d13C responses as well as their amplitudes are difficult to connect between Figures a and b. In reality it is even much more complex due to atmospheric transport variations. Therefore, the way of arguing that it fits together is too easy in my opinion.

We agree with the reviewer on this critique and hence have added $d^{13}C$ simulations with the prior and posterior fluxes using a simple box model.

Figure 5: What are the small plusses in this figure?

They represent trends that are statistically significant at a 95% confidence level. We have added this information in the figure legend.

Figure 6: I'm assuming that this figure shows the diagonal of the averaging kernel. Please mention this somewhere explicitly.

Indeed, the diagonal of the averaging kernel is shown. We have added this information explicitly in the legend.

Figure S11: This figure only shows inventory estimated trends. I was surprised not to see the inversion results in the same figure. Since the inventory trends were not used in the a priori, it would be a great way to independently assess the consistency of the inventories and atmospheric data. The fact, that it the posterior fluxes are not included suggests that the comparing might look very good. In either case, some discussion of it is needed.

We thank the reviewer for this nice suggestion to add the inversion results for comparison. We did not add the inversion results for several reasons: first, it would make the plot very busy so that it becomes difficult to read (adding six versions of inversions); second, this paper does not emphasize the magnitudes of the posterior fluxes compared to the prior (there are systematic zonal differences as illustrated in Fig. 4b), as we would like to focus on the temporal change, adding such a plot would divert the discussion. Figure S11 shows different bottom-up inventory estimates, we have added related discussion in the revised manuscript in section 4.3.

TECHNICAL CORRECTIONS

Page 2, line 21: 'O(1D)' io 'O('D)'

Corrected.

Caption of fig 2: "Deseasoanlized"

Corrected.

Page 15, line 290: "anthropgenic"

Corrected.

Figure 6, caption: 'are shown' io 'is shown'

Corrected.