

Authors' response to anonymous referee #1

In the following, referee's comments are in *italic*, authors' responses in normal font, and references (page, line, figure, and table number) to the revised manuscript in **bold**. Please note that this paper is merged with the accompanying paper, following the referees' comments and with approval from the Topical Editor. A summary of the accompanying paper was included in the Supplementary Material of this paper.

In addition, figures and tables were revised substantially, and the following table summarizes the changes to figure and table numbers. In addition, please note that Fig. 2, Fig. 3, and Fig. 4 of the accompanying paper were moved to the Supplementary Material as Fig. S1, Fig. S2, Fig. S3.

Revised	Original	Short description
Fig. 1	Fig. 1	mTC regions + observation sites
Fig. 2	Fig. 3	Comparison with assimilated observations
Fig. 3	Fig. 4	Global mean XCH ₄ and growth rates
Fig. 4	Fig. 5, S1	Comparison with aircraft observations
Fig. 5	Fig. 6	Model performance in Europe at assimilated sites
Fig. 6	Fig. 7	Comparison with TCCON observations
Fig. 7	Fig. 8	Comparison with GOSAT observations
Fig. 8	Fig. 11, 12, S4	Emission estimates of global and Asian temperate and tropical mTC regions.
Fig. 9	Fig. 9	Growth rates of global emission estimates
Fig. S4	Fig. 2	Land-ecosystem map
Fig. S5	Fig. S2	Comparison with TCCON observations
Fig. S6	Fig. S3	Comparison with GOSAT observations
Fig. S7	Fig. S6	Monthly mean of total emission estimates at latitudinal bands
Fig. S8	Fig. 8, 10, 13	Emission estimates of land mTC regions
Fig. S9	Fig. 14, S5	Emission estimates of ocean mTC regions.
Table 1	Table 1	Inversion set-up
Table 2	Table 2	List of observation sites used in the inversions
Table 3	Table 3	List of aircraft observation sites
Table 4	Table 5	RMSE with TCCON observations
Table 5	Table 6	RMSE with GOSAT observations
Table 6	Table 4	Global and regional emission estimates

General comments

A lot of figures are relegated to the supplemental information, while I would prefer to see them in the main paper. I appreciate this can be a matter of preference, but I have made the case for their promotion in my comments below.

In most of the figures/tables showing either the model run with posterior emissions, only the posterior values are plotted. I would like to see the priors on the figures as well, in order to assess the improvement made by assimilating the data.

We appreciate the reviewer's suggestions, and the figures and tables were revised extensively following the specific comments below. We agree with the reviewer that the figures and tables became more complete by including the prior. The prior is added in the figures and tables in the revised manuscript. Please see below for responses and changes in more detail.

Specific comments

Pg. 5, first paragraph: I found it a bit confusing throughout the paper that the modified Transcom regions and the regions to be optimized were both referred to solely as "regions".

We apologize for the confusion. We agree with the reviewer that the definition of 'regions' was confusing in

many sentences. We have tried to make this clear in the revised manuscript by referring to modified TransCom regions as mTCs and adding the word 'optimization' for the optimization regions where the definition is unclear.

Pg. 5, line 16: What was the variance for the ice region?

The variance for the ice region was 1×10^{-8} . It was stated in the accompanying paper, but we agree with the reviewer that it was not clear only from this paper. In the revised manuscript, the number is explicitly added.

Text is revised: Pg. 6, line 18.

Pg. 6, line 30: I believe that many of these observations are not available for the whole 2000-2012 time period and you might consider giving the date range in Table 2. Does the assimilation system show any effects of observations turning “off” and “on”?

This is an excellent and very interesting point. We indeed acknowledge the sensitivity of the inversions to the set of observations assimilated, and the emission estimates could vary by turning “off” and “on” some of the observations. We surmise that the effect would be mostly in regional emission estimates, and less in continental scale estimates. This is because many “background” sites that constrain large-scale emissions have long-term observations, and the number of “new” background observations is low. The effect was especially prominent in regions that were already poorly constrained by the current background network: sites in South America had a significant effect on the increase in regional emission estimates (see Section 3.4.5). Although we did not test this further for all sites, we also discussed the effect for other observations (discrete vs discrete+continuous (see Supplementary Material), and GSN, AMY in Section 3.4.3), and we feel that this is sufficient for the paper at this point. Finally, we followed the reviewer's suggestion and the date range is added to Table 2.

The date range is added to Table 2.

Pg. 7, line 3: In the companion paper, the minimum mdm is 7.5 ppb. Is this a difference in the approach between the two papers? I'd also suggest you include which category of observation has this value.

Firstly, we thank the reviewer for also carefully reading the accompanying paper. As pointed out by the reviewer, the minimum mdm was different in the two papers. We found that the sites with low mdm (i.e. marine boundary layer or high southern latitude sites) were particularly important to remove the effect of initial 3D mole fraction fields and to constrain background concentrations well. In addition, these sites were important to correct a bias in the latitudinal gradient of the prior emissions. In the long-term inversion we noticed that the constraints introduced by these sites was not strong enough (not shown or published), and therefore, we gave greater weight (=lower mdm) for those sites for the long-term experiment. In the revised paper, the table and description of the accompanying paper regarding mdm was omitted. The main finding of the sensitivity tests in the Supplementary Material does not change due to differences in the mdm of those sites, and therefore, we hope the reviewer agrees that the alternative description of the mdm was not necessary to be included in the Supplementary Material.

Pg. 9, second paragraph and Figure 3: I really cannot make most of these observations out on Figure 3 because of the large range of values used. In fact, I had to stare at the posterior differences for quite a long time before I was sure the three sub-figures were different. I suggest plotting the posterior panels with a different colour scale so the differences can be made out.

We apologize for difficulties that arose by the poor illustration in the figure. This is an excellent suggestion and we revised the figure by plotting posteriors in a different scale.

Fig. 2 (original Fig. 3) is revised.

Pg. 9, third paragraph: A2007 and B2007 are used here (and throughout the paper), but I don't believe they are formally defined.

We acknowledge that the words were not descriptive enough. It was referred to only once in a much earlier

section. To make this clear, the text was revised in the new manuscript, where A2007 is simply written as “after 2007”, and B2007 as “before 2007”.

Pg. 10, third paragraph: I think you've missed a chance to draw some conclusions about the model due to the fact that the agreement with TCCON tends to be better before the averaging kernels are applied. As noted in the paper, applying the kernels does make the comparison more valid, as the effects of the prior used in the TCCON analysis and the instruments' vertical sensitivity are taken into account. The fact that this makes the agreement worse could indicate that there's some compensating effect in the total column, which I think you can see by looking at the comparisons with IMECC in Figure 5.

We agree with the reviewer that the point was missing in the text. This is a very interesting point in regard to the application of averaging kernels. As pointed out by the reviewer, the comparison with IMECC showed that model CH₄ was lower than the observations and agreement was worse at lower altitudes. The weights at lower altitudes were greater than those at higher altitudes when the averaging kernel was applied. Therefore, model XCH₄ decreased and agreement with retrieved XCH₄ became worse when the averaging kernel was applied. Although we acknowledge the effect of the averaging kernel, we decided to remove the text, as all comparisons were done with the averaging kernel applied.

Pg. 10, line 15-16: Why not show the prior?

We acknowledge the importance of also showing the improvements by the inversions for XCH₄ estimates. In the original figures, the prior was not included, because it was already clear from the comparison with the in situ observations that the prior XCH₄ would also be much higher than the observations. Since prior XCH₄ also increases significantly more than the observations and the posterior, the differences between the posterior estimates and that with the observations were less clear when prior estimates were plotted. However, in the revised figures, we included the priors.

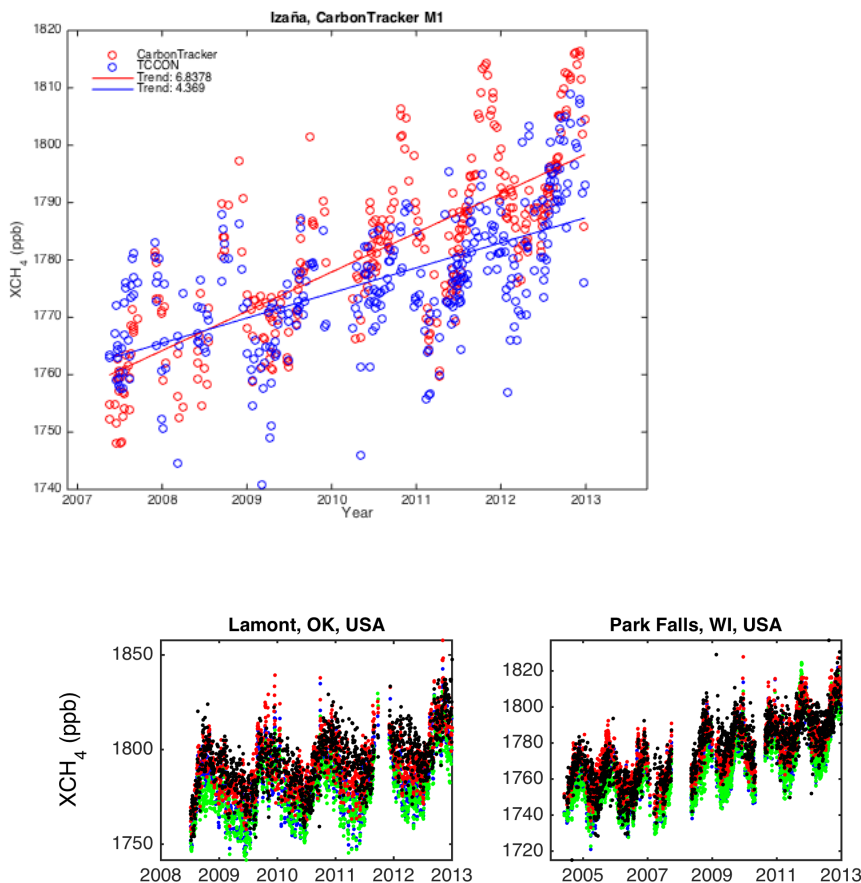
Fig 6 (original Fig. 7) is revised.

Pg. 10, line 24-25: I'm not sure I understand what you're getting at here: why would missing spring observations in one year affect the trend over the study period?

We appreciate the reviewer for noticing this inconsistency. We agree with the reviewer that the missing spring observations were not the reason for the mismatch in trends because the model trend was calculated from the same date and location of the observations, and using the averaging kernel.

We looked at the trend further, and found that the model seems to overestimate the trend (see figure below). XCH₄ before 2010 was lower and after 2010 was higher than the observations. A similar feature was found at other sites, e.g. at Park Falls and Lamont, such that the increasing trend for 2007-2013 was stronger in the model than in the observations. This could be caused by underestimation of emissions in northern temperate regions before 2010 and a significant increase after 2010. This may indicate that our inversion does not estimate the start of the emission increase in temperate regions correctly. The discussion is added in the revised manuscript.

Text is revised: Pg. 12, line 9-11.



Pg. 10, line 32: Can you not be sure that the data was not used by the inversion?

Here, we tried to address that the inversions probably did not learn much from GPA observations because *mdm* was set large (75 ppb). We apologize that the sentence was misleading. We are certain the data were used and assimilated in the inversions, but the high *mdm* limited the effect of the observations on the emissions to some extent. In theory, the effect could be checked by running inversions without the data, although such sensitivity test was not done as it would have been an expensive (CPU time) extra test. The sentence in the new manuscript is revised to address that the data were assimilated in the system.

Text is revised: Pg 12, line 23-24.

Pg. 11, line 1: I know observations are few and far between in Australia, but Cape Grim is in a much different environment than Wollongong! Wollongong is a hard site to model in general – see for example Fraser et al., 2011.

This is a good point. We agree with the reviewer that the XCH₄ cannot simply be compared because the sites are near. As pointed out by the reviewer, Cape Grim is located on an island far from cities and has little local emissions, whereas Wollongong is located in the city of Wollongong, where local emissions could have a large influence. We have added this to the discussion in the revised manuscript.

Text is revised: Pg. 12, line 17-19.

Pg. 11, line 6: I think this is a typo? Darwin is closer to the Equator than Wollongong.

We apologize for the incorrect sentence. The reviewer is correct. Darwin is closer to the Equator than Wollongong. The argument therefore needed to be rephrased. Considering its surrounding environment (see also the above comment by the reviewer), we are now more certain that the model bias in the Wollongong was likely due to an error in local emission estimates rather than long-range transport. We have revised the text in the new manuscript.

Text is revised: Pg. 12, line 17-24.

Pg. 11, last paragraph: The text occasionally refers to Figure 8 when it should be Figure S3 and vice versa.

Pg. 14, line 8: Fig. Sx? I think this figure was dropped from the submitted manuscript.

Pg. 15, line 28: Should this be a reference to Fig. 11?

Pg. 16, line 22: Should this be Fig. S4?

We apologize for the confusion that arose by inconsistencies in the figure references. We revised the references in the new manuscript.

Pg. 12, line 31: Differences are also surely due to the different time periods studied.

We agree with the reviewer that the differences in the increasing rate are also due to the differences in time periods compared. We added further discussion in the revised manuscript.

Text is revised: Pg. 14, line 2-3.

Pg. 19, line 15: Is there something missing after i.e.? I don't understand the reference to i here.

We apologize for the poor formulation in this sentence. The sentence was meant to say that it was not possible to conclude from this study which estimates better capture actual emissions, because the emission estimates for Africa were not well constrained by observations in either study. We revised the new manuscript accordingly.

Text is revised: Pg. 20 line 5-6.

Pg. 22, line 7: I don't understand what is meant here by the extra degree of freedom.

The sentence was meant to explain that estimates of Eurasian boreal were sensitive to estimates of other regions. Due to very sparse observations in boreal Eurasia, the scaling factors of in this mTC region acted more as a compensating effect. We revised the sentence in the new manuscript.

Text is revised: Pg. 22 line 27-29.

Pg. 23, line 10: Here is an example of where I found the term "region" to be confusing, as I thought at first you were referring to the optimized regions here, where in most of your inversions only one of anthropogenic or biogenic emissions were optimized.

Here, the first 'region' was meant to be an optimization region, and the second one to be the mTCs. Both anthropogenic and biospheric emissions were optimized in all mTC regions over land, and the system tends to change optimization regions that have large emissions near the observation sites. In many mTC regions, anthropogenic emissions were greater than biospheric emissions near the observation sites, and therefore, the inversion tends to scale anthropogenic emissions more. This then led to larger changes in the anthropogenic emission estimates for optimization regions and also in mTC regions. We revised the sentence in the new manuscript.

Text is revised: Pg. 24 line 1-2.

Figure 1: Please add a reference here to Table 6 for the names of the regions.

Following the reviewer's suggestion, the caption of Figure 1 is modified by adding a reference to Table 5 (original Table 6).

Caption of Fig. 1 is modified.

Figure 2: I find the colour scheme a little difficult – very hard to make out the green regions in amongst the lighter

blue! You could also consider blocking out the modified Transcom regions with black lines.

We follow the reviewer's suggestion and changed the colour scale of Fig. S4 (original Fig. 2), and mTC and land ecosystem region borders were added.

Fig. S4 (original Fig. 2) is revised.

Figure 3 (again): For the comparisons with the prior I assume it's all surface observations that are used? (As opposed to TCCON etc. . .) Can you also plot the prior difference using only the assimilated observations? It would make it more straight-forward to assess the performance of the inversion.

We revised the figure by plotting the prior differences between assimilated observations. In addition, the colour scale of the posterior comparison changed following the reviewer's earlier comment.

Fig. 2 (original Fig. 3) is revised.

Figure 4: Do the observations appear on the top panel? Can you also plot the differences between the observations and the prior and posterior (for XCH₄ only)? You could consider adding a vertical line at 2007 in both panels.

Observations were not plotted in the figure because NOAA global averages are for the surface, unlike XCH₄. However, we agree with the reviewer that the figure becomes more comprehensive by adding the observations in the top panel. We followed the reviewer's suggestion and added NOAA surface global mean CH₄ with a second y-axis. In addition, a vertical line at 2007 is added to both panels.

We did not include the differences between the NOAA global mean and the simulated XCH₄, as additional complications arise due to sampling. The top panel illustrates global mean XCH₄ from the model, but no satellite products have sufficient global coverage throughout the years to make a fair comparison with it. In Fig. 7 comparison with GOSAT XCH₄ is included, where model XCH₄ was sampled at the locations and times of the observations. We hope the reviewer agrees that the comparison in Fig. 7 is satisfying.

Fig. 3 (original Fig. 4) is revised.

Figure 5: I would use Figure S1 here since it shows the prior concentrations too. It would also be useful to give the prior RMSEs.

We acknowledge the importance of showing the improvements by the inversions for XCH₄ estimates. Following the reviewer's suggestion, we added the prior in Fig. 4 (original Fig. 5), and removed original Fig. S1.

In addition, we agree with the reviewer that it is also important to show the prior RMSE. For that, we found a table was more appropriate and therefore, both prior and posterior RMSE are summarized in Table 3 in the revised manuscript.

Fig. 4 (original Fig. 5) and Table 3 are revised.

Figure 7: I would use Figure S2 here since you discuss many of the sites in that figure in the main paper. (And you don't discuss Park Falls, but it appears here.) I'd also like to see the prior concentrations included.

Figure 8: Again, I'd suggest using Figure S3 here and showing all the regions in the main paper. And I'd include the priors on these figures too.

Response to the comment on Fig. 7 and Fig. 8:

We acknowledge that not only a selection of the sites and regions but all sites and regions are important to illustrate the results. We tried to make satisfactory figures with all sites and regions, but could not find a better illustration. Therefore, we decided to retain the figures, but selecting the sites and regions more carefully based on the results discussed in the text. We hope the reviewer agrees that the new figures illustrate the findings better.

Fig. 6 (original Fig. 7) and Fig. 7 (original Fig. 8) are revised.

Figure 12: This figure is impossible for me to decipher. I'd replace this with the bottom panels from Figure S4. (Or even move S4 to the main paper and remove Asian temperate from Figure 11.)

We agree with the reviewer that the figure in the Supplementary Material illustrates our results and discussion better. Following the suggestion, original Fig. S4 is moved to the main paper with modifications as Fig. 8. The original Fig. 11 is removed.

Fig. 8 (original Fig. S4) is revised.

Figure 13: Since you have the space, you could include Australia in the bottom right.

We agree with the reviewer that the figure is more complete when emission estimates for Australia are added. Following the reviewer's suggestion, the estimates are illustrated in Fig. S8 of the revised manuscript, together with other land mTC regions. We hope the reviewer agrees that the SH emission estimates could be represented in the Supplementary Material. This was done to better balance the manuscript by reducing the number of figures.

Fig. S8 (original Fig. 13) is revised.

Tables 5 and 6: Please include the prior RMSEs as well.

We agree with the reviewer that the tables are more complete when prior RMSE is added. Following the suggestion, prior RMSE is added in both tables.

Table 4 (original Table 5) and Table 5 (original Table 6) are revised.

Typographical comments

Throughout the manuscript, "North" and "South" should be capitalized when referring to the continents (e.g. "North America") but not when referring to regions (e.g. "south Africa").

Pg. 1, line 31: . . . and (b) the parametrization. . ." (remove "on").

Pg. 2, line 31: . . .changes in emission sources. . .

Pg. 7, line 4: . . .20 times mdm because...

Pg. 9, line 4: . . .emissions and if it is actually. . .

Pg. 9, line 23: . . .change much in B2007. . .

Pg. 9, line 2: . . .Fig. 4 are calculated. . .

Pg. 19, line 19: . . .optimized in M1 and M3. . .

Pg. 19, line 32: . . .around the coast were not. . .

Pg. 22, line 12: . . .variation or lack of. . .

Pg. 22., line 31: . . .CTE-CH₄ differ by the . . .

We appreciate the reviewer for the language correction. The manuscript is revised taking into account all the suggested changes.