Review #1

The authors discussed the comments in the response letter. However, only a few of the comments are addressed in the manuscript. I recommend the authors go through all the comments again and at least include most of the comments in the revised manuscript. In addition, I still have some comments that are not well discussed in the response letter. I think at least a major revision is needed.

We thank the reviewer for considering the revised manuscript for publication. We understand that the reviewer considers some of their comments sufficiently covered by our response letter and would like to see more significant changes in the manuscript to complement our response. In addition, some of the comments were not addressed sufficiently in the direct response, which are outlined below.

Other specific comments:

1 Before inversion, the prior emissions were pre-optimized to fit the global mean MCF mole fractions. The authors argue that the pre-optimized MCF emissions can reduce computational costs. But the inversion as shown by equation(1) is to estimate the emissions and OH by combining the information from both bottom-up estimated prior emission inventories and the observations, as well as their errors. The pre-optimize erase the information of prior emission inventories, and only keep the information of observations. I don't think this is the right way to do an inversion. Besides, the question is not answered:" Will the pre-optimization reduce the OH variation estimated by the inversions since the MCF emissions already fit the observations?" In addition, from the author's response to Fig.4, I feel the pre-optimization is somewhat arbitrary, which makes the inversion lost the prior information.

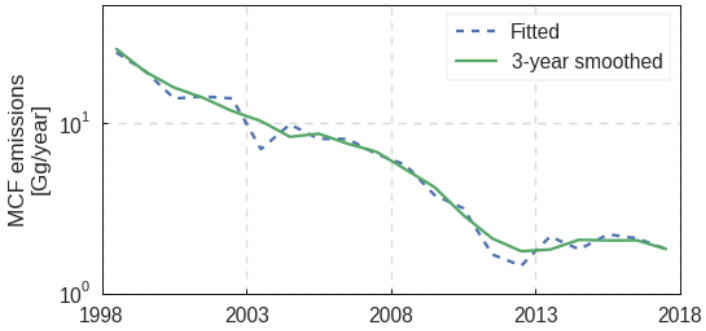

Fig. R1: *The MCF emissions that result from fitting to global mean MCF mole fractions (blue, dashed), and the 3-year smoothed emissions that are used in our inversions.*

Of course, this is an important and valid remark that we gave considerable thoughts. Firstly, we note that after the pre-fitting to annual, global mean MCF mole fractions, prior emissions are smoothed with a 3-year moving average: precisely to avoid pre-fitting interannual variability of MCF too heavily, and because there is no clear bottom-up explanation for strong interannual variations in MCF emissions. Fig. R1 shows that this smoothing helps to remove large interannual variations in emissions, though slower, multi-annual variations remain. The reviewers' remark made us realize that we had not mentioned the emission smoothing before in our response nor in the manuscript but we have now added it in L120-129.

We emphasize that in later years especially, no prior information on MCF emissions is available, and any emission prior therefore requires arbitrary assumptions. Moreover, the global mean MCF mole fraction is only a small part of the observational information that the inverse system leverages to obtain OH and emission variations. Observed spatio-temporal gradients in MCF and transport variations are key to the novelty of our work, compared to previous box model inversions, and this information is completely unused in the prior. We argue that by pre-optimizing emissions, the inversion more quickly starts with optimizing the spatial gradients that are most interesting and bring relevant information to the OH distribution. We now additionally describe and motivate this choice in the manuscript (L120-129).

2 The authors explain why the small correlation coefficient between OH and MCF variations can reflect the OH and MCF are independently derived. But I think a better indicator should be comparing optimized minus prior MCF emissions and optimized minus prior OH. We can see that in Fig.3, the MCF emissions estimated by REF inversion is much higher than prior around 2013, which is corresponding to the large positive OH anomaly around 2012-2013. This may indicate that the inversion system cannot separate the OH and MCF variations.

The reviewer's suggestion is exactly what the correlation coefficient quantifies: it is the correlation between optimized interannual OH variations and interannual variations relative to prior emissions (i.e. as a percentage of the prior emissions). In other words, the inversion derives emission and OH variations that are not correlated. We have now clarified this in the text (L221-223).

Furthermore, we note that the peak in MCF emissions is highest in 2013-14, while the OH anomaly peaks in 2012 and quickly declines into 2013-14. Therefore, while this large anomaly in both quantities is remarkable, the two are not directly coincident in time. Moreover, a positive anomaly in both quantities is undesirable from a cost-function perspective, since positive anomalies in both will cancel out on a global scale, but they will increase the background cost. Therefore, based on our understanding of the inverse system, we consider these variations driven by observational information from the spatial gradients in MCF that require compensating variations in these two state parameters that have distinctly different spatial imprints on MCF.

3 For the convergence problems. From the author's discussion, the 10-year inversions can reach convergence since they require less time per iteration. One problem is that the 10 years inversions are for 1998-2008 when the MCF emissions are higher than 2009-2018 and the corresponding errors are much lower than 2009-2018. If the 10 years inversions focus on 2009-2018, it will be hard to say if the 10-year inversion coverage to similar OH variations since the uncertainties in MCF emissions (reach 200%) are much larger OH during 2009-2018. So I don't think the 10-year inversion for 1998-2008 can prove the robustness of the 20-year inversions, as the author mentioned when discussed my last comment.

This is an excellent point that was insufficiently covered in the manuscript. The spatial signatures of OH and MCF emissions remain distinctly different also in later years, which is part of what drives the system's skill in deriving both OH and emission variations. However, the increased emission uncertainty relative to the atmospheric burden of MCF will likely impact the robustness of the system.

Between 2008 and 2013, the increase in relative uncertainty of emissions is compensated for by a sharp decline in emissions over this period. However, after 2013, prior emissions stabilize and the absolute prior error on emissions becomes significantly larger than the absolute prior error on OH (which can be approximated by assuming that 20% of the MCF burden is removed by OH each year). We now explicitly mention this in the manuscript as a point of caution for the interpretation of derived variations, especially after 2013.

We now emphasize this point in several places in the manuscript (L228-232; L412-417; L503-505)

4 "L352: Why further convergence will result in less realistic OH variations?" The authors answer this question by adding Figure S8 (but show nothing in the manuscript) which showed that the inversions are overfitting. Is this because the inversions use too small observational error?

We do make the case that perhaps the error we use on observations is overly optimistic (L264-267). However, this was motivated by the inability of our coarse-resolution simulations to capture short-term variations in MCF, not by the systematic intrahemispheric biases. We consider these systematic biases to fall outside the bounds of realistic observational and model errors, and therefore propose a changed MCF ocean flux as a more likely explanation. In other words, we consider this "overfitting" a result of including insufficient degrees of freedom in the state, rather than of too-small errors on observations.

We have slightly expanded the reference to Fig. S8 in the new manuscript (L215-216; L331-332).

5 "L372: "Firstly...we generally identified similar tendencies in each." Figure 1 has shown the variations of OH estimated by three inversions are quite different." The authors answer this by

showing the 10 years inversions are similar. But as aforementioned, the three 10-year inversions are similar may not prove the inversions for 2009-2018 can also reach similar results. Here the only thing we see in the main text is that the three 20-year inversions are quite different.

This phrasing was indeed too strong, and we have rewritten this paragraph (L408-417). We mainly view it as important that the derived OH variations are never opposite between inversion set-ups (hence, have the same "tendencies"). Opposite variations would invalidate the premise that the different inversions are moving in the same direction.

6 "L374-379: Here the manuscript tried to prove the robustness of the OH interannual by an additional inversion and a forward simulation. But the details of the two experiments and the results are not given. I suggest include some details in the supplements. E.g. how the one global scaling factors compare with the REF, POP, and TM5OH? Is the forward simulation use the prior or optimized MCF emissions? I cannot understand the logic here, can you clarify why the two experiments can indicate the robustness of the derived OH variations?"

It is still unclear how the authors conduct the two experiments. I think every model experiment established should be introduced in the manuscript or supplements. Since the two model experiments are not shown clearly, the role of the two experiments is certainly unclear.

Our main concern that we aimed to address with these two experiments was that we find only small variations in global mean oxidation (few percent) compared to the systematic spatial adjustments to the OH distribution (tens of percent). Therefore, we doubted how vital these global-scale variations were to the solution and the derived cost-function reduction. We consider this result not important enough for the already complex storyline of our study to extensively discuss them, which is why we chose to remove mention of them.

As noted by the reviewer, it remains difficult to explicitly prove robustness of the REF solution over the 2008-2018 period, and so we now clearly mark this distinction in the manuscript, as noted in point 3. Most notably, we have rewritten the first paragraph of the discussion (L408-417).

Motivated by the new and old reviews, we have made additional adjustments to the manuscript that better highlight the important qualities of our work. For example, we provide a more nuanced perspective in L408-417, and we provide suggestions for improvements in L448-462.

Review #2

I thank the authors for giving the reviewer comments careful consideration.

The authors do not propose too many major changes to the paper as a result of these comments. They acknowledge that there are some limitations to their study (e.g. lack of convergence), but make the case that it is too late to change the experimental design, and argue that there is enough useful information in the paper to warrant publication. I am inclined to agree with this assessment. However, I suggest that the limitations should be spelled out more clearly in the abstract. I suggest adding some text along the following lines (it doesn't have to be exactly this, but I think these caveats need to be noted):

We thank the reviewer for making such helpful and specific suggestions that help improve our manuscript.

- Title: I'm still quite concerned about the lack of convergence. Therefore, I wonder if it's more accurate to remove "inversion" from the title. Perhaps something like: "An investigation of top-down constraints on atmospheric oxidative capacity using methyl chloroform and a global 3D model"

We understand the reviewer's concern. However, the timeseries of OH variations we derive, even if not fully converged in the 20-year inversions, incorporates the spatiotemporal gradients of methyl chloroform observations in a way that only a 3D model inversion can. Recently, the study of Patra et al. (2020) was published, which is also well-described by the suggested title, but did not include a 3D model inversion. The title doesn't suggest any definite conclusions regarding OH variability or trends, which would not be supported by our work. Therefore, we strongly prefer to have 3D model inversion in our title.

If the reviewer still considers this title too bold, we are willing to change it.

- At the end of the first paragraph, or start of the second paragraph, I suggest noting the lack of convergence (e.g. "While our main solutions did not fully converge, they suggest interannual variations in the global oxidative capacity…")

Now noted in L6.

- When talking about the inter-annual variability, I suggest being explicit that the standard deviation of the derived OH concentration was small (< 3% per year) over the 20 year period, but that substantial fluctuations (~±10 %) were derived between certain years.

We have now added a comment that explicitly addresses the rapid change from 2010 to 2012 (8% in annual mean values), in the abstract (L10-12) as well as in the conclusion (495-496). However, as noted in our previous response, we find that the twenty-year OH variability is reasonably well described using a standard deviation of < 3%.

- At the start of the third paragraph of the abstract, it needs to be made clear what the "adjustments" are with respect to (i.e. "compared to a widely used prior OH distribution…", or perhaps even cite the relevant dataset).

Now noted in L14. We do not cite the relevant dataset, because similar adjustments are found for the two different OH distributions.

- At the start of the fourth paragraph of the abstract, clarify (perhaps in parentheses) broadly what the added value of the 3D model is.

We have added clarification in L20-21.

- Final sentence of the abstract: Given the lack of convergence, I think it is too strong to suggest that these particular results be used in studies of the CH4 budget (which is implied), and therefore suggest that this be softened to "Therefore, we consider that variations in OH derived from MCF inversions with 3D models can add value to studies of the budget of e.g. CH4"

We have softened this statement as suggested (L23-24).

In the supplement, line 35: Which section do you mean S2?

Section 2.1.1, it is filled in now.

Reference

Patra, P. K., et al. "Methyl Chloroform continues to constrain the hydroxyl (OH) variability in the troposphere." Journal of Geophysical Research: Atmospheres: *e2020JD033862*.