Author Responses to Second Round of Reviewer Comments

We thank both reviewers for their additional comments. We have addressed all the comments received, including the clarification of several points and changing the manuscript title. We have now included a joint OH-methane inversion as suggested by Reviewer 1. On reflection we decided to report methane emission estimates inferred using OH climatology and the new inversion. This helps illustrates the value of including OH in the state vector on a routine basis. We have also put our results into context of work that has been published since we submitted our original manuscript.

<u>*Reviewer 1*</u>

*First, I think the manuscript is greatly improved and I commend the authors for both adding additional inversions and using simulations that more accurately represent previous work. However, in this reviewer's opinion, the paper needs to be restructured. I am generally reluctant to suggest structural changes to a manuscript, but the current structure resulted in both confusion and misunderstandings by this reviewer about what the authors *actually* did and what they report. Frankly, this reviewer is still confused about which numbers are being reported in the abstract and what contributes to their uncertainties. This reviewer feels the manuscript still needs major revisions before being publishable.*

We thank the reviewer for these additional comments. We have absorbed the text from Appendices D-F into the main text. We have retained Appendices A (supplementary figures), B (box-model calculation), and C (evaluation) to promote readability of the study unencumbered by excessive figures. In doing so, we believe the paper now presents the OH sensitivity calculations more clearly, summarized in Table 2.

To start, this manuscript would greatly benefit from a Table that explains the various simulations including what was held constant, what was perturbed, etc. It could list both 3D inversions, forward simulations, and the Box Model. This should follow a general description of the methodology.

Agreed. We have included Table 2 that summarized the control and OH sensitivity calculations.

The material in the FIVE appendix sections should be moved into the main text. Much of this material is central to their conclusions and, in this reviewer's opinion, should not be relegated to the appendix. The main text references an OH sensitivity run, but the manuscript now includes at least 3 different descriptions of OH: their 5% OH change over CO2 source regions, OH described in Appendix D, and OH inferred in Appendix E.

Agreed, we have now included the experiment descriptions and results in the main paper.

Following this, it is not clear to this reviewer why the authors still include the 5% OH change over CO2 source regions. Both reviewers criticized this sensitivity run because it does not represent what previous work found. Yet this is currently the only OH sensitivity run that is described and discussed in detail in the main text.

We agree that this approach represents an idealised situation but do provide useful insights especially since we have no definitive way of quantifying OH changes during 2020. We have de-emphasized these calculations, but they now provide a useful sanity check for interpreting results from the joint OH-methane inversion, and we now describe in that way. We find our idealized calculations are broadly consistent on the global scale with the joint OH-methane inversion.

Regarding the reported numbers and conclusions, it seems that the authors should be reporting results from their joint inversion of both OH and methane emissions. This seems like the numerical experiment that allows them to make a quantitative statement about the relative importance of sources and sinks, which is the central claim of the manuscript. However, from my reading of the manuscript, I do not think this is the numerical experiment that is used for the numbers in their abstract/conclusions, although I am not actually sure which experiment their numbers come from.

We think we have now struck a balance. We have included the *ad hoc* sensitivity calculations with the OH inversion to build a narrative. We feel that presenting a new methane-OH inversion as a *fait accompli* would not be instructive in this case, and we believe it is much stronger as presented in the context of the fixed-OH baseline calculations and the other sensitivity experiments that produce consistent results. We have, as described above, de-emphasized the ad hoc sensitivity calculations and focused on the results inferred from the formal joint OH-methane inversion.

Regarding the box modeling, does the box modeling reflect the updated findings regarding OH? It seems that the text was not updated even though the authors state: "after considering the effects of methane sinks, we find that a one-box model calculation…" (Line 133).

It is still not clear how much the authors are perturbing global mean OH in their sensitivity runs. This would be important to state. It seems that it is still less than other work reports.

The purpose of the box model calculation was exclusively to help explain the different atmospheric methane growth rates inferred from GOSAT and *in situ* data. We show that using different observation coverage, the resulting estimates for the atmospheric methane growth agree with each other on a multiple-year timescale, but not necessarily individual years. It does not include the influence of OH. The box model calculation explains why by using *in situ* data Peng et al have overestimated the influence of OH in 2020 even though our studies agree on the change in the OH. Similarly a recent study (Qu et al., 2022) based on GOSAT XCH4 retrievals also show larger atmospheric methane increase between 2019 and 2020 than reported by Peng et al. (2022). We have updated the concluding remarks to reflect that point.

I have a number of questions regarding the OH inversion as I think this is the numerical experiment that actually supports their conclusions and claims:

- What do the spatial patterns of the OH inversion look like?

- What about the temporal pattern?

First, we have reduced the state vector so we only report annual OH changes on six 25-degree latitude zonal band. We find that the data can support the independent inference of methane emissions and OH changes over those broad regions. We have added Figure 5 to make that point, which shows that *a posteriori* correlations between OH and methane emission regions are typically < 0.1. Figure 7 shows the annual difference in *a posteriori* methane loss due to OH variations in 2020 and 2021 compared to the inversion that uses OH climatology. We describe those changes in Section 3.

- The authors mention that results in Appendix D and E are consistent but provide no numbers, figures, or really anything to back up that claim. How was this assessed? What is the bar for "consistency"?

Agreed that is confusing. Consistency in this example means that our sensitivity experiments all point to OH representation less than 30% of the observed atmospheric growth rate of methane. This statement remains valid and we have clarified this point, although the sensitivity studies has been de-emphasized in the manuscript.

- The basis functions for OH differ from methane, does that matter?

It is an interesting question, but the data does not contain sufficient information to match the resolution of the methane emission state vector. In the revised manuscript, we have simplified the OH state vector to six 25-degree zonal bands, which we find can be supported by the data (Figure 5). As part of the preparatory calculations for the manuscript, we explored using more scaling factors and we didn't find a significant difference to the result we presented in the OH change between 2020/2021 and 2019 but revealed stronger correlations between state vector elements.

- The authors mention that we do not have sufficient constraints for OH, yet they aim to solve for both longitudinal and latitudinal changes. Are those well constrained? Why not just solve for OH as a function of latitudinal bands?

Yes, this is a great point. On reflection we further simplified our OH state vector and describe in the manuscript the performance of the joint OH-methane inversion. Figure A5 shows we can, indeed, solve independently regional methane emission and zonal band changes in OH.

Another paper was recently published in Nature that uses similar data and reaches similar conclusions (~50/50 sources and sinks; https://www.nature.com/articles/s41586-022-05447-w). This work should be mentioned as it is directly relevant. This paper also included both methane and OH in the state vector (as did Zhen et al., 2022). Therefore I stand by my earlier review that the bar for claiming emissions are responsible for the changes necessitates inverting for both methane and OH

We have now cited this paper in the concluding remarks. Peng et al use *in situ* data so as we now discuss in our paper this introduces a negative bias in emission increase estimates between 2020 and 2021, and consequently a positive bias in the influence of OH on the atmospheric growth rate in 2020; our results for reduced OH in 2020 are remarkably consistent with those reported by Peng et al.

To clarify, Peng et al, do not include OH and methane together in their state vector to determine their top-down methane flux estimate. First, they calculate the magnitude and changes in OH distributions due to reduced emissions using the LMDZ-INCA model and infer OH changes by fitting a 12-box model to HCFC-141b, HFC-32, and HFC-134a measurements. This calculation will of course be subject to errors in inventories but it is reassuring that we get similar results for the reduction in OH.

We are not familiar with Zhen et al, 2022. But we can say is that our paper in its current form does provide different lines of evidence, including a joint methane-OH inversion, that are consistent with increased emissions playing the major role in the atmospheric growth rate in 2020.