Responses to final comments from reviewers

We thank the editor and the two reviewers for their patience with our addressing reviewer comments. Below we address all remaining questions/comments left from the previous reviews.

Based on reviewer comments, we have:

1) Refined the calculations so they now use EDGAR6, which provides methane emission estimates up to 2018 after which we repeat those values. We find our new posterior results are not significantly different from our previous results but nevertheless they have helped us to understand the sensitivity of our result to prior values after 2012.
2) Improved the balance of our reporting of the results across the entire study period.
3) De-emphasised our analysis of the impact of OH. Fundamentally, we wanted to explore the extent to which an alternative change in OH (as suggested by the peer-review literature) would change our conclusions and *not* to infer an OH distribution that was consistent with atmospheric measurements of methane and $d^{13}C\text{-}CH_4$. While we believe that OH changes did not have a significant ongoing influence on the growth of atmospheric methane over our study period, we cannot disprove that with our experiment. We have made that point clearly in the revised manuscript.
4) Extended the comparison between the model and data at different sites.
5) Used language with more care so the reader understands that while collective evidence supports a large role for tropical wetland emissions (as discussed in the conclusions), there is a limit to what we can say with our own analysis.
6) Generally improved our descriptions of the data and methods throughout the paper. We have explained better the estimation of isotopic source signatures.

We have also taken the opportunity to remove the original summary Figure 5. While our results remain the same – tropical emissions of CH_4 have increased with lighter isotopic source signatures (described in our revised Figures 2 and 5) – we found the visual impact of this figure was sensitive to the choice of the baseline period.

Report 1

I would like to thank the authors for their responses to the reviewers' initial comments. They made efforts to improve the presentation of their results and precision of the text. However, their revisions and replies to my comments didn't fully address my major concerns.

We now fully address every remaining concern.

First, it's important to evaluate the inversion results to demonstrate the robustness and weakness. I appreciate that the authors added a supplementary figure (Figure A7) to show good model performance at several un-assimilated sites from north to south. This is useful, although it would be clearer to indicate error statistics for the prior and posterior at each site to show the improvement (the same for Figure A1 & A2). I would still suspect poor model performance at some difficult assimilated sites such as KRS

and BKT due to uncertainties in flux or transport, which was not addressed in the authors' reply or revision. Actually, it would be useful to also show the sites where the posterior doesn't improve much, which could help identify model's deficiency and regions with large uncertainties.

We have now extended Figure A1 and A2 to show the prior and posterior methane mole fractions at more NOAA sites that show a variety of model fits to data. As noted by this reviewer, we have compared the model with data collected at sites where we assimilate data (Figures A1 and A2) and those that are independent of the assimilation process (Figure A7). We have also added mean error statistics for the prior and posterior at each site.

Second, I still don't quite understand the methodology and value of the regional isotopic source signature inversion. According to the description in the inversion methods, the control run of δ13C simulation was built on the posterior emissions from CH4 inversion, which to my understanding were the optimised total CH4 emissions for each region, not emissions for different sectors. Then how did you calculate the regional source signature? Given the large uncertainty of posterior CH4 emissions (as the authors admitted in the paper and responses), how much this could propagate and impact the trends in regional source signature inferred from δ13C? The authors didn't address my previous comment about this in their revisions [reproduced below]. Moreover, I wonder why the authors chose to solve regional source signature rather than CH4 sectorial emissions. Although the inferred trends towards lighter δ13C in the tropics partially corroborated previous studies that reported recent growth in tropical wetland emissions, it didn't provide direct evidence to support this as the contribution from wetlands and other biogenic emissions (such as those from agricultural practices) cannot be differentiated in your inversion.

For the optimization of δ13C signature, I don't quite understand the methodology. It's not clear whether regional methane fluxes and δ13C signatures were solved simultaneously or sequentially? According to the description of methodology in Lines 154–163, it seems that the solution of regional δ13C signatures relies on the solution of regional emissions. I wonder how much errors in estimates of regional emissions would impact the solution of δ13C signatures. Can we trust the results presented in Fig. 3 if the emission trends detected for certain regions are not robust?

The overarching aim of our study was to understand if three separate lines of observational evidence (remote sensing from satellite, in situ mixing ratios and isotope ratios) support a hypothesis on the long-term trends driving the global methane rise. Our approach has therefore purposely not combined all evidence into a single inversion, and we consider the satellite and in situ mixing ratio inversions as the primary lines of evidence for the study.

To address the reviewer comment about whether the isotopic source signatures can be trusted given their reliance on the posterior emissions estimates, we have now edited the text to bring the reviewer's point into the discussion and to make it clear that we interpret the isotopic data as a supplementary line of evidence that is in support of the emissions estimates calculated from the CH_4 inversions. It is well understood that regional isotopic source signatures are very poorly constrained owing to the

uncertainty on the source signatures of individual sectors and how they vary over space and time. The isotope ratio measurements are also sparse relative to the mixing ratio time series. We therefore decided to remove the isotopic data from the first inversion that solves for the regional total CH_4 emissions, which prevented potentially inaccurately assigned isotopic source signatures on influencing the trends in the methane emissions estimation.

By keeping the isotopic evidence separate we can also analyse those results independently (as we have done for remote sensing vs in situ mixing ratios) although only in context of prescribed emissions estimates (as highlighted by the reviewer). We have improved the description of the methodology to communicate our approach and the reasons for it more clearly.

The use of repeated prior emissions after 2012 could also be problematic. As the observation network are sparse globally, the optimised fluxes would be sensitive to the choice of the priors. The use of updated inventories (e.g., EDGARv5 or EDGARv6 over EDGARv4.3.2) are encouraged as errors in earlier versions were corrected in new versions. In Figure A4, the spikes in the emission signatures of Tropical Asia and South America disappeared in the later period of your simulation, probably due to the use of repeated fire emissions after 2012. Have you evaluated the impact of using repeated priors after 2012 on your inversion results? The authors claimed that the trends towards isotopically lighter sources was started since 2012 (Figure 4 and the corresponding text), could it be somehow related to the specific configuration of the priors?

This has now been addressed. We have used EDGARv6 that describes emissions from 2004 to 2018, after which we repeat 2018 emission estimates. Figures A1 and A7 show that the prior inventory already does a good job after 2012, better than what we used previously. We find our posterior results are not significantly different from our previous results but nevertheless they have helped us to understand the sensitivity of our result to prior values after 2012.

Third, the sensitivity test results using only one particular OH scenario (decreasing by 0.5%/yr for 2004-2019 and 5% for 2020) were mostly within the posterior uncertainty of the control run using the climatological OH. In my opinion, this does not imply that the variations in OH was not likely to have significant impacts on the atmospheric methane growth for the study period. It only reflects the fact that the inversion system is not well constrained (so that changing OH trends and variabilities doesn't change the inversion results too much). The authors should do better to prove this.

We have de-emphasized our OH experiment, as suggested by this reviewer. We wanted to explore the extent to which an alternative change in OH (as suggested by the peer-review literature) would change our conclusions. While we believe that OH changes did not have a significant ongoing influence on the growth of atmospheric methane over our study period, we cannot disprove that with our experiment.

We agree with this reviewer that because our inversion does NOT include OH in the state vector so we cannot comment on the ability of the *in situ* data to estimate

methane emissions and OH. This involves more in-depth calculations that are outside of the paper. However, recent work by Feng et al, 2023 showed that GOSAT and *in situ* data together can independently constrain estimated geographical changes in OH and methane emissions on coarse spatial regions.

Overall, the methodology and results presented in this paper are not adequate to support the main conclusions (i.e., multi-decadal shift in the global methane budget towards natural tropical emissions, particularly wetland emissions; limited role of OH variations in atmospheric methane growth), despite some consistency with former studies claimed by the authors. In terms of writing and the presentation of results, the revised version has been improved, but still lack precision and details.

We have gone through the paper and improved the text for detail and precision. These changes can be seen in the tracked changes version of the manuscript.

For clarity, we say that our results are *consistent* with result studies that have highlighted a growing role for wetland emissions. We agree with the reviewer that we cannot say anything more with the data that we have reported in this manuscript. In the revised conclusions, we have attempted to bring together and interpret the different lines of empirical evidence.

Report 2

Overall, the manuscript's structure has improved, and the introduction now sets out more clearly what is being studied and why. However, I have a few major and minor comments on the current version.

Below, we describe how we systematically address all the remaining comments from this reviewer.

Major comments:

1. P8 L 228 ff: The results part only discusses emission changes in 2020. It is still not clear to me why the authors are interested in the COVID-19 lockdown year 2020, instead of evaluating the whole period 2004-2020 and the trend of emissions after 2007. Is this not the actual purpose of the study? In my opinion, these are two different scientific questions (i.e. 1. the changes in CH4 source contribution after 2007, 2. changes in CH4 emissions/OH during the COVID-19 lockdown).

We agree with this viewpoint. The focus on this study was not the recent changes by the longer-term change since 2004. We now briefly discussion 2020 but since this has now been addressed by a few papers so we can put our study into context of recent findings.

However, the sensitivity study considering a 5 % drop in the OH concentration in 2020 testing the robustness of the presented results is fine. Although I miss explanations on the results (see my previous comment 52). A discussion of the OH uncertainties could also be added here (as in my previous comment 14).

Based on this comment and the comment from Report 1 we have de-emphasised this sensitivity run but for completeness we do discuss the results we find and include a discussion of OH uncertainties.

2. Table 1: The emissions table is shown again, without any reference to the results of this study. Can a posteriori estimates be compared to those values shown in the table? Are they within the uncertainty ranges of Saunois et al.?

We apologize for this oversight. We have now included our a posteriori estimates into Table 1 and commented on their agreement with Saunois et al.

3. Unlike the first version, the authors now distinguish between biogenic natural and biogenic anthropogenic emissions. A posteriori estimates indicate a shift towards more tropical biogenic emissions. In their conclusion, the authors claim that these biogenic emissions originate from wetlands. How is this conclusion derived exactly? Which satellite data do you use to identify wetland regions? This should be discussed in more detail. Would the increasing seasonality in tropical regions shown in Figure 4A support an increase in natural emissions, e.g. from wetlands?

We agree with both reviewers that we cannot attribute the observed shift in methane emissions exclusively to an increase in tropical wetland emissions. The most we can say that the location of increased methane emissions and isotopically lighter $\delta^{13}C$ is consistent with the known locations of large wetland regions. We have now made that clearer throughout the manuscript.

Minor comments and technical corrections:
1. Some of my comments have misleadingly resulted in these points now being described in too much detail, rather than the actual point being made briefly and more precisely. E.g.:

Comment 2: Instead of explaining the GWP, the sentence "CH4 is the second most important GHG in terms of anthropogenic radiative forcing" would have been fine, too.

Comment 6: I only missed the explanation on the usage of the isotopic signatures, such as the authors now describe on P3 in L82 ff (the exact explanation of the delta value was not necessary).

However, I leave it to the authors to decide how much of the newer details they want to keep in current version.

We have now addressed these points. See tracked changes.

2. P2 L21: delete second "from"

Done.

3. P2 L29: Do Lan et al. (2021) really assume a decrease in thermogenic sources?

The reviewer is correct. Analysis reported by Lan et al (2020), including measurements of $\delta^{13}C\text{-}CH^4$, suggest that thermogenic sources are unlikely to be the dominant driver

of the post-2006 global mean increase in atmospheric methane. We have now clarified that point.

4. P2 L51: "…OH proposed by Turner et a. (2017) are…"

Done.

5. P4 L116: I guess, that the inversion has no problem with upscaling yearly repeated emission of 2012. However, the a posteriori uncertainty would be larger. Could you briefly state the effect on the systematic underestimation in the a priori emissions from 2012 instead of those between 2012 to 2020?

This is a point also raised by the other reviewer. We have now addressed this directly by using EDGARv6.0 that describes methane emission from 2004 to 2018; after 2018, we repeat 2018 emissions . We find our posterior results are not significantly different from our previous results that used 2012 emission in years later than 2012, but nevertheless it helped us to understand the sensitivity of our result to prior values after 2012.

6. P8 L23-24: I do not understand the authors' explanation on why they only consider decreasing OH trends between 2004 and 2019. Where is the connection to an OH decline during COVID-19 in 2020?

First, we have reduced our emphasis of changes in OH in the paper. We report the results from an idealised experiment, but nevertheless if provides us with some idea of the robustness of our results.

We argue that there is gradual, long-term geographic shift of methane emissions to the tropics that is independent of the perturbation in 2020.

The underlying explanation for the increase in atmospheric methane in 2020 is a combination of increased emissions and a decrease in OH (due to reducing emissions of nitrogen oxides from the widespread shutdown of manufacturing during Covid-19).