Author responses to reviewer comments on acp-2022-561

We thank the two additional reviews of the paper. We respond below to all comments.

REVIEWER 1

Comments on acp-2022-561

Atmospheric data support a multi-decadal shift in the global methane budget towards natural tropical emissions. Drinkwater et al.

Explaining why methane is rising so rapidly is important. We simply don't know what is going on. What's more, methane is simultaneously becoming relatively richer in 12C, reversing its centuries-long trend towards 13C. Why? – the simplest answer is that the rise is driven by new inputs of biogenic methane, though it is also possible that changes may be happening in the methane sins.

But this puzzle is not like most scientific puzzles. Figuring out the exact life cycle of a graptolite or the exact origins of an ancient volcanic ash in an ice core can be solved in a leisurely way. But understanding methane is urgent. It very directly affects the hopes for the UN Paris Agreement and the climate future of us all.
Drinkwater et al. make a very good attempt to address this great problem. Yes, some individual assumptions and parameter choices they make can be debated – that's what science is about – but the work is sound.

First I will list my own very minor requests for changes. Then, as requested third referee, I shall comment on the earlier assessments of the earlier version of this paper, and the responses of the authors.

***Minor comments**.*

1.) Two new papers are relevant and should be considered:
*Oh, Y., et al. (2022). Improved global wetland carbon isotopic signatures support post-2006 microbial methane emission increase. Communications Earth & Environment, 3, 159, 1-12. Zhang Z, et al. (2023) Nature Climate Change. **13**, 430–433*

Good suggestion by the reviewer. The Oh et al study is relevant to our discussion about the tropical wetland signature of carbon isotopes, and we have now included it in the revised manuscript. The Zhang et al reference is less relevant for our work, although we appreciate it contains a useful model-led message.

2.) Abstract lines 4 and 5, and in the main text conclusions – It would help general readers to have some idea of the total increase in emissions over the 17 year period – the 'acceleration' (Tg/yr/yr) is given but not the total change (i.e. how much greater emissions were in 2020 than in 2004.). That should be given in Table 1 perhaps, and mentioned in the concluding section 4. Indeed, what exactly does 'Annual Mean Emissions (This study)' convey in Table 1? Maybe that's because it's being compared with Saunois et al, but it's like saying your speed as you accelerate down a freeway is some mean between when you entered from the junction and now when you're whizzing along, foot flat on the pedal.

We have now addressed this suggestion by tweaking the abstract and adding Table 2. For the abstract, we have stated that we use the Siegel linear estimator to determine the linear trend. With respect, we don't think reporting the emissions from 2004 and 2020 is insightful – some regions show a progressive increase with time while other regions show a large year to year variation but with an overall increase/decrease – so that reporting values for two years is not sufficient to reconcile with the overall linear trend being reported. Annual mean emissions from individual regions are reported in Figure 3. In response to this reviewer comment, we have added Table 2 that includes the information requested.

The reviewer is correct that we reported the mean statistic because it is reported by Saunois et al. We compared our results with Saunois et al, following a previous reviewer suggestion.

3.) Line 125 – maybe some discussion of Oh et al 2022 would be useful?

Agreed. Now addressed.

4.) Line 139 – Any thoughts on the OH KIE puzzle? Cantrell? Sauressig?

For the OH KIE, we used *a* value based upon Saueressig et al, which differs from values proposed by Cantrell et al. We use this value because it is the recommended value from JPL Chemical Kinetics studies, version 19 (Saueressig et al is the latest comprehensive study on the OH and $^{13}CH_4$ reaction). We understand that discrepancies between studies are not resolved and that more work is required, but a discussion is not within the scope of this paper.

5.) Line 334 – Several other recent papers have also come to fairly robust conclusions that OH, while important, is not the primary driver of growth.

Agreed. We have now contextualized our result but acknowledge (following a prompt by a previous reviewer) that our paper is not the right study to highlight the role for OH.

6.) it would help to make Table 1 more detailed, or perhaps to create an entirely new Table 2 to list all the changes in emissions and growth rates over the study years. (see comment above on the Abstract).

We have put together a new Table 2 in response to this reviewer comment.

Comments on the authors' responses to earlier remarks

Referee 1 comments on 1. the need to assess both the robustness and weaknesses of the inversion; 2. is concerned about regional isotopic signatures; 3. is worried about the sparseness of the observational network and thus the sensitivity of the optimised fluxes to the priors; and 4. is concerned about OH.

The authors have responded with significant revisions and perhaps a softening of their conclusions, that their "results are consistent with result studies that have highlighted a growing role for wetland emissions".

As third referee, I agree with the good points raised by Ref 1 over the initial submission, but I also consider the authors have responded well to the comments and have made appropriate revisions. The methane problem is unconstrained – we have too few real data, whether in the measurement network or in the source signature, so we have to do the best we can. We can't put the problem off for a decade until we get more stations and better measurements.

Agreed. We appreciate this input from the reviewer.

Referee 2 *also makes helpful comments.*
1. Question about 2020. This year was extraordinary for methane. So it's well worth detailed attention. Note that 2021 was also extreme. Although covid obviously had impacts on air chemistry, these dramatic growth events in 2020 and 2021 were probably not primarily because of covid. Factors like the unusual triple dip La Nina and the behavior of the Indian Ocean Dipole were surely more significant. Indeed, if the growth in 2007-2018 was interesting, the changes since 2019 seem to be of a different order.

We resisted a detailed discussion about 2020 and 2021, given previous reviewer comments, and as this reviewer acknowledge we have softened some of our conclusions. We have added a statement in section 4 referring the reader to more detailed studies on this subject.

2. Table 1 – see comment 6 above on ref. 1

We have now included a new Table 2 including the information requested.

3. Biogenic natural vs biogenic anthropogenic. Of course rain feeds cows as well as wetlands and these two are almost indistinguishable. Note the Z. Zhang et al. (2023) revision of wetland emissions – we very badly need new real in situ observations from wetlands, especially tropical wetlands, not models.

Agreed. There is a possibility of a climate feedback mechanism involving ruminants, but anomalous flooding (associated with increased wetland emissions of methane) can also lead to the loss of cattle as they are swept away. There is evidence that local communities are taking advantage of larger flooded areas in S. Sudan to cultivate rice, which add the difficulties associated with robust source attribution.

Conclusion

This is an important paper that has been well debated in review, has responded well to helpful comments, and now deserves to be published, perhaps after some small further changes. The topic is important and urgent and the work is sound, as far as can be achieved given our lack of measurements, especially in the topics. This contribution needs to be published, to become part of the wider debate.

We appreciate that this reviewer appreciates the importance of our work as part of the wider debate.

REVIEWER 2

The analysis use state-of-the-art techniques, although the complex inverse modelling results, using also carbon isotope signatures, are difficult to check by an reviewer. The real test would come from independent modelling trying to replicate these model results, and for this reason it is important that the authors make available the exact model assumptions, and data selections used in this modelling. My minor suggestion here refer to some clarification issues in the abstract and conclusions that make the interpretation of these results more accessible to a larger audience.

We agree that sufficient information should be provided to the reader so they can reproduce our calculations.

375-380 an extended discussion of the above could be included here and summarized in the summary.

Table A2 and Figure 1 show the sites chosen for the ground-based inversion. The sites were chosen as ones that had complete monthly coverage over most of the time period examined (2004-2020). Model assumptions are described in section 2.2, including details about the isotopic simulations (Table 1). Assumptions about the inversions are described in section 2.3. We believe there is sufficient information in the manuscript to reproduce our work.

To address this comment, we have added references to the site selection in section 4 and have linked our consistency with previous studies to confidence in our model assumptions and data selection.

l. 8 an increased biogenic source - aka wetland source- in the tropics. Explain here what main mechanism(s) are responsible for this: temperature, rainfall, wetland extents.

Done.

l. 10 heavier isotope sources: what could that mean on the ground wrt source changes in China?

Heavier isotopic signatures in China are indicative of greater proportion of sources from thermogenic or pyrogenic sources than is suggested by the prior emissions inventories.

l. 15 what is meant with 'robust against', does it mean 'not sensitive to'? What is it that is not consistent with the global growth- and could there be an additional missing process that could make it consistent.

We have now clarified this point. Our use of 'robust' means that the agreement is within the uncertainty of the estimate and/or the observations.

We have revised that last statement so it is now clearer. Changing OH too much is not only inconsistent with observed changes in methane but other trace gases that have OH oxidation as their dominant loss process. A detailed discussion is outside the scope of this paper, as noted by a previous reviewer.

What is the consistency of this statement with the recent paper of Stevenson et al. https://doi.org/10.5194/acp-22-14243-2022 that suggested a strong role in 2020 for methane concentration growth? (for discussion).

The analysis of Stevenson et al is an anomaly within the wider debate – it appears very little observational data was used to support the conclusions being reported and so it is difficult to reconcile it with a growing number of studies that have used a variety of data to test the hypothesis about a larger role for OH in determining atmospheric methane growth in 2020. In section 4, we have now pointed the reader to more detailed data-driven studies of the methane growth rate in 2020 and the role of OH.