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Comment on acp-2022-317

Anonymous Referee #2

Referee comment on "Estimating emissions of methane consistent with atmospheric measurements of methane and δ^{13}C of methane" by Sourish Basu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-317-RC1, 2022

Comments on Basu et al.

Estimating emissions of methane consistent with atmospheric measurements of methane and d^{13}C of methane.

First, a disclaimer: in the small closely-collaborating global methane community I have published with several of the co-authors in this paper. My review thus needs to be editorially assessed in terms of potential conflict of interest.

General comments

This is a major contribution to a very important global debate. The work is thorough, has no obvious major errors, and is well-presented.

Basu et al. address key challenges to the UN Paris Agreement – our very poor knowledge of the global methane budget, and our lack of understanding why methane is rising so fast.

If the methane burden is not brought under control soon, the Paris Agreement will fail. Conversely, halting methane growth and then reversing it may be the most immediately cost-effective way of reducing greenhouse warming. There are two linked but separate puzzles – 1) determining the overall methane budget, and 2) understanding the causes of recent growth. Thus this paper, which seeks to answer both questions, is globally important if correct.

Hitherto most global methane budget studies have not used isotopes to constrain the global budget. Hence many budget assessments simply do not tally with the direct isotopic measurements in the air and thus must be wrong. Although some regional budgets have used isotopic balance as a powerful constraint, very few global inversions are consistent with the isotopic record.

The recent exception to this lack of isotopic balance is the important paper by Zhang et al. (2022) Anthropogenic emission is the main contributor to the rise of atmospheric methane during 1993–2017." *National science review* 9.5: nwab200. Zhang et al domake use of the isotopic data. They inverted a two-box model representing the troposphere in each hemisphere. In contrast, Basu et al. use global atmospheric inversion based on TM5 4DVAR to assimilate methane and $d^{13}C$ in methane. As explained in their paper, this is intrinsically a much more powerful method, avoiding the necessary simplifications of box models.

Both studies, Zhang et al and Basu et al., use similar evidence but different analysis. Zhang et al (2022) find that the dominant cause of recent methane growth is anthropogenic inputs. They further comment that "the hypothesis that a large increase in emissions from natural wetlands drove the decrease in atmospheric δ13C-CH4 values cannot be reconciled with current process-based wetland CH4 models".

In contrast, Basu et al. come to somewhat different conclusions, perhaps challenging current wetland process models, especially in the tropics. Their work is thoughtful and well-crafted, and persuasive. This is an intelligent, important and scientifically very productive debate, that has wide-reaching social implications.

Thus Basu et al. should be published after minor revisions.

Specific comments

Line 7. Tropics – for future studies it would be interesting to include the extra-tropical monsoon regions.

Line 50. Box models. Major comment here is that Zhang et al. (2022) should be discussed and its different methodology assessed explicitly. Basu et al's analysis is very persuasive, but the differences of methodology between Basu et al and Zhang et al. should be made clear, and the debate should be returned to in the discussion in Section 4.

Line 60. The F parameter (presumably Flux?) is not specified. It would be good to have a small table of parameter identification to identify all the variables.

Line 94. Maybe later in the paper (in section 4) the power of the TM5 4DVAR approach could be re-emphasised. It's what makes this paper potent.

Line 102. Here we have fluxes termed 'x'…see line 60.

Line 136. Is Bergamaschi et al 2007 the last word on Termites? If so, maybe more work is needed. Maybe also relook at older work, such as by Zimmerman.

Lines 137 and 142. The ice core work by Petrenko's team suggests very strongly and persuasively that natural fossil emissions are much smaller than found by Etiope et al 2019. The Petrenko team's papers (e.g Hmiel et al https://doi.org/10.1038/s41586-020-1991-8) need to be considered explicitly here. It's OK if Basu et al disagree with Hmiel et al, but they must explain why they refute the pre-industrial ice core results.

Line 157. Presecribed sinks. OK, understand, but might be worth adding a line of discussion on the impact of this decision to prescribe sinks.

Line 165 paragraph. Upland soil sinks especially in the moist aerated tropical woodlands – very little information on them. Might be worth a discussion here or later in paper?- e.g likely climate change response?

Line 195 I tried the doi but had no answer. All I got was advice on making a cake. I quote: It "looks like there aren't many great matches for your search. Try using words that might appear on the page that you're looking for. For example, 'cake recipes' instead of 'how to make a cake'."

Figure 3 right – shows how important it is to get isotopic measurements from Africa and S. America and South Asia. The sole site in Africa, in Namibia, samples southern ocean air. This lack of tropical observation should be stressed in the conclusions.

Line 285 There might also be a discussion here of the difference between Basu et al's findings and the implications of tropical wetland process models mentioned by Zhang et al..

Line 263 C4 signature chosen seems a bit heavy – yes for pure C4 tropical grass fires but many are perhaps a bit lighter as there are lots of bushes etc in the fires also. Note also that tropical wetlands have varying C3/C4 ratios – dominantly C4 in equatorial areas (papyrus) but more C3 reeds in outer tropics. This gradation also shows in land plants – tropical ruminants are typically as much browsers as grazers.

Line 289. Also L 348. Inundation – yes, but temperature also. It's likely T is a key factor, with an Arrhenius relation to methane flux. But there is very little work on T impact in tropical wetlands. Lots of models simply cite Gedney Q10, based on a few much older studies.

Note that a key factor may be inundation depth – if the plants are rooted or floating. If they are rooted in ooze (e.g. 4m tall papyrus) then they advect a lot of methane from the anaerobic bottom direct up the stalks. But floating plants in oxygenated shallow open waters allow water methanotrophy to eat the ebullition. So flooding increases area of inundation but may reduce emission from the deeper parts. Conversely the T in hot dry years increases emissions provided there is enough water (and wetland responses have a long, 6-12 month lag time after preciptation anomalies, but an immediate response to Temperature change.).

Line 300. Nice to cite Tans 1997. Key paper!

Line 325 – core finding of the paper.

Line 344 – estimate of pyrogenic emissions not changing? That's a bit surprising?

Line 390 – major finding…shows how important the isotopic data are.

Line 395. It would be interesting to model what I'd call the monsoonal tropics, rather than limit by geometric latitudes – i.e the region within the sweep of the Inter-tropical convergence zone.

Line 466. Pyrogenic. Surprising.

Line 508. Maybe there should be a mention here of Feng et al. (2022)Tropical methane emissions explain large fraction of recent changes in global atmospheric methane growth rate." *Nature communications* 13.1 : 1-8.

Line 525 The paragraphs where Basu et al. paper discuss 'GCP' findings need to be rewritten and the important work of Zhang et al needs to be addressed. There might also be a discussion of the conflict between Basu et al's findings and the implications of tropical wetland process models (compare Zhang et al., who accept the modelling). Note that many process models share a major weakness in their dependence on a Q10 assumption that is based on very little observational evidence.

Line 560 More generally, perhaps in the discussion section, maybe the paper could discuss the wide range of needs: more tropical isotopic measurements, better information on isotopic signatures (but I note Line 503), better knowledge of tropical soil uptake, better assessment of Cl and OH sink fractionation, and the potential usefulness of D/H isotopic monitoring. It might also help to add here a table 6 of the methane budget findings by source.

Summary.

Basu et al have a very strong and convincing case: This is an excellent paper and a major contribution. Only minor revisions are needed.