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## Reply to anonymous reviewer 2

Sourish Basu et al.

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Author comment on "Estimating emissions of methane consistent with atmospheric measurements of methane and  $\delta^{13}\text{C}$  of methane" by Sourish Basu et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-317-AC1>, 2022

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We thank the reviewer for taking the time to carefully read through the manuscript and the generally positive comments. The work presented here is a culmination of almost five years of effort, and it is gratifying to know that others in the community consider it a step forward. We are happy that the reviewer thinks this work is an important contribution to the field and appreciate the suggestions for improvement. Please find below the reviewer's comments italicized, with our responses in normal font.

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

We defined the "Tropics" as the region where the angle of solar declination can be  $90^\circ$  at least once a year. We are aware that there are several other definitions, such as the region where trade winds are primarily easterly ( $\sim\pm 30^\circ$ ), or the region where the incoming solar radiation exceeds the outgoing terrestrial radiation ( $\sim\pm 35^\circ$ ). Some of these definitions indeed encompass extra-Tropical monsoon regions that are of interest for the global methane budget. We will take the reviewer's suggestion and include a larger region ( $\sim\pm 30^\circ$ ) into our definition of the "Tropics" in future work.

Note that when we compare our estimates to the GCP estimates, we do evaluate our emission totals for this larger band to be comparable to the GCP definition. However, we are unable to change our definition of the Tropics throughout the current work because it would involve significant additional computation to calculate the emission uncertainties for that new definition.

*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.*

We have added Zhang et al (2021) to the references here for  $\text{CH}_4$  and  $\delta^{13}\text{CH}_4$  box modeling, and have discussed results from Zhang et al (2021) in the "Conclusions and Discussion" section. We note here that none of the scenarios of Zhang et al (2021) reproduce the atmospheric  $\delta^{13}\text{CH}_4$  very well (consider Figures 1B and 2A of that paper), therefore their estimate of microbial (fossil) contribution to the recent  $\text{CH}_4$  growth is likely to be an underestimate (overestimate). In response to another reviewer, we have also added comparisons to a few other publications to the same section.

*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.*

Explanation of F added.

*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.*

We are happy that the reviewer appreciates the significance of using a 3D atmospheric inverse model as opposed to a box model. We have indeed mentioned the advantages of a 3D inverse model at different places in the manuscript. However, we are unsure of how exactly to further emphasize the power of a 3D inverse model, or whether such emphasis is needed.

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

Fair point. The difference is that in general "x" is a linear vector of all fluxes to be optimized, while  $F_s$  earlier denotes source specific fluxes that have spatiotemporal structure. Calling the vector "F" at this point would be mathematically inaccurate. To resolve this, we have added a description for "x" here, "the set of all  $F_s$  in 2.1".

*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.*

Zimmerman et al (1982) estimated 150 Tg CH<sub>4</sub>/year emission from termites, which does not fit our current knowledge of the methane budget. Bergamaschi et al (2007) does not actually construct estimates of termite emissions, they use a database from Sanderson (1996) which gives a much more realistic estimate of 19.5 Tg CH<sub>4</sub>/year. We have not found a more recent update after that. Also, we have changed the citations for termite and wild animal emissions to Sanderson (1996) and Houweling et al (1999) respectively in the text.

*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.*

We cannot in fact refute the smaller geologic emission estimates from Petrenko's group. We can only estimate the **total** fossil emission, and therefore have been very careful not to report anthropogenic fossil emissions. The referee is correct that if the true geologic emission in fact is ~5 Tg CH<sub>4</sub>/year or lower (Hmiel et al., 2020), then our estimate of the anthropogenic portion of fossil emissions would be significantly higher and even further from GCP estimates. We have added a discussion of this in the section where we compare our emission estimates to the GCP budget. Note that even with a lower estimate of geologic emissions, our attribution of the recent growth of atmospheric methane still stands, because we do not expect geologic sources to change over decadal time scales.

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

This is of course a long-standing debate in the methane community. We do not currently have a method – based solely on CH<sub>4</sub> and  $\delta^{13}\text{C}_{\text{CH}_4}$  measurements – to disentangle the sources and sinks. Therefore, we keep the sink fixed. In a previous publication (Lan et al., 2021) we explored the possibility that the entire post-2007 methane growth was due to a

trend in OH, and concluded that such a scenario did not fit the atmospheric  $\delta^{13}\text{CH}_4$  data. However, we cannot currently rule out that sink variations play a relatively minor role in the renewed growth. Such a variation, however, would have to be mechanistically plausible, ruling out large trends in atmospheric OH (Anderson et al., 2021; Nicely et al., 2018). In the future we would like to explore ways to use  $\delta\text{D}$  of  $\text{CH}_4$  measurements and models to disentangle the influences of sources and sinks. We have added a discussion of this in the new section "Future needs".

*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?*

The referee is correct that the total magnitude of and trend in upland soil sinks are highly uncertain (Murguia-Flores et al., 2021; Ni and Groffman, 2018). While we have used a soil sink map generated by the same biogeochemical model as our wetland fluxes (so they're at least internally consistent), we do not claim that it is perfect in any way. We have added this as an area of improvement in the newly-added section "Future needs" especially for  $\delta^{13}\text{CH}_4$  analysis. We are currently setting up inversions with alternate process-based realizations of the soil sink (Oh et al., 2020).

*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'."*

We are unsure why the link did not work for the reviewer. Clicking on <https://doi.org/10.15138/64w0-0g71> gets us to a NOAA website which asks for a password to download the dataset. The password was provided to the referees during submission, and the dataset will be made public once the paper is accepted for publication.

*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.*

We agree with the reviewer and stress this in the new section "Future needs".

*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..*

The separation of microbial fluxes into wetland and anthropogenic by Zhang et al (2021) relied on their wetland process model and not on atmospheric data. We note this in the revised manuscript in the section where we compare results from the two works. We also note elsewhere in the revised manuscript that current biogeochemical models have several deficiencies (such as inability to correctly model tree stems and transpiration), which could prevent them from simulating a large increase in natural wetland emissions.

*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.*

The reviewer may have a point. From our  $\delta^{13}\text{C}-\text{CH}_4$  inventory v2020 (Sherwood et al., 2021), the C4 signatures from four studies (grass fire from Brazil, Zambia and Zimbabwe)

we compiled seems to be lighter, at around -17‰. However, we were concerned that they presented a very small sample size, and perhaps the material burned in those studies were not pure C4 grass. Therefore, we decided to use Cerling et al (1998), which included more than 800 plant varieties.

We would also like to emphasize here that we do not assume a grid cell to be purely C3 or C4. We calculate source signature maps for biomass burning by multiplying C3 and C4 signatures of -26.7‰ and -12.5‰ respectively (Cerling et al., 1998) with the C3/C4 fraction for each 1°×1° grid cell. This results in variation of the source signature from gridcell to gridcell depending on the C3 and C4 fractions (although, to be honest, the C3/C4 map may have unquantified errors). Bush is normally C3; so a grid cell with (say) 40% bush and 60% tropical grass will have a source signature of  $-26.7‰ \times 0.4 + -12.5‰ \times 0.6 = -18.18‰$ .

*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.*

We agree that temperature is also a key factor that controls wetland methane emissions, and Q10 is sensitive to soil and vegetation properties. In fact, the version of TEM model that we used for this paper optimized Q10 for different latitudes and wetland types separately, and we used observations from seven independent sites to optimize wetland-specific Q10 for tropical regions (please refer to Table 2 of Liu et al (2020)).

Liu et al (2020) also thoroughly investigated the sensitivity of different inundation and meteorology inputs (including different temperature datasets) in wetland emissions (Figure 4 and Table 8), and the results show that the uncertainty from different inundation extent is much larger than the uncertainty from different temperature inputs. Based on this analysis, we decided to test the impact of inundation extent on our emission estimates.

*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 precipitation anomalies, but an immediate response to Temperature change.).*

We agree that inundation and plant root depths, and lags in wetland emissions are all very important but missing parameters/processes in current biogeochemical models. Also, current biogeochemical models do not accurately simulate the methane emissions from aquatic sources (Rosentreter et al., 2021), tree stems (Barba et al., 2018), and transpiration (Helfter et al., 2022) properly. These missing processes/parameters cause major uncertainties in biogeochemical models, which probably explains why current process-based wetland models cannot explain the large increase in emissions from natural wetlands (Zhang et al., 2021).

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

Yes, it is a bit surprising. Our GFED 4.1s prior does have a small downward trend in pyrogenic emissions, but that is taken out when we assimilate  $\delta^{13}\text{CH}_4$  data. In the revised manuscript, we have compared our results with a few others in recent literature that have also used  $\delta^{13}\text{CH}_4$  data (McNorton et al., 2018; Thompson et al., 2018; Zhang et al.,

2021). They all infer a downward trend in pyrogenic emissions, which then requires an upward adjustment in fossil (since that is the only other enriched source) to balance the  $^{13}\text{C}$  budget. However, we think it likely that in those studies the pyrogenic trend was forced by the prior and not the data. McNorton et al (2018) did a test where they took all trends out of their prior fluxes, and their resultant estimate did not have a pyrogenic trend in the posterior. This is in line with our finding of a negligible trend in pyrogenic emissions.

*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.*

Agreed. As mentioned in an earlier response, we would like to do this in future work, potentially aided by addition  $\delta^{13}\text{CCH}_4$  samples from the Tropics.

*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."*

Feng et al (2022) pin ~80% of the recent growth of atmospheric methane on Tropical emissions. This is actually a larger fraction than suggested by our Figure 11.

*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.*

We have added a section specifically comparing our results with Zhang et al (2021).

*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.*

We have added a "Future needs" section under Discussions to address these issues.

## References

Anderson, D. C., Duncan, B. N., Fiore, A. M., Baublitz, C. B., Follette-Cook, M. B., Nicely, J. M., and Wolfe, G. M.: Spatial and temporal variability in the hydroxyl (OH) radical: understanding the role of large-scale climate features and their influence on OH through its dynamical and photochemical drivers, *Atmospheric Chem. Phys.*, 21, 6481–6508, <https://doi.org/10.5194/acp-21-6481-2021>, 2021.

Barba, J., Bradford, M. A., Brewer, P. E., Bruhn, D., Covey, K., van Haren, J., Megonigal, J. P., Mikkelsen, T. N., Pangala, S. R., Pihlatie, M., Poulter, B., Rivas-Ubach, A., Schadt, C. W., Terazawa, K., Warner, D. L., Zhang, Z., and Vargas, R.: Methane emissions from tree stems: a new frontier in the global carbon cycle, *New Phytol.*, <https://doi.org/10.1111/nph.15582>, 2018.

Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Dentener, F., Wagner, T., Platt, U., Kaplan, J. O., Körner, S., Heimann, M., Dlugokencky, E. J., and Goede, A.: Satellite cartography of atmospheric methane from SCIAMACHY on board ENVISAT: 2. Evaluation

based on inverse model simulations, *J. Geophys. Res.*, 112, D02304–D02304, <https://doi.org/10.1029/2006JD007268>, 2007.

Cerling, T. E., Ehleringer, J. R., and Harris, J. M.: Carbon dioxide starvation, the development of C4 ecosystems, and mammalian evolution, *Philos. Trans. R. Soc. Lond. B. Biol. Sci.*, 353, 159–171, <https://doi.org/10.1098/rstb.1998.0198>, 1998.

Feng, L., Palmer, P. I., Zhu, S., Parker, R. J., and Liu, Y.: Tropical methane emissions explain large fraction of recent changes in global atmospheric methane growth rate, *Nat. Commun.*, 13, 1378, <https://doi.org/10.1038/s41467-022-28989-z>, 2022.

Helfter, C., Gondwe, M., Murray-Hudson, M., Makati, A., Lunt, M. F., Palmer, P. I., and Skiba, U.: Phenology is the dominant control of methane emissions in a tropical non-forested wetland, *Nat. Commun.*, 13, 133, <https://doi.org/10.1038/s41467-021-27786-4>, 2022.

Hmiel, B., Petrenko, V. V., Dyonisius, M. N., Buizert, C., Smith, A. M., Place, P. F., Harth, C., Beaudette, R., Hua, Q., Yang, B., Vimont, I., Michel, S. E., Severinghaus, J. P., Etheridge, D., Bromley, T., Schmitt, J., Faïn, X., Weiss, R. F., and Dlugokencky, E.: Preindustrial <sup>14</sup>CH<sub>4</sub> indicates greater anthropogenic fossil CH<sub>4</sub> emissions, *Nature*, 578, 409–412, <https://doi.org/10.1038/s41586-020-1991-8>, 2020.

Houweling, S., Kaminski, T., Dentener, F., Lelieveld, J., and Heimann, M.: Inverse modeling of methane sources and sinks using the adjoint of a global transport model, *J. Geophys. Res.*, 104, 26137–26160, <https://doi.org/10.1029/1999JD900428>, 1999.

Lan, X., Basu, S., Schwietzke, S., Bruhwiler, L. M. P., Dlugokencky, E. J., Michel, S. E., Sherwood, O. A., Tans, P. P., Thoning, K., Etiope, G., Zhuang, Q., Liu, L., Oh, Y., Miller, J. B., Pétron, G., Vaughn, B. H., and Crippa, M.: Improved Constraints on Global Methane Emissions and Sinks Using  $\delta^{13}\text{C}\text{-CH}_4$ , *Glob. Biogeochem. Cycles*, 35, e2021GB007000, <https://doi.org/10.1029/2021GB007000>, 2021.

Liu, L., Zhuang, Q., Oh, Y., Shurpali, N. J., Kim, S., and Poulter, B.: Uncertainty quantification of global net methane emissions from terrestrial ecosystems using a mechanistically based biogeochemistry model, *J. Geophys. Res. Biogeosciences*, 125, e2019JG005428, <https://doi.org/10.1029/2019JG005428>, 2020.

McNorton, J., Wilson, C., Gloor, M., Parker, R. J., Boesch, H., Feng, W., Hossaini, R., and Chipperfield, M. P.: Attribution of recent increases in atmospheric methane through 3-D inverse modelling, *Atmospheric Chem. Phys.*, 18, 18149–18168, <https://doi.org/10.5194/acp-18-18149-2018>, 2018.

Murguia-Flores, F., Ganesan, A. L., Arndt, S., and Hornibrook, E. R. C.: Global uptake of atmospheric methane by soil from 1900 to 2100, *Glob. Biogeochem. Cycles*, 35, e2020GB006774, <https://doi.org/10.1029/2020GB006774>, 2021.

Ni, X. and Groffman, P. M.: Declines in methane uptake in forest soils, *Proc. Natl. Acad. Sci.*, 115, 8587–8590, <https://doi.org/10.1073/pnas.1807377115>, 2018.

Nicely, J. M., Canty, T. P., Manyin, M., Oman, L. D., Salawitch, R. J., Steenrod, S. D., Strahan, S. E., and Strode, S. A.: Changes in Global Tropospheric OH Expected as a Result of Climate Change Over the Last Several Decades, *J. Geophys. Res. Atmospheres*, 123, 10,774–10,795, <https://doi.org/10.1029/2018JD028388>, 2018.

Oh, Y., Zhuang, Q., Liu, L., Welp, L. R., Lau, M. C. Y., Onstott, T. C., Medvigy, D., Bruhwiler, L., Dlugokencky, E. J., Hugelius, G., D'Imperio, L., and Elberling, B.: Reduced

net methane emissions due to microbial methane oxidation in a warmer Arctic, *Nat. Clim. Change*, 10, 317–321, <https://doi.org/10.1038/s41558-020-0734-z>, 2020.

Rosentreter, J. A., Borges, A. V., Deemer, B. R., Holgerson, M. A., Liu, S., Song, C., Melack, J., Raymond, P. A., Duarte, C. M., Allen, G. H., Olefeldt, D., Poulter, B., Battin, T. I., and Eyre, B. D.: Half of global methane emissions come from highly variable aquatic ecosystem sources, *Nat. Geosci.*, 14, 225–230, <https://doi.org/10.1038/s41561-021-00715-2>, 2021.

Sanderson, M. G.: Biomass of termites and their emissions of methane and carbon dioxide: A global database, *Glob. Biogeochem. Cycles*, 10, 543–557, <https://doi.org/10.1029/96GB01893>, 1996.

Sherwood, O. A., Schwietzke, S., and Lan, X.: Global Inventory of Fossil and Non-fossil  $\delta^{13}\text{C}\text{-CH}_4$  Source Signature Measurements for Improved Atmospheric Modeling, <https://doi.org/10.15138/qn55-e011>, 2021.

Thompson, R. L., Nisbet, E. G., Pisso, I., Stohl, A., Blake, D., Dlugokencky, E. J., Helmig, D., and White, J. W. C.: Variability in atmospheric methane from fossil fuel and microbial sources over the last three decades, *Geophys. Res. Lett.*, 45, 11,499–11,508, <https://doi.org/10.1029/2018GL078127>, 2018.

Zhang, Z., Poulter, B., Knox, S., Stavert, A., McNicol, G., Fluet-Chouinard, E., Feinberg, A., Zhao, Y., Bousquet, P., Canadell, J. G., Ganesan, A., Hugelius, G., Hurtt, G., Jackson, R. B., Patra, P. K., Saunio, M., Höglund-Isaksson, L., Huang, C., Chatterjee, A., and Li, X.: Anthropogenic emission is the main contributor to the rise of atmospheric methane during 1993–2017, *Natl. Sci. Rev.*, 9, <https://doi.org/10.1093/nsr/nwab200>, 2021.

Zimmerman, P. R., Greenberg, J. P., Wandiga, S. O., and P. J. Crutzen: Termites: A potentially large source of atmospheric methane, carbon dioxide, and molecular hydrogen, *Science*, 218, 563–565, <https://doi.org/10.1126/science.218.4572.563>, 1982.