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Comment on acp-2022-317

Anonymous Referee #1

Referee comment on "Estimating emissions of methane consistent with atmospheric measurements of methane and $\delta^{13}C$ of methane" by Sourish Basu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-317-RC3, 2022

This study investigates the use of d13C methane measurements to constrain the global sources and sinks of methane in a new 3D global inverse modelling setup based on TM5-4DVAR. d13C has been used been used in several past studies to investigate the causes of the renewed increase in methane after 2007. This study takes advantage of a 3D atmospheric transport model, whereas several of the previous studies used simplified box models. Doing so, it is concluded that the methane increase is dominated by increasing microbial sources, as opposed to fossil sources which did not change much. The authors acknowledge the many sources of uncertainty in the use of d13C and spend considerable effort quantifying their possible impact, highlighting the need for better constrained atmospheric sinks – notably tropospheric chlorine. Despite these uncertainties, fairly strong conclusions remain concerning the role of microbial versus fossil and the Tropics and high-latitudes, challenging the GCP view on how the methane budget evolved. The manuscript is well written and present a thorough analysis, attempting to take optimal benefit of a highly valuable dataset of d13C measurements. This is all fine, however, I remain concerned about the validity of the conclusions given the uncertainties and choices that were made in the setup, as explained further below. These will need to be carefully addressed to make this manuscript publishable.

GENERAL COMMENTS

As explained in the introduction section, this study is motivated by the large uncertainties in the global methane budget. More specifically, a mismatch of 160 Tg/yr between top down and bottom up in the GCP budgets is mentioned. The procedure for specifying prior uncertainties, however, adds up globally to 7% (40 Tg/yr) which doesn't quite reflect this uncertainty. The difference may be more than a factor 4, as the prior budget uncertainty probably reflects only a single year. The setup of prior fluxes mentions a rescaling to make sure that the prior budget is consistent with the observed trend. In sensitivity tests also alternative rescaling is applied to restore the source-sink balance in the a priori budget. It is unclear why this is needed, given that the measurements used in the inversion contain this information (strictly speaking measurement information is double counted in the setup that is used). My guess is that the division of the full time series into blocks and the initialization of these blocks requires the rescaling. If the prior is over constrained for methodological reasons this raises the question how the choice of rescaling might have

steered the solution in the direction of the main outcomes and how suitable the proposed time splitting methodology is.

The section on future work mentions some of the limitations of the current setup that will need to be improved on. The question is how these limitations may affect the conclusions from this work on the relative importance of changes in microbial and fossil emissions. Here, attention is raised for uncertainties in OH, rightly so I would say. A study of Zhao et al (ACP, 2020) concludes that sink uncertainties are about as important as emission uncertainties. As Lan et al (2021) and others have shown, changes in the sink are unlikely to explain the post 2007 increase. However, this doesn't mean that the sink should have remained constant. There is little consensus on the longer-term OH trend, particularly between the atmospheric chemistry community and those who study methyl-chloroform. Worden et al (2017) tried to quantify the impact of hypothetic OH trends on the microbial / fossil emission trend partitioning. The question is how sensitive the conclusion on microbial versus fossil emission increases is to the possibility of a non-zero OH trend that is within the uncertainty of methyl chloroform. I realize that this is a difficult question to answer, but to draw conclusions on emission scenarios postponing the discussion on OH to future work is too easy in my opinion.

Besides comparisons to the GCP inversions there must be some specific comparison with other methane modelling studies that have used isotopic measurements. Some of those also used 3D models (e.g. Thompson et al, 2018 and McNorton et al, 2018). In the case of McNorton et al (2018), the largest contribution to the emission increase was attributed to the energy sector. Apparently, within the uncertainty of different models and setups it is possible to arrive at different conclusions regarding the contribution of microbial and fossil sources to the CH4 increase. It is important to understand what explains this, which may be difficult, but to compare and acknowledge the different outcomes is easy and should be done.

SPECIFIC COMMENTS

Line 69: Another number should be used for r_std. Footnote 1 is well taken, but the use of the outdated R_PDB=0.0112372 causes confusion in the literature that is better avoided.

Equation 8 and 9: What is the advantage of modelling CH4 and CH4 delta' over modelling CH_4 and $^{13}CH_4$? Both choices provide all the information that is needed to compute model analogues of CH4 and d13C measurements given sources, sinks and atmospheric signatures, right?

Line 108: What time step is used in B?

Table 1: Does "microbial" make a distinction between natural and anthropogenic microbial

emissions? Emissions from the main contributors ruminants and natural wetlands would have rather different uncertainties.

Figure 2b: What motivates the use of this scheme over a single long sequential inversion? (memory / max. run time of a job?)

Line 183: This assumes that the scale conversion error can be represented by a random uncertainty, changing from sample to sample with zero mean. Whereas, in reality this scaling error would be systematic. Then how appropriate is the treatment of this uncertainy?

Line 194: How about model representation errors, particularly in measurements from tower sites?

Line 230: A table would be useful that summarizes the adjustments that are made in the sensitivity tests.

Line 240: This assumes that the relative importance of Cl for the oxidation CH4 and MCF is the same, which is not the case.

Line 259: Why would systematic errors in fractionation factors be non-gaussian?

Line 305: As shown in Houweling et al (2017) what matters more for the initialization than the gradient are the initial atmospheric burdens of CH4 and 13CH4 – which take longer to equilibrate. It is unclear how the burdens differ between the scenarios that are tested.

Figure 5: It is obvious that the CH4-only inversion will not get the isotopic source/sink balance right (it would be a coincidence if it did). But if you would account for mean offsets in this balance (e.g. by a different choice of fractionation factors), then I wonder how well it might reproduce the observed d13C time variation (towards the end of the time record it seems to be getting some of the observed variability right).

Figure 6: It is obvious that the CH4 only inversion is performing worse than the CH4 + d13C inversion. But it is not obvious that the CH4 + d13C inversion is doing better than the prior. Some further quantification of this would be useful to evaluate the performance of the isotope inversion.

Figure 8: The perfect separation between the two sensitivity tests in several of the panels puzzled me initially. The way I understand it now is that the shaded region is the difference between the reference inversion and the sensitivity test. The puzzling plots show impacts of two tests that point systematically in opposite direction of the reference. It would help to add a clarifying sentence in the text.

Line 400: Why "this suggests"? You have all the numbers needed to quantify the contribution of the Tropics to the total, right?

Line 401: The trouble is that the Tropical continents have almost no measurement coverage, especially for d13C. Could this outcome just be explained by larger a priori uncertainties in the Tropics making the flux adjustments the least costly?

Section 4, discussion: To end the paper with this section title suggests that the paper has no conclusions, whereas this section actually contains a numbered list of conclusions. Due to this, the conclusions appear to be somewhat hidden for a reason that is unclear to me. I would suggest to avoid any confusion, particularly among those readers who quickly want to jump to the conclusions, by changing the title into 'Discussion and conclusions'.

TECHNICAL CORRECTIONS

Line 65, This suggest that L refers to a lifetime, whereas instead probably something like "Loss" was meant. The use of "lifetime" is confusing because that would be 1/L. I realize that the sentence can be read differently, but it is better to avoid possible confusion.

Line 116, "eq. (12)" i.o. "(12)"

line 212: 'errors' i.o. 'variations'? 'Variations' is ambiguous in this context in the sense that it could refer to real variations as opposed to errors.