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Reply to anonymous referee 1

Sourish Basu et al.

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-AC3>, 2022

We thank the reviewer for taking the time to carefully read through the manuscript and appreciate the suggestions for improvement. Please find below the reviewer's comments italicized, with our responses in normal font.

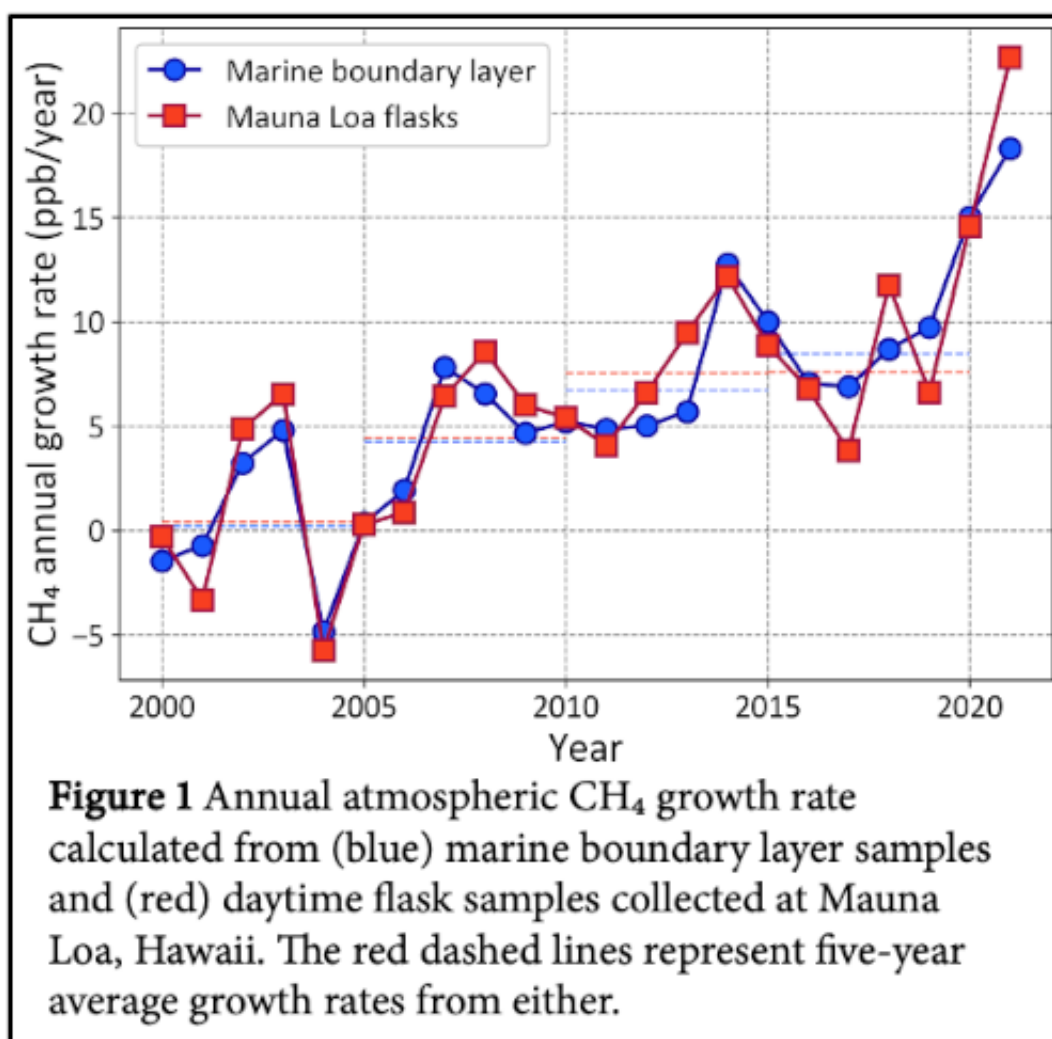
As explained in the introduction section, this study is motivated by the large uncertainties in the global methane budget. More specifically, a mismatch of 160 Tg/yr between top down and bottom up in the GCP budgets is mentioned. The procedure for specifying prior uncertainties, however, adds up globally 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 first step in performing any inversion is a good forward model, which includes a realistic prior flux. Since most bottom-up estimates for long-lived greenhouse gas fluxes do not reflect the atmospheric growth rate, it is standard practice, especially for long model runs, to adjust these fluxes to get the correct decadal atmospheric growth for CO_2 , CH_4 or SF_6 . See, e.g., Houweling et al (2017) for an overview for CH_4 , Chevallier et al (2010) and Weir et al (2021) for implementation for CO_2 , and Patra et al (2011) for implementation for SF_6 . Our approach of starting with priors that match the global growth rate is entirely in line with this philosophy. In fact, we would go even further and argue that starting with a prior that has a known bias in something as basic as the decadal growth rate **violates** one of the basic premises of Bayesian estimation, which is that the prior is unbiased. For short inversions this may result in posterior biases that are negligible. However, for long inversions, starting with a biased prior will usually result in a biased posterior (e.g., Bruhwiler et al (2014)) and an atmospheric tracer field that moves farther away from the truth with time. This is clearly not desirable for an atmospheric inversion.

Regarding the referee's observation that our 1σ prior error of 40 Tg/year is four times

smaller than the gap of 160 Tg/year between top-down and bottom-up GCP estimates, this is correct but not entirely relevant to the setup of an atmospheric inversion. The 160 Tg/year gap is a **bias** (there is no reason to believe that the GCP bottom-up estimates are normally distributed around the truth), and the prior error covariance is not supposed to be a measure of the bias in the prior, nor the spread between a set of bottom-up fluxes. The prior error covariance is supposed to be the covariance of (prior - truth). Since we have scaled the prior to match the decadal growth rate (i.e., mitigated the prior bias), we only need to provide enough "slack" in the prior to encompass the range of flux estimates that may arise from reasonable bottom-up models. We note that with a 1σ error of 40 Tg/year, 95% of the bottom-up flux estimates are supposed to lie within a range of $\pm 2\sigma$ or 160 Tg/year.

Finally, regarding the referee's concern that we are using the same data twice in creating a balanced prior, while theoretically true, is a red herring that makes little difference in practice. Figure 1 shows the annual growth rate of CH₄ derived from NOAA's marine boundary layer (MBL) sites and from daytime flask samples at Mauna Loa (not included in MBL). The differences are very small at the annual scale and ~ 1 ppb or less for five-year averages. In this work, we used MBL-derived growth rates (blue circles) to adjust our prior and did not assimilate Mauna Loa flask CH₄ data. Figure 1 suggests that if, instead, we had just used (non-assimilated) Mauna Loa flask data (red squares) to calculate the growth rate and adjust our prior, it would have made very little difference to the prior and consequently even less difference to the posterior. Technically, this is because we are using up exactly one degree of freedom from the data to adjust the prior - the annual growth rate - leaving many, many more for the inversion to interpret.



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 hypothetical 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.

The reviewer makes a valid point. While a purely OH-driven explanation for the recent growth in CH₄ has been ruled out by Lan et al (2021), smaller variations in the CH₄ budget are entirely possible due to variations in OH. Before getting into the possibilities for OH changes, we would like to make two comments. First, while the global methane emissions depend strongly on the total sink, the split between microbial, fossil and pyrogenic sources based on $\delta^{13}\text{CH}_4$ has a very weak dependence on the total sink (no dependence at steady state). This is because the total sink strength does not factor into the relationship between the emission-weighted source signature and the atmospheric $\delta^{13}\text{CH}_4$ at steady state (Miller, 2004). Second, OH is not the only sink of atmospheric methane. It is the largest sink but has the smallest discrimination between ¹²C and ¹³C. Therefore, a downward trend in OH makes atmospheric CH₄ **heavier** because the other sinks become (relatively) more important. Most studies that try to attribute the recent growth of methane to sink changes (e.g., Rigby et al (2017), Turner et al (2017)) miss this fact because they assume OH is the only sink of methane. In the presence of other sinks (chlorine, oxygen, soil sink), if even part of the upward CH₄ trend were to be explained by a downward trend in OH, it would require even more microbial contribution than what has been reported in our work to match the $\delta^{13}\text{CH}_4$ time series.

There are two kinds of possible variations in OH, trend and interannual variability with no long-term trend. Not only would a decreasing OH trend be insufficient to explain the recent trends in CH₄ and $\delta^{13}\text{CH}_4$ for reasons detailed above, neither is there any mechanistic explanation for why such a trend should exist for the past fifteen years (Nicely et al., 2018; Anderson et al., 2021). In fact, oxidation of CH₄ by OH may have increased in recent times due to the reduction in CO emissions (Gaubert et al., 2017). On the other hand, interannual variability in OH is a very real possibility that is simulated by most atmospheric chemistry models. We have hesitated to use OH directly from an atmospheric chemistry model because their simulation of the decay of methyl chloroform or the north-south gradient do not match constraints from observations (Montzka et al., 2011; Patra et al., 2014). However, in the near future we plan to impose OH **variations** calculated by several different atmospheric chemistry models to see what difference that makes on our methane emission estimates.

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.

Thompson et al (2018) use a 2D model without zonal structure, and not a 3D model. However, we take the reviewer's point about comparison with other top-down studies that use CH₄ and δ¹³CH₄ data. In the revised manuscript, we have added sections to compare our results to Thompson et al (2018), McNorton et al (2018) and Zhang et al (2021). The most significant difference we find between our work and these three previous studies is that unlike us, all three studies estimate a significant downward trend in pyrogenic emissions. Since pyrogenic emissions are the heaviest of the three categories, their reduction necessitates a larger increase in fossil emissions to reproduce the δ¹³CH₄ trend. We note that our prior pyrogenic emissions from GFED 4.1s do have a negative trend (van der Werf et al., 2017), and therefore our smaller posterior trend must be driven by atmospheric data. It is possible that the larger negative trend in pyrogenic emissions seen by these three studies were driven more by their assumed prior emissions than atmospheric data. This is supported by the "INV_FIXED" sensitivity test of McNorton et al (2018), where they do not impose a prior trend on pyrogenic emissions and consequently see no trend in the posterior.

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.

As we note in that footnote (and the reviewer seems to agree), the exact value of r_{std} affects neither our formulation nor our conclusions. However, since we have used the long-standing value 0.0112372 from literature (Craig, 1957), it would be incorrect of us to modify the text and simply substitute that value with another value (which one?) from more recent literature. Furthermore, even though the meat of our conclusions would not change, since there are several places where conversion between δ's and tracer fluxes are performed, some of the exact numbers in our figures and tables might. Adopting a new value of r_{std} consistently would entail reprocessing our observations and prior fluxes and redoing every single one of our model runs. We think that this is too much of an ask at this stage, and therefore respectfully decline the reviewer's recommendation.

Equation 8 and 9: What is the advantage of modelling CH4 and CH4 delta' over modelling CH4 and 13CH4? 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?

The most straight-forward set up of an atmospheric inversion has the form $d(\text{conc})/dt = \text{fluxes} - \text{loss}$. This lets us assimilate measurements of 'conc' and estimate 'fluxes'. If we set up the problem where 'conc' is either CH₄ or ¹³CH₄, then the second mass balance equation would lead us to estimate fluxes of ¹³CH₄. This is one step removed from what we want, which are fluxes of CH₄. It is true that at each iteration, we could calculate the adjoint emissions of ¹³CH₄, then transform them into adjoint emissions of CH₄ through partial derivatives and assumed surface maps of the isotope signatures. However, that is one additional transformation we would have to build into the variational loop. Formulating our mass-balance the way we have (equations 8 and 9) lets δ¹³CH₄ measurements directly influence and optimize CH₄ surface fluxes without that additional transformation.

Line 108: What time step is used in B?

The time resolution at which fluxes are optimized, i.e., monthly.

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.

It does not. We are aware that this is a limitation of our current work, and we have mentioned this at several places in the text. In principle, CH₄ emissions from agriculture, waste and wetlands can have small differences in their $\delta^{13}\text{C}$ signatures that vary geographically. In practice, neither our source signature maps nor our $\delta^{13}\text{CH}_4$ measurements are extensive or detailed enough to reflect or utilize such differences.

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

The limitation is indeed the maximum allowed run time of a job on the clusters we had access to. In a 1997–2017 inversion performed as a single run, we would need to perform 21-year TM5 forward and adjoint runs, either of which would exceed the 8 hours allowed per job on our cluster.

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 uncertainty?

The reviewer is right that a different scale means a systematic difference in measurements. However, we use the scale conversion factor to correct for that, and what is left, i.e., the “uncorrected” error, is assumed to be mostly random. We have accounted for the uncertainty in the scale conversion factor in the total measurement uncertainty. Random uncertainty also exists during scale propagation for labs on the same scale (the absolute scale uncertainty is typically larger, but if all measurements are on the same scale, the absolute scale uncertainty is not a problem). We estimate this uncertainty using the reproducibility of standards, i.e., changes in measurements of the same standards over a long period of time. The scale propagation uncertainty is also included in the total measurement uncertainty.

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

We do include a model representation error, calculated for each measurement, based on modeled tracer gradients. This is mentioned on line 107 of the original manuscript with Meirink et al (2008) as the reference. Note that this model representation error, being specific to TM5, is not included in the released dataset.

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

We agree that there are quite a lot of sensitivity tests. However, since the tests are exploring very different axes (e.g., source signatures, initial fields, atmospheric chemistry, wetland inundation, to name some), we could not come up with way to summarize them in a table efficiently. We have therefore taken the current approach of grouping them by motivation and listing them as separate subsections.

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

Not really. For each instance of tropospheric chlorine, we came up with an OH scale by repeatedly running TM5 forward with that tropospheric chlorine and different scalings of OH, until (iteratively) we arrived at a scale that gave the same CH₄ lifetime in our model. No equivalence between CH₄ and MCF was assumed.

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

We are not sure if the reviewer has put down the appropriate line number for this comment. In the original manuscript, line 259 is under §2.6.3, which discusses systematic errors due to source signatures, while errors in fractionation factors are covered in §2.6.2. Since the term “non-Gaussian” occurs in the original manuscript in §2.6.3, we will assume that they are talking about source signatures and not fractionation factors.

In a Gaussian distribution, values that are equidistant from the mean on either side are equally likely. However, if the assumed source signature of a region in our inversion is (say) -45‰ , and a second source signature map reports -50‰ for that region, we cannot simulate that with a Gaussian perturbation on the first map. Any Gaussian perturbation around a mean of -45‰ will yield -50‰ and -40‰ with equal likelihood. That is, we cannot design a covariance matrix that, under random Gaussian perturbations around -45‰ , will yield -50‰ more often than -40‰ . This is what we mean by “non-Gaussian”, and this why we have not explored source signature errors in our Monte Carlo ensembles.

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.

This is true, and was pointed out earlier by Tans (1997). For each of the scenarios that would result in different atmospheric CH₄ and ¹³CH₄ burdens, we did a long forward run from 1984 to equilibrate the atmospheric burdens by 2000.

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 $\delta^{13}\text{C}$ time variation (towards the end of the time record it seems to be getting some of the observed variability right).

This is possible in principle. The thing to note here is that even in the CH₄-only inversion, the initial field and prior fluxes are constructed to match the atmospheric CH₄ and $\delta^{13}\text{C}$ trends, yet in the absence of $\delta^{13}\text{C}$ data they progressively stray away from observed $\delta^{13}\text{C}$. We agree with the reviewer that if they did not, that would be a coincidence.

Figure 6: It is obvious that the CH₄ only inversion is performing worse than the CH₄ + $\delta^{13}\text{C}$ inversion. But it is not obvious that the CH₄ + $\delta^{13}\text{C}$ inversion is doing better than the prior. Some further quantification of this would be useful to evaluate the performance of the isotope inversion.

The reviewer is correct that the prior seems to do as well as the joint inversion in Figure 6, while the CH₄-only inversion always does worse. In fact, if we calculate the mean mismatch in $\delta^{13}\text{C}$, only for the HIPPO comparisons does the joint inversion do significantly better than the prior. The table below shows the mean difference between modeled and observed $\delta^{13}\text{C}$ values from the three campaigns. As in Figure 6, the main point is that the CH₄-only inversion does significantly worse and is therefore likely to yield incorrect emissions.

Model run	CONTRAIL	HIPPO	<u>ATom</u>
Prior	0.48 ‰	0.46 ‰	0.24 ‰
CH ₄ only	-9.56 ‰	-2.18 ‰	-1.41 ‰
CH ₄ + $\delta^{13}\text{C}$	0.76 ‰	0.31 ‰	0.23 ‰

We think the prior is doing well in this comparison because the prior fluxes, scenario

"C_WL+" of Lan et al (2021), were constructed to match the background CH₄ and δ¹³CH₄ fields quite well. They did not match the observed high-latitude seasonality in δ¹³CH₄, but the aircraft data shown in Figure 6 do not have the samples to pick that up.

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.

A stronger OH fractionation makes the atmosphere heavier, requiring a larger (smaller) fraction of microbial (fossil) to match the same observations. Since chlorine has the strongest discrimination among all sinks, a smaller chlorine sink makes the atmosphere lighter, requiring a smaller (larger) fraction of microbial (fossil) to match the same observations. Therefore, a stronger OH fractionation and a smaller chlorine sink affect the fossil/microbial partitioning in opposite ways, resulting in the structure seen in Figure 8. This is now explained in the figure caption.

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

Correct. We were just being cautious, as in "suggests" instead of "proves", because our inversion is not the last word on the methane budget. We have changed the phrasing to "Therefore, the largest contribution to the global increase in microbial emissions between the two periods comes from the Tropics."

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?

We would of course love more δ¹³CH₄ measurements (also more CH₄ measurements) in the Tropics. However, the point here is that we only get this majority attribution to the Tropics if we use δ¹³CH₄ data. If it were just a function of the prior flux uncertainty, we would also see this attribution in a CH₄-only inversion.

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'.

This suggestion is well taken. We have completely reworked this section, calling it "Conclusions and discussion", with a subsection called "Enumerated conclusions". Comparisons with other published papers and future directions are other subsections of this section now.

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.

We called it "inverse lifetime". However, we agree that this can be confusing, so in the revision we say "can be denoted as a loss rate or inverse lifetime".

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

Corrected.

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.

Fair point. Changed "variations" to "errors".

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