Response to Editor's Comments

Joël Thanwerdas[1,a*], Marielle Saunois[1], Antoine Berchet[1], Isabelle Pison[1], and Philippe Bousquet[1]

[1]Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ, IPSL, Gif-sur-Yvette, France.

[a]now at: Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland.

Again, we thank the two referees and the editor for reading attentively our paper and making comments that allow us to improve it. We provide here a response to the comments received. Comments are in red. For each comment, an answer is provided in normal text, *citations from the text are in italic* and **the modifications from the new version of the manuscript are provided in bold and small text**. Note that modifications have been included only when deemed substantial enough. Also, the incorrect sentence spotted by Referee #1 has been removed, it was a proofreading mistake from last revision.

Attached to this response, we also provide the new version of the manuscript and a track-changes document.

1) I understand that a mathematically rigorous uncertainty analysis is computationally expensive, but even so, some uncertainty estimate is required. I don't expect every number in the paper to have an uncertainty, but at the least the values in the abstract and conclusions (and the values they originate from) should come with some "educated guess" of how reliable they might be. As an example, the 51 %:49 % split between fossil and agriculture/waste source suggests a level of uncertainty of the order of 1 %. Is that realistic? Or could it be 60 %:40 %? Or 40 %:60 %? Similarly, for the relative increases stated further below. To be clear, the "educated guess" should also appear in the abstract itself.

An estimate of the uncertainty calculated using the sensitivity tests performed for this study has been added to the abstract, the main text and the conclusion. Here is the sentence added to the abstract:

Additionally, some other sensitivity tests have been performed. While prescribed OH inter-annual variability can have a large impact on the results, assimilating δ(D, CH_4) observations in addition to the other constraints have a minor influence. Using all the information derived from these tests, the net increase in emissions is still primarily attributed to fossil sources (50 ± 3 %) and agriculture and waste sources (47 ± 5 %).

2) The potential value of deuterium isotope ratio measurements as constraint on sink magnitudes and sink partitioning (between OH and Cl) should be discussed (see comment from referee #2).

A discussion has been added at the end of Section 3.7.

As including δ(D, CH_4) in the inversion doubles the computational cost compared to a setup like INV_REF, we recommend not assimilating δ(D, CH_4) in our system until either the computational cost can be reduced, more observations become available or lower uncertainties are established. However, a hypothetical network of δ(D, CH_4) measurements, obtained at a reasonable frequency and spanning a longer period of time could efficiently complement δ(^{13}C, CH_4) observations and provide a wealth of information (Rigby et al., 2012). More specifically, reactions with OH, O1D and Cl have fractionation coefficients that depend on the isotope. Therefore, incorporating δ(D, CH_4) constraints might help to disentangle the effects of the associated sinks and provide additional insights into the global sink and its mixture. However, optimizing the sinks introduces additional degrees of freedom and complexifies the inverse problem. With the current system and at such a high resolution for the optimized variables, we recommend against the simultaneous optimization of both the source signatures and the sinks. However, a coarser resolution for the optimized variables, or at least for the sink, might be able to accommodate a simultaneous optimization.

3) The compensating effects of variations in source magnitude and isotopic signature should be discussed (see comment from referee #2).

A discussion has been added at the end of Section 3.4.

Figure B4 and Figure B5, in Appendix B, show the full temporal variations for prior and posterior source signatures. For FF, high variations indicate a change in activities associated to fossil fuel extraction, e.g. switching from one location with a specific signature to another, transitioning from one fuel type (oil, gas, coal) to another or a combination of both. For example, the substantial shift around 2009 in the United States was caused by a large increase in emissions from the extraction of natural gas. As we chose not to prescribe temporal error correlations between different years, the system is free to optimize each year independently to better fit δ(^{13}C,CH_4) observations. For certain continental regions, such as Africa, Temperate Asia or South Asia, the inter-annual variability of the source signature adjustments is large and rather unrealistic, especially when compared to the emission adjustments in the same regions (see Fig. 7). It is unlikely that these changes occurred without detectable changes in emissions in the same areas, especially considering Temperate Asia, which exhibits larger emissions from FF than in the United States. These results suggest the need for prescribing yearly temporal error correlations to dampen this artificial inter-annual variability. However, the example from the United States also indicate that large changes can occur and it is reasonable to assume, considering the lack of isotopic data, that such changes might go unnoticed by the prior data for other regions. Therefore, while implementing stronger temporal correlations could be a solution to mitigate unrealistic inter-annual variability for this category, it diminishes the likelihood of detecting potential substantial changes that remain undetected by the prior data. Nevertheless, it might be sufficient to reduce the prescribed uncertainties in the source signatures in order to balance out the pressure applied by the system on the emissions and the source signatures. Overall, the same reasoning applies to AGW, although there is no evidence in the prior data that AGW source signatures can change as rapidly as FF. Due to the scarcity of existing data on the temporal variability of source signatures, designing a data-driven methodology to estimate potential temporal correlations, especially at the regional scale, remains highly challenging. Investigating the correlations that the system creates between the uncertainties associated to source signatures and fluxes could offer a promising avenue for extending the analysis. Due to the high computational cost of an inversion performed with our inversion system, it is impossible to derive robust posterior uncertainties. This impossibility is a major drawback and additional studies with this system cannot be performed in the future without tackling this issue.

4) Data sets cannot just be available on request. Please make all relevant data available in a suitable permanent repository (i.e., the input emissions and the other relevant data from this study.)

As requested, a permanent repository with a DOI (10.5281/zenodo.10390430) has been created to store all the relevant data. Two datasets obtained from the authors of Wang et al. (2021) and Saunois et al. (2020) were not stored on permanent repositories. After contacting them, these authors agree to store their data on our permanent repository, provided that the origin of this data is properly acknowledged. For some reason, the modification of the data availability paragraph does not appear in the track-changes document.

5) In section 3.2 and the caption of Fig. 5, please replace XCH4 with column-average CH4 amount fraction.

- In Eqs. 1 and 2 and the line below, replace square brackets with X(12CH4) etc. (with X in italics)
- Replace the y-axis label of Fig. 4 d with X(CH4)/(nmol mol–1)
- l. 164: Write "atmospheric X(CH4)" before the delta symbols
- In section 2.5.2, define $\bar{X}$(CH4) or <X(CH4)> as column-average amount fraction, but explain in the text that it is "often referred to with the symbol XCH4", and use either $\bar{X}$(CH4), <X(CH4)> or XCH4 in the remaining text.

- Change the title of section 3.2 to "Comparison of model-optimized with satellite-derived column average CH4 amount fractions”

All these comments have been taken into account and modifications have been made. Note that the change from XCH_4 to $\bar{X}(CH_4)$ does not appear in the track-changes manuscript because of the Latex command.

6) The differences in the linear regressions applied to different periods of time in Fig. B1 do not appear to be statistically significant. What are the uncertainties associated with the slopes? These should be added to section 3.1 and the legend of Fig. B1, e.g., (-0.23±0.25) ‰

We agree that this had to be done. We apologize for not providing it in the last revision. We added the uncertainty (standard error) associated to each slope of the regression lines, both in Fig. B1 and in the text to show that these trends are significant.

7) Please cite the final version of Wang et al.’s paper (https://acp.copernicus.org/articles/21/13973/2021/), not the Discussion document.

We apologize for this mistake. We changed the citation.

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