Reply on RC1
Malika Menoud et al.

Author comment on "New contributions of measurements in Europe to the global inventory of the stable isotopic composition of methane" by Malika Menoud et al., Earth Syst. Sci. Data Discuss., https://doi.org/10.5194/essd-2022-30-AC1, 2022

RC1: General comments

An important number of new measurements are reported to help inform a global methane isotope ratio database. These measurements were made as part of a European project on methane, accruing a large number of measurements over the project’s period. Interesting and significant findings are explained regarding the variability of isotope ratios over the region and within source categories. These findings are then put in the context of the global measurements made and reported across the literature.

The continuing improvement of our understanding of isotopic source signatures for methane isotope ratios is vital if atmospheric measurements are to make accurate conclusions about the global methane cycle or about regional emissions estimates.

There is obviously significant overlap with the work of Sherwood et al. (2016&2021). This is not necessarily an issue as a significant new number of measurements have been reported here. However, how will the process of improving a ‘definitive’ source signature database evolve from here? Sherwood et al., 2017 provided a very detailed set of averages (Table 5 in their paper), however, this sort of detailed breakdown has not been made in this paper. Table 3 provides one mean value for fossil (does it also include oil?) but it is not clear how exactly this was calculated – it is an important number and surprisingly small uncertainties are reported. Table 3 is important but incomplete and needs thorough revision in relation to how averages are reported from this study (more on this in specific comments below).

The paper would benefit from some restructuring, significant clarification in places, and some further and transparent quantitative analysis.

A: Thank you for your feedback.

We agree that the description of the quantitative analysis could be improved, including the information on how we processed the data. In the revised version we have explained it in more detail in section 2.2.1. Section 2.2.1 was also adjusted by adding a Table that lists the variables reported in our database.
**RC1: Specific comments**

The paper would benefit from clearer structure and also clearer language in places. Section 2.2.2 might be better placed in 2.1.3?

A: Thank you for the suggestion. We have changed the order of these paragraphs. We have also changes the titles of the paragraphs in section 3 (previously section 2.3).

RC1: Is section 2.2’s ‘Update of the global database’ a correct section title? A ‘global database’ suggests a single point of data collection, however, this is not the case here? It appears that this is a separate database to the Sherwood et al. 2017/2021. Perhaps a better section title is ‘Additional measurements contributing to the global isotope ratio dataset’. 2.2.1 could then be ‘Structure of this database to include previous and new measurements’. Then 2.2.3 ‘Additional data sources from previous published literature’. These are suggestions to help clarify the work.

A: We agree with this suggestion and have adjusted the title of section 2.2. We have also adjusted the titles of sections 2.2.1 and 2.2.2 to more accurate statements.

RC1: Section 2.3 ‘results and discussion’ is six lines long. Is there an error in the section structure? Section 2.4 is ‘The European Methane Isotope Database’ but section 2.2.1 is ‘Structure of the database’ Is this the same database? I think so but the understanding of the work would benefit significantly from improved paper structuring.

A: There was indeed a mistake in the numbering of the paragraphs, that we have corrected now. The titles have been changed to make it clearer for the reader.

RC1: Table 3 – why is Sherwood not included here? For coal Sherwood calculate a mean of -49.5 per mil published in 2017 (see their Table 5). The value in this paper in table 3 is -50.7 per mil. What has shifted the methane slightly lighter in this new database? Given the similarity in the goals of these papers it is important that the reader can make direct comparisons between them and intuitively understand the reasons for discrepancies/changes and whether they are significant or not. How have the new measurements from this specific work shifted the global averages from Sherwood et al.? A more complete analysis of these averages and a discussion of why they have shifted would be very beneficial. Currently there is very little arithmetic behind what has actually changed between the two databases, and the significance.

A: Thank you for your suggestions. A more detailed comparison with the values from Sherwood et al. is now included (Table 4 and paragraph 3.2.2) and indeed it makes our study much more pertinent.

RC1: At least one of the citations in Table 3 is not relevant – I don’t think Rigby et al made any conclusions on the global methane cycle, only using a value of -40 in the demonstration of the potential usefulness of isotope ratios. There is no reference to later Rigby and Turner papers here that both use 13C/12C in their global box model analyses.
What about McNorton et al. 2018 etc etc? There are many other papers. It would be an interesting and very useful table if fully complete. As it stands it is highly selective of literature sources and out of date.

A: The original goal here was to provide a quick overview of the changes in the signatures that were assigned to the fossil fuel category, regardless of the purpose of the modelling in each study. We also wanted to show that it evolves through time, from the first global models using stable isotopes. Reacting to this referee comment we have included more references now (see Lassey et al. 2000, Neef et al. 2010, Gosh et al. 2015, McNorton et al. 2018), in an attempt to cover more of the relevant literature, even if the values that were used were the same.

RC1: Table 1 – it would be good to understand the difference between carbon and hydrogen isotopes in these quantities.

A: We included the numbers of hydrogen signatures in the table.

**RC1: Technical corrections**

Below are some corrections and clarifications that are needed. This is likely not comprehensive and the authors are encouraged to improve the general readability of the manuscript.

RC1: Line 1: Rather say ‘Carbon and hydrogen isotope ratio (δ13C and δ2H) measurements...’

A: This has been added.

RC1: Lines 8-9 are confusing: Previous studies are not additional literature?

A: We have rephrased the sentence.

RC1: Line 9: fossil fuel, not fossil fuels

A: This has been corrected.

RC1: Line 13: abbreviate methane as before

A: This has been corrected.

RC1: Line 17: Worth abbreviating GHGs?
A: We think it is not necessary because we use this expression only twice. It would affect the readability to introduce another abbreviation.

RC1: Line 29: ‘Statistical indicators’ is not a common phrase – is this correct?
A: We've replaced "indicators“ with "methods”.

RC1: Line 31: Not fully compatible? Use as simple language as possible – ‘not in full agreement’?
A: This has been changed.

RC1: Line 36: Not a complete list. State they are selected/example publications if not citing full list.
A: We added “e.g.” before the reference list.

RC1: Line 64: Updated version? Rather ‘latest version’?
A: This has been changed.

RC1: Line 69 ‘eight’
A: This has been corrected.

RC1: Lines 78+: state what each of the CRDS analysers is used to measure. Were all these analysers used in a single mobile setup? Why?
A: Samples were taken on several mobile surveys with different and varying setups, that included one or two analysers. We have clarified this in the text (l.87-96).

RC1: Lines 104-105: Fig A1 doesn’t illustrate different sampling procedures. I don’t think there is any value in this figure, it is not scientific and doesn’t provide useful information. Do any of the techniques used in the study need to be better explained. Are there any sampling schemes that could explain lines 78-85, for example?
A: We have decided to remove Fig A1, as it does not provide useful information.

RC1: Line 113 ‘beetween’ = ‘between’
A: This has been corrected.

RC1: Line 115 ‘available’.
A: This has been corrected.

RC1: Lines 138-140: These two sentences don’t make sense
A: We have rephrased these sentences.

RC1: Lines 142: Literature database? This is slightly confusing. Isn’t it the other way round? The literature values are incorporated into your ‘European Methane Isotope Database’. This is where the use of database terms gets a bit confusing. Please clarify in the text throughout and be consistent. As I understand this the European Methane Isotope Database actually includes a global inventory of isotope signatures. Is it worth abbreviating this new database and then referring to it throughout as that?
A: We have changed the title to make this less confusing. We have followed your suggestion of abbreviating the term ‘European Methane Isotope Database’ into ‘EMID’, and we now refer to it as such throughout the text.

RC1: Line 148 – do you mean ‘we extended the pyrogenic...’?
A: No, the category was “biomass burning” and we extended it to include all pyrogenic sources. The sentence was rephrased to clarify this.

RC1: Line 154: I don’t think per mille is strictly a unit. You could say ‘Values are published in per mille’
A: This has been rephrased.

RC1: Line 156+, Section 2.2.3 – It might be good to cite in this main text all the additional literature on top of that used by Sherwood et al. or state how many additional sources there were.
A: We have included the number of additional literature sources in section 2.2.2, and listed them in a footnote.

RC1: Line 173: delete ‘necessarily’
A: This has been done.
RC1: Line 190: prefer ‘..generally have enriched δ13C values than..’
A: This has been changed.

RC1: Line 208: Fig 5a (not 5A)
A: This has been corrected.

RC1: Line 219: ‘number of measurements’ not ‘amount’
A: This has been corrected.

RC1: Lines 297+: This paragraph is confusing and it is not clear the message that is trying to be communicated. Applying appropriate weighting arithmetic is essential for what? This study provides further evidence for the values and uncertainties that are needed on source signatures if these measurements are to be used properly in to-down studies. I think that should be the main takeaway.

A: Applying appropriate weighting arithmetic is essential “for deriving a representative concept of the isotopic composition of CH4 sources and the associated uncertainty”. We have rephrased this paragraph, the last paragraph of the conclusion and the abstract to include your suggestion as the main takeaway.

RC1: Line 318: Suggestion: ‘The present database can be used in CH4 source attribution studies at local and regional scales, and to derive global source signatures for input to global methane cycle modelling studies’
A: Thank you for the suggestion. We have implemented it.

RC1: Line 530: ‘CH4 Sotopic’ = isotopic
A: This has been corrected.