Earth Syst. Sci. Data Discuss., referee comment RC1
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Comment on essd-2022-30

Anonymous Referee #1

Referee 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-RC1, 2022

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

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?

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.

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.

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.

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.

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

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.

Line 1: Rather say 'Carbon and hydrogen isotope ratio ($\delta^{13}C$ and $\delta^{2}H$) measurements…'

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

Line 9: fossil fuel, not fossil fuels

Line 13: abbreviate methane as before

Line 17: Worth abbreviating GHGs?

Line 29: 'Statistical indicators' is not a common phrase – is this correct?

Line 31: Not fully compatible? Use as simple language as possible – 'not in full agreement'?

Line 36: Not a complete list. State they are selected/example publications if not citing full list.

Line 64: Updated version? Rather 'latest version'?

Line 69 'eight'

Lines 78+: state what each of the CRDS analysers is used to measure. Were all these analysers used in a single mobile setup? Why?

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?

Line 113 ‘beetween’ = ‘between’

Line 115 ‘available’.

Lines 138-140: These two sentences don’t make sense

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?

Line 148 – do you mean ‘ we extended the pyrogenic…’?

Line 154: I don’t think per mille is strictly a unit. You could say ‘Values are published in per mille’

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.

Line 173: delete ‘necessarily’

Line 190: prefer ‘..generally have enriched $\delta^{13}C$ values than..’

Line 208: Fig 5a (not 5A)

Line 219: ‘number of measurements’ not ‘amount’

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.

Line 318: Suggestion: 'The present database can be used in CH_4 source attribution studies at local and regional scales, and to derive global source signatures for input to global methane cycle modelling studies'

Line 530: 'CH_4 Sotopic' = isotopic