

General comments

The article presents the data of methane isotopic air samples collected in Europe, their analysis, and discussions in respect to previous studies of global inventories/data. The isotopic information for various sources in methane are useful in understanding the methane budgets better. Atmospheric inverse models (top-down methods) would especially benefit from such information – not only closing the total budgets based on total methane concentration data, but also allow to separately estimate emission magnitude of different source sectors. As authors point out, the previous studies on methane isotope measurements were mainly based on samples from US. The additional data from Europe therefore increases geographical representation, and is highly valuable. The paper presents important information and data for carbon cycle community, and is worth publishing. I also appreciate the authors for making the data open access.

I have a few suggestions below to improve the presentation of the manuscript that would possibly increase the value of the paper.

- Please consider rephrasing the title. To me, updating global inventory is a by-product, and the European data collected/presented here is the most important part.
- Please consider focusing more on Europe. You could, for example, add information and discussion about European data from previous studies, i.e. validation and updating information on Europe. It is unclear from the current manuscript how much information/data were available in Europe previously, and what were the isotopic values. You can also add comparison of the European data to global data, i.e. discussion on geographical differences/similarities of isotopic composition. In addition, geographical bias still remains within Europe. Please comment on it – what should we still improve in Europe? Where would be critical locations, and from which sources that we should sample data from, and why?
- Please consider focusing on new information brought by newly sampled and collected data. A few suggestions/comments on this are found under Specific comments below.
- Please add temporal information about the collected data in MEMO² and those compared (collected from previous studies). If those are from very different period, the differences in isotopic composition may indicate temporal changes in underlying processes. Please comment on such if any.
- Modern microbial section: in many cases, CH₄ emissions from natural sources (e.g. peatlands) are separately estimated from anthropogenic sources (e.g. waste, agriculture). It would be more helpful if you could analyse those separately. Could you separate those e.g. when comparing to previous studies (incl. Section 2.5)?

Specific comments

Please check language in general. Sometimes informal structures/phrases are used.

Please check the journal criteria for citations. There are a few “in review” papers that are cited several times, but the manuscript/preprint are not available. Note that those should not be included in the final version of this manuscript. Please acknowledge that some comments maybe senseless because of this, and I apologies for that. In addition, some are cited within a page, and some at the end of the manuscript. I think all should be given at the end of the manuscript.

P1 L2: “measurements” → “isotopic measurements”

P2 L44: What do you refer to by “them”?

L3 L60-61: “numerous CH₄ sources could be sampled for isotopic measurements”

I think what you sample is air, and not the sources. Please revise the sentence.

Method:

In the excel file, I see that in some locations, only ¹³C or H isotopes are sampled/analysed (for MEMO² data). Why both were not sampled at all locations? Could you add information about selection criteria or sort if there was such?

Section 2.1.1

- Please consider adding a table about different sampling methods, and give focus to the differences/similarities in those methods in the text. This way, you could reduce amount of details and avoid duplicates in the text.
- Was there any location where different sampling methods were applied at the same place? What kind of differences would occur in the measured values due to the differences in sampling methods?

P5 L116-119:

I am now confused. In the beginning of this section (2.1.2), it is written that “the mass spectrometry measurements were performed at **two** laboratories”. However, is it so that actually four labs analysed the air samples? You have included detailed comparison and measurement precision on IRMS, but how about CRDS compared to IRMS? Was there any differences between UHEI and LSCE measurements?

P5 L133: “we did not work towards a uniform procedure”

Why? Did you allow e.g. each lab to calculate based on their choice of method? All methods being “statistically **valid**” does not mean that there are no differences. So I do not see why this is a valid statement to use different procedures.

P6 L139: “our objective concern only values ... of emitted CH₄”

Do you mean that the forests, where samples are taken from, also emit CH₄? Most of forests are net sink of CH₄, but in some cases they emit CH₄. Sorry for this picky comment, but I am simply interested in. Could you provide the references where those samples were taken from?

P6 L141-143:

I do not see country/region in Table 1. What is the “region” in those sentences?

P8 L164: (2017-2020)

Is this the project period or actual period when data was collected? As in the earlier comments, it would be informative to state clearly the sampling period.

P8 L165-167: “The first version was made accessible on October 1st 2020”

Now that it is updated, is this information needed?

P8 L166: “The European data was used in several publications”

This gives an impression that the data has been published already. Do you mean the first version of the data was used in those publications? If so, why did you need to update? Please also consider moving this to Introduction by add a bit more details about those studies, e.g. how and what the data (e.g. which isotopologue/country) were used, and what are the main findings. This would then show the importance of the European dataset and what new information it can bring.

P9 L183-187:

What were then the isotopic signature values for those coal reservoirs where natural gas of microbial origin are present? You mention “a relatively enriched $\delta^2\text{H}$ (>-250 ‰), and relatively depleted $\delta^{13}\text{C}$ (<-60 ‰)”, but are those values those measured in this study or from Milkov and Etiope (2018)?

P10 L190-202:

Similar to the comment above. What is the value found in this dataset? You mention that “microbial fermentation range specified in previous reviews”, but how were the waste-related source signatures compared to fermentation range measured in this study? What are the mean and range of signature values in sewage treatment plants and biogas plants? How are those values compared to those in other types of plants?

P10 L207-P11 L208: “This distinction is also visible in the histograms of the European Methane Isotope Database in Fig. 5.A”

- “Fig. 5.A.” → Fig. 5(a)”
- From the figure, those from Poland and Romania have two peaks. Does this mean that within Poland and Romania, there are different types of plants, one similar to those in the UK/Netherlands and another that is microbial origin?

P10 L208-209: “In western Europe, $\delta^{13}\text{C}$ allows for a good separation between microbial and fossil fuel sources”

Is it true? I see a quite much overlap still.

P11 L216-217: “increase in number of measurements”

I do not see from the figure how much data is increased, but only the total. Is the Figure caption or text wrong?

Section 2.5:

Much of the text seems to be more suitable as Introduction. It is unclear what is the results of this study in addition to Sherwood et al (2021). Did representativeness increased by additional data found in this study?

Section 2.6:

There are a lot of discussion where newly sampled and collected data are compared to Sherwood et al. (2017, 2021) values. However, I feel that it is difficult to comprehend the differences from the current figures and tables. Could you consider adding figures corresponding to e.g. Fig. 4 and 5, but showing e.g. differences to the previous study? Such figures could be in the supplementary, but would be helpful for readers who does not remember all the details in Sherwood et al. (2017, 2021).

P13 L255: “mainly following the fermentation pathway”

Fermentation pathway applies to agricultural sources, but not for e.g. wetlands. Please revise the sentence.

P13 L256: “They show a normal distribution”

Have you checked whether they really are normally distributed?

P13 L256-258:

I see quite much overlap in waste sector to, e.g. agriculture, also in MEMO² data.

P13 L271-273:

I am not sure what you wish to emphasise here. Why $\delta^{13}\text{C}\text{-CH}_4$ from fossil fuel burning being “more variable than biomass burning” lead to “smoother” distribution?

P13 L273-276 (and P13 L264-267):

I feel this relation and need of additional data on $\delta^2\text{H}\text{-H}_2\text{O}$ is more suitable to be mentioned in Conclusions.

P14 L280: “weighted average”

What is the weight used? If it is emission weighted, did you also measure emissions at the same locations?

P15 L297-301:

This is more suitable to be mentioned in Conclusions.

Conclusions: Now that the MEMO² project has ended (if I understood correctly), is there a plan to continue activities on isotopic measurements?

Figures and tables

Figure 2: The literature values seem to be illustrated by boxplot-type. Are those squares mean/median? What does the bar length illustrate? The letters presenting the types of shaded areas (e.g MF) can be bigger, and the letter “T” is better to be straight. I cannot see clearly the area “A”, but is it a white space on the right side?

Table 3:

- Please add/separate global to European means.
- What is “sem”?

Please consider adding A2 in the main text.

Technical comments

P2 L17: “the earth’s” → “the Earth’s”

P5 L115: “deliverable report publically availabe” → “deliverable report, and publically available”