In this paper, Hoheisel et Schmidt describe new continuous CH4 and δ(13C,CH4) measurements retrieved between 2014 and 2020 In Heidelberg (Germany). After introducing the experimental setup, they analyze the temporal variability of this data and apply the Miller-Tans method to derive estimates of the mean isotopic signature that could cause these variations. These determined estimates are then compared to bottom-up estimates using two different inventories.

Overall, the paper is well presented and well written. The structure is clear and it is easy to understand where the authors are leading us. Also, the scientific questions addressed in this study are well within the scope of ACP and the analysis conducted to answer these questions is detailed, elaborate and tackles very interesting points, both for experimentalists and atmospheric modelers. Last but not least, this new continuous data is invaluable to better investigate methane sources and will likely be utilized in the future by the rest of the atmospheric community.

Most of my comments only call for additional clarity in the methodology and the presentation of results. Also, a few additional details in the methodology and in the results would be beneficial both for the reproducibility of the study and the comprehensiveness of the analysis. However, these comments are very minor and I can already recommend this paper for a publication in the journal ACP.

Specific comments

Line 1: I recommend not using the abbreviation δ(13CH4) in the abstract. Use δ(13C,CH4).

Line 2: You write that it is a 6-year time-series, since 2014. It may give the reader the impression that the measurements stopped in 2020. I suggest a small revision: “Between 2014 and 2020, the time series shows an increasing trend of (6.8 ± 0.3) nmolmol−1 a−1 for the CH4 mole fraction.”

Line 6: At present, it seems you are using δ(13C,CH4) (abbreviated as δ(13CH4)) for atmospheric isotopic composition and δ13C (also an abbreviation of δ(13C,CH4)) for isotopic signature. In my opinion, it’s okay to keep it that way but you should not use the abbreviations in the abstract and also introduce the abbreviation δ13C in the main text.

Line 9: Sentence not clear. As far as I understand, the mean estimated δ(13C,CH4) source isotopic signature exhibits a seasonal variation, with a peak-to-peak variation -6.2 ‰. If it is the case, you should reformulate. I suggest replacing the sentence “This annual cycle in 13C-CH4 sources…”, with “This annual cycle in the mean source isotopic signature source, with a peak-to-peak amplitude of −6.2‰, can only be partially explained by seasonal variations in the 13C-enriched emissions from heating.”

Line 25: After this sentence, you should introduce the isotopic scale δ(13C,CH4) with a formula. Because after that you provide typical source signature values for different source categories (e.g. −55‰ to −70‰) , but the reader does not know what scale you are referring to. You could also be talking about 14C rather than 13C.

Line 30: Give the recent references for these values also here (e.g., Sherwood et al. 2017; 2021; Menoud et al., 2022). Also, the thermogenic range you provide appears slightly inconsistent (too small and too enriched) when compared to the information presented in these references.

Line 59: “To our knowledge, our time series is the longest in situ δ(13CH4) record, with high temporal resolution, reported to date.”. You mean the longest for Heidelberg or the longest ever in the world ?

Line 64: A continuous six-year time series between when and when ?

Line 99: Would be worth mentioning the value you are using for the reference 13C/12C ratio because there is sometimes confusion between PDB and VPDB values.

Line 119: I think two significant figures is not enough for δ(13CH4). I would recommend three, as you do in the rest of the paper. If we look at the bulk, I mostly see values between -49.0‰ and -47.5‰. If you also want to include extreme values, then it is approximately -49.5‰ to -47.2‰

Line 123: Please plot the trend on Figure 3. I think if you increase the size of the figure and increase the transparency of the 1day averages, then it won’t decrease the overall clarity.

Line 127: Less enriched compared to what ? To the mean ? Overall, I think some confusion can arise from the fact that you use the mean over the full time series as the center of your annual cycle, rather than using zero. Line 128 and 129 suggest that the values are always -48.3‰ in early autumn and -47.9‰ in spring, while these values have an interannual variability, due to the trend and the variations in the seasonal cycle. It’s okay to keep it that way but you should comment on that.

Line 139: After the analysis in Section 3.3, can you think of a reasonable explanation ?

Line 146: Could you provide the details of the Mace Head Observatory (altitude, longitude, latitude) ? Also, please explain why Mace Head can be considered as a “background” station.

Line 156: In this paragraph, you use both “emissions in Heidelberg” and “emissions in the catchment area of Heidelberg”. Is it supposed to mean the same thing (I suppose so) ? Or did Levin et al. (2011, 2021) only analyze emissions in the city of Heidelberg (without the surroundings) ?

Line 166: You should very briefly introduce the abbreviation δ13C the same way you introduced it for δ(13CH4) for the atmospheric isotopic composition.

Line 182: If you want to make a comparison between the Miller-Tans method and the Keeling plot, which is a good idea, you should briefly introduce the Keeling plot method as well.

Line 187 and line 200: Which CH4 range ? Apologies for being confused here. You discard every data point where the difference between C_{bg} and C_{obs} is below 100 nmol mol^{-1} ? If it is the case, for the night data set, do you discard the full night if one of the data points does not satisfy this criteria ?

Line 193: Please, reformulate. What can be assumed ? That it is constant ?

Line 213: What do you mean by ‘directly adjacent in time’ ? What amount of time do you consider to be ‘adjacent in time’ ? And what percentage of the 18% are during night time ?

Line 222: Here and throughout the text, you often use “more/less depleted” or “more/less enriched”. Usually, a value is depleted/enriched compared to a point of comparison, which is often the atmospheric value (around -48.07‰ in your case), as you state it very clearly in the following sentence. For instance, in this situation, I would use “more depleted” rather than “less enriched” because the source signatures for the moving Miller-Tans and night time approaches are already depleted compared to the atmospheric composition. Therefore, the third one is even more depleted.

Line 224: It seems that you are suggesting there is a causality between the fact that the estimated source isotopic signature is more depleted than the atmospheric composition and the fact that biogenic sources play a dominant role. I do not think it is true. You can suggest biogenic sources are dominant because the estimated source signature is low and close to what could be expected if biogenic sources (typically between -55‰ and -70‰) were dominant. Or to the contrary, it would be too low compared to a situation where only pyrogenic and thermogenic were dominant (although some thermogenic sources can have a source isotopic signature as low as -60‰, see Sherwood et al., 2017).

Line 271: Please reformulate. Do you mean both explanations are plausible ? Is it exclusive ? The too at the end is a bit misleading. In general, I do not understand how long an “event” is. Therefore, it is difficult to confirm that a night time increase can influence the event detected by the Miller-Tans approach. It would be nice to show somewhere the typical length of an event. Do these events happen mostly during the night ? As far as I understand, you can access this information with your methodology.

Line 280: You suggest two explanations but as far as I understand, both explanations are closely linked. Small pollution events of the first explanation can be the ones from distant sources from the second explanation. If it is correct, maybe you could mention it at the end of the paragraph.

Line 292 and onwards: Again “more depleted” compared to what ?

Line 303: “The monthly values vary on average between 0.1‰ and 0.8‰”. What does the percentage represent ?

Line 319: Give the exact location of the station.

Line 340: You have only one subsection 3.4.1 under section 3.4. Shouldn’t section 3.5 and 3.6 be subsections of sections 3.4 ? Or at least section 3.5 ?

Line 354: Why does it “seem” to decrease ? If you are not confident, where does this number 7%, without any uncertainty, come from ?

Line 389: It is not clear what you needed.

Technical comments

Figures: Although they have a good resolution, I would have preferred the figures to be larger, i.e. fitting the width of the page.

Figure 1: In the caption, map data on from → map data from

Figure 3: There is a problem with the x-axis ticks of the top-right panel. Please, make it similar to the bottom-right panel. Also, please add in the caption a note saying that the y-axis ranges are not the same for the left and right panels.

Line 67: to CH4 **total** emissions.

Line 152: Replace with “This was different” or “this is different for the 1990s”

Line 180: Put these subscripts in the same format as in the equation.

Line 214: Do you mean 2014 instead of 2011 ?

Line 266: Although → However

Line 274: less enriched → more depleted

Line 294: remove the space between the first parenthesis and “see”

Line 331: go one step further → extend the effort ?

Line 378: This is supported by the fact that the amount of emissions from sectors, such as livestock farming, with well studied emission factors and accurate statistical data are comparable for both inventories → This is supported by the fact that the amount of emissions from sectors with well studied emission factors and accurate statistical data are comparable for both inventories, such as livestock farming, are comparable for both inventories.

References

Sherwood, O. A., Schwietzke, S., Arling, V. A., and Etiope, G.: Global Inventory of Gas Geochemistry Data from Fossil Fuel, Microbial and Burning Sources, version 2017, Earth Syst. Sci. Data, 9, 639–656, https://doi.org/10.5194/essd-9-639-2017, 2017

Menoud, M., van der Veen, C., Lowry, D., Fernandez, J. M., Bakkaloglu, S., France, J. L., Fisher, R. E., Maazallahi, H., Stanisavljević, M., Nęcki, J., Vinkovic, K., Łakomiec, P., Rinne, J., Korbeń, P., Schmidt, M., Defratyka, S., Yver-Kwok, C., Andersen, T., Chen, H., and Röckmann, T.: New contributions of measurements in Europe to the global inventory of the stable isotopic composition of methane, Earth Syst. Sci. Data, 14, 4365–4386, https://doi.org/10.5194/essd-14-4365-2022, 2022.