

Answer to reviewer #1 for “Source attribution of methane emissions from the Upper Silesian Coal Basin, Poland, using isotopic signatures”

We would like to thank the reviewer for the suggestions to improve the manuscript. Below you find our answers to their comments. The reviewer’s comments are written in normal font, our answers in italics.

The manuscript “Source attribution of methane emissions from the Upper Silesian Coal Basin, Poland, using isotopic signatures” by Alina Fiehn et al., calculates the isotopic source signatures of $\delta^{13}\text{C}$ and $\delta^2\text{H}$ from the airborne samples and the ground-based samples using the Keeling method which later helps to constrain the different source contributions for coal and biogenic emissions.

Overall, this work provides important information and data about isotopic source signatures. The isotopic signatures analysis is presented in a straightforward way which is easy to understand. The manuscript is written and structured well. Therefore, I suggest it to be suitable for publication in ACP after addressing specific and technical comments as listed below.

We thank the reviewer for this positive statement.

Specific comment:

1. L210: how to obtain the uncertainty of $\delta^{13}\text{C}$ showed in Figure 3? $\delta^{13}\text{C}$ is the interception for the fitting line and how do the authors define the uncertainty?

The standard errors of the intercepts or $\delta^{13}\text{C}$ signatures are a result of the ODR algorithm. We used the `scipy.odr` package. We added this information to the manuscript in Section 2.3, L203: “The regression was calculated with the Python `scipy.odr` package, which calculates the intercept as well as its uncertainty as standard deviation from the uncertainties of the input data.”

2. L224: authors mentioned that “the signature of all inflow samples...indicates that the CH_4 enhancements in the upwind boundary layer are mostly biogenic, but with a fossil influence”. But from Figure 4, the IN symbol and its error bars are all located in green shaded area, i.e., EMID modern microbial. I do not fully understand why there is a fossil influence.

True, the IN symbol and error bars are fully located in the green shaded area, but the signature is shifted towards the fossil fuel signatures compared to the FT (free troposphere) signature. If we assume that the free troposphere methane is from biogenic sources, then the inflow methane either is mainly biogenic with fossil influence or from a different type of biogenic sources. So, it could be that inflow sources are rather from waste management, which has more positive signatures, than agriculture, wetlands or ruminants, which are more negative. We added this to the text.

3. L 250, Table 1: are the values of the isotopic composition and standard error originated from all flasks during each flight or only from the flasks in PBL? Please specify it.

I suppose the symbol “#” presents the number of flasks. Maybe No. is better for understanding.

Only flasks in the PBL are considered here. We specified this and changed # to No.

4. L253, Figure 4: I think the individual source signatures (with black symbols and numbers) represent the results originate from the flasks in PBL. If so, please specify this information, otherwise, it is misleading.

We also specified this here.

5. L267: “ $\delta^{13}\text{C}$ signatures from all samples are in the same range (not shown)”. It would be interesting to see the results. Maybe put it in the appendix.

We added Figure A1 in the appendix to show the Pniowek samples from inside and outside the ventilation shafts.

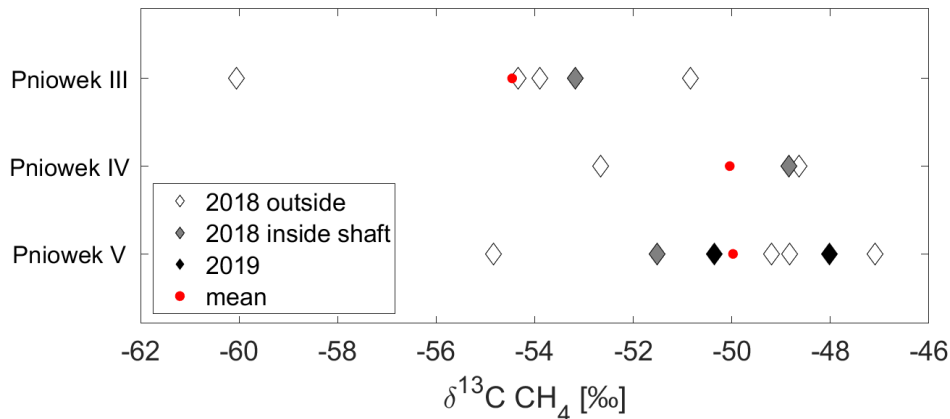


Figure A1: Comparison of coal mine signatures from samples taken outside and inside the ventilation shafts.

6. L269: “...with one signature at -38 ‰.” I assume that you mean this value is out of the main range. Perhaps change it to “with one biased signature at -38 ‰”.

Good idea. This is not exactly a biased signature though, but an outlier. We specified this.

7. L278: “The aircraft samples showed average $\delta^2\text{H}$ signatures for the southern region, but the southeastern mines had lower $\delta^2\text{H}$ signatures than the entire USCB.” I assume the aircraft samples indicate the dots in Figure 3, and the cyan square symbols in Figure 4. If so, in my opinion it would be beneficial to show another figure (like in the appendix) to better present the statement. The coordinate-related information cannot be found from Figure 3 or 4. If the “aircraft samples” represent the isotopic source signatures in PBL shown in Figure 4., it is the southwest but not the southeastern mines having lower $\delta^2\text{H}$ signatures. Please comment.

Indeed, this refers to the isotopic source signatures in the PBL shown in Figure 4 and should name the southwestern mines with the lowest $\delta^2\text{H}$ signatures. We simplified and corrected the sentence to: “The $\delta^2\text{H}$ source signatures in the PBL derived from aircraft samples also showed that the southwestern region had the lowest $\delta^2\text{H}$ signatures (Figure 4).”

8. L339: “For comparison, the EMID includes $\delta^2\text{H}$ signatures from 7 landfills (-275 ± 21 ‰) and signatures from 6 wastewater facilities (-323 ± 14 ‰) across Europe. The average signature over all these data points is -297 ± 30 ‰.” I assume that the average signature from the 7+6=13 sites is calculated based on weighted average. How about the uncertainty? Is the error propagation or other method used? Please comment.

The average was calculated from the 13 individual values. Also, the uncertainty gives the standard deviation of all 13 signatures. We agree that it makes more sense in this case to use a weighted average and error propagation, because these are two different source categories and the combination of their signatures would not increase the uncertainty range. We changed our method and the new values are 297 ± 18 ‰. We changed this in the text: “The weighted average of the signatures of the two sectors is -297 ± 18 ‰. The uncertainty was calculated through error propagation”

9. L343: “we assume that the USCB $\delta^2\text{H}_{\text{bio}}$ signature for waste emissions is $-300 \pm 20 \text{ ‰}$ for our study”. Is the mean value of -300‰ from the global modeling as mentioned in L341? This value does not match with the mean value of biogenic signatures shown in Figure 7.

Additionally, from where the uncertainty of 20‰ come? Please specify.

The average signature of -300‰ is a combination of the global modeling value and the EMID value. The 20‰ uncertainty is taken from the EMID observation uncertainty ranges. We added this to the manuscript.

10. L351: CAMS-REG-GHG inventory has been updated to v5.3 in 2022. There might be no huge difference between v3.1 and v5.3. It is for your information.

It would be also beneficial to compare the spatial distribution of gridded inventory and the results here. Will the CAMS inventory in the southwest area tends to have more biogenic sources?

We conferred with the creators of CAMS. The emission inventory v5.3 is only the global inventory. The regional inventory CAMS-REG-GHG has also been updated to a version 4.1, which includes the year 2018, when our measurements took place. Although the data is not publicly available yet, it was provided to us. There are no big changes.

That the southwest USCB has more biogenic sources is not a result or conclusion from our study. The lower signatures in $d^{13}\text{C}$ and $d^2\text{H}$ in the southwest are probably due to regional gradients in the coal mine methane signatures rather than sectorial fractionation changes because the gradient is observed both in the ground-based as well as the airborne data.

Technical comments:

11. L100: the subscript in the “ $\delta_2\text{H}$ ” should be superscript. – *Changed.*

12. L162: not fully understand the sentence “For each of these categories we determined the mean isotopic signature from all flights combined and for PL samples also for individual flights.” Please rephrase.

We rephrased to: For each of these categories we determined the mean isotopic signature for the entire campaign. Using the PL samples from each flight individually, we calculated the isotopic signatures of the individual target regions.

13. L199: please keep consistent format for δ_x in equation (3) and afterwards. The δ_{obs} , δ_{bg} are in bold in previous text.

We changed all the bold face into normal font in the equations and text.

14. L264: I think this sentence has a grammatical error. “This variability may result from different areas of the mine being exploited as longwalls at different depths of the mine are opened up or shut down during excavation.”

Maybe change to “This variability may result from different areas of the mine during longwall exploitation at different depths of the mine which are opened up or shut down during excavation” Please rephrase.

We changed this to: Within one mine the isotopic signatures differ due to the geographical structure. The signature of the ventilated methane then also varies with time, because longwalls at different depths of the mine are opened up or shut down during excavation.