

Answer to referee comment 2 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 authors use airborne and ground-based air samples to attribute methane emissions in the region to sources, with a focus on distinguishing coal/fossil sources from biogenic/waste sources. The ground-based samples are used to identify source signatures and then, the airborne samples are used to determine the contribution of the two source types. Overall, the study is nicely written and presented but can benefit from some editing to clarify the analysis and results.

An interesting finding from this paper is the relatively large contribution of biogenic sources in the Upper Silesian Coal Basin. Given this finding, there is a need for some introduction of these biogenic sources in the Introduction and annotations in Figure 2. Although there is some text on this towards the end, additional text earlier on would be helpful.

It appears that a lot of the data being used in this paper have been published in previous papers. However, it’s unclear if there’s new data presented in this paper. This should be clarified (see detailed comments below).

Thank you for the positive feedback and good suggestions. We improved the introduction of biogenic sources and clarified where data has been published previously. All airborne isotopic measurements are unpublished, but ground-based observations have been published previously with different focus.

Below are detailed line-by-line comments:

L20: Replace "growth" with "concentration accumulation". – *We decided to use the term “increase of atmospheric methane levels”.*

L20: Replace "on global" with "at global". – *Done.*

L24: Are the "other anthropogenic sources" related to the coal mine or something else? Landfills and wastewater should be listed here in the abstract given the findings of the paper. – *Done.*

L30: What are all the sources in the region? This speaks to an earlier comment about clarifying the sources being studied.

In this sentence the focus is not on the individual sources or even what kind of sources there are. We want to convey that the airborne methane plume contained a well-mixed plume of methane containing all emissions from within the study region. We changed the phrase to: “...methane from the entire region...”

L35: "allowed for the determination of the source signatures..."

We changed this to: “We determined the source signatures of individual coal mine ventilation shafts using ground-based samples.”

L40: I think the main point is that the d^2H is important for source attribution and that the d^2H of the ventilation shafts differ from the regional d^2H values. Therefore, it would be worth rephrasing this sentence to highlight this point. The fact that the d^2H of ventilation shafts match previous studies is relevant but should be the secondary point.

This is true and we changed the sentence accordingly omitting the comparison with previous studies. This secondary point is now only mentioned in the Discussion and Conclusions.

L41-42, 34: Because wetlands and ruminants were mentioned in L34, I expected these to be significant. Suggest revising L34.

We omitted the naming of source categories in line 34 to avoid confusion. Wetlands and ruminants are probably significant in the upwind and free tropospheric source signatures though.

L45: It would be good to provide some quantitative comparison of underestimation. Either state that the common inventories estimate 6% biogenic or state the difference as a factor.

We added the fractions given in the inventories (0.4-14%). This is also stated in the Conclusions.

L50: “at limiting” to “to limit” – *Done.*

L52: The Global Methane Pledge is specifically for methane emission reductions, not all greenhouse gas emission reductions. Replace “greenhouse gas” with “methane”. – *Done.*

L53: Replace "localize" with "locate". – *Done.*

L116: Define what is meant by "regional perspective". What is the scale of "regional"? It sounds as though the authors are re-analyzing existing data. Either way, this needs to be clarified. A clearer description of what new analysis is being performed here would be helpful.

The region considered here is the USCB. We changed the paragraph to: In this study, we present isotopic methane sample analysis for the USCB. We analyze unpublished samples taken on a small aircraft and compare to already published ground samples to determine the contributions of coal mining and waste treatment to the total USCB methane emissions.

L136: Pniowek is not shown in Figure 2. It should be identified. – *We marked it in the Figure.*

L141: Define FUB at first mention. – *We spelled out “Freie Universität Berlin”, because it only appears once in the text.*

L153-154: Were the data published already? This needs to be clarified.

The airborne data from the DLR Cessna Caravan has not been published previously. We added this information to the manuscript.

L171: "...concentrations and isotopic compositions of CH_4 were..."

This sentence seems correct to us.

L218: how can the standard errors be small if the variance is large?

What we meant here is that we have a large range in the concentration values and thus could constrain the source signature well. We rephrased to “large range of concentrations”.

L226: the location of Silesia needs to be shown in Figure 2. – *We marked it in the Figure.*

L261: the locations of the cow farm, landfills, manholes and wastewater facilities need to be shown in Figure 2. – *We marked the cow farm and landfill 2 in the Figure. The biogenic sources are not on the map, but 100 km to the east in Krakow.*

L266: what is meant by vicinity? In the conclusions, 1-2 km is mentioned. It's surprising that the samples taken directly within the shafts are similar to those taken 1-2 km away. *We added the correct distance of 1-2 km also here in the text. The similarity of isotopic signatures from inside and outside the ventilation shafts seems logical. Although the concentrations in the samples were different, the enhancements outside the mines were large enough for a thorough Keeling analysis. The resulting source signatures are free of the influence of the background methane and independent of the sample concentrations. The signatures are not identical, though, because they were not sampled on the same day.*

Figure 5: Specify where in the mines the "mining" samples were taken. – *Done.*

Figure 5: Is the data from the MEMO2 dataset? Also specify that this is from ground-based studies.

The ground-based data in our analysis is from the MEMO2 dataset, which in turn is part of EMID. We clarified this here.

Figure 6: Specify whether the data is from ground-based measurements in the caption. – *Done.*

Figure 7: specify that EMID fossil fuel data is ground-based.

Overall, there are a lot of acronyms that are used interchangeably. For the ground-based data, it's referred to as MEMO2, EMID, and "ground-based". I suggest simply calling it "ground-based", if possible, and not using multiple names for the same datasets.

We now only use two names for the ground data: “ground-based” are all samples in the USCB that were used in this study. “EMID” signatures and data applies to all samples taken all over Europe. “MEMO2” has been eliminated except for in the Acknowledgements.

L324: Given the importance of the waste sector in this region, there is a need to describe these sources more. How big are the landfills and wastewater treatment plants both spatially and in terms of waste volumes? Also, is there an inventory of all methane sources in the USCB?

There really is not a lot of information available on the waste sector in the region as there does not exist an emission database dedicated to the waste sector. However, there are some information where the landfills are located and how much waste is being deposited every year. Population density normally gives an idea of the amount of wastewater produced in a certain region. This information needs to be translated into methane emissions though. A rough estimate gives emissions in the order of 60-80 kt/year for the landfills. This will be topic of a planned future publication.

There are the Industrial Reporting (IR) emission database (ED) (former E-PRTR), the CoMet v4.01 ED (Galkowski et al., 2021) and scientific gridded ED like EDGAR and CAMS-REG. Within CoMet v4.01 ED, 32 landfill locations are listed. Most of them are not reporting emissions of CH₄ to IR. We did not investigate this in depth, but in most cases, they are relatively small and their individual emissions fall below reporting threshold. Of those that did report emissions to E-PRTR, the reported numbers were around 3 kt/year for 8 to 14 reporting landfills. That is an order of magnitude smaller than what is estimated based on the amount of trash deposited. Additionally, 24 individual wastewater treatment plants are identified within CoMet v4.01 ED. None was reporting CH₄ in 2018. The numbers from gridded data like CAMS-REG should be consistent with E-PRTR.

We added some of this information to the manuscript.

L333: the better discrimination of d²H signatures than d¹³C is interesting. Therefore, it would be helpful to see what the error would be if only d¹³C was used in source attribution. *The source attribution is not really possible with δ¹³C. The uncertainties of the signatures overlap considerably. Although we did calculate an average signature for the USCB coal mine emissions from the ground-based data, this value is very uncertain in δ¹³C (-49.8 ± 5.7 ‰). The error using this value against the total USCB signature of -50.9 ± 0.7 ‰ would be around 500%.*

L359: can the locations of these landfills and wastewater treatment plants be shown in Figure 2?

Krakov and the landfills and wastewater treatment plants are not in the area depicted by Figure 2, but about 50 km to the east of the eastern USCB border. We highlighted landfill 2 in the new Figure 2.

Figure 8: The authors assume that waste is the dominant biogenic source in the area. However, there may be natural (and agricultural) sources that may be contributing more than assumed. The authors are probably right but it may be worth pointing out that there still are uncertainties in the biogenic methane source.

A contribution from natural sources is possible. Especially agricultural sources may also be underestimated in the gridded inventories. This would influence our results because their isotopic methane signatures are at the lowest edge of the spectrum.

L398: how much do the signatures vary over time? How does this affect uncertainties and the comparisons/analysis presented in this paper?

Ground-based samples taken in 2018 and 2019 show no trend in the signatures from the coal mines (Figure 5). The δ¹³C signatures of previous studies (Kotarba, 2001) have been lower than our values (Figure 7), but the δ²H is in the same range. Since we only use δ²H in the source apportionment we do not expect an influence of this shift. It is also hard to determine whether this is a real shift over time or if it is a result of the different techniques used.

We only have airborne signatures for the year 2018 and cannot deduce if the signature of the total emissions of the USCB changed over time.

- Gałkowski, M., Fiehn, A., Swolkien, J., Stanisavljevic, M., Korben, P., Menoud, M., Necki, J., Roiger, A., Röckmann, T., Gerbig, C., and Fix, A.: Emissions of CH₄ and CO₂ over the Upper Silesian Coal Basin (Poland) and its vicinity, ICOS ERIC - Carbon Portal, 10.18160/3K6Z-4H73, 2021.
- Kotarba, M. J.: Composition and origin of coalbed gases in the Upper Silesian and Lublin basins, Poland, *Organic Geochemistry*, 32, 163-180, 10.1016/S0146-6380(00)00134-0, 2001.