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Interactive comment

Interactive comment on "Characterisation of δ^{13} CH₄ source signatures from methane sources in Germany using mobile measurements" by Antje Hoheisel et al.

Anonymous Referee #2

Received and published: 3 November 2018

This paper investigates a method for unbiased isotopic measurements of methane using a commercially available cavity ring-down spectrometer in a mobile setup. It specifically provides guidance on the correction for potential biases from ethane and water in the measured gases. These methods are then used to measure and characterize methane isotopic signatures from different sources in Germany.

Given the increasing interest in isotopic measurements of methane for source attribution studies and the availability of relatively low-cost analyzers, this paper is a useful addition to the literature to help researchers improve their measurements. The tables and figures are well presented. No major shortcomings were noticed. However, below





is a list of detailed comments that may help clarify arguments and language, and correct potential errors. Additionally, I suggest altering the title to something like "Improved method for mobile characterisation of δ 13CH4 source signatures and its application in Germany." The current title sells short of a major contribution of the paper.

Detailed comments follow:

1. P 1, In 3: Remove "Therefore"; This sentence doesn't logically follow from the previous sentence. It is detail in addition to the previous sentence.

2. P 1, In 5: Explain gas matrix or replace with less jargon.

3. P 1, In 7: Abundant in many natural gases, but not all. Dry gas regions can contain only very small traces of ethane.

4. P 1, In 16: A 2.8 per mil difference begs the question whether the above mentioned up to 3 per mil ethane bias could have played a role here.

5. Switching between "CH4" and "methane" throughout the MS. Check for consistency.

6. P 1, In 20: Use original data references instead of reviews, e.g., https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2009GL039780

7. P 2, In 1: You probably mean biomass burning.

8. P 2, In 2: More accurate to say "e.g. a sub-category of fossil fuel extraction". Some gases extracted by the fossil fuel industry is biogenic.

9. P 2, In 7: Do you mean to say that the isotopic signal was used to diagnose methane emission reductions? Not clear as written.

10. P 2, In 13: What do you mean with "all of these seasonal variations"?

11. P 2, In 26: Briefly describe the importance/use of a storage tube (first mention).

12. P 2, In 26: Nine facilities in 21 campaigns? Do you mean 21 measurement days?

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13. P 3, In 15: Perhaps that's explained later, but how do you get so close to the source that you're able to sample between 50 and 90% CH4?

14. P 3, In 17: Specify the composition of synthetic air.

15. P 3, In 33: Do you mean a 20-25 sec lag time between air sampling at inlet and fully arriving in the cavity? Sounds like it, but not fully clear.

16. P 4, In 11: What do you mean with representations have the same width? They don't. It's a bit confusing. The next sentence already states that both peaks represent the same emission plume.

17. P 4, In 13: Any hypotheses why this may be the case? This seems to be an important observation.

18. Table 1: Is the unit for ethane sensitivity to water (ppm) a typo? Units of ppb would make more sense. For reference, atmospheric ethane background concentrations are in the order of < 1 ppb.

19. P 5, In 5: Natural gases on a continuum between 0 to 40% ethane have been measured. See Sherwood et al., 2017 (already in your refs). Hence, the importance of ethane correction varies: very important for wet gas basins (meaning lots of associated gas) and less important for very dry gas regions (mature thermogenic dry gas).

20. P 5, In 10: where does the 3 per mil value come from?

21. P 7, In 3: What is the sign of the drift? Is the drift due to fractionation in the bag or due to leakage of background air into the bag?

22. P 7, In 28: What are simulated data?

23. P 10, In 4: I'm confused. Here it says no significant seasonal cycle has been observed. In the next paragraph, it says the values are more depleted in winter than in summer. Are there enough samples to determine this correctly?

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24. P 10, In 13: What are the uncertainties for the ethane-to-methane ratios?

25. P 12, In 2: Why were the plumes expected to be smaller than on the other farm?

26. P 12, In 11: Why are seasonal differences for the biogas plant expected?

27. P 12, par. 1 & 3: Is C3/C4 diet information available for the Ladenburg and Kleve farms?

28. P 14, In 11: How small the fluxes were actually depends on the size of the facility (is it possible to detect all plumes at once?) and the wind conditions in addition to the measured methane concentrations.

29. P 14, In 20: Did your measurements include incomplete combustion from the compressor turbines? This could be detected by the associated CO2 or CO signal. It is still an open question whether high heat leads to isotopic fractionation of the source.

30. P 14, In 25: Do you use "open" and "in service" interchangeably? Not clear. If yes, it's not a surprise that a mine currently in service produces more emissions than a closed mine.

31. P 14, In 30: The Bottrop mine shaft measurements do not match coal bed gas samples except for the closed mine with a large error bar and only one AirCore measurement.

32. Conclusions: In the first paragraph, it's important to highlight again that these results (including the ethane bias) are specific to the CRDS instrument used.

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