

We would like to thank the reviewer for the helpful comments and suggestions. We answer each of them here after, with the original comment in blue and our response in black. We added the modifications done in the revised version in italics.

Specific Comments:

[Referee #1]

One clear omission is the C_2H_6 / CH_4 signatures from the individual plume measurements. A great deal of important work is done in calibrating the C_2H_6 channel from the instrument, but then the field data from this channel is not presented. Reporting these ratios would serve to validate that the instrument is reporting reasonable values; conversely, if the ratios do not make sense (e.g., if the landfill C_2/C_1 ratio is not zero within uncertainties), then that may point to an additional unknown systematic error in the system. For example, these data could be easily added to Table 2.

Similarly, for the bag samples of the local natural gas source, the measurements of the ethane content would be an interesting addition to the manuscript.

Furthermore, the ethane content in the natural gas infrastructure is generally recorded by the natural gas distribution company; the values obtained from the analysis could be compared to the known values.

[Hoheisel et al.]

Thank you for pointing this out, we have added an additional section (see below) and another figure.

“ C_2H_6 to CH_4 ratio of direct samples and mobile measurements

The C_2H_6 to CH_4 ratio of gas samples from the natural gas distribution network in Heidelberg varies between 0.027 and 0.072 with lower values in winter, 0.04 ± 0.01 , and higher ones in summer, 0.06 ± 0.01 . This result can indicate that the percentage of Russian gas is higher in winter than in summer taking into account, that Nitschke-Kowsky et al., (2012) reported a C_2H_6 to CH_4 ratio for Russian natural gas to be 0.014 while for North Sea gas it is 0.078. Also the isotopic signatures of our natural gas samples support this trend with more depleted values in winter than in summer.

Gas emitted by other CH_4 sources like landfills, biogas plants and wastewater treatment plants do not contain C_2H_6 . The C_2H_6 to CH_4 ratio of the gas samples taken directly at the gas collecting systems of these sources are zero within the errors and can be clearly separated from the natural gas samples (see Fig.7). The separation due to C_2H_6 to CH_4 ratio works well with direct gas samples, but unfortunately not yet for mobile AirCore measurements. In contrast to the direct sample measurement, in mobile AirCore measurements CH_4 and C_2H_6 emitted by the source are diluted in the background. To determine the ratio, a linear fit of the measured C_2H_6 concentration to the measured CH_4 concentration is used.

However, due to the high uncertainty of the C_2H_6 measurement without averaging as it is possible for the direct sample measurement, in combination with the very small or not existing changes in C_2H_6 the fit does not provide reasonable data.”

[Referee #1]

A second aspect of the manuscript that could use more attention is the calibration of $\delta^{13}C-CH_4$. Hoheisel (2017) is cited, but at least a brief description should be provided here (and/or a more detailed description in the supplemental material). For example, there are terms in Fig 3 (e.g., $\delta^{13}CH_4$ _Nominal) which are not even described in the text. This is critical information if these data are to be used in partitioning of global or regional methane emissions. It is not clear from the text or the figure whether standards with a range of isotope ratios was used in the calibration; please clarify.

[Hoheisel et al.]

We understand the point of the referee. For a better understanding, we deleted “and calibration” on page 4 line 22, since in this section we focus on corrections. In addition, we added some explanations in section 2.3.3 and in the caption of Figure 3. This section also provides more information about the used standard.

“All data measured with the CRDS analyser in the laboratory or during mobile campaigns was corrected using the factors from Table 1 and following Fig.3 prior to the one point calibration calculation.”

“The gas cylinder used for calibration was chosen according to the experiment to ensure a similar composition and similar concentrations for sample and standard.”

“Scheme to correct and calibrate C_2H_6 and $\delta^{13}CH_4$. $\delta^{13}CH_4$ _Nominal is the nominal (known value of the standard) and $\delta^{13}CH_4$ _Standard the measured (or interpolated) value of the calibration standard.”

Detailed Comments and Questions:

[Referee #1] P1L3: "Therefore..." poor word usage. Rewrite.
[Hoheisel et al.] Agreed, we have changed it.

[Referee #1] P3L17: polypropylene
[Hoheisel et al.] Yes, corrected.

[Referee #1] P3L27: 260 amp-hr. check units
[Hoheisel et al.] Done.

[Referee #1] P3L28: Supplier & Model Number for the inverter
[Hoheisel et al.] Done, we now have included the Supplier and model number:
"e-ast HighPowerSinus HPLS 1000-12".

[Referee #1] P4L3: How were the valves triggered for "replay" mode? Add a description.
[Hoheisel et al.] We switch the valves manually after passing a methane plume. For clarification, we now have added the following phrase.
"by switching the valves manually".

[Referee #1] P6L24-25: Relative increase of which uncertainty? This statement is unclear.
Please clarify.
[Hoheisel et al.] Ok, we have changed it.
"the uncertainty of $\delta^{13}\text{CH}_4$ "

[Referee #1] P7L29: Please provide a brief description of the York fit.
[Hoheisel et al.] Thank you for pointing this out. We have now included a short description.
"York's solution is the general LSE (least-squares estimation) solution: his equations provide the best possible, unbiased estimates of the true intercept a and slope b in all cases where the points are independent and the errors are normally distributed" (Wehr and Saleska, 2017)

[Referee #1] P8L12: Why do the Keeling and MT methods produce identical source signatures and uncertainties (as shown in the figure)? I assume that this is an error in the preparation of the figure. If not, please explain.
[Hoheisel et al.] We appreciate the recommendation for clarity and we looked again into the matter. However, with both methods (KP and MT) the isotopic signature is determined identical within the first few decimal places when using the measured data and also for simulated datasets.

[Referee #1] P10L13: It's surprising to me that north sea gas is high in ethane content and heavy in isotope ratio. Generally heavy isotope ratios are associated with low C2+ content. Please confirm these values.
[Hoheisel et al.] We have checked it again and the values are correct.

[Referee #1] P11L33 (for example). The plume peak heights should be reported above the local baseline value, rather than absolute methane values. Please clarify in the description (e.g., "Peak height above baseline")
[Hoheisel et al.] We generally agree with the reviewer. However, in the text we wanted to give an expression of the CH_4 concentrations measured around the different sources. To clarify it, we change the phrase "peak height", when we report maximum CH_4 concentrations measured in the plume. Since it is correct, that the CH_4 peak heights above the background are mostly driving the fit error, we decided, to change the maximum values reported in Table 2 and in the supplements to peak heights above baseline.

- [Referee #1] P12 Landfill section: The isotope signature of landfills are complicated by a) the isotope signature of the source material in the landfill, this fractionation that occurs by the anaerobic bacteria, and the further fractionation that can occur in the landfill cap where the methane is consumed aerobically.
- [Hoheisel et al.] We now have added an additional sentence at the beginning of the section.
“In addition to the source material and fractionation during the production the isotopic signature of gas emitted by landfills depends also on fractionation processes in the upper soil layers of the landfill. Due to the presence of oxygen in the upper soil layers, aerobic bacteria oxidate parts of CH₄ which diffuses through the soil cover and shift the isotopic composition to more depleted values. Bergamaschi et al. (1998) measured these different isotopic signatures.”
- [Referee #1] P13: WWTP: The methane emissions from WWTPs can come from various locations and processes within the facility, and these processes may have different isotope signatures. Further, different WWTP plants employ different techniques to digest the waste; some discussion of these processes and their likely impact on your measurements would be helpful.
- [Hoheisel et al.] We understand the concern of the referee and added a few minor explanations (see below). However, more details about the specific processes lead too far for this rather technical paper. Indeed, the study will be continued and another paper is planned that will describe the different processes in more detail.
“There, the sludge treatment inside the digestion towers takes place in three septic tanks with a volume of 2500m³ each under anaerobic mesophilic conditions, that means without oxygen at 37°C.”

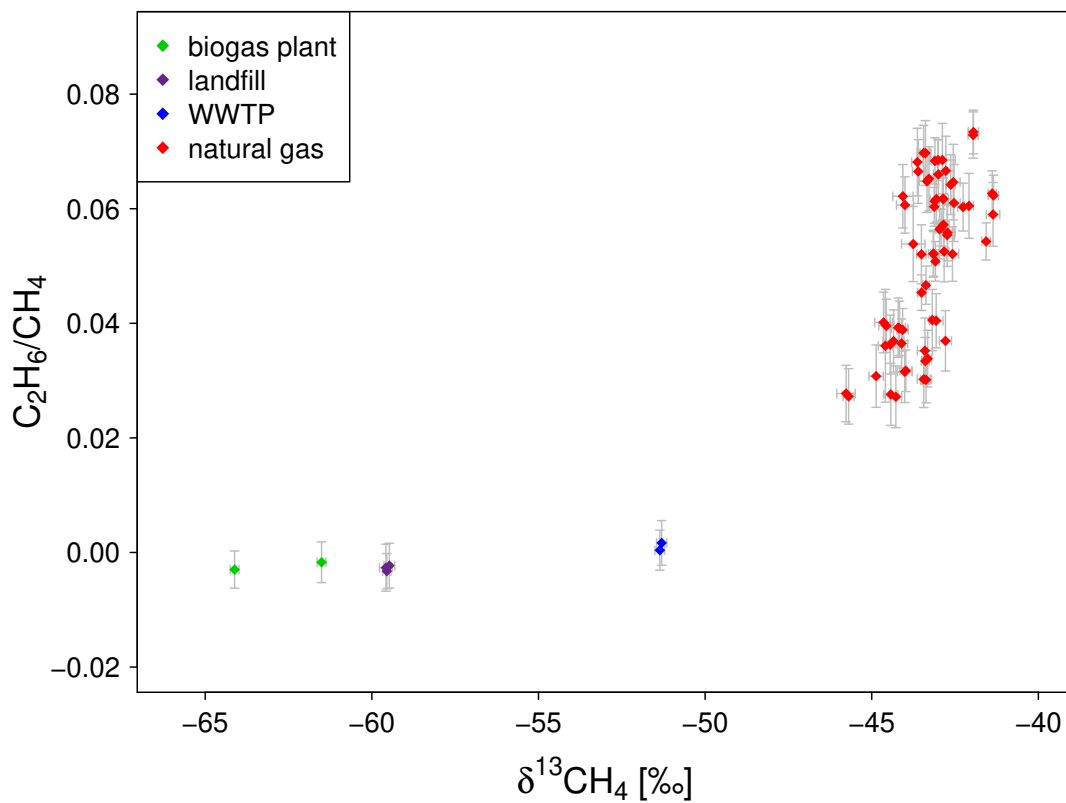


Figure 7. Isotopic signature and C_2H_6 to CH_4 ratio of gas samples from a biogas plant, a landfill, a WWTP and the natural gas distribution system in Heidelberg measured between end of 2016 and November 2018.