

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.

#### General Comments

[Referee #3] It is nevertheless regrettable that the  $C_2H_6$  to  $CH_4$  ratios are not presented along with the isotopic signatures. I strongly suggest to add it in order to give the paper more visibility.

[Hoheisel et al.] We appreciate the recommendation and we added the short section  *$C_2H_6$  to  $CH_4$  ratio of direct samples and mobile measurements* as well as one 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,  $0.04 \pm 0.01$ , and higher ones in summer,  $0.06 \pm 0.01$ . 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."*

#### Specific comments

[Referee #3] The language used in the manuscript is not always precise. For instance, the terms " $^{13}C$  values" and " $^{13}C$  signatures" are not the same and sometimes confusing as used in the MS. Also, the  $^{13}C$  signatures are not directly measured by the CRDS but determined after data treatment from the measurements. Please correct through the all manuscript.

[Hoheisel et al.] Yes, we clarified it.

[Referee #3] Most of the time, the uncertainties are presented, but without any explanation on how they are calculated and which parameters are used to computed them. It is also not clear what is the difference between uncertainties and precision. For instance, page 9 line 5/6, it is stated that increasing the number of data point improves the uncertainties on signatures. The next sentence, "uncertainties" has been replaced by "precision". It is confusing. Please clarify through the all manuscript.

[Hoheisel et al.] Yes, we corrected it.

[Referee #3] The G2201-i instrument can be used in different mode, which drives the measurement frequency of each species. Also, the methane is reported by the instrument in high range (HR) and high precision (HP) mode depending on the level of the measured mole fractions. This drives the instrumental precision. Please, add some details on these two points.

[Hoheisel et al.] We agree that this is an important point and added the following text.  
*"All measurements with the CRDS analyser were done in the combined  $CO_2/CH_4$  mode to measure  $CH_4$  and  $CO_2$  parallel. In addition, the High Precision (HP) mode for  $CH_4$  is chosen to provide the most precise  $CH_4$  measurements for  $CH_4$  concentrations up to 12 ppm."*

[Referee #3]	Part2.1.2 and figure1: here is a list of questions/comments which should be addressed:
[Referee #3]	- how are the valves switched?
[Hoheisel et al.]	We switch the valves manually, after passing a methane plume. For clarification, we now have added the phrase “ <i>by switching the valves manually</i> ”.
[Referee #3]	- how is the flow measured and/or controlled?
[Hoheisel et al.]	The flow is adjusted by needle valves and measured by a flowmeter from time to time.
[Referee #3]	-why the flow presented here differs so much from the laboratory setup (160ml/min vs 20 to 80ml/min)?
[Hoheisel et al.]	In the laboratory we use a rotary valve to switch between cylinder and ambient air measurements and we will have a small flow rate. In the mobile setup, we would like to have a higher flow rate for a better time resolution and a shorter lag time between air sampling at inlet and measurement in the cavity of the CRDS analyser. Therefore, we bypass the rotary valve.
[Referee #3]	- what is the typical air flow going through the AC in monitoring mode?
[Hoheisel et al.]	The typical air flow through the AirCore in monitoring mode is 0.8 l/min.
[Referee #3]	- what is the typical vehicle speed while in monitoring mode?
[Hoheisel et al.]	The speed depends strongly on the road. We tried to drive as slowly as possible, but without constraining other vehicles. Typical vehicle speeds are 10-50 km/h.
[Referee #3]	- in figure1, the blue and green arrows are difficult to differentiate. Please, make them thicker.
[Hoheisel et al.]	Done.
[Referee #3]	Part3: how do you make sure that the direct samples are not mixed with ambient air? This could bias the isotopic signatures.
[Hoheisel et al.]	We take gas samples for example directly from the gas collecting system of the landfill and the WWTP. Therefore, the CH <sub>4</sub> concentration of these samples is between 50-90% and a potential mixture with small amounts of ambient air would be negligible. We now have clarified it and changed the text. “Gas samples taken directly from different installations (e.g. natural gas pipelines, biogas plants, gas collecting systems of landfills and wastewater treatment plants) need to be diluted before the measurement with the CRDS analyser, because such samples usually consist of between 50 and 90% CH <sub>4</sub> .”
[Referee #3]	Part 3.2: it has been shown in figure5 that the CH <sub>4</sub> peak height above the background is mostly driving the fit error. It would make much more sense to present all the peak heights above the background instead of the absolute CH <sub>4</sub> value.
[Hoheisel et al.]	We generally agree with the reviewer. However, in the text we wanted to give an expression of the CH <sub>4</sub> concentrations measured around the different sources. To clarify it, we changed the phrase “peak height”, when we report maximum CH <sub>4</sub> concentrations measured in the plume. Since it is correct, that the CH <sub>4</sub> peak height above the background is 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 #3]	Conclusion: the Miller-Tans and Keeling approaches give the same results. Why do you suggest using one instead of the other one?
[Hoheisel et al.]	We agree that this is misleading. In our study we have seen that when using the York fit, it does not matter if we use the Miller-Tans or the Keeling approach. So we changed it in the conclusion.
[Referee #3]	P1, line 6: C <sub>2</sub> H <sub>6</sub> only affects the <sup>13</sup> C measurements.
[Hoheisel et al.]	Yes, we changed it.
[Referee #3]	P1, line 11: <sup>13</sup> CH <sub>4</sub> signatures instead of values.
[Hoheisel et al.]	Done.

[Referee #3] [Hoheisel et al.]	P2, line 1: "biogas burning" is not a CH <sub>4</sub> source. Yes, corrected to "biomass burning".
[Referee #3] [Hoheisel et al.]	P2, line 11: do you mean "due to its origin,"? Yes, we have changed it.
[Referee #3] [Hoheisel et al.]	P2, line 15: introduce IRMS here and not line 17, and add what GC stands for. Done.
[Referee #3] [Hoheisel et al.]	P2, line 16: I suggest to delete "has been"! as shown by Röckmann et al. Done.
[Referee #3] [Hoheisel et al.]	P3, line 1: replace signature by ratio. Signatures are not directly measured by the CRDS. Corrected.
[Referee #3] [Hoheisel et al.]	P3, line 13: it is surprising to observe such a flow range (factor 4). Could you explain why and give more details? Is the flow controlled somehow? Ok, we have made several changes: <i>"The gasflow to the analyser is typically 25 to 35ml/min for calibration gas, target gas and sample bag measurements. For some applications like ambient air measurements the flow is higher with values around 80ml/min to resolve shorter temporal variabilities. The flow is measured by an electronic flow meter (model: 5067-0223, Agilent Technologies, Inc., Santa Clara, CA) before entering the analyser."</i> Tests have shown that in the flow regime of 25-80ml/min the measurement did not depend on the flow. An electronic flow meter measures the flow but the flow is not controlled.
[Referee #3] [Hoheisel et al.]	P3, line 17: what do you mean by synthetic air? Have you checked it is CH <sub>4</sub> free? We now have added the composition (20.5±0.5% O <sub>2</sub> in N <sub>2</sub> ). We had also checked that it is CH <sub>4</sub> free.
[Referee #3] [Hoheisel et al.]	P3, line 20: do you keep the 15min or is there a stabilization time? Yes, there is a stabilisation time, so we cut off the first 5 minutes. We have included the following sentence. <i>"However, only the last 10min were used to take into account the stabilisation time."</i>
[Referee #3] [Hoheisel et al.]	P3, line 25: decabon. Done.
[Referee #3] [Hoheisel et al.]	P3, line 33: 160ml/min to be consistent with the previous part. Changed as suggested.
[Referee #3] [Hoheisel et al.]	P4, line 5: How the measurement precision can be better in replay mode than in monitoring mode with the same instrument? Please clarify? The measurement precision of the analyser is the same in replay and in monitoring mode. What we wanted to point out is that in replay mode we have a higher time resolution and so more data points to describe the peak. The higher amount of data points also leads to a higher precision of the determined isotopic source signature. Since the sentence is misleading, we removed the phrase " <i>and a better precision</i> ".
[Referee #3] [Hoheisel et al.]	P4, line 30: have you tested the Nafion for potential fractionation? Yes, our tests did not show a fractionation.
[Referee #3] [Hoheisel et al.]	P5, line 29: Assan et al. 2017 showed that the intercept changes in time due to baseline drift. Have you regularly checked it? During the testing phase the intercept stayed constant.

[Referee #3] [Hoheisel et al.]	P5, line 33: please, check the unit. Corrected.
[Referee #3] [Hoheisel et al.]	P6, line 1: what do you mean by fully? We replaced the phrase “fully” with “yet”.
[Referee #3] [Hoheisel et al.]	P6, line 6: you cannot get a concentration range with a single cylinder. Please reformulate. Yes, we changed it. <i>“The gas cylinder used for calibration was chosen according to the experiment to ensure a similar composition and similar concentrations for sample and standard”.</i>
[Referee #3] [Hoheisel et al.]	P6, line 8: as I understood, a one point calibration strategy is used, meaning that you assume that the instrument has a linear response through the all measurement scale (mobile and sample measurements). Then why two different cylinders are used as calibration gases? We only used one calibration cylinder for each calibration. We use a cylinders for samples with atmospheric CH <sub>4</sub> concentrations and a different one for samples around 10ppm CH <sub>4</sub> since we have noticed that the instrument drift in $\delta^{13}\text{CH}_4$ is stronger for lower CH <sub>4</sub> concentrations.
[Referee #3] [Hoheisel et al.]	P6, line 10: have you seen some changes in the CRDS regime before and after each experiments? What was the maximal drift observed and how are they taking into account? The maximal drift of the CH <sub>4</sub> concentration was around 0.3ppb. We now added the phrase: <i>“To take into account possible drifts during the measurement we determined the time function of the standard (<math>\delta^{13}\text{CH}_4</math> Standard), used in the one point calibration, for each measuring point with a linear interpolation between the two calibration measurements.”</i>
[Referee #3] [Hoheisel et al.]	P6, line 4 to 11: it is not clear to me how the CH <sub>4</sub> is calibrated. There is no CH <sub>4</sub> calibration factor in Table1. Yes, we have changed the text for better understanding and we added a short explanation according the one point calibration. <i>“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.”</i> <i>“The gas cylinder used for calibration was chosen according to the experiment to ensure a similar composition and similar concentrations for sample and standard.”</i>
[Referee #3] [Hoheisel et al.]	P6, line 25: Please, detail how these uncertainties are calculated. We calculated these relative increase by comparing the error of $\delta^{13}\text{CH}_4$ before and after the correction and calibration. We changed the text to: <i>“Due to the correction and calibration <math>\delta^{13}\text{CH}_4</math> there is a relative increase in the uncertainty of <math>\delta^{13}\text{CH}_4</math> of approximately 3 to 12% for H<sub>2</sub>O concentrations below 1.3% and atmospheric CH<sub>4</sub> concentrations.”</i>
[Referee #3] [Hoheisel et al.]	P6, line 32: is it the last 10min over the 15min measurements? Yes, corrected.
[Referee #3] [Hoheisel et al.]	P7, line 5: is it a linear drift? The uncertainties is larger than the drift itself. Yes, because the uncertainties are larger than the drift itself we did not make a drift correction. We only use it to have an estimate how long a sample can be stored in the sample bag and in addition to quantify if a bag is leaky.
[Referee #3] [Hoheisel et al.]	P7, line 19: please clarify which uncertainties you are talking about. Ok, we have corrected it to “fit uncertainties”.
[Referee #3]	P7, line 23/24: were you driving while analyzing the AC? Micro-vibrations due to vehicle motion degrade the CRDS performances.

[Hoheisel et al.] No, the vehicle stands while analysing the AC. We also noticed that especially the measurement of  $C_2H_6$  is not as good as in the laboratory.

[Referee #3] P7, line 25: do you mean uncertainties? Is that calculated only from the fits?  
[Hoheisel et al.] Yes, we changed it to "*fit uncertainty*".

[Referee #3] P8, line 17: isotopic signatures are not directly measured.  
[Hoheisel et al.] Yes, we have changed it to: "*the isotopic signatures of  $CH_4$  from the AirCore measurements*".

[Referee #3] P8, line 33: do you mean the fit error? Or is there more parameters used to derived the uncertainties?  
[Hoheisel et al.] Yes, we meant fit error. Corrected.

[Referee #3] P9, line 10: what precision?  
[Hoheisel et al.] Clarified. We added "*the precision of the determined  $\delta^{13}CH_4$  signature*".

[Referee #3] P10, line 17/18: these criteria are already described earlier, no need to state it again here.  
[Hoheisel et al.] Ok. We still keep the criteria, because we find them importance enough to remind them again in this context.

[Referee #3] P10, line 20: why is the daily mean used and not the single signatures? Same p13 line 6.  
[Hoheisel et al.] We discuss the single signatures as well as the averaged daily mean values for each source. Because we had taken a different number of AirCore measurements per day and per site, the simple average over all samples can be biased compared to the average over the daily mean values.  
By changing the phrase in P10 line 20 (see below) and including the phrase "*averaged daily mean*", we try to make it easier to understand in the.  
"*During each measurement day one to five AirCore measurements were carried out at selected  $CH_4$  sources. In addition to the single signatures the averaged daily mean isotopic signatures of each  $CH_4$  source were calculated (see Fig.9, Table 2, and Supplement TableS1)*".

[Referee #3] P11, line 30: what is the peak height of the third AC? Please replaced value with signature.  
[Hoheisel et al.] Done. The three peak heights are 8.3, 8.5 and 8.9ppm.

[Referee #3] P14, line 10; replace the dot with and.  
[Hoheisel et al.] Done.

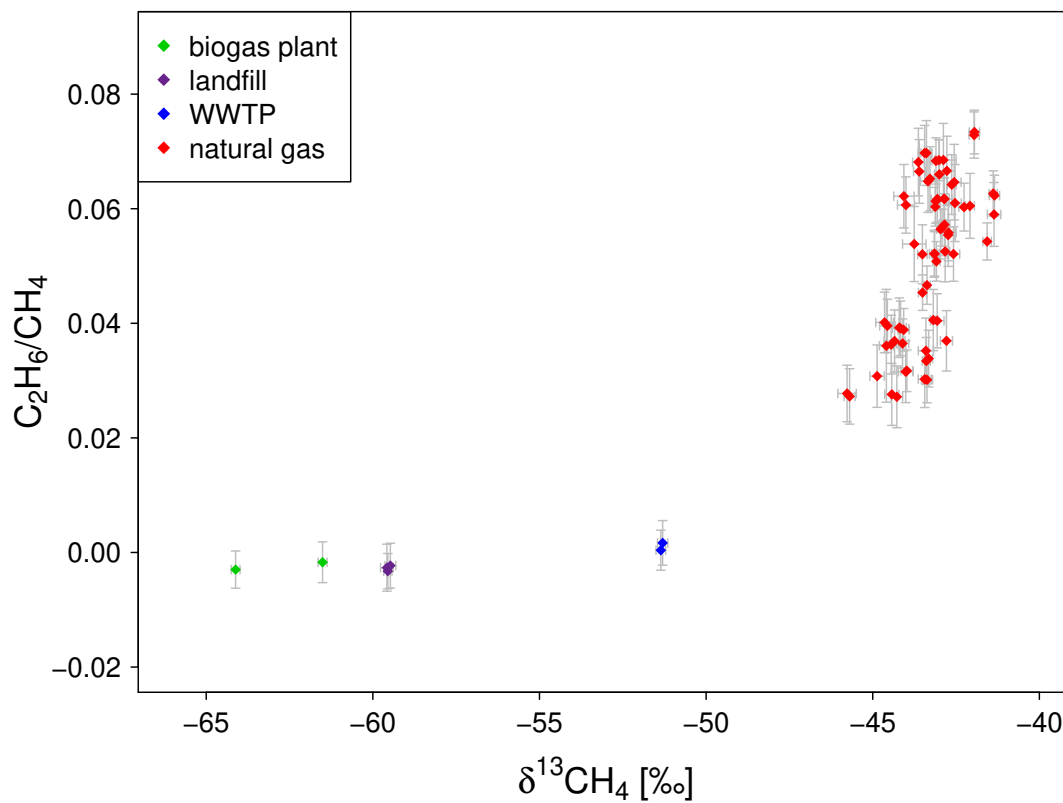
[Referee #3] P14, line 11: I suggest to replace monitor by sample.  
[Hoheisel et al.] Yes, corrected.

[Referee #3] P14, line 15: choose between "always" and "mostly".  
[Hoheisel et al.] OK, I removed both.

[Referee #3] P14, line 16: only peak heights are measured, not fluxes. I would then delete "therefore" and add "from these natural gas facilities".  
[Hoheisel et al.] Yes, corrected.

[Referee #3] P15, line 3: what is the detection limit of the system? Are you sure there is plumes coming out?  
[Hoheisel et al.] Ok, we changed the text.  
"*Around the opencast mine Hambach, the  $CH_4$  concentration varied only slightly between 1.94 and 1.98 ppm when we measured upwind as well as downwind of the pit. Therefore, it was not possible to identify an emission peak.*"

[Referee #3] P15, line 6: check for typo.  
[Hoheisel et al.] Done.



**Figure 7.** Isotopic signature and  $\text{C}_2\text{H}_6$  to  $\text{CH}_4$  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.