Atmospheric methane isotopes identify inventory knowledge gaps in the Surat Basin, Australia, coal seam gas and agricultural regions

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Replies to Reviewer 1

Comment 1

- This paper describes inflight measurements of atmospheric methane, which are particularly challenging, but can provide insights on the regional methane budget and on the main local methane sources. The application of the multi-Keeling model regression is of great interest and has been found useful to define the local background, given the difficulties in specifying a background in an area with such a multitude of sources.

- Authors' response

   We thank Reviewer 1 for spending considerable time reviewing the manuscript and for the well-considered comments. In the replies below we have discussed how we have used many of the constructive comments to refine the manuscript and improve the scientific insights.

- Authors' changes

   No change required.

Comment 2

- One of the aims of this study is the attribution of new isotopic signatures to methane sources, as stated in few parts of the paper. However, I would strongly weaken this statement, as the little CH4 enhancements between samples lead to a very high uncertainty and therefore a large isotopic range. I would focus the study more on the identification of potential new sources that are not accounted in the inventories and on the quality of the measurement technique. I would also add a paragraph explaining how to better constrain the source isotopic signatures (e.g. collecting more samples to constrain better the keeling line? Is it possible to collect a smaller bag than 3L ? Perhaps explain better the reason why more samples could not be collected. I am not an expert of inflight measurements, I would need more clarification in the text).

- Authors' response

   We have added in the Abstract, Introduction and Summary that the primary aim was to identify inventory knowledge gaps, and that the secondary aim was to investigate whether IFAA samples collected downwind of predominantly similar sources were useable for characterising the $\delta^{13}C_{CH4}$ signature of CH$_4$ sources, and to identify mitigation opportunities.

Under methods in Section 2.4 we added a paragraph about the sampling logistics and constraints. In the new summary, we say that this secondary objective had mixed results.

- Authors' changes

In the Abstract we now write:

"Secondary aims were to investigate whether IFAA samples collected downwind of predominantly similar inventory sources were useable for characterising the isotopic signature of CH_4 sources ($\delta^{13}C_{CH4(s)}$), and to identify mitigation opportunities."

Added to Section 2.4

"When collecting IFAA samples there are many sampling and logistical challenges. We collected 3 L samples of air to enable both on-site testing and accurate laboratory measurements, and we used SKC FlexFoil PLUS bags to reduce the cost of the project. Also, because the air samples were collected manually and stored in the cockpit, the number of samples collected in each sampling run was limited to a maximum of ~15. A purpose-built sampling system that rapidly fills 1 L canisters would potentially enable in-plume higher mole fraction IFAA samples to be collected. The smaller canisters would also allow for more samples to be collected each flight. More in-plume samples with higher CH_4 mole fraction values would reduce the uncertainty in the derived $\delta^{13}C_{CH4(s)}$ signatures. However, if the plume is heterogenous there is also a risk that rapidly filling the canisters will not sample the highest mole fraction portions of the plume."

Added to Summary

"An objective of this study was to use IFAA samples **to investigate whether** we could characterise the $\delta^{13}C_{CH4}$ source signature of emissions from facilities that could not be sampled during the ground campaign (Lu et al. 2021), especially the CSG regions that are remote from public roads. **To achieve this objective, we had to produce a BU inventory of both point and diffuse CH_4 sources for the region.** This inventory enabled us to sort the IFAA samples into sets based on the predominant 2-hour upwind inventory source of CH_4 (e.g., one sample per feedlot, for multiple feedlots). We were then able to determine the $\delta^{13}C_{CH4(s)}$ signature for a single source category. **The method worked with mixed results.**

A concern after the measurements of the IFAA samples in the laboratory was that the lack of $CH_{4(a)}$ enhancement above $CH_{4(b)}$ (less than 0.04 ppm) would not allow for the interpretation of these data using the Keeling plot method. Establishing $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$, as traditionally done from the collated data sets, was not possible by fitting the Keeling model (Eq. 1) or the Miller-Tans model (Eq. 2) to individual data sets (this is demonstrated in Appendix B). We overcame this challenge with careful sample quality control and by using multi-Keeling-model regression with shared $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$. An interpretation in alignment with other ground and continuous airborne observations was possible only after applying this regression algorithm. Importantly, despite the low $CH_{4(a)}$ enhancement of less than 0.04 ppm the derived values for background air $CH_{4(b)}$ = 1.826 ppm (CI 95 % ± 0.037 ppm) and $\delta^{13}C_{CH4(b)}$ = −47.3 ‰ (CI 95 % ± 0.3 ‰)

match independent observations. Being able to assign a well-constrained value to $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$ was central to the interpretation of all IFAA samples.

The derived $\delta^{13}C_{CH4(s)}$ values for the 250–350 mAGL IFAA sample sets (Figs 5 (a), 6 (a) and 6 (b); Table A6) where the inventory was dominated by CSG facilities or grazing cattle were close to those determined from the ground-based analysis of plumes (Lu et al. 2021). **It can be concluded that the upwind inventory for these samples was reasonably well characterised.**

For IFAA samples collected downwind of the feedlots the derived multi-Keeling-model regression $\delta^{13}C_{CH4(s)}$ signature was isotopically lighter than expected by approximately 5 ‰. **However, this category was poorly constrained and had a large 95 % confidence interval ranging from -92.2 ‰ to -47.0 ‰. A better data set is required to characterise the population statistics for feedlot CH₄ emissions, especially since there are no uniform procedures for feedlot design and waste management.**

The results for the 100–200 mAGL altitude IFAA samples where the inventory was dominated by CSG facilities or grazing cattle did not match expectations and were isotopically lighter than expected (Figs 5 (a), 6 (a) and 6 (b); Table A6). There are many possible explanations that cannot be resolved using currently available data. The mismatch could be due to there being more than one dominant source category in the upwind region **(with potential inputs from beyond the 2-hour back trajectory)**, incomplete mixing of all sources, sources missing from the BU inventory, the applied emission factors used for source apportionment not being precise for the individual source, or the $\delta^{13}C_{CH4(s)}$ signatures from the few plumes sampled as part of the ground-based studies not being representative of the complete population statistics.

To constrain the interpretation, for each CH₄ source the population distribution for both $\delta^{13}C_{CH4}$ and δD_{CH4} needs to be better characterised. These data would enable the statistical modelling of inventories for better comparison with IFAA sample $CH_{4(a)}$ and $\delta^{13}C_{CH4(a)}$ data and be useful for atmospheric transport isotope mixing model studies, which have the potential to yield more insights about inventory knowledge gaps compared to the pragmatic methods used in this study. **Due to the low enhancement in the mole fraction and the small number of samples collected with predominantly one source category upwind, the derived $\delta^{13}C_{CH4(s)}$ signatures have large uncertainties. For the methods presented in this study to work more effectively, more samples are needed downwind of each source category, and the sampling containers should be filled as rapidly as possible.**

A primary aim of the study was to see if the IFAA samples would be useful for identifying overlooked sources of CH₄ and this was achieved. In Fig. 3 (c) **three points of interest** were identified for their relatively low $\delta^{13}C_{CH4(a)}$ values: IFAA samples 1604, 1906 and 2103. **Although this is a small subset, the insights obtained are important.**

- Another issue that I think should be addressed more is the mismatch between the samples collected at different heights. It looks that in some cases there is a mismatch between the calculated footprint area and the observed area, some sources might have entered the domain and some other maybe not included. It is not the scope of this paper, but for few sources, forward modelling would help to see if some emission plumes would have been captured during the flight.

- Authors' response

    We extend the comments about the mismatch as shown below. Please refer to the supplementary material in Neininger et al. (2021) where we discuss forward modelling of the BU inventory. For the forward modelling, we only modelled the CH_4 mole fraction, not the mixing of many sources with different isotopic signatures, because the population statistics for the $\delta^{13}C_{CH4(s)}$ signatures are not well characterised for the region as discussed in this paper and Lu et al. (2021). We also modelled the back trajectories for each IFAA sample. We show this for all IFAA samples collected on the 16th Sep 2018 in Neininger et al. (2021) supplementary material Figure SF28. We used these probability contribution calculations in Neininger et al. (2021) to determine the pragmatic 2-hour limit used in this paper. We made the switch to HYSPLIT in this paper, so that others could replicate the procedures presented in this manuscript.

    When updating the figures for revised manuscript, we discovered an error with the map placement of sample 1808. This has been corrected and all calculations were redone. We can now explain the difference in the derived $\delta^{13}C_{CH4(s)}$ signatures for *Grazing Cattle >50 % BTF BU inventory, 100–200 mAGL and Grazing Cattle >50 % BTF BU inventory, 250–350 mAGL.*

    We still cannot explain the difference between the *CSG >50 % BTF BU inventory, 100–200 mAGL and CSG >50 % BTF BU inventory, 250–350 mAGL* sets. As we discuss in the manuscript, further research is required to better understand the methane source and mixing processes in the region.

- Authors' changes

    We added at the base of section 4.1.3 (now 3.3.3)

    " An additional possibility is that the air upwind of the 2-hour limit is really a blend of background and other upwind sources, and that the extent of enhancement of the air entering the 2-hour limit was enough to invalidate the assumption of predominantly two-endmember mixing. Thus, an apparent source signature has been determined (Vardarg et al. 2016). This possibility could be examined using a multisource transport model."

    We have added the following new section

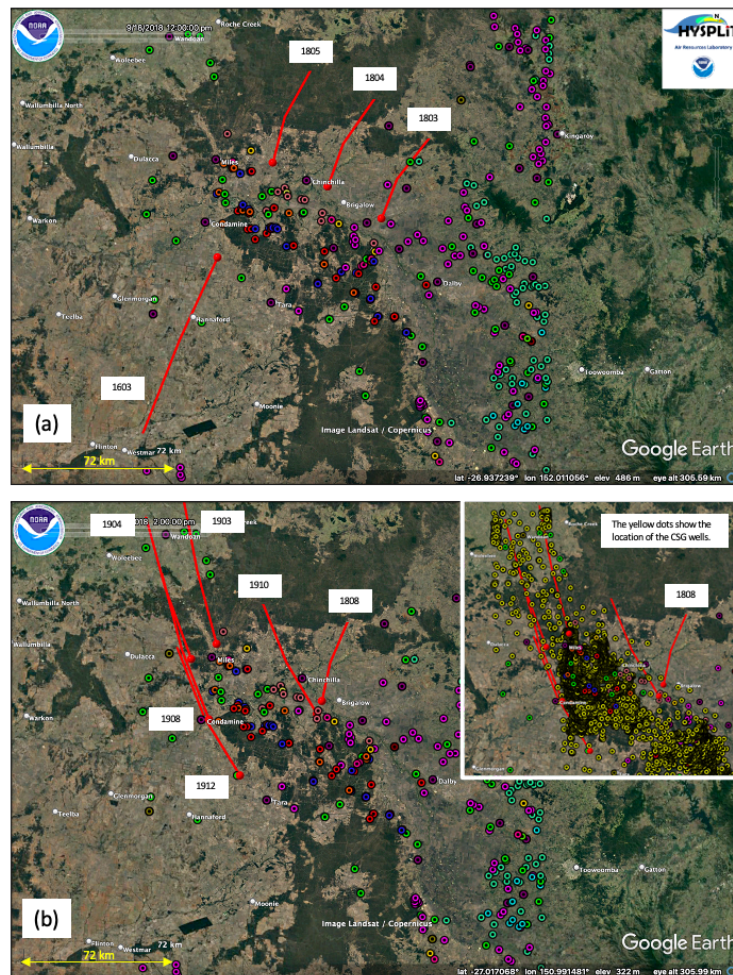
    3.3.5 Grazing Cattle >50 % BTF BU inventory, 100–200 mAGL

    The multi-Keeling-model regression $\delta^{13}C_{CH4(s)}$ signature for the category *Grazing Cattle >50 % BTF BU inventory, 100–200 mAGL* was −53.8 ‰ (CI 95 % ± 17.4 ‰, Figs. 5 (a) and 6 (b) red line). This is too isotopically heavy for cattle and is closer to the expected value for CH_4 emissions from CSG. Referring to Figs. 1 (a) and A4 (b) there are three possibilities that need further investigation.

The most likely explanation consistent with the source being within the 2-hour BTF area is that there are numerous CSG production wells and associated gas pipelines and co-produced water pipelines, which have many high-point vents, immediately upwind of IFAA samples 1903, 1904, 1908, 1910 and 1912. Thus, there are numerous locations where venting could have been occurring on the day. In support of local CSG production causing the heavier than expected signature, IFAA sample 1808 plots on the grazing cattle line in Figs 5 (a) and 6 (b) and it has no CSG wells upwind (refer to the upper right inset Fig. A4 (b)).

The second potential explanation is larger than expected urban CH_4 emissions. IFAA sample 1910 is downwind of Chinchilla (population ~6000), and 1912 is downwind of the towns of Condamine (population ~400), and Drillham (population ~130). In Table 2 four domestic sources of CH_4 could be contributing to the heavier than expected $\delta^{13}C_{CH4(s)}$ signature.

The third possible explanation is that CH_4 emissions from the north-western Surat Basin CSG facilities have been sampled in the north of the study area on 19th Sept 2018. Just beyond the 2-hour back trajectories shown in Fig. A4 (b) the air parcels would have travelled over the largest northwest Surat Basin gas fields near Woleebee Creek, which contains CSG plants, distribution hubs, and water treatment facilities. However, with reference to the modelling in Neininger et al. (2021), this is less likely compared to the first explanation that there are greater local CSG emissions than estimated in the inventory."

New Figure A4. The yellow dots are coal seam gas wells.

Comment 4

- Overall, the method and results are thoroughly described, and given the importance of the findings included in this study, I would recommend this manuscript for publication after addressing the issues that I mentioned above and the following comments:

- Authors' response

    The authors thank Reviewer 1 for carefully reading the manuscript, the constructive comments and for recommending publication.

- Authors' changes

    No changes are required for this comment. We have made many changes in response to the other comments below.

Comment 5

- Abstract: it is too long. I am not sure there is a word limit but I think it could be heavily shortened.

- Authors' response

    The original abstract was 700 words. We have reduced the abstract to 497 words.

- Authors' changes

The revised abstract is below.

"In-flight measurements of atmospheric methane ($CH_{4(a)}$) and mass balance flux quantification studies can assist with verification and improvement of UNFCCC National Inventory reported CH_4 emissions. In the Surat Basin gas fields, Queensland, Australia, coal seam gas (CSG) production and cattle farming are two of the major sources of CH_4 emissions into the atmosphere. Because of the rapid mixing of adjacent plumes within the convective boundary layer, spatially attributing $CH_{4(a)}$ mole fraction readings to one or more emission sources is difficult.

The primary aims of this study were to use the $CH_{4(a)}$ isotopic composition ($\delta^{13}C_{CH4(a)}$) of in-flight atmospheric air (IFAA) samples to assess where the bottom-up (BU) inventory developed specifically for the region was well characterised, and to identify gaps in the BU inventory (missing sources, or over- and underestimated source categories). Secondary aims were to investigate whether IFAA samples collected downwind of predominantly similar inventory sources were useable for characterising the isotopic signature of CH_4 sources ($\delta^{13}C_{CH4(s)}$) and to identify mitigation opportunities.

IFAA samples were collected between 100–350 m above ground level (mAGL) over a 2-week period in September 2018. For each IFAA sample the 2-hour back trajectory footprint area was determined using the NOAA HYSPLIT atmospheric trajectory modelling application. IFAA samples were gathered into sets, where the 2-hour upwind BU inventory had >50 % attributable to a single predominant CH_4 source (CSG, grazing cattle, or cattle feedlots). Keeling models were globally fitted to these sets using multiple regression with shared parameters (background air $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$).

For IFAA samples collected from 250–350 mAGL altitude, the best-fit $\delta^{13}C_{CH4(s)}$ signatures compare well with the ground observation: CSG $\delta^{13}C_{CH4(s)}$ −55.4 ‰ (CI 95 % ± 13.7 ‰) versus $\delta^{13}C_{CH4(s)}$ −56.7 ‰ to −45.6 ‰; grazing cattle $\delta^{13}C_{CH4(s)}$ −60.5 ‰ (CI 95 % ± 15.6 ‰) versus −61.7 ‰ to −57.5 ‰. For cattle feedlots, the derived $\delta^{13}C_{CH4(s)}$, −69.6 ‰ (CI 95 % ± 22.6 ‰), was isotopically lighter than the ground-based study ($\delta^{13}C_{CH4(s)}$ from −65.2 ‰ to −60.3 ‰), but within agreement given the large uncertainty for this source. For IFAA samples collected between 100–200 mAGL the $\delta^{13}C_{CH4(s)}$ signature for the CSG set, −65.4 ‰ (CI 95 % ±13.3 ‰), was isotopically lighter than expected, suggesting a BU inventory knowledge gap or the need to extend the population statistics for CSG $\delta^{13}C_{CH4(s)}$ signatures. For the 100–200 mAGL set collected over grazing cattle districts the $\delta^{13}C_{CH4(s)}$ signature, −53.8 ‰ (CI 95 % ± 17.4 ‰), was heavier than expected from the BU inventory. An isotopically light set had a low $\delta^{13}C_{CH4(s)}$ signature of −80.2 ‰ (CI 95 % ± 4.7 ‰). A CH_4 source with this low $\delta^{13}C_{CH4(s)}$ signature has not been incorporated into existing BU inventories for the region. Possible sources include termites and CSG brine ponds. If the excess emissions are from the brine ponds, they can potentially be mitigated. It is concluded that in-flight atmospheric $\delta^{13}C_{CH4(a)}$ measurements used in conjunction with endmember mixing modelling of CH_4 sources are powerful tools for BU inventory verification."

Comment 6

- Line 82: BU, I don't think the acronym has been explained in the text above. Expand for readers who don't know what you are referring to e.g emission factors x statistics.

- Authors' response

    We added some definitions in the sentences before line 82.

- Authors' changes

    "Inventories prepared using the national and IPCC emission factors are commonly called bottom-up (BU) emission estimates (Neininger et al. 2021), and an emission factor is a coefficient that quantifies the emissions or removals of a gas per unit activity (IPCC, 2006, 2019)."

Comment 7

- Line 122: how can these challenges been tackled?

- Authors' response

    With currently readily available sampling systems, tackling these challenges is not easy or cheap. We have extended the paragraph to address this comment.

- Authors' changes

    "To reduce the uncertainty in the derived $\delta^{13}C_{CH4(s)}$ signatures, ideally many samples would be collected in a plume from a known source, and these discrete samples would be rapidly collected (as fast as possible). However, when collecting IFAA samples there are often numerous CH_4 sources upwind; it takes time to fill the sample collection bags (resulting in a sampling window in the order of kilometres); assumptions must be made about the mixing of air parcels within the convective boundary layer; and it is often not possible to sample enough points to minimise the uncertainty of $\delta^{13}C_{CH4(s)}$ signature estimates."

Comment 8

- Line 197: "distributed sources". These are explained later in the text, but I would move some details here as the reader might be confused by the term "distributed".

- Authors' response

    We have added an example to explain distributed sources.

- Authors' changes

    "In Fig. 2 (a) all point sources (CSG facilities, feedlots, coal mines etc) are presented as an emission intensity map, and in Fig. 2 (b) the distributed sources are shown. Distributed sources are multiple small sources spread evenly over a subregion. For example, we know the total number of cattle within a statistical district (Condamine, Burnett-Mary, and Queensland Murray-Darling Basin) but not their locations, so the emissions are spread evenly using the population density. Comprehensive details about how the emissions from distributed sources were determined are discussed in Neininger et al. (2021) supplementary material Section S."

Comment 9

- 209: refer to the Figure including also the symbol color to help the reader i.e. "The largest individual source in an open pit....red square in Fig 2a.

- Authors' response

    The colour description has been added.

- Authors' changes

    "The largest individual source is an open-pit coal mine (27.28° S, 151.71° E, **red square**), which emits 843 kg h^{-1} (4.1 % of the UNSW inventory total). The second largest source is a feedlot (27.42° S, long. 151.14° E, **orange square**), which emits 563 kg h^{-1} (2.7 % of the UNSW inventory total). The largest CSG source is a raw water pond (26.96° S, 150.49° E, **light green square**), which emits 221 kg h^{-1} (1.1 % of the UNSW inventory total)."

Comment 10

- 215: I was wondering how Fig 2b was created. Then you explained that later in the text. I would mention briefly about the isotopic signatures attribution here and then describe more in detailed in the following paragraph.

- Authors' response

    Please refer to the reply for comment 8.

- Authors' changes

    Changes were incorporated into the reply for comment 8.

Comment 11

- 239: see my previous comment

- Authors' response

  We assume you are referring to the comment about "One of the aims of this study is the attribution of new isotopic signatures to methane sources, as stated in few parts of the paper. However, I would strongly weaken this statement, as the little CH4 enhancements between samples lead to a very high uncertainty and therefore a large isotopic range."

  We acknowledge there is large uncertainty in the determined isotopic source signatures, and this is fully documented throughout the paper. At line 239 in the submitted manuscript we write "examine if". We make no claim that this is the best way to determine the isotopic signatures. But there is merit in exploring if the sampling and analysis methodology works. In the accompanying paper Lu et al. (2021) we demonstrate how to obtain the isotopic signature of these sources with low uncertainty. We make no changes here, but we have added a paragraph to the summary.

- Authors' changes

  We added to the summary

  "To constrain the interpretation, for each CH_4 source the population distribution for both $\delta^{13}C_{CH4}$ and δD_{CH4} needs to be better characterised. These data would enable the statistical modelling of inventories for better comparison with IFAA sample $CH_{4(a)}$ and $\delta^{13}C_{CH4(a)}$ data and be useful for atmospheric transport isotope mixing model studies, which have the potential to yield more insights about inventory knowledge gaps compared to the pragmatic methods used in this study. Due to the low enhancement in the mole fraction and the small number of samples collected with predominantly one inventory source category upwind, the derived $\delta^{13}C_{CH4(s)}$ signatures have large uncertainties. For the methods presented in this study to work more effectively, more samples are needed downwind of each source category, and the sampling containers should be filled as rapidly as possible."

Comment 12

- 245: why? Can you include a reference?

- Authors' response

  We now briefly explain why the shallow coals contain methane with a biological signature and references have been added.

- Authors' changes

  "this is due to the displacement of the original CH_4 in coal seams nearest the ground surface with biologically derived CH_4 (Iverach et al. 2015, 2017)."

Comment 13

- 260: perhaps there are no studies on termite in this area, but I think there are some isotopic values in literature that you can refer to and you can include here (Monteil et al. 2011?).

- Authors' response

    Monteil et al. (2011) is not a primary source reference for termite emissions or their isotopic signature. In Table 1 of that paper, it lists a value of -57 ‰ for termites, which is high. Monteil et al. (2011) cite Sanderson et al. (1996) for their termite data, but Sanderson et al. (1996) did not locate any permil values for termite emissions. We now cite Sugimoto et al. (1998), which has data for Australian termites.

- Authors' changes

    We have added "For worker termites collected from mounds near Darwin, Australia, Sugimoto et al. (1998) reported $\delta^{13}C_{CH4(s)}$ values ranging from -88.2 ‰ to -77.6 ‰."

Comment 14

- Figure 3 b: what do the lines represent? The confidence bands? State that in the Figure caption.

- Authors' response

    In the caption for Figure 3 (b) we had described the confidence bands:

    "The linear regression fit highlights the moderate correlation ($R^2 = 0.59$) between the two variables. The grey zone is the 95% confidence level."

- Authors' changes

    We now provide a little more clarity:

    "The linear regression fit highlights the moderate correlation ($R^2 = 0.59$) between the two variables. The grey zone **between the two orange** lines is the 95% confidence level."

Comment 15

- 349: instead of using only a visual identification of outliers, I would quantify them using a statistic approach, so that the identification is more solid. It is not clear to me just looking at Figure 3 how these outliers have been selected.

- Authors' response

    As defined in the paragraph starting line 336 these are not all outliers based on statistical distributions, rather "IFAA samples of interest are those that have relatively high $CH_{4(a)}$ or different than expected $\delta^{13}C_{CH4(a)}$ (for brevity called outliers) because these samples may indicate over- or underestimation of CH_4 emissions in the BU inventory."

    We are simply looking at some of the samples with the highest mole fraction values, and a set with very light $\delta^{13}C_{CH4(s)}$ signatures. Because the term "outlier" has strong statistical associations, we have replaced the word outlier with "points of interest"

- Authors' changes

    Throughout the manuscript we replace "outlier" with "points of interest"

Comment 16

- 373: add "see appendix X".

- Authors' response

  This cross reference has been added.

- Authors' changes

  The sentence now reads:

  "Regression of a single source data set is poorly constrained, resulting in large uncertainties in the derived $\delta^{13}C_{CH4(s)}$ due to the low enhancement above background, less than 0.040 ppm, and the small number of samples in each category (**Appendix B**)."

Comment 17

- 415: include here the Neininger background value.

- Authors' response

  The background values reported in Neininger et al. (2021) are reported lower in the same paragraph.

- Authors' changes

  We deleted the sentence:

  "Here, we provide further information as context for background $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$ for comparison with the Keeling model results."

  This edit then joins the two sentences about the background values reported in Neininger et al. (2021) to read:

  "Subsequent analysis of all the IFAA samples indicated that none of the IFAA samples matched the low CH_4 mole fractions recorded in Neininger et al. (2021). The background CH_4 mole fraction recorded in continuous airborne surveys in Neininger et al. (2021) was stable between days and varied between 1.822 ppm and 1.827 ppm."

Comment 18

- 533: again refer to the figure colors. "within the range listed in table A2, grey in Fig 6"

- Authors' response

  This is a sensible suggestion.

- Authors' changes

  The colour cross references have been added for all categories.

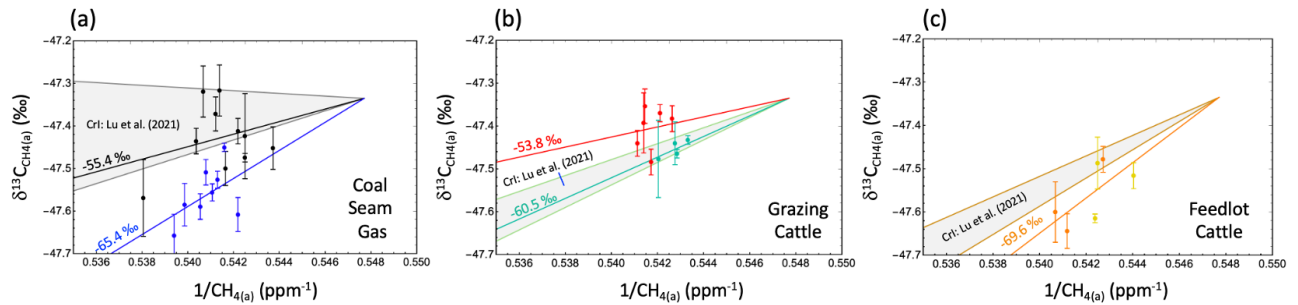
Comment 19

- Fig 6: include a figure title for each plot "Gracing Cattle; Feedlots…"

- Authors' response

    The journal style is not to use titles at the tops of a figure. We have added inset labels.

- Authors' changes

    Labels have been added to the plots.

Comment 20

- Line 555: the isotopic signature was…(blue line)

- Authors' response

    Refer to comment 19

- Authors' changes

    Refer to comment 19

Comment 21

- Line 562: maybe for the high altitude samples the footprint is different and you see different sources? See my previous comment

- Authors' response

    We discuss this point in the paragraphs that immediately follow line 562. Also refer to Figures A3 to A5 and Table A3 (now Tables A3, A4 and A5), which all show that the upper and lower altitude samples do not sample the identical sources.

- Authors' changes

    No change was made.

Comment 22

- 588: again include the line color

- Authors' response

    Refer to comment 19

- Authors' changes

    Refer to comment 19

Comment 23

- 526: Fig 5 a?

- Authors' response

  This error is a legacy cross reference from where we changed the order of the figures compared to an earlier draft.

- Authors' changes

  Fig. 5 (b) corrected to Fig. 5 (a).

Comment 24

- 740: also some atmospheric transport modelling would address this issue.

- Authors' response

  We fully agree that atmospheric transport modelling would be a useful tool for providing additional insights. We did extensive forward and inverse modelling, and the results are presented in Neininger et al. (2021). However, we did not have enough data for reliable atmospheric transport isotope mixing modelling. We have added a comment about this to the end of that paragraph.

- Authors' changes

  Added to the summary

  " To constrain the interpretation, for each CH_4 source the population distribution for both $\delta^{13}C_{CH4}$ and δD_{CH4} needs to be better characterised. These data would enable the statistical modelling of inventories for better comparison with IFAA sample $CH_{4(a)}$ and $\delta^{13}C_{CH4(a)}$ data and be useful for atmospheric transport isotope mixing model studies, which have the potential to yield more insights about inventory knowledge gaps compared to the pragmatic methods used in this study."

Reviewer Acknowledgement

All authors would like to thank reviewer 1 for providing constructive comments. We hope the edits documented above have addressed your concerns and enhanced the value of the manuscript. Your help with refining the lucidity of the manuscript will enhance the impact of the scientific outcomes.