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

Bryce F. J. Kelly, Xinyi Lu, Stephen J. Harris, Bruno G. Neininger, Jorg M. Hacker, Stefan Schwietzke, Rebecca E. Fisher, James L. France, Euan G. Nisbet, David Lowry, Carina van der Veen, Malika Menoud, Thomas Röckmann

https://acp.copernicus.org/preprints/acp-2022-552/

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 CH_4 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 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 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 CH_4 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.**

Comment 3

- 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 Neinginger 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

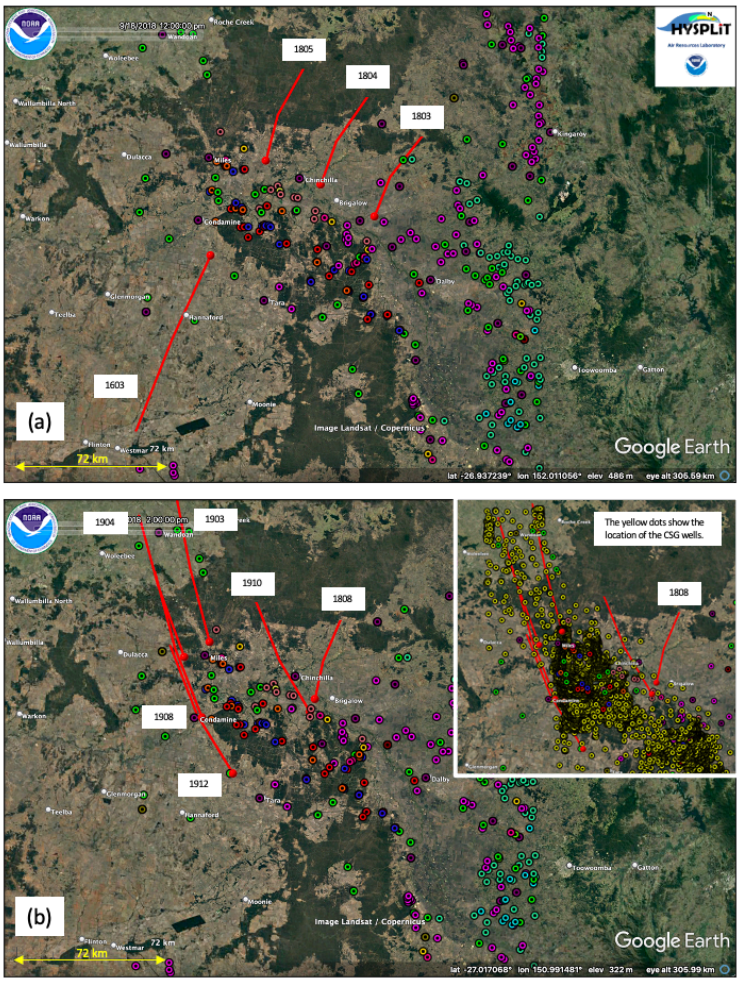
  The multi-Keeling-model regression $\delta^{13}C_{CH4(s)}$ signature for the category *Grazing Cattle >50 % BTF BU inventory, 100–200 mAGL* was −53.8 ‰ (CI 95 % ± 17.4 ‰, Figs. 5 (a) and 6 (b) red line). This is too isotopically heavy for cattle and is closer to the expected value for CH_4 emissions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

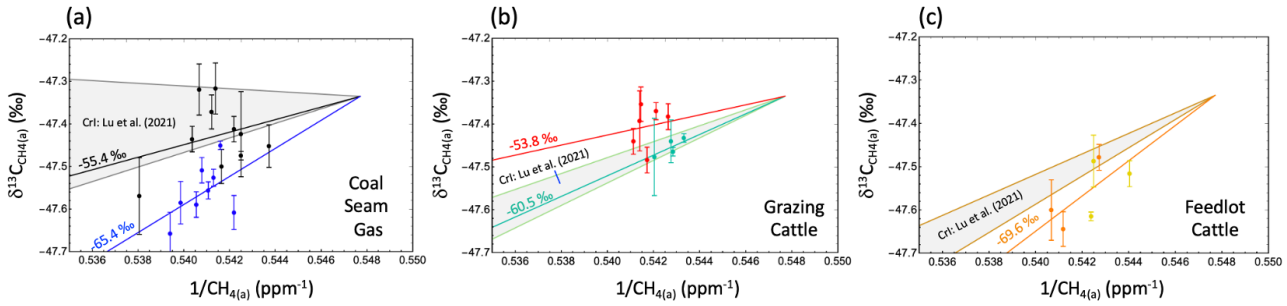
Comment 19

- 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.

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Replies to Reviewer 2

Comment 1

- Review of "Atmospheric methane isotopes identify inventory knowledge gaps in the Surat Basin, Australia, coal seam gas and agricultural regions" by Kelly et al., for publication in Atmospheric Chemistry and PhysicsThe authors used measurements of CH4 and δ13CCH4 from samples collected during airborne surveys across the Surat Basin in Australia to study CH4 emissions from different types of sources in this region. This work is a follow up of two other studies: Lu et al. (2021) that estimated δ13CCH4 of single methane sources in the region using ground-based surveys and Neininger et al. (2021) that developed a bottom-up (BU) inventory of the CH4 emissions in the region. In this study, the authors identify in-flight atmospheric air samples with a predominant type of CH4 source based on a combination of HYSPLIT back trajectory footprints and the BU inventory developed in Neininger et al. (2021). They regroup samples with the same predominant CH4 source and sampling height into different sets. These sets are used in a multi-Keeling-model regression to estimate CH4 and δ13CCH4 of the background first, and then these background estimates are used to estimate δ13CCH4 of the different sets of samples. This manuscript is very ambitious as it aims at assessing the quality of the BU inventory, extending the knowledge of δ13CCH4 from sources difficult to access with ground-based surveys, and identifying mitigation opportunities using these samples containing low signals from a mix of CH4 sources. This is a laudable goal and will be of great interest to the journal's readership as the state of knowledge of coal seam gas (CSG) and cattle farming emissions and isotopic signatures is not very advanced. However, the manuscript currently requires multiple improvement before final publication. I recommend publishing it after addressing the comments listed below:
- Authors' response

  The authors thank Reviewer 2 for spending considerable time reading and commenting on the strengths and weaknesses of the study. Below we address your concerns. We appreciate your support for noting that this ambitious project will be of interest to the atmospheric and greenhouse gas inventory communities. Your comments have enhanced the clarity of the manuscript and made our scientific discussion more precise.
- Authors' changes

  No change is required.

Major comments

Comment 2

- The manuscript needs a bit of reorganization:
    - The end of the introduction needs to be reworked.
- Authors' response

    We deleted the last paragraph and shortened the 2nd last paragraph of the introduction.

- Authors' changes

    We reduced the last two paragraphs of the introduction to read:

    "A well-established method to determine the $\delta^{13}C_{CH4(s)}$ signature is to collect air samples within the plume downwind of the source and analyse the data using a two-endmember mixing model (Keeling, 1961; Pataki et al., 2003; Miller and Tans 2003). However, the airborne surveys were not designed to track individual plumes; the flight tracks were designed to optimise the results for regional mass balance estimates of CH_4 emissions (Neininger et al. 2021). For aircraft surveys that intersect multiple plumes we present an alternative method. Multiple IFAA samples were collected downwind of a predominant inventory source category, for example CSG or cattle feedlots, and these samples were analysed in sets, which is analogous to multiple samples in a plume. We demonstrate how to analyse these IFAA samples using a detailed BU inventory (presented in Neininger et al. (2021)), hybrid single particle Lagrangian integrated trajectory (HYSPLIT) modelling (Draxler et al., 1998) and multi-Keeling-model regression with shared parameters (background air $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$).

    We moved "(refer to Neininger et al. (2021) supplementary material Figure SF26 for an example of the more detailed back trajectory modelling, used to guide the HYSPLIT settings)." to Section 2.5.

Comment 3

- Some subsections from results could go to the "Material and methods' section.
- Authors' response

    This is a non-specific comment that we have addressed as part of Comment 4. We have kept this in mind with our responses to later Comments.

- Authors' changes

    No change was made directly relating to this comment. However, the results and discussion sections have now been combined and reorganised as requested in comment 4.

Comment 4

- The 'Results' and 'Discussion' sections should be merged to avoid the repetitions of the results and the discussion teasing (all of the sections talking about the background could be together: 3.1, first part of 3.3 and 4.1.1 for example).
- Authors' response

  To address any repetition, we have moved Section 3.1 and combined it with Section 4.1.1. The results and the discussion have been combined.

- Authors' changes

  Section 3.1 has been moved and added to Section 4.1.1 (now 3.1), and the results and discussion have been combined. This resulted in one paragraph of listed results being deleted, and we deleted the opening paragraphs for the old discussion section.

Comment 5

- Getting different isotopic signatures for a given source depending on the sampling height is very concerning regarding the ability of the method to correctly assess δ13CCH4. It is even more concerning since this problem occurs for the two type of sources. I would expect IPAA samples collected at lower heights to have higher methane signals since they are sampled closer to the sources and therefore making it a bit easier to distinguish methane sources with isotope techniques, not the other way around.
- Authors' response

  The data set is small, and we have good agreement with the ground observations for three out of five categories. Clearly the method has promise, and the methods will be refined in future studies. We see no issues with getting different $\delta^{13}C_{CH4(s)}$ signatures for the set at different altitudes. Each IFAA sample has collected air that has travelled over different sources.

  Please refer to Figure 3a. This figure plots the IFAA sample altitude (mAGL) versus the IFAA sample CH_4 (ppm). The simple relationship suggested by the reviewer of "I would expect IPAA samples collected at lower heights to have higher methane signals since they are sampled closer to the sources and therefore making it a bit easier to distinguish methane sources" is not present in the data collected. We note that two of the highest mole fraction readings were recorded for samples collected at ~300 mAGL, and the lowest was recorded for a sample at ~130 m. Throughout the campaign we had warm sunny days. There was rapid uplift and mixing of the plumes, and the IFAA samples were collected at various distances from the source. There is also the complication that the slow sample bag fill blends the air over many kilometres. We are not analysing discrete plume samples; we are analysing complex blends.

  It should also be noted that for the CSG samples the isotopically heavier samples were collected at the higher altitudes, and for the grazing cattle districts, the isotopically heavier samples were

collected at the lower altitudes. Many confounding factors are influencing the derived $\delta^{13}C_{CH4(s)}$ signatures. In the manuscript, we discuss at length the multiple interpretations.

We added a paragraph to the summary on how to improve the source signature characterisation.

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.* When updating the figures for the revised manuscript, we discovered an error with the map placement of sample 1808. This has been corrected and all calculations were redone. This also resulted in a closer examination of the lower altitude grazing cattle set. Please see the text below for details about why the two altitude sets have different $\delta^{13}C_{CH4(s)}$ signatures.

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 discussed in the manuscript, further research is required to explain these results.

- Authors' changes

  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)).

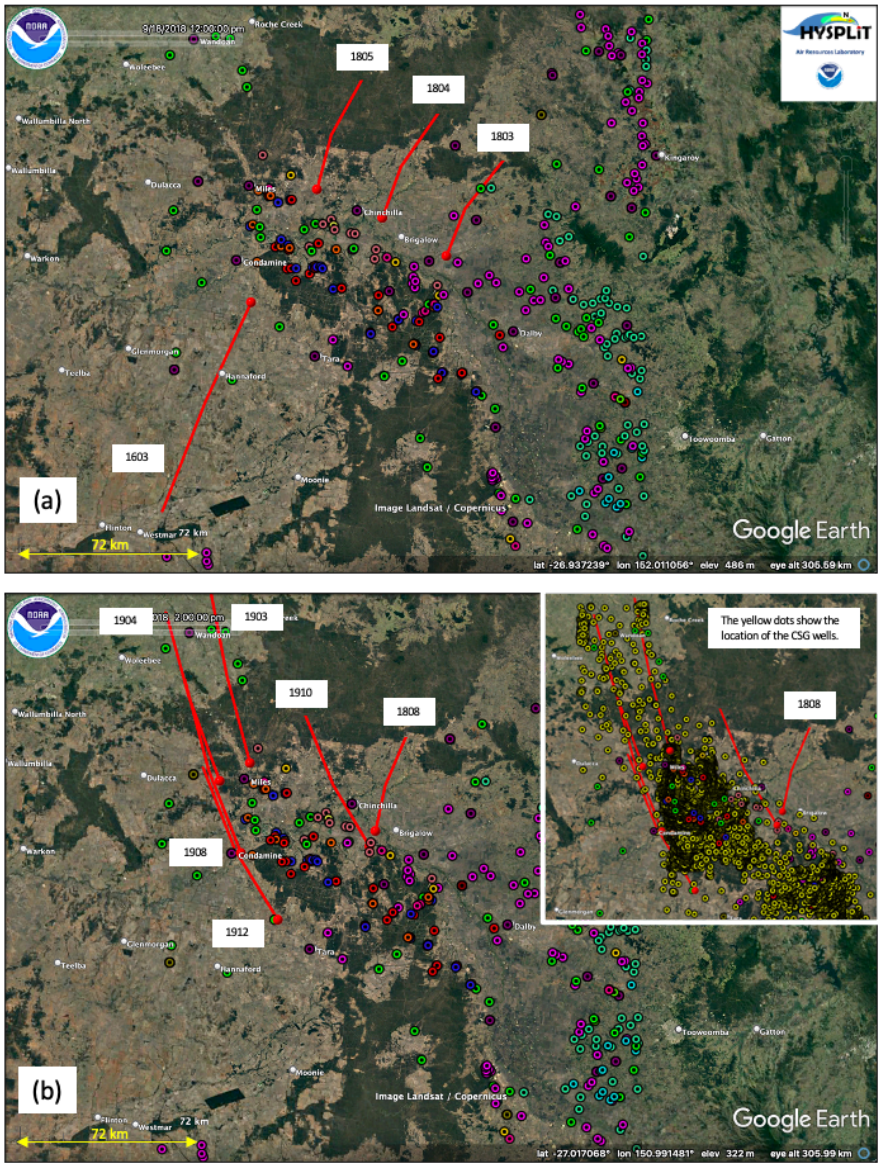
  The second potential explanation is larger than expected urban CH_4 emissions. IFAA sample 1910 is downwind of Chinchilla (population ~6000), and 1912 is downwind of the towns of Condamine (population ~400), and Drillham (population ~130). In Table 2 there are four domestic sources of CH_4 that could 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.

The following has been 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."

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



Comment 6

- Additional analysis/discussion should include:
    - The uncertainty on the estimated background values with the multi-Keeling-model regression (1.825 +/- 0.037 ppm and -47.3 +/- 0.3 ‰): Are these uncertainty include in the Keeling regression of the different sources? How does that affect the Keeling regressions for the different categories? If δ13CCH4 of the background signature was -47.0 ‰ or -47.6 ‰, this would impact all of the other sources signatures.
- Authors' response

    All the uncertainties are fully characterised, and this is comprehensively documented in the main text, in the graphs for the input data and in Tables A4 and A5 (Tables A6, A7 and A8 in the revised manuscript). We hope the following visual helps to clarify how the algorithm works. To find the optimal set and characterise the uncertainty the five input data sets (2 CSG, 2 Grazing Cattle, and 1 Feedlot) are analysed as a combined set. For each of the original data sets (now a subset) a line is defined by Equation 1 that will pass through the original subset (a spoke for each subset) and the shared $\delta^{13}C_{CH4(b)}$ and $CH_{4(b)}$ (the hub). The algorithm then adjusts simultaneously the position of each spoke and the position of the hub, to minimise the distance between each spoke and its associated data subset. As all the spokes are attached to a single hub there can only be one optimal solution set.

    In Mathematica the script line is:

    fitALL = ResourceFunction["MultiNonlinearModelFit"][inputData, {concBack (delBack - delSource1) x + delSource1, concBack (delBack - delSource2) x + delSource2, concBack (delBack - delSource3) x + delSource3, concBack (delBack - delSource4) x + delSource4, concBack (delBack - delSource5) x + delSource5}, {{delBack, seedValue}, {concBack, seedValue}, delSource1, delSource2, delSource3, delSource4, delSource5}, {x}]

    In the above script line, x is the $1/[CH_4]$ axis for the Keeling plot. The seed values are any random set for $\delta^{13}C_{CH4(b)}$ and $CH_{4(b)}$ that will start the global optimisation root mean square minimisation search near the global optimal solution (a guiding nudge into the real solution domain). For example, we could use -47 for delBack, and 1.8 for concBack. The algorithm is robust, so the seed values can be further from the final solution if preferred. No seed values are required for delSource1, delSource2 etc. There are many optimisation methods and thresholds that can be set. These are all explained in the Mathematica documentation.

    For the above fit Mathematica will produce a report with all the basic statistical measures including the standard error, confidence intervals, t-statistic, P value, among others.

    Regarding " If δ13CCH4 of the background signature was -47.0 ‰ or -47.6 ‰, this would impact all of the other sources signatures."

This is correct, and this is reflected in the large uncertainties returned for all reported $\delta^{13}C_{CH4(s)}$ and discussed in the text and presented in the result tables. In Figure 6, we have added the error bars for background air to highlight the large uncertainty in the derived values $\delta^{13}C_{CH4(b)}$ and $CH_{4(b)}$. Although the uncertainties are large, if you focus on the most probable value, insights about the inventory and overlooked sources are obtained.

- Authors' changes

  In Figure 6 we added the error bar for $\delta^{13}C_{CH4(b)}$ and $CH_{4(b)}$. A copy of all the revised figures are included at the end of this document.

  In the methods section we now write:

  "This algorithm **globally optimises** $\delta^{13}C_{CH4(s)}$ for each category and returns the shared values for $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$. Comprehensive details about the Mathematica MultiNonlinearModelFit function for fitting multiple data sets to multiple expressions that share parameters are available from the Wolfram function repository (Smit, 1986).

  When the multi-Keeling-model regression with shared parameters is **applied globally** to all category data sets the values for ……"

Comment 7

- Footprint calculation: is 2 hours enough? A footprint calculated from 250-350 mAGL will likely be larger than 100-200 mAGL due to the increase of wind speed with altitude. Maybe 2-hour BTF is not enough to properly capture sources that influence measurements at lower heights.
- Authors' response

  It would be a large separate study to find the optimal back trajectory cut-off for each IFAA sample. As discussed in the text, in Neininger et al. (2021) we did both extensive forward and inverse modelling. We used these results to guide setting the pragmatic 2-hour limit. We showed in Figure 3 (b) that there is a moderate correlation ($R^2 = 0.59$) between the BTF BU Inventory (kg h^{-1}) versus IFAA Sample CH_4 (ppm). If the 2-hour limit was an unreasonable cut-off, you would not get this moderate correlation.

- Authors' changes

  We added to Section 2.5 "Also refer to Neininger et al. (2021) supplementary material Figure SF26 for an example of the more detailed back trajectory modelling, used to guide the HYSPLIT settings."

Comment 8

- The IFAA samples contain a mix of methane coming from different types of sources, I am wondering if the 50% threshold to attribute a sample to a category is not too low (see below).
- Authors' response

  The category subset is already small, and this request makes them even smaller. Increasing the threshold to 70 % results in fewer points in the multi-Keeling-model regression, which dramatically increases the uncertainties. For example, for background air a 70% threshold yields $CH_{4(b)}$ 1.818 ± 0.094 (ppm) and $\delta^{13}C_{CH4(b)}$ -47.3 ± 1 ‰. Another tie-in point is that we know we had clean plume sampling for the feedlots. The 50% threshold returns values that are closer to those expected for both $CH_{4(b)}$, $\delta^{13}C_{CH4(b)}$ and $\delta^{13}C_{CH4(cattle)}$, and confidence intervals are smaller. The 50% threshold choice was guided by the better results for background air, the 250 to 350 mAGL grazing cattle districts (where there are few other sources), and the feedlot results.

  With respect to assigning a signature to the light isotope set (1604, 1906 and 2103) it does not matter if you use the 50% or 70% threshold. Both identify that the $\delta^{13}C_{CH4(s)}$ signature of the set will be ~-80 ‰.

  No matter what threshold is set, none of the IFAA samples will have clean two-endmember mixing. The BU inventory is a guide that there will be a predominant single inventory source. The 50% threshold used is a compromise, which allows for useful insights and reduced uncertainties in the derived values. Because the data set is small, and the derived $\delta^{13}C_{CH4(s)}$ signatures are sensitive to arbitrary thresholds, we have added statements on this issue throughout the manuscript. Ideally, we would have more IFAA samples, and footprint regions with a single predominant source above the 90% threshold. We originally aimed to analyse more samples, but due the low CH_4 enhancement above the background CH_4 mole fraction and to exclude any potential impacts from shipping the samples from Australia to the UK, we set a low 1 % threshold for the QA step. This tight QA threshold reduced the size of the data set analysed by approximately 50 %. The positive impact of setting this threshold is that for the samples analysed we have excellent agreement for the CH_4 readings on two analysers. We analysed a high-quality data set.

- Authors' changes

  We added to Section 2.7

  "The >50 % threshold was set to achieve a balance between reducing the uncertainty in the regression and having a predominant CH_4 source type in the upwind inventory. Ideally a higher threshold would be used, but this would require the collection of a greater number of IFAA samples than done in this study. The derived $\delta^{13}C_{CH4(s)}$ signatures for each category will be affected by the threshold, but the relative insights about a category being isotopically heavier or lighter will not."

Comment 9

- Could be interesting to try to see how different would be the results if 100-200 mAGL and 250-350 mAGL sets were merged for CSG and grazing cattle.
- Authors' response

  Blending the data from different altitudes is not ideal, as you are reducing the resolution of the information. We only blended for the feedlot samples to get enough data points to obtain the only insight possible, and these plumes were in several cases better isolated.

  We now show in the revised paper that the IFAA samples in the north have an input that is not grazing cattle. A re-examination of these trajectories highlights 3 potential sources: CSG production wells and associated pipelines (now considered the most likely source), urban emissions, or input from beyond the 2-hour trajectory (potentially the NW gas field). With this knowledge, blending the 100-200 mAGL and 250-350 mAGL sets for cattle is mixing source types. When you mix different source types, you get an apparent $\delta^{13}C_{CH4}$ signature (refer to Vardag et al. (2016) for a good discussion on blending sources and the impact on the derived $\delta^{13}C_{CH4(s)}$ signature). Although we never have clean two-endmember mixing for the data sets analysed in the paper, we have been careful with the filtering of the data to maximise one source mixing with background air for the analyses.
- Authors' changes

  No change was made to the text because this request does not enhance the interpretation.

Comment 10

- The story about the outliers and the identification of potential mitigation opportunities is a bit wobbly… I am not sure that associating these 3 outliers because they seem to align is really convincing.
- Authors' response

  It is clear from Figure 3 (c) that these points all have low $\delta^{13}C_{CH4(a)}$ values. It is also apparent in Figure 3 (c) and Figure 5 (a) that the three points all sit on a similar Keeling line. To find out the value of that Keeling line to get insights about these isotopically light samples we group them and fit a single Keeling line. The points may or may not have a common source type, but it is indisputable that they are all isotopically light samples. We answer the question of what the source signature for these three points is, and we determine that it is –80 ‰. As we discuss in the paper, we had no sources in the BU inventory with such a low signature. We then discuss possible overlooked sources, and we acknowledge that the hypothesised sources can only be verified with further field studies. This was one of the central aims of the research: we have identified an inventory knowledge gap. We now know from this study that there is at least one significant source in the region with an $\delta^{13}C_{CH4(s)}$ signature of approximately –80 ‰.

These three points do suggest that a source of isotopically light CH_4 is missing from the inventory. That is a useful insight for this region, and we plan to do further studies to verify the insights from this study.

- Authors' changes

  No change was made to the text.

Detailed comments

Comment 11

- L35: I would add 'based on a bottom-up inventory developed specifically for the region' after '… could be attributed to a single source (CSG, grazing cattle, or feedlots)' or something like that.
- Authors' response

  That line has been removed because of Reviewer 1's request for a shorter abstract. We have incorporated aspects of the request in the new paragraph.

- Authors' changes

  The relevant paragraph in the new abstract now reads

  "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 sources were useable for characterising the isotopic signature of CH_4 sources ($\delta^{13}C_{CH4(s)}$), and to identify mitigation opportunities."

Comment 12

- L60: Introduce the notation CH4 for methane here instead of L66.
- Authors' response

  The notation for methane is introduced in line 1 of the Abstract and line 1 of the Introduction.

- Authors' changes

  See Authors' response above

Comment 13

- L63: Introduce the notation CSG for coal seam gas here instead of L66.
- Authors' response

  CSG is now introduced in line 20 in the Abstract and line 54 of the Introduction

- Authors' changes

  See Authors' response above.

Comment 14

- L82: Introduce the BU abbreviation as bottom-up here instead of L99.
- Authors' response

  BU is now introduced in line 25 in the Abstract and line 146 of the Introduction.
- Authors' changes

  See Authors' response above.

Comment 15

- L148-150: "However, multiple IFAA samples were collected…" This sentence seems to imply that several IFAA samples were collected downwind of plumes coming from only one source category but looking at Fig. 4 I only see maybe 2 samples from grazing cattle with 100%. I would rather say with "… multiple IFAA samples were collected downwind of a **predominant** source category…"
- Authors' response

  We have used this suggested edit.
- Authors' changes

  The sentence now reads:

  "Multiple IFAA samples were collected downwind of a **predominant** inventory source category, for example CSG or cattle feedlots, and these samples were analysed in sets, which is analogous to multiple samples in a plume."

Comment 16

- L150-151: "One aim of this study…" I would remove this sentence, it does not add much.
- Authors' response

  This sentence was removed when reducing the length of the introduction.
- Authors' changes

  The last paragraph of the introduction now reads:

  "A well-established method to determine the $\delta^{13}C_{CH4(s)}$ signature is to collect air samples within the plume downwind of the source and analyse the data using a two-endmember mixing model (Keeling, 1961; Pataki et al., 2003; Miller and Tans 2003). However, the airborne surveys were not designed to track individual plumes; the flight tracks were designed to optimise the results for regional mass balance estimates of CH_4 emissions (Neininger et al. 2021). For aircraft surveys that intersect multiple plumes we present an alternative method. Multiple IFAA samples were collected downwind of a predominant inventory source category, for example CSG or cattle feedlots, and these samples were analysed in sets, which is analogous to multiple samples in a plume. We demonstrate how to analyse these IFAA samples using a detailed BU inventory (presented in

Neininger et al. (2021)), hybrid single particle Lagrangian integrated trajectory (HYSPLIT) modelling (Draxler et al., 1998) and multi-Keeling-model regression with shared parameters (background air $CH_{4(b)}$ and $\delta\,^{13}C_{CH4(b)}$)."

Comment 17

- L151-156: This is the introduction, there is no need to give too many details about the HYSPLIT footprints, it should rather be developed in the 'Material and methods' section. I would just say: "Predominant upwind sources were identified using a combination of the BU inventory presented in Neininger et al. (2021) and back-trajectory footprints (BTF) modeled with the Hybrid single particle Lagrangian integrated trajectory (HYSPLIT)."
- Authors' response

  The last two paragraphs of the introduction were combined and reduced (refer comment 16).
- Authors' changes

  Please see comment 16.

Comment 18

- L158-162: I would develop a bit on the multi-Keeling-model regression, this is the main part of your paper and it seems like a detail here. Maybe better link it to the sets of IFAA samples that you are mentioning L149-150 (we do not understand what these sets are for otherwise). The last sentence of this paragraph does not really belong to the introduction section, these types of conclusions are more for the abstract, the results/discussion or summary sections.
- Authors' response

  The last two paragraphs of the introduction were combined and reduced, which removed this discussion. Please see comment 16.
- Authors' changes

  Please see comment 16.

Comment 19

- Figure 1: There is a lot of information on these maps and their legend. I would try to simplify it a bit by removing the TD domain: it is a bit confusing at first because we do not really know what is this domain and it is just briefly mentioned in section 2.2. For the methane sources, I would already clearly separate point sources and distributed sources in different columns in the legend. I do not think that it is very useful to have the three types of grazing cattle (17a, 17b and 17c), grazing cattle should only be one category with one color. I would also remove the numbers in front of the methane sources, I did not see them anywhere else in the paper.

- Authors' response

  We feel that although the discussion about the airborne measurement TD domain is short, we would like to keep that TD discussion as part of this manuscript because it highlights the link to the research published in Neininger et al. (2021). It is also only within the TD domain that we established in Neininger et al. (2021) the good correlation between the BU inventory and the aircraft-based flux estimates.

  In Table A3 (A3, A4 and A5 in the revised manuscript) the three statistical regions are used. This manuscript, Lu et al. (2021), and Neininger et al. (2021) are all part of one study in the Surat Basin. We have attempted to keep the naming of the categories consistent between the publications.

- Authors' changes

  We have added to Section 2.2, paragraph 2:

  "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 20

- L198: I would start by explaining that there are two types of sources: point sources and distributed (or diffuse) sources, it is a bit difficult to understand the difference during the first read.

- Authors' response

  See the "Author's changes" comment 19

- Authors' changes

  See the "Author's changes" comment 19

Comment 21

- L203-206: As I mentioned earlier, I would remove any mention of the TD domain. It just got me confused and wondering how it was defined and why some IFAA samples are outside of it. I would just say that the area surveyed by the airborne platform has a much higher proportion of CSG and a lower proportion of grazing cattle.

- Authors' response

  Please see the author's response comment 19

- Authors' changes

  Please see the author's response comment 19

Comment 22

- L233: Remove coma after 'The'.
- Authors' response

  Done
- Authors' changes

  Comma removed.

Comment 23

- L237-240: I would remove this last part of the paragraph, it is already stated in the introduction that this study aims at extending the knowledge of isotopic signatures from various methane sources in the Surat Basin. The part about gaining access to a wider range of farms/CSG facilities vs. one sample for the ground-based measurements can go in the discussion section.
- Authors' response

  The last four lines of that paragraph have been removed.
- Authors' changes

  We deleted the following text from the first paragraph of Section 2.3

  "For many source types only one $\delta^{13}C_{CH4(s)}$ signature was determined in Lu et al. (2021). Gaining access to a wide range of farms and CSG facilities is difficult due to operational procedures, and health and safety concerns. Therefore, an aim of this study was to examine if IFAA samples can be used to extend our knowledge of the $CH_{4(s)}$ signatures from various sources in the Surat Basin."

Comment 24

- L258-261: I would move this part to the end of Section 2.3 or merge it with the last paragraph as they are both talking about source categories obtained from the literature.
- Authors' response

  The last two paragraphs in Section 2.3 have been merged.
- Authors' changes

  The last paragraph for Section 2.3 now reads:

  " For CH_4 source categories listed in the BU inventories that were not sampled during the mobile survey, $\delta^{13}C_{CH4(s)}$ signatures were obtained from the literature. These include the $\delta^{13}C_{CH4(s)}$ signatures for kangaroos (-80 ‰, Godwin et al., 2014), on-farm water bodies (dams) (-51.2 ‰, Day et al., 2016), and domestic wood heaters and native vegetation wildfires (-22.2 ‰, Ginty, 2016). There are also numerous termite mounds in the region, but there have been no studies on the rate of CH_4 emissions from these mounds, nor has $\delta^{13}C_{CH4(s)}$ been characterised for termites in the region. 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 ‰. A major gas distribution line passes through the region; this transports conventional gas from the fields to the west of the study area to the LNG terminals on the coast and for the domestic market at Brisbane (Jemena, 2021). The $\delta^{13}C_{CH4(s)}$ population statistics for this gas are not known."

Comment 25

- L279: PU = polyurethane?
- Authors' response

  We replaced PU with "polyurethane"
- Authors' changes

  As above.

Comment 26

- Section 2.5: Which meteorological archives were used with the HYSPLIT model? What is its resolution? Did the authors try different meteorological archives to see how it could affect the definition of the BTF polygons? I don't know if it changes much at such scales but it is worth checking. HYSPLIT can also produce footprints (HYSPLIT dispersion model), did the authors try to simulate footprints from HYSPLIT and compare to their polygons?It is not stated very clearly in the text that the back-trajectories start at the mid-point of the sample collection interval (it is only mentioned in the legend of Figure A1).

- Authors' response

  We used GDAS 0.5 degree, global 09/2007-06/2019, the normal trajectory, and modelled the vertical velocity. We compared these back trajectories against the higher resolution modelling in Neininger et al. (2021), and there were only minor differences. At the scale of the analysis, and given the many large uncertainties with the inventory, there would be no significant difference in the positioning of the polygons and the resulting outcomes.

  Please refer to the supplementary material in Neininger et al. (2021); we did dispersion modelling at a higher resolution than possible in HYSPLIT. Unfortunately, we did not have enough control over the population statistics for the $\delta^{13}C_{CH4(s)}$ signatures of all sources to do precise isotope mixing modelling using dispersion modelling. Thus, the pragmatic methods presented in this paper were developed to glean insights about inventory knowledge gaps from the IFAA samples. We switched to HYSPLIT in this paper, so others could replicate the methods.

  Regarding "It is not stated very clearly in the text that the back-trajectories start at the mid-point of the sample collection interval (it is only mentioned in the legend of Figure A1).

In line 297 it reads “on each side of the IFAA sample collection mid-point”.

- Authors’ changes

  We added more details in the methods section about HYSPLIT.

  In this study, we determine the contributing CH_4 sources (from the UNSW BU inventory in Neininger et al. 2021) of an IFAA sample within a BTF based on the 2-hour HYSPLIT back-trajectory starting at the IFAA sampling height, and at the mid-point of the IFAA sampling interval. The HYSPLIT back trajectory calculations were done using the global data assimilation system (GDAS) 0.5-degree meteorology option (GDAS 0.5 degree, global 09/2007-06/2019, using the normal trajectory, and for the vertical motion we selected model the vertical velocity).

Comment 27

- L294-298: The explanation of how the BTF inventory polygons are estimated is not easy to understand, figure A2 really helps!
- Authors’ response

  Thank you for the positive comment about Figure A2.

- Authors’ changes

  No change is required.

Comment 28

- L310: relative difference?
- Authors’ response

  Done

- Authors’ changes

  We changed ”had a difference” to “had a relative difference”

Comment 29

- L311: 49 IFAA samples were useable out of the 92 collected?
- Authors’ response

  The CH_4 enhancement above background air was only 0.040 ppm. Thus, we set a low threshold of 1 % allowable difference between the infield Picarro CH_4 mole fraction measurements and the laboratory RHUL CH_4 mole fraction measurements. This low threshold was set to exclude any impacts due to shipping the samples from Australia to the UK.

- Authors’ changes

  No change is required.

Comment 30

- Figure 3: I am not a big fan of Figure 3b, or rather the linear regression between the IFAA concentrations and the BTF BU inventory emissions. Our lives would be so much easier if the relationship between methane concentrations and emissions in the atmosphere was linear… But it depends on so many other parameters: meteorological conditions (these points were collected on different days with different conditions), distance from the source (the samples were collected at different altitudes: 100-200 mAGL vs. 250-350 mAGL) and so on… I don't think this linear regression means much to be honest. Also, it would be nice to have the same markers than in the middle plot in the two other plots of this figure. IFAA sample 2111 is not mentioned in the legend like the other outliers are.

- Authors' response

  Although the underlying processes may be nonlinear, it is still valid to examine the statistical strength of the correlation between the mole fraction measured in all the IFAA and the 2-hour upwind inventory. In part this plot addresses one of your other concerns about the 2-hour back trajectory not being enough. The R^2 value of 0.59 highlights that there is a moderate correlation, and that the core assumption in the paper that the mole fraction value of the IFAA sample is a function of the 2-hour back trajectory inventory is valid.

  Exploring other back-trajectory times may enhance or reduce the determined correlation. Collecting more samples, which is a costly exercise, may allow for the characterisation of nonlinear mixing processes and the analysis of the nonlinear correlation strength. However, for the pragmatic task of identifying bottom-up inventory knowledge gaps, the methods presented have achieved the goal.

- Authors' changes

  To highlight that we are aware that it is a complex relationship we now write:

  "A plot of the BTF BU inventory emissions (kg h^{-1}) versus IFAA sample $CH_{4(a)}$ (ppm) shows that there is a moderate correlation (R^2 = 0.59) (Fig. 3 (b)). This moderate correlation is expected **because the mixing of multiple CH_4 sources under turbulent atmospheric conditions is not a linear process,** the inventory is calculated using annual data, and the rate of emissions for many CH_4 sources in the inventory will vary either throughout the seasons (agriculture) or daily (for example, CSG production or grazing cattle location). In Fig. 3 (b) three samples have relatively high $CH_{4(a)}$ values (IFAA samples 2103, 2105, and 2111) and these points are discussed in detail below. IFAA sample 1817 is highlighted, as it is discussed in Section 3.4."

Comment 31

- L350: I am wondering if 50% is a good enough threshold to attribute a sample to a category of if it is not too low. Let say we have a sample with 55% of CSG (-54.5 ‰), 20% of grazing cattle (-59.7 ‰) and 25% of feedlot (-62.9 ‰), the resulting signature will likely be much lighter than the CSG typical signature and I do not see how this point could be useful in the Keeling inversion. It would be interesting to see how the results are changing with a threshold of 70-75% instead (based on Fig 4b, there should still be enough points for CSG and grazing cattle).
- Authors' response

  Please see the reply to comment 8.
- Authors' changes

  No change is required.

Comment 32

- L406-408: "A subset of visually identified outliers **with low δ13CCH4(a) values (1604, 1906, 2103)** is analysed using the results of the multi-Keeling-model regression. Using the values for CH4(b) and δ13CCH4(b) derived from the multi-Keeling-model regression, the Keeling model (Eq. 1) is fitted to **this** outlier subset to determine **its** δ13CCH4(s)."
- Authors' response

  We have incorporated this suggestion.
- Authors' changes

  The text now reads:

  "A subset of visually identified points of interest **(1604, 1906, and 2103)**, **all with low $\delta^{13}C_{CH4(a)}$ values,** is analysed using the results of the multi-Keeling-model regression. Using the values for $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$ derived from the multi-Keeling-model regression, the Keeling model (Eq. 1) is fitted to this subset to determine its $\delta^{13}C_{CH4(s)}$. For this subset a similar result could be obtained using Eq. 2."

Comment 33

- L427: Should be section 3.2 instead of 3.1.
- Authors' response

  The section numbering has been corrected, as part of combining the results and discussion.
- Authors' changes

  As above.

Comment 34

- L440: “IFAA samples 1604, 1817, and 1906 are also highlighted for later discussion.” I would remove “for later discussion” and just directly continue with what is in the next paragraph.
- Authors’ response

  We made this edit.

- Authors’ changes

  We improved this paragraph. It now reads:

  ”In Fig. 3 (b) three samples have relatively high $CH_{4(a)}$ values (IFAA samples 2103, 2105, and 2111) and these points are discussed in detail below. IFAA sample 1817 is highlighted, as it is discussed in Sect. 3.4.

  The IFAA samples are shown in a Keeling plot (Fig. 3 (c)). In this graph three points with relatively low $\delta^{13}C_{CH4(a)}$ measurements are highlighted: 1604, 1906 and 2103. These three points were not included in the initial Keeling analysis but are analysed using insights from the multi-Keeling-model regression.”

Comment 35

- L449: Why do the authors show 3-hour back-trajectory if the footprint calculation is based on 2-hour trajectories?
- Authors’ response

  We presented the 3-hour trajectories, because some of the emissions captured in the IFAA samples will come from beyond the 2-hour polygon and we thought 3-hours was a useful visual guide, especially where the trajectories curved. As requested, we now plot the 2-hour back-trajectory line in all figures.

- Authors’ changes

  All figures have been updated using 2-hour trajectories.

Comment 36

- L474: It would be even easier to see if the BTF polygons for each IFAA sample were also displayed on these figures.
- Authors’ response

  We originally had a version of the maps with all the polygons for each IFAA shown, but the figures were far too cluttered. For example, visualise all the overlapping yellow polygons in Figure A1 or A3. The back trajectories plots are mainly to highlight the regions where the air has been sampled and the spatial context for each subset used in the regression analyses. All the details about what

CH_4 point and diffuse sources are captured within a BTF polygon are presented in Table A3 (now A3, A4 and A5).

- Authors' changes

  No changes were made.

Comment 37

- Section 3.3: If I understand correctly, the system uses the different sets to estimate CH4(b) and δ13CCH4(b) first and then use these background values in the Keeling regression of each set. Is the uncertainty on CH4(b) and δ13CCH4(b) estimates taken into account for the Keeling regression of each set in the second step? Looking at Figure 5, there is no error bar for background point and it seems like all the regression lines are exactly passing by this point even if δ13CCH4(b) has a standard error of 0.1 ‰. I think this +/- 0.1 ‰ can have a big impact on the calculated δ13CCH4(s) for the different categories.

- Authors' response

  We hope below provides some clarity about "the system uses the different sets to estimate CH4(b) and δ13CCH4(b) first and then use these background values in the Keeling regression of each set"

  There is only ever one shared $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$ set, and all Keeling lines must converge to this one point. Please also see the reply for comment 6. Due to the low enhancement above background CH_4 and the small number in each subset the uncertainties will be large, and these are all determined as part of the multi-Keeling-model regression.

  To help readers we have added on all appropriate graphs that the error bars are one standard deviation. When we discuss the $\delta^{13}C_{CH4}$ signatures in the text, we always write the 95% confidence interval. In Figure 5 the uncertainties for $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$ are large relative to the $CH_{4(a)}$ and $\delta^{13}C_{CH4(a)}$ values. But the focus should not be placed on the large uncertainties, rather the focus should be on the most probable value. Although the most probable values for $\delta^{13}C_{CH4(s)}$, $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$ have high uncertainty there are still many insights gained from interpreting the results. Ideally the uncertainties should be smaller, but that would require many more samples than those collected during the campaign.

- Authors' changes

  We now say in several locations that we have done global optimisation. In graphs that display data we have added at the end of the caption a sentence that states that the error bars are one standard deviation. We have placed the error bars on the $CH_{4(b)}$ and $\delta^{13}C_{CH4(b)}$ estimates in all plots.

Comment 38

- L500-501: “Below we also discuss…” I don’t understand this sentence, what is going to be compared? It is not clear to me what is the difference between the 2-hour upwind BU inventory estimates and the expected values based on the BU inventory.
- Authors’ response

  This sentence was removed when combining the results and discussion.
- Authors’ changes

  There is a new combined results and discussion section.

Comment 39

- L501-502: “For the IFAA samples discussed below details about the sample location, day and time of collection, and the upwind inventory are listed in Table A2.” There are many commas missing in the paper but sentences usually stay understandable, unfortunately it does not work here.
- Authors’ response

  This sentence was removed when combining the results and discussion.
- Authors’ changes

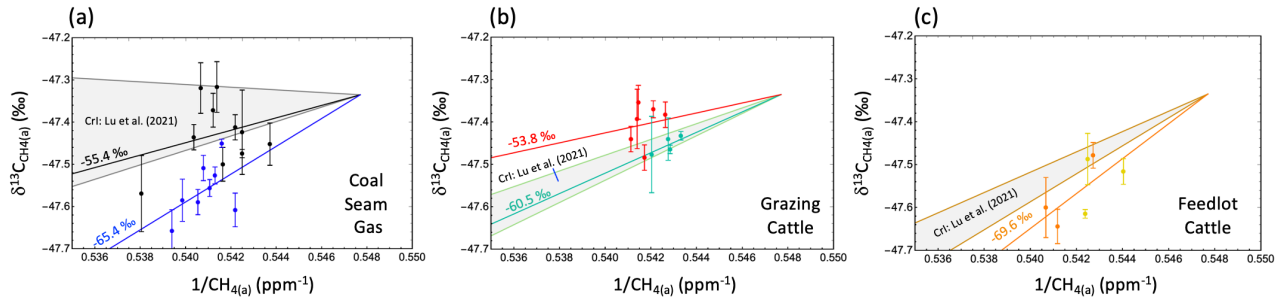
  There is a new combined results and discussion section.

Comment 40

- Figure 6: Add titles with the source category for each plot. L547: there is a missing “)” after “including derived source signatures”.
- Authors’ response

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

  We added “)”.
- Authors’ changes



Comment 41

- Section 4.1.3: Looking at Figure 6a, 100-200 mAGL points seem to better align than 250-350 mAGL points but somehow both sets end up with similar standard error. Also, it seems like 4 or 5 points (out of 9?) collected at 250-350 mAGL could fit the 100-200 regression line, this is a bit concerning…
- Author's response

  The standard error is in part a function of the $1/[CH_4]$ data range and the number of samples (points), and these are small and similar for both the 250-350 mAGL the 100-200 mAGL CSG sets.

  All determined $\delta^{13}C_{CH4(s)}$ values have large uncertainties and overlapping uncertainties given the small spread in both the x and y directions is expected. What is more important is that for CSG the 100-200 mAGL set is systematically offset to lower $\delta^{13}C_{CH4(a)}$ values compared to the 250-350 mAGL set. We suspect that in some of the CSG back trajectory regions there are more cattle emissions than captured in the inventories. This applies to both altitude sets.
- Author's changes

  No change has been made.

Comment 42

- L595-598: I don't think excluding sample 1808 because it was collected on a different day is a good reason! Different days have been used in all the previous categories and it was not a problem. Several points of CSG 250-350 mAGL falls into the CSG 100-200 mAGL δ13CCH4(s) signature regression line and have not been excluded! Excluding sample 1808 seems very random, how different is δ13CCH4(s) with this point?
- Authors' response

  We thank the reviewer for this comment, as it resulted in an important revision to the manuscript.

  Please refer to new Figure A4 at comment 5. With respect to 1808 being sampled on a different day, this was a minor consideration when this was removed. The main reason we excluded 1808 was because it was clearly far from the other samples and in a very different landscape setting where farming is far from CSG activities. However, the position of 1808 is wrong in the submit Figure A4. We discovered this when updating the images with the 2-hour trajectories, so this set of calculations turned out to be a nice QA step for the results. When making the original Figure A4 the wrong back trajectory was loaded. It is correct in Figure A1, and we used the correct polygon region for the calculations of the BTF inventory. We have updated Figure A4 (see comment 5). We have also added 1808 back into the Grazing Cattle 100-200 mAGL $\delta^{13}C_{CH4(s)}$ set and redid all the calculations, because everything is linked in multi-Keeling-model regression. The impact of adding 1808 back into the calculations was minimal and had no significant impact on the scientific

discussion. These revisions prompted a close inspection of the sources in the back trajectories, and we also checked our field notes, and this resulted in an extended interpretation for this set.

Including 1808 in the calculation resulted in updating Figures 4, 5, 6, A4, and B1. We also updated the results in Tables A4 (now A6) and in numerous places in the text where there was a minor change in the value.

- Authors' changes

Figures 4, 5, 6, A4, and B1. Have all been updated. In the revised manuscript it now reads:

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 there are four domestic sources of CH_4 that 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.

Comment 43

- L603-606: Not clear if the problem is that 2-hour back trajectories are not enough or if the BTF polygons have portions out of the BU inventory map. Maybe the border of the BU inventory map should be displayed on figures A3, A4, A5 (as well as the polygons). If the problem comes from the 2-hour back trajectories not being enough for this case, then why should it be enough for the other cases?
- Authors' response

  The edge of the image is the border of the inventory. As a result of correcting the error for sample 1808 we have revised the interpretation. We can now explain the observation using local CSG emissions. Please refer to the updated section 3.3.5 at comment 42. In all cases there could be input from beyond the two hours, and we made a comment about that in several locations throughout the original manuscript.
- Authors' changes

  Please refer to comment 42

Comment 44

- L626: Figure 5(a)
- Authors' response

  We made this edit.
- Authors' changes

  Both here and in other places Fig. 5 (b) changed to Fig. 5 (a).

Comment 45

- Section 4.2 and 4.3: These sections should be merged, most of what is said in section 4.2 is repeated in the beginning of section 4.3.
- Authors' response

  We have merged the results and discussion and removed the duplication in reported values. However, Section 4.3 was not discussed in Section 4.2. Section 4.3 is a separate discussion about the isotopically light samples.
- Authors' changes

  The results and discussion have been merged.

Comment 46

- Section 4.3: Reading this section, it seems like it is difficult to draw any conclusions from these outliers… Outliers 1604 and 1906 are potentially sampling termite emissions and are associated with outlier 2103 whose signal is potentially coming from brine water ponds. Altogether, they end up having an isotopic signature of -80.5 ‰ but outliers 1817 and 2111 sampling these same brine water ponds don't get the same isotopic signature…

- Authors' response

  Samples 1604, 1906 and 2103 all sit close to a Keeling line with a signature of –80 ‰. We discuss potential sources, but we do not claim that these must be the sources. Water bodies and wetlands may or may not emit CH_4 with an $\delta^{13}C_{CH4(s)}$ signature of approximately –80 ‰. It may still be that 2103 is capturing termite emissions. In the paper we presented some plausible ideas that matched likely sources in the region. We write that further fieldwork will confirm or dismiss the suggested potential sources.

  The back trajectories for 1817 and 2111 pass directly over the "raw water ponds", not the "brine ponds". The raw water ponds and the brine water ponds have different water types and likely have different microbial communities. Only the back trajectory for 2103 passes over the brine ponds (Orana 2,3, and 4 in Figure 8). We also show in Figure 8 that the back trajectories for 2111 and 1817 only pass over the raw water ponds. There is a paragraph in the original manuscript section 4.3 that discusses this in detail.

- Authors' changes

  No changes made for this comment.

Comment 47

- L733-734: Sentence not clear.
- Authors' response

  This sentence was removed when rewriting the summary.
- Authors' changes

  This sentence has been removed.

Comment 48

- L745: “For all three samples, termite emissions may have been sampled.” There is no mention of termite emissions at all in the paragraph discussing outlier 2103 (L671-677) in section 4.3.
- Authors’ response

  In Section 4.2 at lines 633 to 634 there is reference to the possibility of emissions from termites. ”For IFAA sample 2103 both the brine water ponds and termites could be the missing biological source with a low $\delta^{13}C_{CH4(s)}$ signature.”
- Authors’ changes

  No change is required.

Comment 49

- Figure A1: I would only show the 2-hour back-trajectories rather than the 3-hour BT. Most of the time, it is difficult to see where is the 2-hour point on the red lines. This comment applies to all the other figures with back-trajectories.

- Authors’ response

  We have replaced the 3-hour trajectories with 2-hour trajectories. As discussed in one of the earlier comments, these maps are not for detailed inspection of the sources. They are to show the locations of the samples, and the subregions over which the airmasses have moved. Details about what sources are in the 2-hour back trajectory polygon region are presented in Table A3 (in the revised manuscript Tables A3, A4, and A5). Updating the back trajectories turned out to be a useful quality assurance step. When updating the trajectories, we identified that 1808 had been wrongly located. This changed the interpretation for the 100 to 200 mAGL grazing cattle data set.

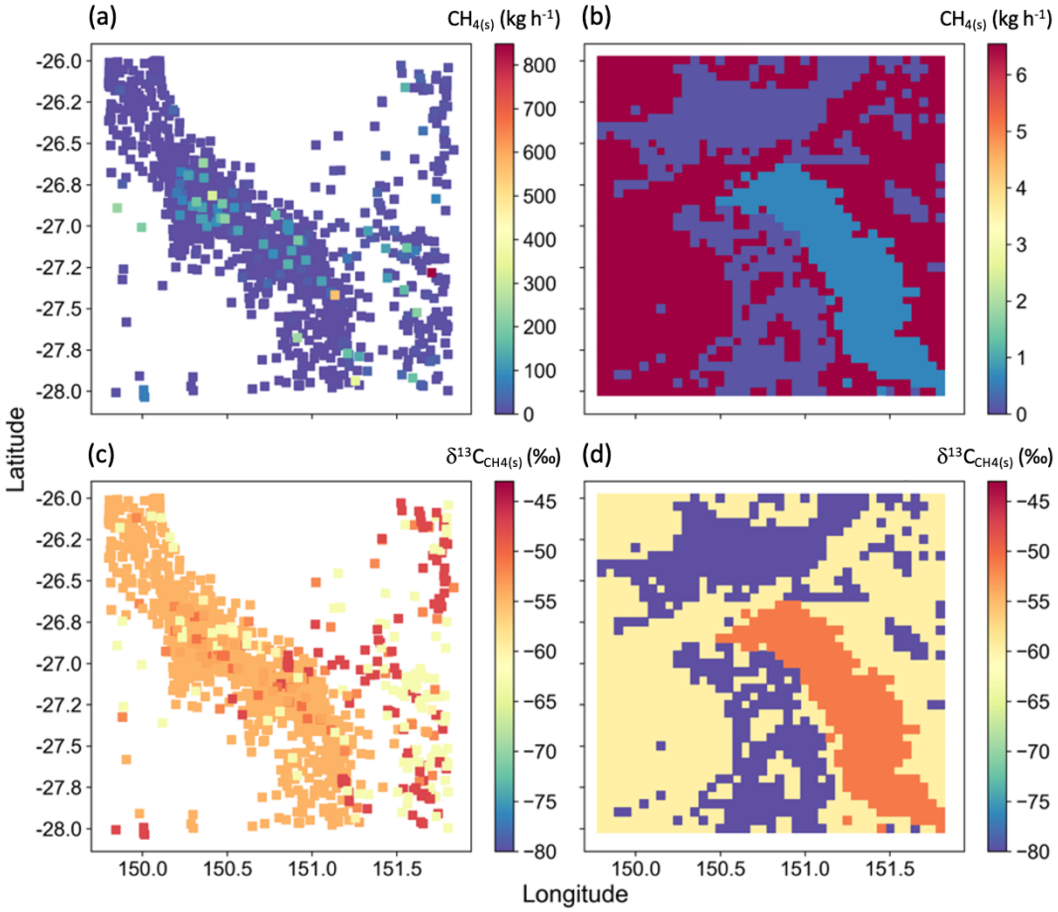
- Authors’ changes

  We updated the back trajectories shown in Figures A1, A3 (a), A3 (b), A4 (a), A4 (b), and A5.

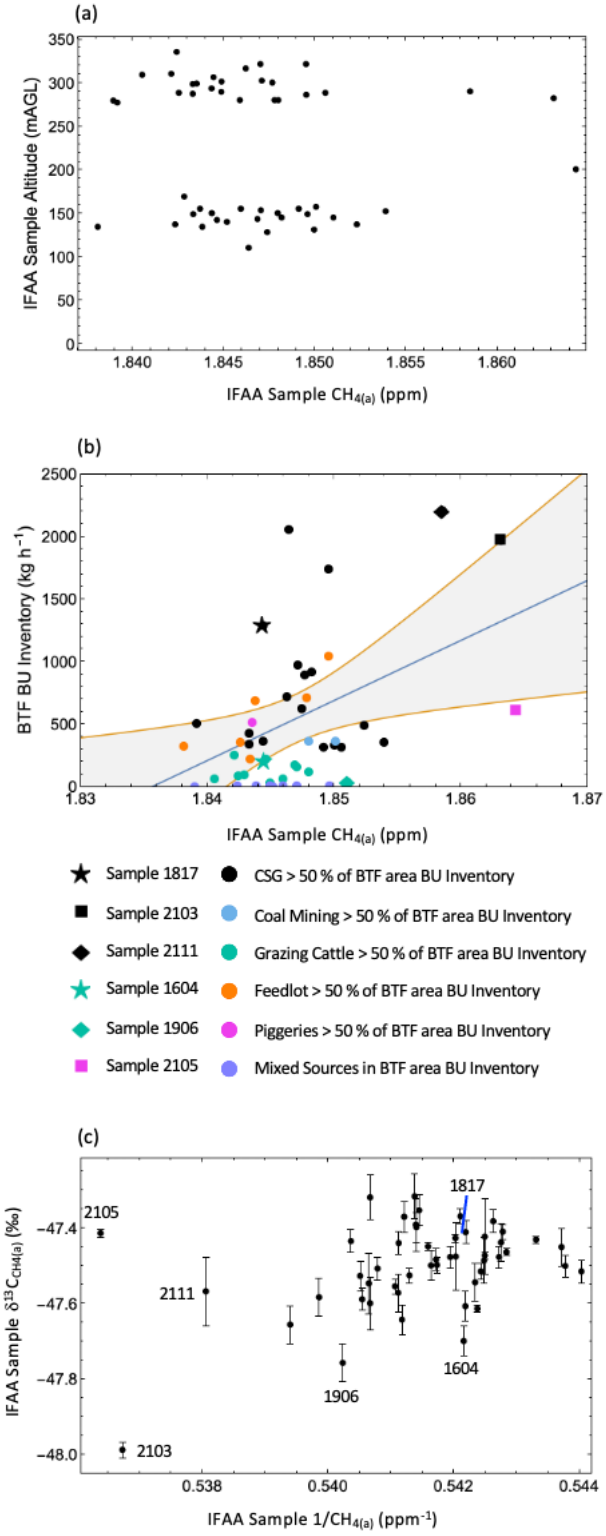
Reviewer Acknowledgement

All authors would like to thank reviewer 2 for carefully reading of the manuscript. We hope our replies above have addressed your concerns and improved the quality of the manuscript. This research is complex and important. Your help with improving the clarity of the manuscript will help the scientific outcomes have impact. Thank you.

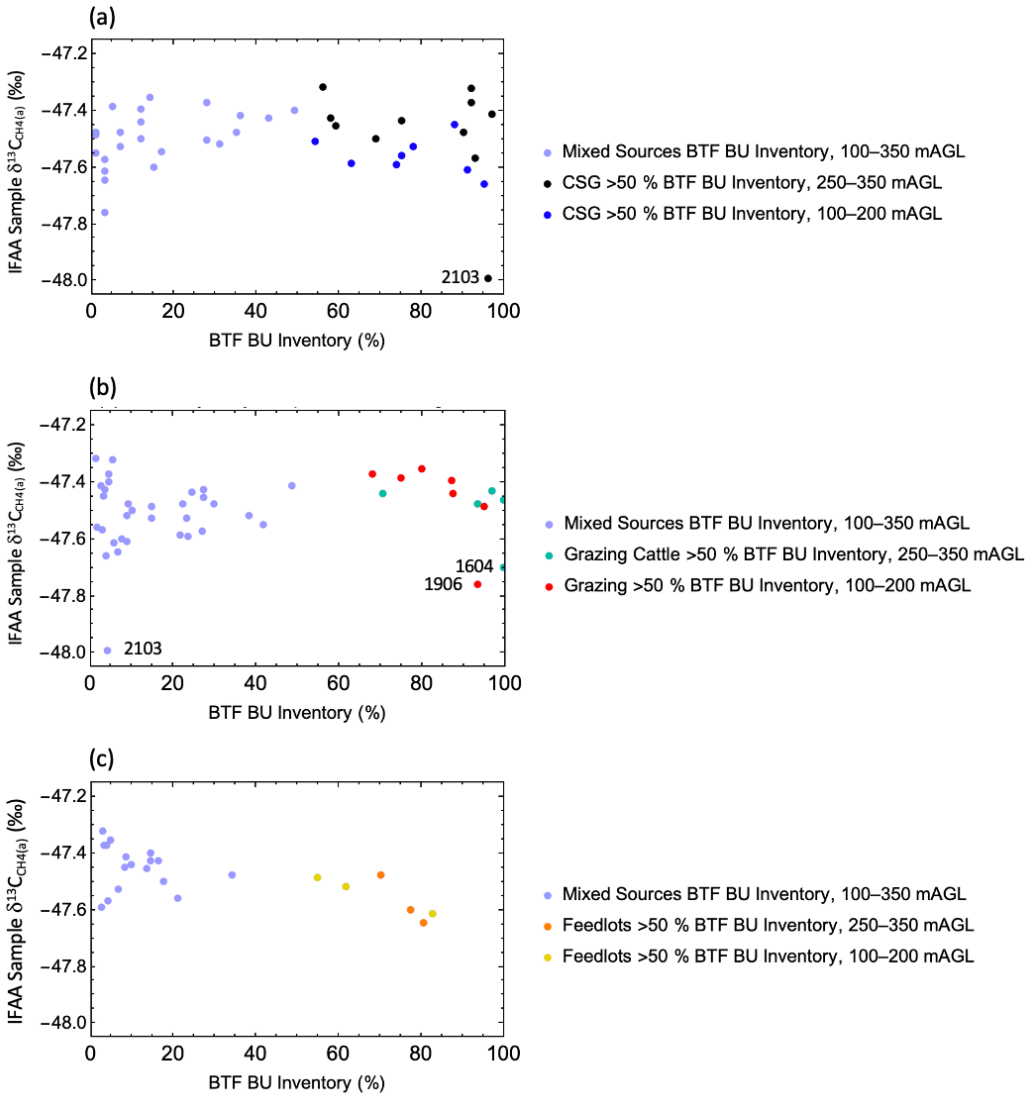
Revised Figures



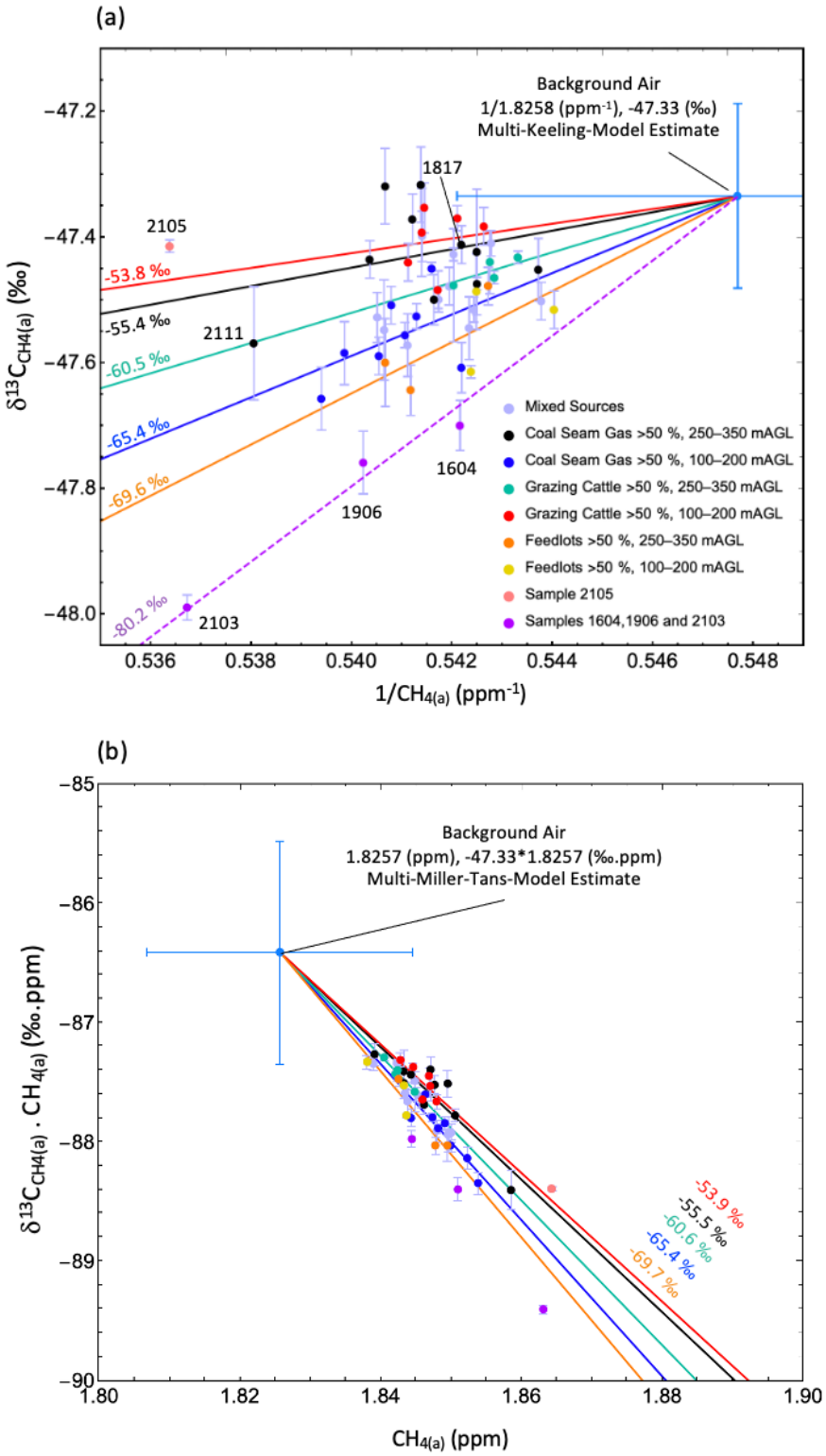
Revised Figure 2.



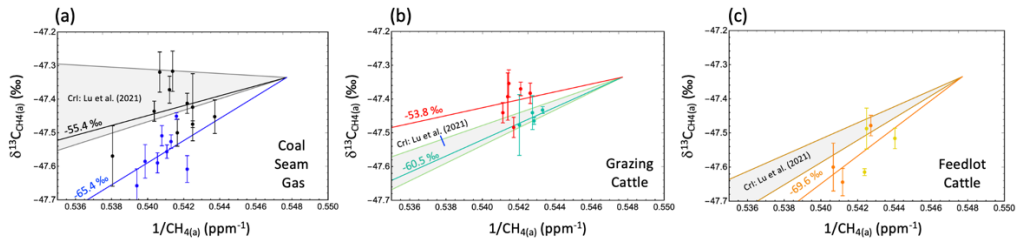
Revised Figure 3.



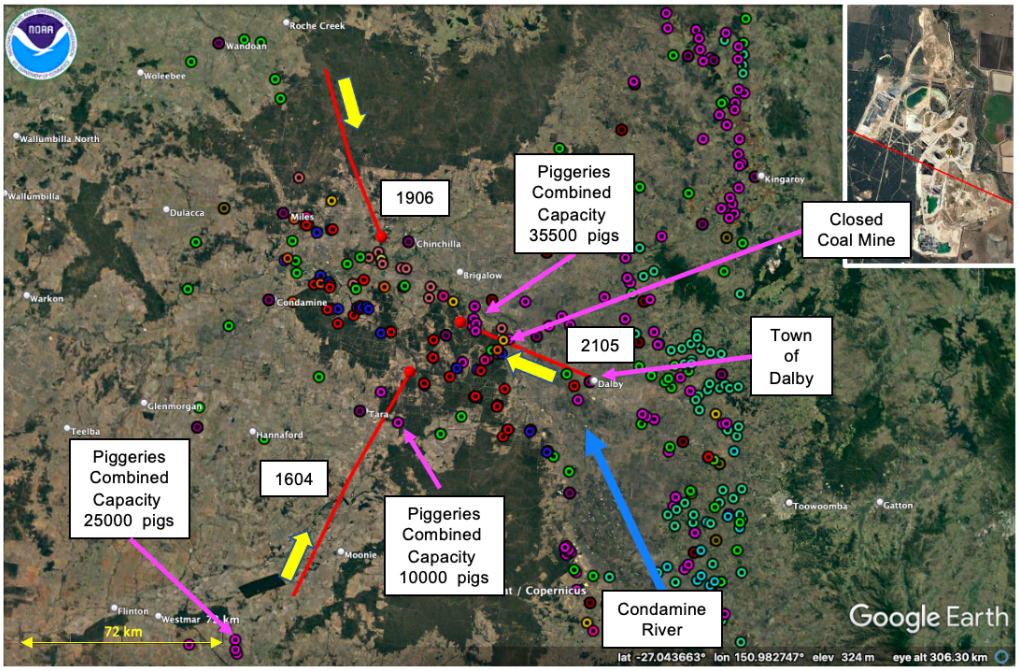
Revised Figure 4.



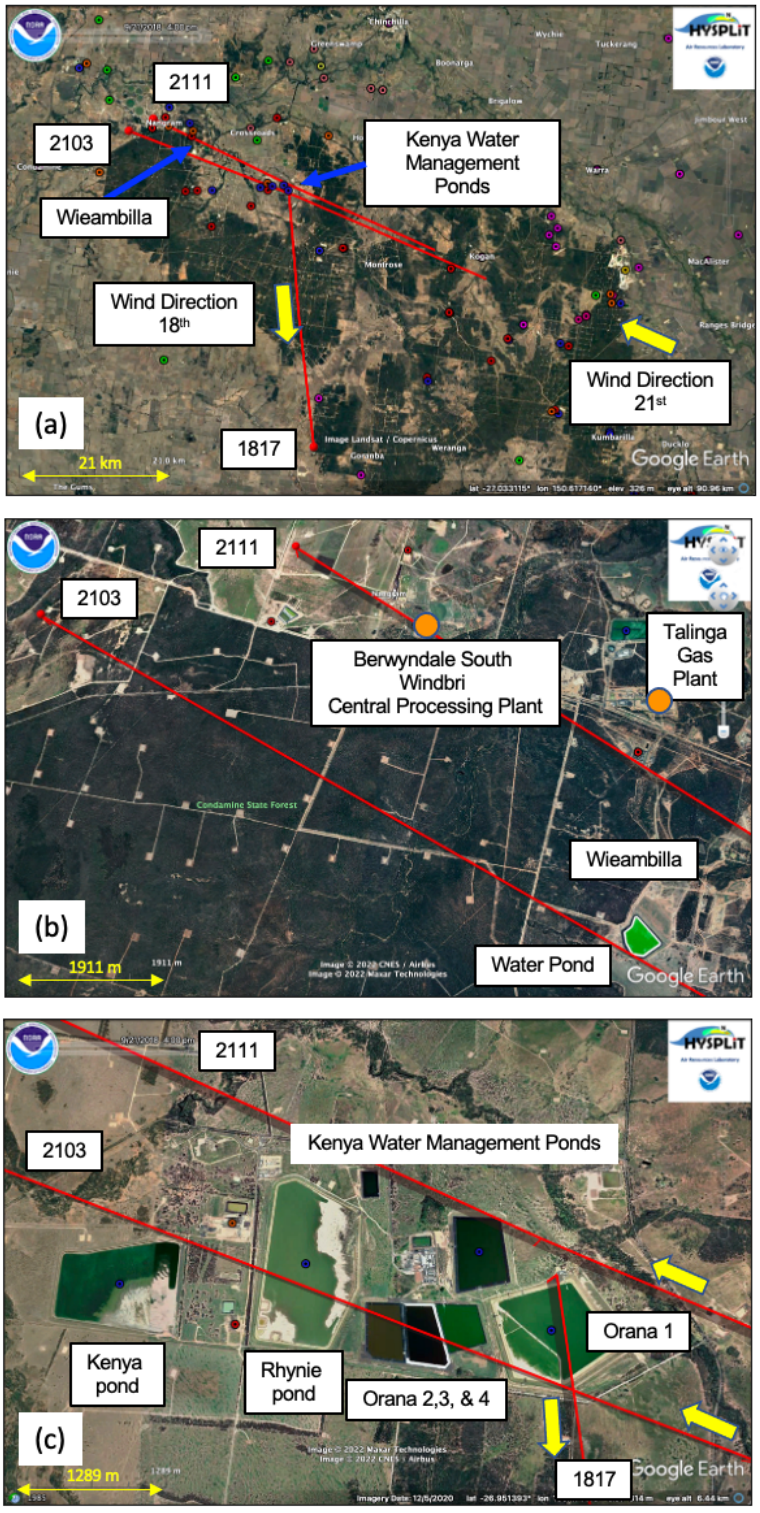
Revised Figure 5.



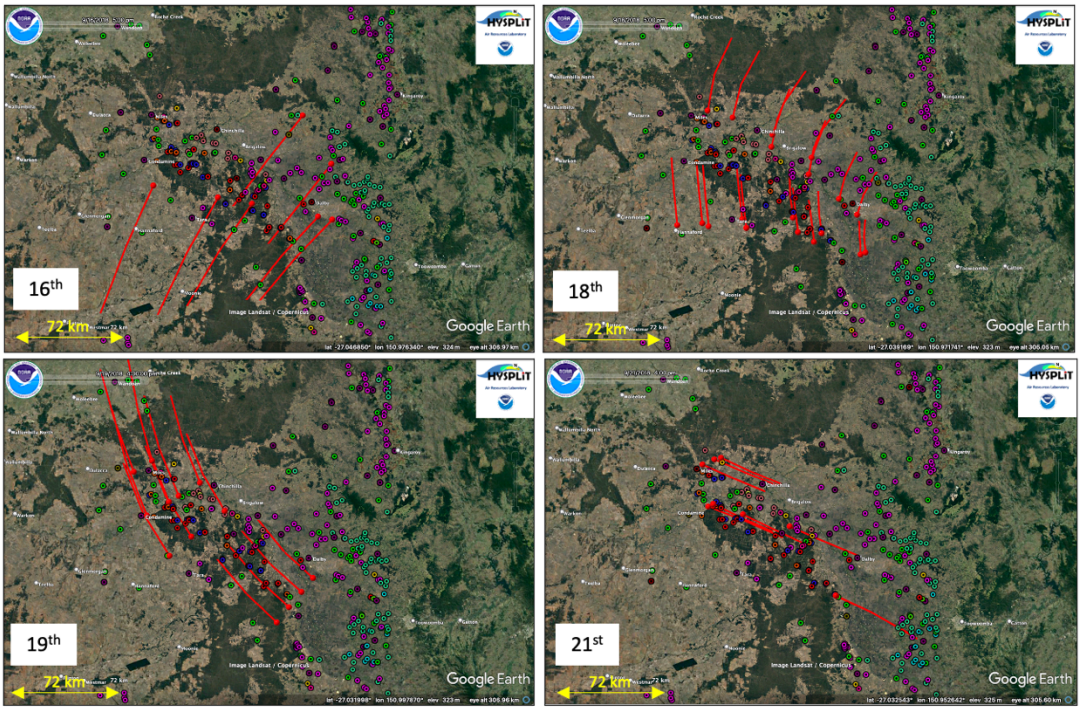
Revised Figure 6.



Revised Figure 7.



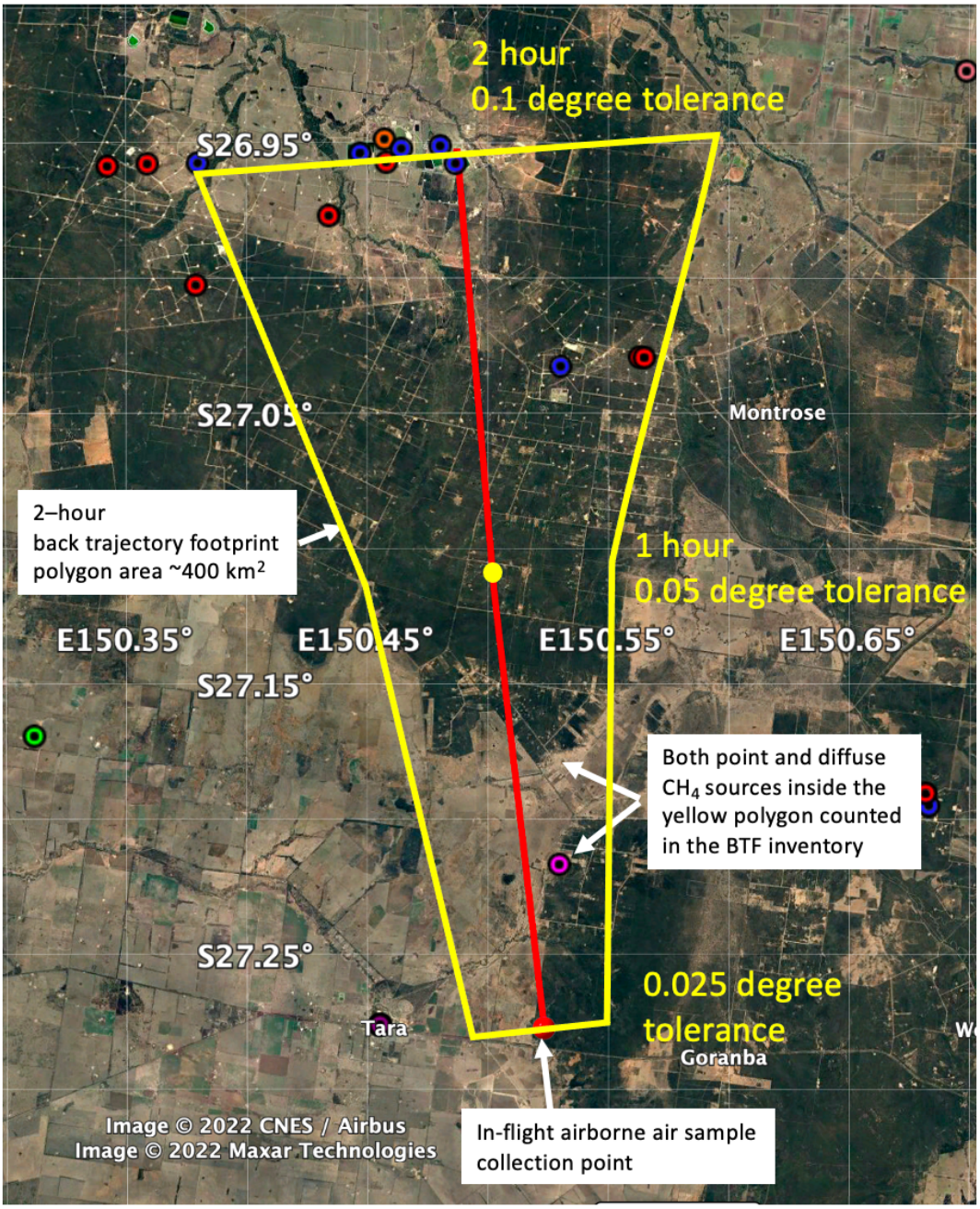
Revised Figure 8.



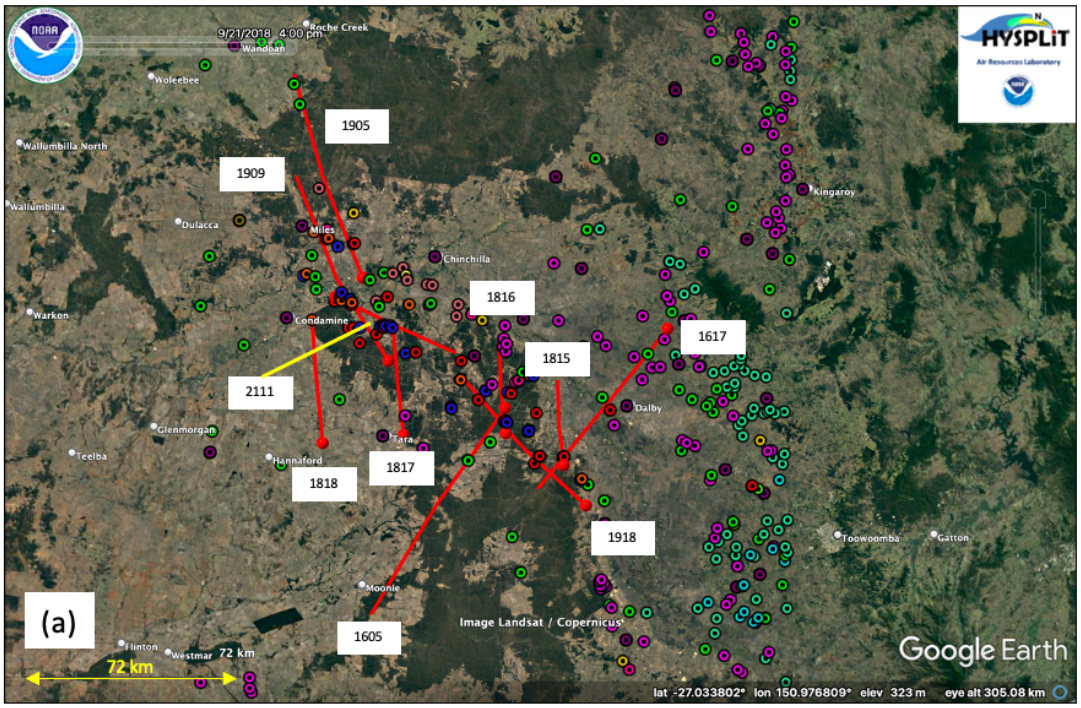
Point Sources

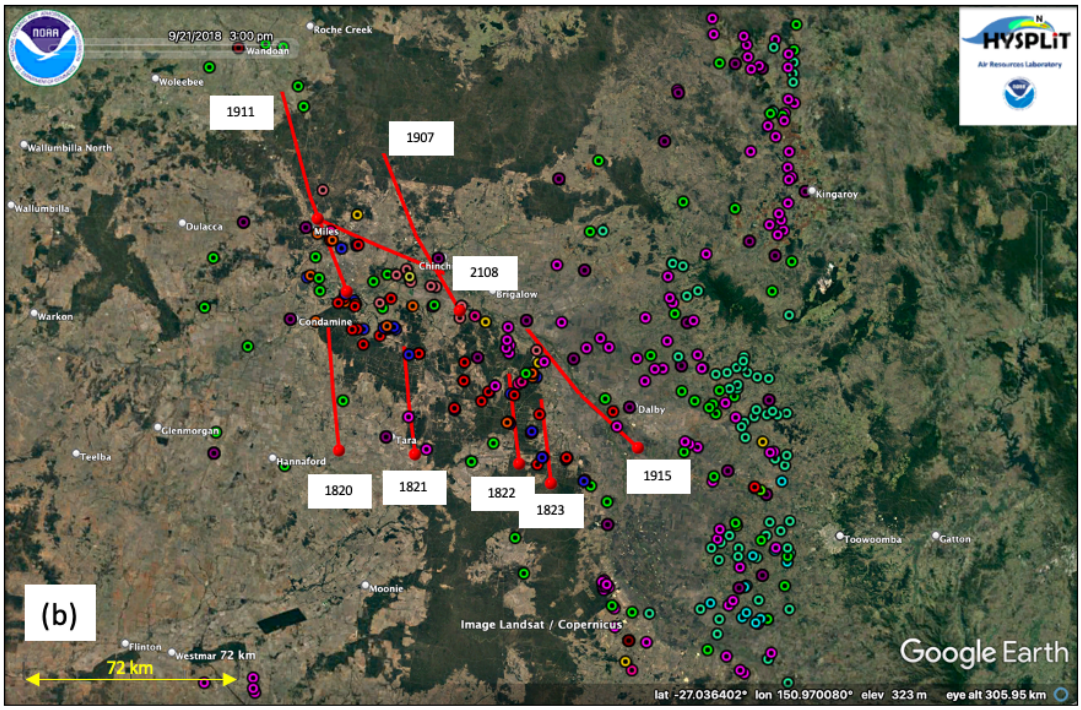
- CSG processing plants
- CSG compressor stations
- CSG raw water ponds
- Coal mines
- Power stations
- Ground seeps
- River gas seeps
- Cattle feedlots
- Cattle dairy
- Piggeries
- Poultry farms
- Landfills
- Wastewater treatment plants
- Domestic wood fires

Revised Figure A1.

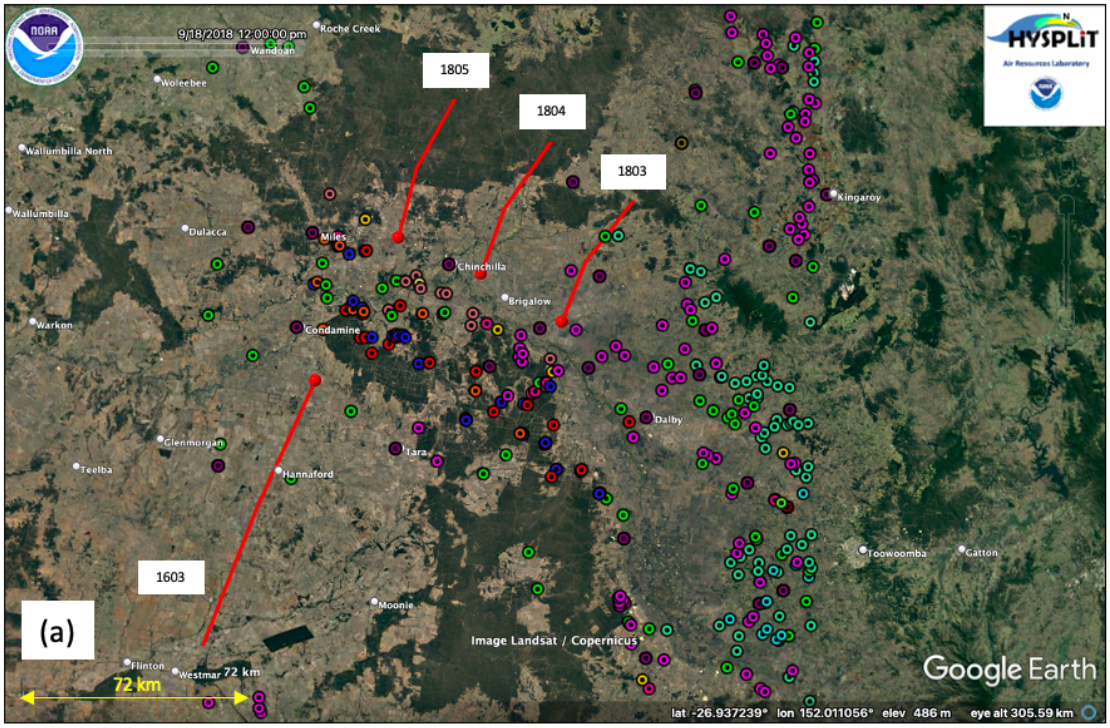


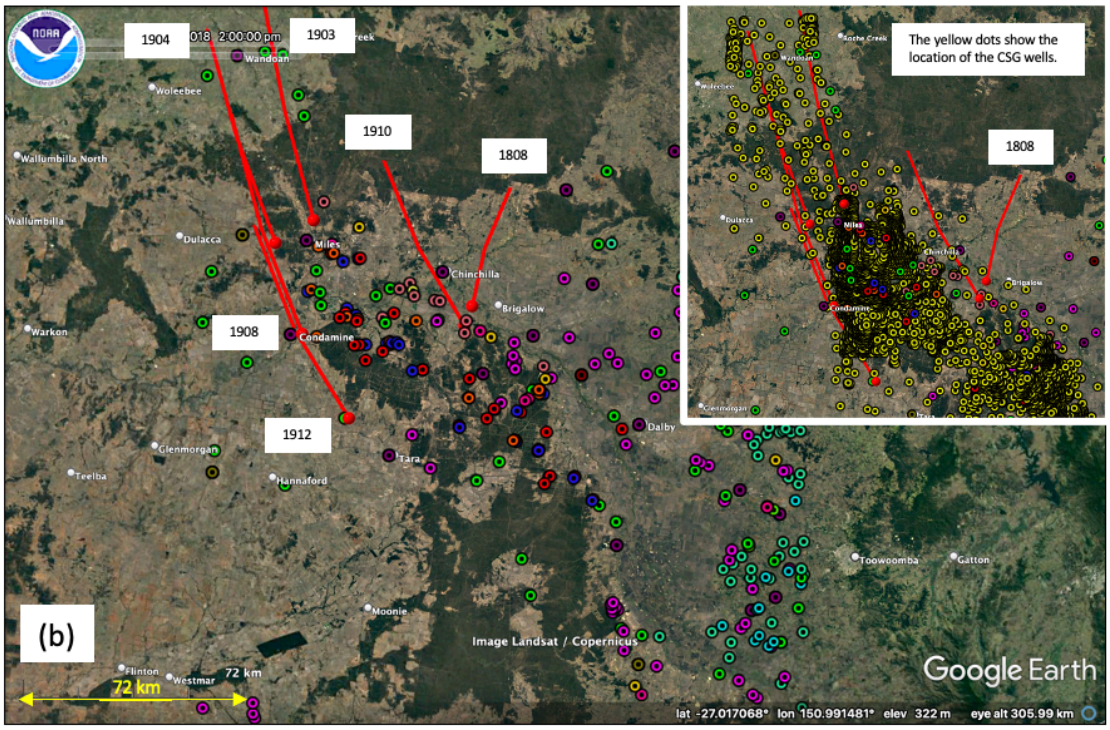
Revised Figure A2.



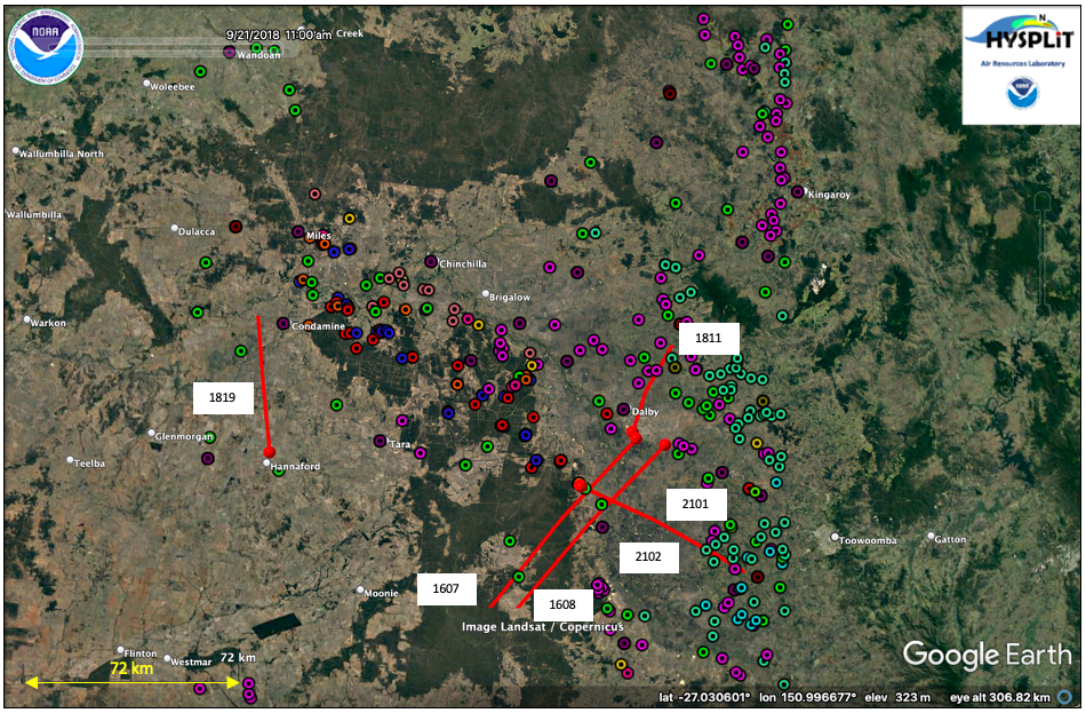


Revised Figure A3.

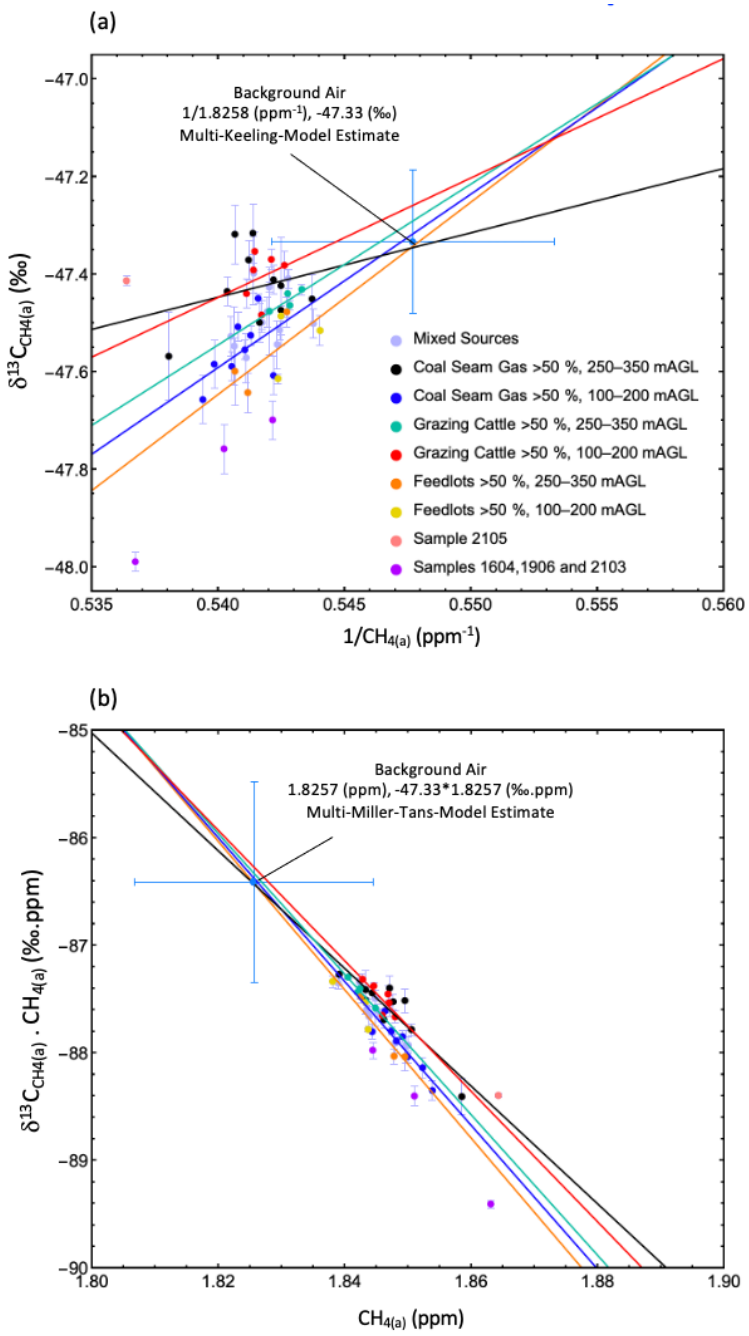




Revised Figure A4.



Revised Figure A5.



Revised Figure B1.