Comment on acp-2021-76

Anonymous Referee #2

Referee comment on "Isotopic signatures of major methane sources in the coal seam gas fields and adjacent agricultural districts, Queensland, Australia" by Xinyi Lu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-76-RC2, 2021

General comments:

In this manuscript, Lu et al. present mobile atmospheric methane measurements conducted in 2018 an 2019 and subsequent laboratory analyse of the δC and δD in CH_4 signature of 17 methane sources in the Surat Basin. During the campaigns, mobile GHG analysers are used to identify emission plumes of different important CH_4 sources and whole-air samples taken within those plumes are later analysed in the lab by suitable means. The results are then compared with previous studies from Australia and globally. The authors also highlight the added value of double isotope analysis, i.e. δD and δC in CH_4

Overall, the manuscript is well written and nicely structured, which makes it easy to follow. The issue of source apportionment of anthropogenic and natural methane sources continuous to be of great importance in this field of research with possible future policy implications. This study reflects important incremental progress and the fact that it emphasis the value of double isotope analysis make it very relevant beyond the immediate study region. The quality and scope of the paper is, therefore, fully suitable for publication in Atmospheric Chemistry and Physics. However, there are two general issues and a few very minor comments that should be addressed before publication.

- Representativeness of results:

Without further information on the type of landfill and WWTP for example it seems impossible to judge if the found isotopic signatures can be used for other such facilities in the regions. The same problem arises for the piggery and especially the abattoirs where the source mix/methane producing process seems unclear. Are the isotopic signatures presented here representative for the each class of facility in tje region or will future studies have to be conducted to characterize each piggery and abattoir seen in Figure 1? This should be discussed in more detail in the conclusion section.

- Calculation of uncertainties:

The uncertainties reported seem to only rely on the uncertainty of the Miller-Tans fit. However, for some plumes e.g. the river seep (Figure A2) only 4 data points are available and any sub-sample of 3 of those data points could yield different results. As the isotopic signatures rely on very few data points per site (some on only 3 data points) it seems reasonable to use leave-one-out-validation to check if the Miller-Tans fit uncertainty is reasonable. For some sites the current uncertainties could just reflect the lower bound of the true uncertainty, which is not highlighted in the conclusion section.

Specific and technical comments:

L5: If appropriate, consider spelling out the name of the University of New South Wales Sydney here

L17: Why is this limited to 'warm and hot climate regions''? Even in temperate and colder climates, nighttime GHG levels are known to be enhanced compared to afternoon values due to less vertical mixing in a lower PBL.

L19: Were all 17 plumes analysed for δD_{CH4}? Only 14 sites are given in Figure 3 (and the tables) what happened to the δD_{CH4} data from the other sites (e.g. the Chinchilla landfill)?

L40: Nisbet et al. 2020 is a great reference here, but IPCC AR5 (you cite Myrhe et al. 2013 later on) and others (e.g. Ganesan et al. 2019) could be/should be mentioned as well.

https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2018GB006065

L41: Suggest rephrasing to "not **always** possible" or "not **easily** possible" as it is indeed possible to identify sources from atmospheric mole fraction measurements when sources are **not** juxtaposed and isolated enough and/or when suitable calibrated atmospheric modelling can be performed.

L152 (and following): Why is 'coal seam gas' not abbreviated as CSG here? Suggest to consistently use CSG throughout the manuscript. Possible exception at beginning of new sections if readers 'jump' directly to the conclusions it might be useful to reintroduce CSG.

L157: Further information on the power stations seems necessary, how many are gas-fired and how many are coal-fired? Also, they account for 4.7% of GHG emissions from the electricity sector, but is this mostly due to their CO2 emissions or do they report

significant loss rates for CH4 as well?

L174: Correct to "fa**r**ming"

L199: Here the authors nicely outline which factors determine landfill emissions, but none of the suggested parameters controlling landfill CH4 production is reported for the Chinchilla landfill.

L201: The amount of (organic) waste deposited at this landfill site would seem a critical parameter to add here (its disposal area might be of secondary importance).

L205: What kind of waste treatment is used at the Miles WWTP, e.g. are sludge digesters used?

L220-L230: Suggestion to cite appropriate peer-reviewed studies on instrument performance rather than only manufacturer specifications. Previous studies have investigated the performance of G2201i and UGGA instruments e.g.

https://www.sciencedirect.com/science/article/pii/S0956053X17309698

https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/331683/SC130034_Report.pdf

https://amt.copernicus.org/articles/8/4539/2015/

https://amt.copernicus.org/articles/10/2077/2017/

L224: The cited precision of 8cm is likely only for static measurements. Typically, the limiting factor for the resolution on mobile platforms is the GPS frequency. Was the 10Hz or the optional 20Hz version of the A326 used here?

L230: see L224

L319: Are those uncertainties only based on the uncertainty of the fit or was a bootstrapping method used as well to check that individual data points do not overly bias the slope? Was any data selection applied, i.e. only accepting slopes of fits with an R2 above a threshold value?

L325 – Figure 2: The three reference levels of CH4 (3, 2 and 1.8ppm) are rather confusing and make the figure more difficult to follow. As all major peaks are labelled with their concentration this seems unnecessary (or maybe just have one reference level). The grey ribbon is hardly visible with blue and green levels on the figure.

L330 – Table 1: Please add the range or maximum CH4 mole fraction for each plume, additionally the Pearson's R or R2 of the Miller-Tans fit might be informative. Furthermore, it seems important to highlight if the samples were part of a daytime or nighttime survey.

L339 – Could ground migration/stray natural gas migration be important here? If yes, this means emissions can be significantly displaced from the actual leaking infrastructure.

L387 – Figure 4: The current figure makes it difficult to compare literature sources and this study. A two-panel figure with the same size and same X&Y scaling could make it easier. Alternatively, adding PM, SM and T areas to the main part of Figure 4 would achieve the same.

L395 – The abbreviation ROM seems not to be used elsewhere in the manuscript.

L440 – Why are no δDCH4 values reported for abattoir A?

L469 – Please state more clearly what your theory is about the source of the CH4 from abattoir A and B. There is a discussion of different options whichs are likely not the source and eventually, I think, you are suggesting that there is a biogas generator on site or a waste lagoon that is causing the emissions?

Also, if one of the abattoirs actually has an integrated feedlot and we cannot be sure what the source is, are the values found for this abattoir applicable to other abattoirs?

L523 – What kind of aerobic and/or anaerobic treatment is implemented at the Miles wastewater treatment plant? A simple description in section 2. might be helpful to understand why it is more similar to some WWTPs from previous studies.

See: https://www.wdrc.qld.gov.au/living-here/engineering-services/utility-services/wastewater-and-sewerage/regional-sewerage-networks/#miles-sewerage

L530: Was the consumer grade natural gas used in the city measured or were samples from wood stoves taken? If not those seem important sources to add in future studies.

L590-L600: Seems to be a summary, rather than contain conclusions. Other parts of this section also seem to repeat previous results. Therefore, renaming the section to "Summary and conclusion" might be appropriate.

L607: Are the samples for piggeries and abattoirs representative of other facilities of the same type in Queensland or even Australia (see general comments).

L625 – Suggest to add Pearson's R or R2 for each Miller-Tans fit in Figures A1, A2. A3, A4 and A5