Author's response for Ref: egusphere-2025-377

Ref. No.: egusphere-2025-377
Title: Isotopic signatures of methane emission from oil and natural gas plants in southwestern China.

Journal: Atmospheric Chemistry and Physics.
Authors: Dingxi Chen, Yi Liu, Zetong Niu, Ao Wang, Pius Otwil, Yuanyuan Huang, Zhongcong Sun, Xiaobing Pang, Liyang Zhan, and Longfei Yu.

Dear Editor,

We would like to express our sincere appreciation to you, and the anonymous reviewers for your constructive comments and suggestions on our manuscript (egusphere-2025-377) submitted to Atmospheric Chemistry and Physics. We have carefully considered all reviewers' comments and have responded to each point in detail.

Regards,
Longfei Yu
On behalf of all coauthors
May 14, 2025

- **Reviewer comments**
- Author's response

RC3

1. **This manuscript presents new methane isotopic data for Chinese oil and gas infrastructure measured on a CRDS instrument. It concludes that it can distinguish between sources. Unfortunately, the data presented can only imply what the averaged fossil fuel signature might be. Overall the data shown are not convincing because: a) the emissions categories are not characterized at source, or close to individual emission points, and b) the source signature calculations mostly have very large errors and rely on a 1-point calibration of raw data.**

R: Thank you very much for your comments and critical points. We agree partially with the reviewer 3 that uncertainty in ground-based or UAV-based isotopic measurements of CH_4 in environmental atmosphere is generally larger than direct measurements of source isotope signatures next to singular ONG sources. However, such study design was planned on purpose, to evaluate the CH_4 leakage from the site-levels, which are believed to be valid for those intensive ONG processing facilities with distributed leakage points which is impossible to quantify one by one. Such work has also been conducted in other countries or regions (Menoud et al., 2022; Leitner et al., 2023; Fosco et al., 2025). In addition, we have also measured CH4 emission (with portable TDLAS instrument for CH4 mixing ratios) next to significant leakage points(Chen et al., 2024), via mobile monitoring platforms (vehicle-based monitoring on ground and UAV-based monitoring in the atmosphere). This work would provide important support to validate our isotope measurements for appointing ONG sources. In addition, for ONG sites under operation, it is highly difficult and challenging to conduct in situ monitoring or sample collections, not to mention even more restrictions to get access directly to the emission source. Therefore, our research campaign and collected data was highly valuable and scarce considering all the obstacles and restrictions in conducting systematic research from ONG entrepreneurs. Regarding close-to-source measurements, as noted in our SI (Table S2), those were the only possibilities that we could sample in body distance to the production well. And the isotope signature from this well was generally in very good consistency with our source partitioning results based on isotope measurements.

Regarding the measurements, we are sorry for the misunderstanding in our dataset and analyses. The CRDS system from Picarro used in our research has been widely applied for measuring CH_4 isotopic signatures from various sources (Menoud et al., 2022; Ars et al., 2024; Al-Shalan et al., 2022; Geum et al., 2024; Rella et al., 2015; Lu et al., 2021). As for the laboratory measuring our samples, the initial calibration was conducted only with 1-point calibration, mainly due to that the G2132-i system is rather stable throughout the time. Actually, we should note that, we have used two international primary standards (based on VPDB; Std1 and Std2, -68.6‰ and -40.0‰) and one secondary standards (House-standards, Hstd, -46.89‰) which had been cross-calibrated. Along each measurement sequence (see more details as demonstrated in the figure 1), we measure all three standards together with the standards (5 samples for each sequence), during which Std 1 is used for calibration and correction for sequence-drift, Std 2 used for quality control and Hstd used for constraining long-term drift. As for our case, the measurements were finished within less than a week, so we didn't correct for long-term drift. For simplicity consideration, we only adopted Std 1 for calibration (both delta calibration and short-term drift correction). Of course, the short-term drift was minor compared with delta calibration. Nevertheless, we value the comments from reviewer 3, and will recalibration and evaluate our datasets with 2-point calibration scheme. As shown in the figure 2, given the good correlation among all three standards, we believe that our Picarro G2132-i system is rather robust and will not significantly alter our final results and main findings.

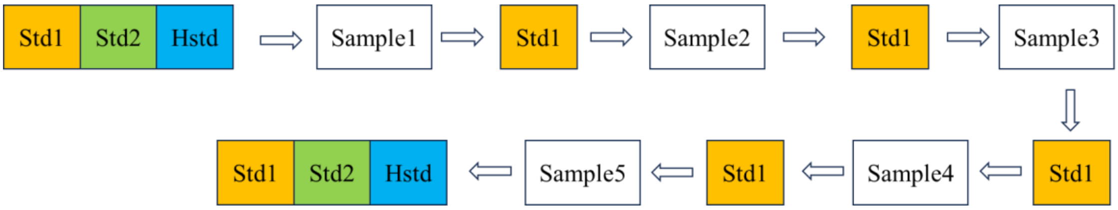

Fig.1 Methane Isotopic Analysis Procedure for Gas Samples

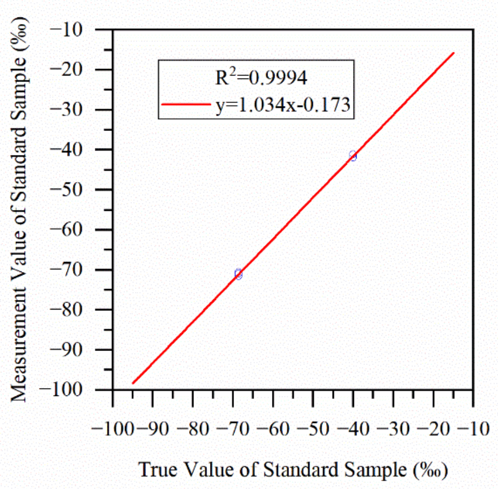

Fig.2 Calibration of the instrument was conducted by linear regression between the certified values and measured isotopic values of Std1 and Std2, ensuring accuracy and consistency in $\delta^{13}C$ measurements.

2. **To emphasize further comment a) above, it is important that you give a clear distinction between the facilities that are related to oil extraction with residual gas, as this gas component could be highly fractionated, or flared on site, with those that solely focus on gas upgrading and delivery to the network, where there is likely to be less isotopic variability. Your table of facilities does not make this clear. Combustion of fossil fuels will increase ^{13}C in any emitted residual CH_4, so you need to be sure that this is not part of the activities.**

R: Thank you for your further comments. As shown in Table 1, sites S1-S2 are for Natural gas processing, including membrane separation, adsorption, desulfurization, dehydration and other processes; sites S3-S6, and S10 are for Gas Collection and transportation; sites S7-S8 are for C 3+ component of natural gas was recovered by low temperature separation process. We will do our best to add information for description of our sampling points and major facilities in the nearby. However, as indicated in the 1st response from the major comments, we are restricted from directly approaching singular sources except for two production wells (Table S2). Another source we did not measure is CH_4 emission from flare exhaust, which is not easily accessible for UAV or man. Hence, we do not consider the contribution from flare exhaust, but focus more on average emission scenarios from these sites. The work is designed to measure mixing atmospheric samples from the ONG site, for more effectively evaluating CH_4 leakage from site-level and quantifying source signatures. For more details, please refer to the 1st response to the major comments from RC3.

3. **To emphasize further comment b) above, the manuscript shows very limited understanding of the measurements and instrumentation. Calibration gases are measured by either metrology or by isotope ratio mass spectrometry and assigned a ratio. Each mass spectrometer behaves differently and so needs at least a 3-point calibration to slope correct the measured data across a range of isotopic signatures, say -75 to -20‰. The slopes for CRDS instruments show even greater calibration slope correction factors, so correcting data with one standard at -69‰ will not give the correct calibration at -10‰. Additionally, the authors refer to instrument accuracies, when they are actually showing precisions. Accuracy can only be assessed after calibration using at least 3 reference points, and applying the calibration equation to measurements of the calibration gases as unknowns. As the isotopic calibration will not change within error of measurement in the long-term it should still be possible to retrospectively provide a correction to the data by adding calibrants that are close to atmospheric background isotopic signature and with a more ^{13}C-enriched signature.**

R: As mentioned in the 1st response to the major comments from RC3, we have indicated in full details regarding our calibration scheme and evaluations of our isotope results. We actually have laboratory standards spanning the ^{13}C range of -68.6 ~ -40.0‰, which may not be perfect but suitable for delta calibrations during our analysis. In addition, we only have two samples from the production wells to be in the range of -15‰~ -24‰, which were not even used for keeling plot evaluations as they were next to the singular sources. Regarding the reviewers' comments on the failure of calibration for -10‰, our analytical results were indeed rarely lying in this range (95% of the data points were between -68.6 and -40.0‰). Instead, the keeling plots results were higher, with some source signatures close to the range of -20.0‰. But this won't necessarily interfere with our calibration for the isotope data of the collection air samples.

We are also aware and have the possibility of conducting 2-point calibrations for reevaluating our analytical results. As indicated previously, we will recalibrate the isotope datasets and be more careful in interpreting our results. Regarding the mixing of precision and accuracy, we will double-check our texts and describe our analytical quality in clearer format.

4. **The authors also discuss results of vertical profiles without showing the measurement precisions, but from the calibration gas precision it is clear that for some vertical profiles the whole of the variation seen is within the measurement precision of the instrument, so these results have no meaning. This is also true for more than half of the source signature graphs presented.**

R: We thank the authors for the critical suggestions on the vertical profiles. It is important to note, for environmental atmospheric measurements, one would not expect a significant change or fluctuations of isotope signatures in CH_4, unless the sampling is not conducted in an open environment. Alternatively, the isotopic signatures will be clearer if next to source, but this is technically difficult and not feasible in most ONG-sites. Based on our dataset, we are able to confirm the clear and consistent source signatures of ground-based ONG CH_4 leakage. Certainly, there are uncertainties in the measurement itself as well as some influence of the surrounding environment or meteorological conditions. Nevertheless, based on our further detailed explanation of our analytical precision and reliability as well as the already discussed influence from meteorological conditions, we believe our results are able to reflect the CH_4 leakage from the ONG region in SW China.

5. **UAV sampling of air for laboratory isotopic measurement is not new, and there are papers at least back to 2016. For heights of a few hundred metres there is also the possibility to use AirCore sampling and get the full vertical profile rather than just at 3 or 4 heights.**

R: We thank the authors for the critical suggestions on the vertical profiles. It is important to note that this is the 1st study of site-level CH_4 leakage from Chinese ONG sites based on stable isotope techniques to our knowledge. This information could be of high value for unraveling the global debating on the variations of CH_4 isotope signatures from global background atmosphere. We are aware of the AirCore sampling methods (Karion et al., 2010)， which will be able to give the full vertical profile of CH_4 measurements. However, for the ONG sites under operation of CNPC (China National Petroleum Corporation), we only get short time windows to conduct in situ measurements, and the local site condition/facilities did not allow us to install such devices and long gas tubes for continuous measurement of CH_4, particularly considering these many sites we have visited (11 sites). Therefore, our study design was carefully planned based on the local situation, and the UAV method is most convenient and mobile (from site to site).

6. **There is no clear statement on what the Ground Open Area category is, or how it can be assigned an isotopic signature with any confidence. I am presuming that it is a mix of no sources, farmland sources and wetland sources, but this**

could give a signature anywhere between -70 and -55 ‰ depending on what the sources are and their relative proportions.

R: Thank you for your detailed comments. We are sorry for the unclear information for the surrounding environment in these rural areas. Around all ONG sites, the major land-use type is rural roads and paddy fields (scattered), through which small ditches or streams run. In those paddy fields and other rural areas, no livestock farm or landfill was present, thus excluding the possibilities of cows/pigs or waste in affecting CH_4 sources. In addition, during our sampling period (April), we didn't spot any biomass burning, which is also forbidden by law in China. Another important information to note is, our UAV sampling points are mostly located near middle or at least not close the edges of the ONG sites, which are much larger in area than the scattered paddy fields in the surroundings. For the revision, we will add the corresponding information to the methods and also the Table 1.

In addition, in a parallel study of our team conducted in the same region, we collected several ambient atmospheric samples for ^{13}C- CH_4 close to the paddy field (1.5 m above surface, 10-20 m from the borders of paddy rice fields). The isotopic values ($\delta^{13}C$-CH_4= -47.2 ± 0.2‰; unpublished) were quite similar to the global background. The little influence from paddy rice on the atmospheric measurements could be due to small CH_4 emission from the sampling periods (relatively dry in April for Sichuan region). Secondly, all stations are surrounded by high walls, which further reduces the impact of other methane sources on near-surface methane mixing ratios.

We agree that ground open is may not be precise enough to indicate the sampling spots. A few criteria for the ground-open area: no significant observation of CH_4 concentration elevation compared with ambient background (Chen et al., 2024)(based on portable TDLAS methane mixing ratios monitor); at least 20 m from any facility; no spot of pipelines or valves.

7. **I also found that the selection of literature being reviewed was quite limited and focused on a few geographical areas, missing a lot of key isotopic studies on methane.**

R: Thank you for your constructive comments. We will enrich the isotopic study in the revised manuscript to cover a broader geographical area.

Detailed Comments:

Abstract:

1. **Line 33 – should have some explanation how a different isotopic signature can suggest a global overestimation.**

R: Thanks. For global estimation of CH_4 emission from anthropogenic activities, current methods are depending on either top-down or bottom-up estimates based on atmospheric monitoring or emission inventories. Large uncertainties remain, particularly on the debate between biological sources (wetland and so on) and industrial sources (e.g., ONG and coal). While flux measurements cannot resolve such problem on a global basis, more and more work has recently adopted isotopic measurements to further indicate the likely contributions from either biological and industrial source(Schwietzke et al., 2016; Milkov et al., 2020). On top of such information derived from isotopes, researchers may further reduce the uncertainties in global estimation of CH_4 emissions. We will further elaborate this in the revision.

Introduction:

2. **Lines 102-104 – Start a new paragraph for the UAV topic. UAV have been used to collect samples for isotopic analysis using air sampling bags much earlier than the studies that you mention.**

R: Thanks. We agree with your comment. We will start a new paragraph for the UAV section. It is worth noting that UAV-based sampling is an earlier-developed technique; however, our intention is to emphasize the application of UAV monitoring at the station scale.

In the revision, we will re-screen the literature to cover the classic references.

3. **Lines 104-106 – Don't mix up UAV and large aircraft sampling. France et al. 2021 used a large aircraft, not a UAV.**

R: Thanks. We agree with your comment, and we will make the revision.

4. **Line 108 – studies concentrated in foreign countries. This statement depends on who is reading the text. You mean in countries outside of China.**

R: Thanks. We sincerely apologize for the lack of clarity in our wording; we will make the necessary revisions in the updated manuscript.

5. **Lines 111-114 – an isotopic signature is that which is calculated for the source. It cannot be used for the measurement of an atmospheric mixture. The signature will not change due to meteorology or sampling method. The measured values can be different due to sampling distance from the source and dispersion meaning that there is a larger component of the ambient air in the mixture, but the source signature will remain the same.**

R: Thanks. We agree with your comment. We sincerely apologize for the confusion in the description of isotope data. We agree that isotope signature reflects the source itself, but not the mixing isotope results. We will revise the manuscript accordingly.

6. **Lines 125-126 – what do you mean by Upper Atmosphere? You sampled at 200-300m. This is not upper atmosphere. That would be the mesosphere.**

R: Thanks. In this study, the term "upper atmosphere" refers to the 50 m, 100 m, 200 m, and 300 m heights. Alternatively, this refers to the atmosphere above these O&G plants. We will correct this in the revision.

Method:

7. **Table 1 – needs better formatting to avoid splitting words between lines. The column on surrounding environment needs more detail for farmland. Does it contain rice growing, ruminants or both?**

R: Thanks for the valuable comments. We will improve our formatting regarding the tables. For surrounding environment, as we noted previously in the response, it was mainly rural roads and paddy fields (scattered), through which small ditches or streams run. In those paddy fields and other rural areas, no livestock farm or landfill was present, thus excluding the possibilities of cows/pigs or waste in affecting CH_4 sources. For more details, please refer to our response to RC1.

8. **Figure 1 – needs a better picture to show how the bags and pump are connected to each other and to the UAV and to the sampling inlet. Perhaps drawing a schematic would be better than the top right picture.**

R: Thanks for the valuable comments. We will draw a schematic to show the UAV sampling system.

9. **Line 207 – as above, 200-300m is not high altitude.**

R: Thanks for your comments. We will provide a more accurate description.

10. **Lines 213-215 – precision not accuracy; you can only get accuracy if you calibrate against isotopic standards of known composition across the known measurement range. The Picarro G2132 is a discontinued instrument before 2024. Are these precisions quoted from Picarro for the G2132 or the newer replacement G2201i?**

R: Thanks for your comments. We will change “accuracy” to “precision”. These precisions were quoted from the Picarro G2132 model, originating from the manufacturer.

11. **Line 216 - you cannot have a 1-point calibration for isotopes, as the isotopic calibration slope is very different for the CRDS between say -70 and -30‰, probably making a difference of about 1‰ for every 10‰ change in measured isotopic value compared to the slope defined by metrology and isotope ratio mass spectrometry. Therefore, if you calibrate at -70‰ you will be then incorrect by 4‰ at -30‰. You need at least 3 calibration points across your range of measured values.**

R: As mentioned in the 1st response to the major comments from RC3, we have indicated in full details regarding our calibration scheme and evaluations of our isotope results. We actually have laboratory standards spanning the ^{13}C range of -68.6 ~ -40.0‰, which may not be perfect but suitable for delta calibrations during our analysis. In addition, we only have two samples from the production wells to be in the range of -15‰~ -24‰, which were not even used for keeling plot evaluations as they were next to the singular sources. Regarding the reviewers’ comments on the failure of calibration for -10‰, our analytical results were indeed rarely lying in this range (95% of the data points were between -68.6 and -40.0‰). Instead, the keeling plots results were higher, with some source signatures close to the range of -20.0‰. But this won't necessarily interfere with our calibration for the isotope data of the collection air samples.

We are also aware and have the possibility of conducting 2-point calibrations for reevaluating our analytical results. As indicated previously, we will recalibrate the isotope datasets and be more careful in interpreting our results. Regarding the mixing of precision and accuracy, we will double-check our texts and describe our analytical quality in clearer format.

12. Lines 257-258 – need to check some of the writing to avoid sentences like 'Backward trajectories were used ….. to calculate ….. back trajectories'.

R: Thanks for your comments. We will review and revise accordingly.

Results:

13. Line 283 - this is not the range shown on box and whisker plots (1.7 ppm and below -50‰).

R: Thanks for your comments. We will revise the box and whisker plots.

14. Figure 2 - how do you measure a CH_4 mixing ratio of 1.7 ppm that is well below the lowest measurement at any background site, using South Pole as the lowest?

R: Thanks for your comments. The lower end of the box-whisker plot was representing the large variation range of the sampling points, not necessarily indicating that the measured mixing ratio was 1.7 ppm. Instead, Table S2 shows all the results, with a minimum value of CH_4 mixing ratio to be near 1.88 ppm.

15. Lines 302-321 - you cannot make statements about changes with altitude when they are smaller than the instrument measurement precision over the measurement period of 120 sec, which at near background mixing ratios (1.9-2.2 ppm) is about ±1‰ at best. Also, you should show the precisions on these values. Your isotopic data will be compared to those made by IRMS at <0.1‰, so the reader needs to believe that your measurements are actually showing real variations and not just measurement noise.

R: As mentioned in the 1st response to the major comments from RC3, we believe that our Picarro G2132-i system is rather reliable and would allow us to interpret the variations along altitude. We would show the precision in the revision.

16. Lines 332-333 – you do not explain what is ground open area (also no explanation in the supplementary material), so what is it and how can you realistically assign an isotopic signature to it? Are we to presume that this is a mixture of no sources, natural wetland sources and farmland sources?

R: Please refer to the 6th response to the major comments from RC3 for details.

Discussion

17. Figure 5 – a problem with the low precision of CRDS for isotopes at close to background mixing ratios means that you could put any slope through most of these graphs, particularly as you have avoided to show any error bars on the measurements. Only those with trend errors of <2.5‰ are strongly correlated but again mostly by joining 2 clusters rather than a series of points along the line, an important factor to reduce source signature calculation errors.

R: We fully agree with the reviewer that, if data points are not evenly spread along the line, the estimation of source isotopic signatures with keeling plot approach may be relatively more uncertain, which is also the major drawback from the classic keel plot approach. However, in actual field applications, the interpretation is based on the natural extent of data collected by our sampling campaign and could not be manipulated for perfect regression. Therefore, as we indicated in the previous response to RC3, we believe that our data analysis and interpretation of source isotope signature is valuable and constructive for understanding O&G CH_4 emission sources, as long as we prove that our analytical precision and sampling method is reasonable. In the revision, we will further incorporate the errors from measurements and provide more quality assurance/discussion on the keel plot evaluation.

18. Lines 422-424 – the statistical analysis in Figure S4 is not valid as the interpretation hinges on 1 point and without it there would be no correlation.

R: We agree that such regression may be uncertain given the wide spread of all data points. However, we can not simply remove this point with replicate measurements from a field measurement study, as we believe the spread of data points as found in our *in situ* work is important and valid for interpretating the pattern particularly for field practice. In the revision, we will describe such relationship based on Fig. S4 with more cautions.

19. Lines 425-427 - presumably you measured upwind of each study area and collected a sample before sampling on site. If there was an influence from a wetland source for example you would see a higher CH_4 background and a depletion in ^{13}C.

R: Thanks for your comments. We fully agree with your comment. As described later in the manuscript, we used the S7 station as an example to illustrate the impact of wind direction on methane distribution. Based on our results, the wind effect was mostly a connection with the background atmosphere and not mixing with another source (higher CH4 mixing ratio).

20. Lines 435-437 - you should specify that this statement is for fossil fuel extraction sites. This does not necessarily apply to other sources, and a big pipeline leak (500-1000 kg/hr) can come from a small point source.

R: Thanks for your comments. We will specify it in the revision. It is true that, for fossil fuel extraction sites or even major pipeline leaks, a small point would result in very large emission, leaving the site size to be less important. In the revision, we will mention this together with the discussion of site size effect on the measurements.

21. **Lines 471-485 - the isotopes of small % gas residue from an oil well are easily fractionated. Purely gas production wells tend to have much more homogenised signatures that are better correlated with the temperature that the gas is produced at, the hotter the gas production window the more enriched in ^{13}C it is.**

R: Thank you for your comment. We agree that the enrichment of methane isotopes is related to temperature. Due to different formation conditions, methane from different geographical locations can exhibit significant differences in the concentration of ^{13}C.

22. **Figure 6 - you should say which studies you used for this compilation. There are far more studies outside of North America and China that you do not show here, so what you show is not representative. Additionally, ONG covers a lot of different source types. How can you be sure that you are comparing like with like? So, it is not an overview figure. If you are using this data you should focus on oil and gas production areas (which includes Romania), but not include studies on refined gas distribution networks in European cities.**

R: Thank you for your valuable comment. We will further enrich our coverage of literature and data sources in more regions, providing a detail table indicating the data sources (including whether the site is a production site or processing site). We need to clarify that the collected values were all from oil and gas production areas, without including studies on refined gas distribution networks.

23. **Lines 507-511 - you cannot pluck this statement out of the air with no explanation of how it is calculated (and even with the SI it is not very clear what are your defined parameters), and you should not compare gas as an oil field residual product, with gas production wells, or with a global average of -44‰ that includes coal, oil and gas sources with a range of 50‰ or more.**

R: Thank you for your comment. Firstly, we will revise the description in the SI to make it clearer and easier to understand.

We thank the reviewer again for the very constructive suggestion. We do agree that it is best to separate CH_4 isotopic signature for production and processing sites; however, due to limited data and site coverage in one project, this is difficult and not practical. In addition, we used our measurements to represent the CH_4 isotope signature from Chinese O&G industry, mainly because this is the very first site-level study and may not be comparable with other studies investigating geological sources. On the global basis, the studies on CH_4 isotope from coal, oil and natural gas industry are rather scare. In addition, the sensitivity practice was mainly used for indicating how CH_4 isotope signature evaluation from Chinese O&G industry may assist in flux estimations; more work is certainly necessary and indispensable for better constraining CH_4 emission estimates from Chinese O&G activities. Methane source isotopic data with broader coverage and higher resolution (including methane from all sources) will contribute to global methane emission inversion and lead to more accurate emission estimates.

24. Lines 514-515 - what is the relevance of this sentence? Yes, ponds have been overlooked, but most sources are not correct in the inventories, and there is much more work needed to reduce the uncertainties.

R: We sorry for the unclear wording that caused the misunderstanding. Our intention was to highlight that microbial methane has been underestimated in the accounting inventory, which is consistent with the findings of this study and serves to confirm the results presented in the manuscript. We will leave this out in the revision.

25. Line 521 - who says that it is an overestimation? If you increase biogenic sources by 30% and fossil fuel sources by 10% the result is a decrease in global ^{13}C.

R: Thank you for your valuable criticism. If you vary both biogenic and fossil fuel sources, the results will be different, and can even result in different directions. However, our sensitivity analysis was conducted based on the hypothesis that the global mean isotope signatures of CH_4 from biogenic sources remain constant. Therefore, to avoid confusion, we will clearly indicate that our sensitivity analysis is based on an ideal scenario.

26. Lines 524-525 - it does a lot more than just separate microbial and fossil sources and can distinguish a wide range of biogenic processes, such as between oxidation and reduction, between different feedstocks and diets. Microbial from fossil is just scratching the surface.

R: Thank you very much for your comment. We understand that more processes are interplaying in the regulation of global CH_4 isotopes and trends. Nevertheless, much remains unknown and is still complexed in the global CH_4 science community. Such topics may be out of the scope for our current work.

27. Line 529 - Unfortunately the data shown are not convincing, as the emissions categories are not characterized at source, or close to individual emission points.

R: Thank you for your critical comments. As mentioned in the 1st response to the major comments from RC3. We should note that we had a few samples collected near production wells, which is close to individual emission points. We believe, as we noted in the responses to RCs, our *in situ* work provides important field evidence of CH_4 emission and isotope characteristics from the O&G sites in China, and will likely bring significant scientific value to the research community.

Summary:

28. Line 541 – Unfortunately the work does not distinguish between sources, it only implies what the averaged fossil fuel signature might be.

R: Thank you for your feedback. We will clarify our descriptions in the summary. We agree that we mainly examined the CH_4 source from O&G industry, and this is not related with multiple source partitioning. The original purpose of source distinguishing refers to the tracing of source from different production units or the comparison of sites.

29. **Line 556 - The global CH_4 isotope databases of Schweitzke, Sherwood and Menoud contain measurements from direct sampling of sources and measurement by isotope ratio mass spectrometry to high precision. Measurements of source signatures with large errors should not be included in any database that will be used for global and regional modelling.**

R: We believe that our isotope data quality is useful for indicating the CH_4 emission patterns as well as source characteristics from Chinese O&G sites, as mentioned for several times in the responses to RC3. Also referred to the response for line 33, we used the isotope results from our study to mainly indicate the sensitivity of CH_4 isotope data in estimating industry CH_4 emission from oil and natural gas production/processing. This is similar to other global or regional studies(Fisher et al., 2017; Nisbet et al., 2016).

References：

30. **Burnham et al. is not correctly formatted.**

R: Thanks. We will revise it.

31. **There are 9 references to web sites, all ending in the word 'last' and all incorrectly placed within the list.**

R: Thanks. We will revise it.

Supplementary Data：

32. **S2 – how robust is the signature of -21.95‰? Signature is not based on correctly calibrated data so how do you know that it is typical? The precisions on many of the 11 station source calculations are very wide so these should not be used in averaging. Global fossil fuel includes a significant component of coal, whereas your ONG sites do not, so is this a good comparison?**

R: We apologize for the confusion particularly to the reviewer. We have explained in much detail in the previous replies, that our calibration is reliable for interpreting the source isotope signatures, although we unfortunately omitted the details in the earlier version. The overall measurement precisions for samples across the 11 sites were actually consistent and follow similar source characteristics as validated in the keel plot with all ground data points (-17.8‰; see the figure 3 below). In the revision, we will use the isotope signature obtained from the overall keel plot estimates and assess uncertainties in our sensitivity analysis.

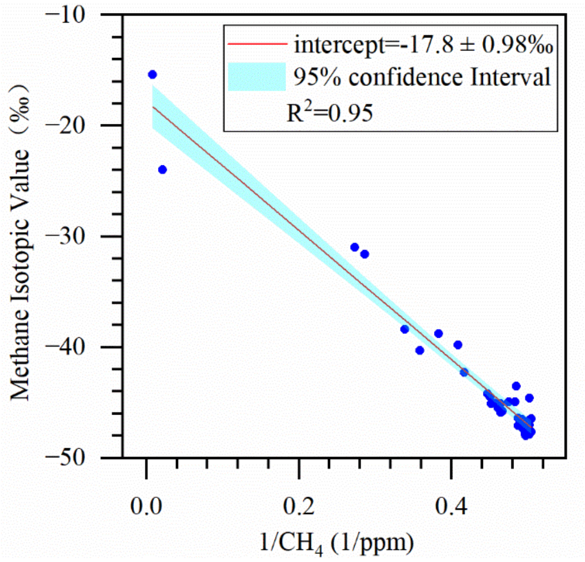

Fig.3 Analysis of Methane Source Isotopic Signature. The Keeling plot approach was applied using ground-level sample data from all stations.

33. S2 – you correctly use Saunois et al., 2024 here for the budget calculations, but in the main paper you use the older Saunois et al 2016 which has the budget to 2012.

R: Thanks. We will revise it to ensure the use of the latest data and maintain consistency.

34. Table S1 – basins are normally predominantly gas only, or oil with a bit of gas, and the gas only tend to have narrower isotopic ranges typical of mature gas. It would be useful to know what is the proportion of oil to gas extraction in these basins.

R: Thank you for your comment. We will make every effort to seek out relevant data.

35. Table S2 – Time column - this is not a time and looks more like a date. Presume 13 April. Need to show the sampling height for pipeline and production areas.

R: Thanks. We will change “time” to “date”. Regarding the sampling height, it is indicated in Table S2—for example, “H:100 m” denotes a sampling height of 100 meters.

36. Table S4 – I still have no idea what is Ground and how it can be assigned an isotopic signature.

R: As mentioned in the 6th response to the major comments from RC3.

37. Figure S4 - this statistical interpretation is not valid. Remove 1 data point and there is no correlation.

R: Please see our response 18 to RC3 as referred to line 422-424.

38. Figure S5 - so why was the plume to the SW of site S7 not sampled for isotopic analysis, to get a more precise source signature? The plume could have been

sampled at multiple points to give a spread of data on the Keeling plot and points above 10 ppm with much better CRDS precision.

R: Thank you very much for your constructive comment. It would be great if we could sample paired isotope data together with the plume measurement. However, this was conducted by another instrument by the other research team focusing on vehicle-based mobile monitoring (the data shown was collected at static mode).

39. **Figure S8 – why is this included? The global trend is not part of the study and this trend has been reported in many other studies that have thoroughly analysed the reasons behind this trend.**

R: Thank you for your comment. Figure S8 in our Supplementary Information is intended to provide background for our analytical calculations and to help better contextualize and understand our hypothetical estimates. We will remove it and only mention this in the contents instead.

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