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

RC1

General comments:

This paper provides important new isotopic measurements from methane emitted by China's Oil and Gas sector. China is the world's largest emitter of anthropogenic methane and isotopic data are essential if Chinese emissions are to be quantified by sector. Thus these new measurements are very valuable indeed. The paper should certainly be published. That said, there are a number of problems with the manuscript as it stands at the moment and it needs to be revised before final acceptance.

R: Thanks for your valuable comments and suggestions. Please find our detailed response below.

Specific points:

1. The introduction needs to be heavily rewritten between lines 37-107. It reads rather like something written at the start of the project some years ago and lightly updated. This is a very active field and many important recent references are missing, while a lot of good but very elderly papers are still cited. I would strongly suggest shortening this section (L37-107) by perhaps half and making it much more modern. I have added a list of papers that might be considered below. In particular I would draw attention to the ongoing work by Saunois et al, most recently in 2024/5. Maybe the International Energy Agency should be cited for China's total methane emissions. The state of methane should be updated to 2024 – see Michel et al. 2024 and Nisbet et al 2025. Given the focus on field measurement of isotopes, maybe there is one older reference (Dlugokencky et al. 2011) but many new papers.

R: Thanks for your suggestion. Regarding the introduction section from L37 to L107, we agree that more citations should be provided. We will further revise it and improve the conciseness.

2. From line 106-129 the introduction gets specific. That's good, but maybe there should be a paragraph on the power of isotopes.

R: Good idea. We have actually indicated the potential of CH_4 isotope techniques in quantifying CH4 source contributions in L69-76, before we move on to the introduction of ONG-related CH_4 monitoring or assessment. In the revision, we will add contents describing the application of methane isotopes in assessing CH_4 emissions from ONG industry.

3．Line 148 – Paddy fields – more information needed here. This is important because the isotopes help discriminate between ricefield methane and fossil methane. Also, how many cows and how much pig manure is in the region, and how many landfills. Another major factor is biomass burning, that can give very heavy methane (as in some later results in the paper).

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 CH4 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).

In the revision, we will discuss more carefully on the possible influence of surrounding environment on our isotope measurements based on UAV sampling.

4．Line 167 – maybe have a paragraph break here.

R: Thank you for your comment. Paragraph separation has been made accordingly.

5．Line 196 to 208 in Section 2.2 – no information is given about time of day and diurnal variation in the height of the boundary layer, yet this is obviously important to the later discussion.

R: Thank you for your comment. During our sampling campaign, the air sample collections based on UAV were conducted consistently around the noon time following consecutive days, as this is also according the regulations from ONG site managements (for external visitors). Mostly, the samples were taken between 11:00 am and 2:00 pm. We will indicate the time in the methods. Regarding the diurnal variation in the height of the boundary layer, we will add the information in the methods as well as in the discussion. However, we believe there won't be too much difference considering the short time window for our sampling.

6．Line 243-261 Hysplit - How much local diurnal understanding is there for

the movement of the boundary layer? Is there any information about the stability of the air masses during UAV sampling? Pasquill stability classes?

R: Thank you for your comment. This is a very good suggestion. As we noted previously, our sampling was conducted all in the daytime, along a rather short time window. Therefore, diurnal variability of boundary layer would not likely exert a large impact on the air mixing. We have relooked into our Hysplit-model analysis, and further computed stability index. It shows that the Pasquill stability class during UAV sampling predominantly was C (Slightly unstable conditions).

7 . Line 228 – Keeling plot. What line regression is being used? Maybe see Akritas and Bershady (1996) as used in France et al 2016 (see below for details)

R: Thank you for your comment. We will indicate more clearly in the method that "general linear regression method is used for keeling plot calculations, while the uncertainties were evaluated based on the ordinary least squares (OLS) method".

8 . Section 3.1 is the core of the paper and very valuable.

R: Thank you very much for your comments and recognition.

9 . Section 3.2 has no mention of time of day or diurnal evolution of the boundary layer. Also there is no real discussion of other local sources including rice and animals (isotopically light) and crop waste and other biomass fires (heavy). Some of the heavy values (e.g. in L356 could be from local fires. However the very heavy value directly measured in L361 is indeed interesting. Overall I think this section 3.2 of the paper needs a fairly major reevaluation.

R: Thank you for your comment. We fully agree that we should add more discussions on the variabilities of boundary layer regarding vertical mixing conditions as well as other sources contributed from non-ONG fields. For more details, please refer to our earlier response to RC1.

For the exceptionally heavy value as found for the well (YJ-01&02; Table S2), it was sampled next to leakage source (ground) and also observed with high CH4 mixing ratio, confirming that this is not influenced by surrounding environment. In addition, all the ONG sites were fenced with high walls for security reason.

Overall, the isotopic source signature of CH4 leakage from our studied sites is generally consistent either for individual sites or total average, supporting that the ONG sites in SW China represents a CH_4 leakage source that is distinctive in isotope signals. This can be attributed to both the geothermal sources or processing procedures from the local.

10 . Line 331 percentages are quoted to a precision far beyond the real uncertainty. About half and about a fifth to a quarter might be a more accurate

statement.

R: Thank you very much for your comments and suggestions. The necessary revisions will be made in the manuscript to make the description more accurate. We will also be more cautious in the discussion.

11．Line 365 onwards. The discussion should take into account other local sources – rice, animals, fires, and perhaps coal use. Fig 5 would be useful also a Table. Line 401 linear regression method not specified – see France et al / Ahritas and Bershady method.

R: Thank you very much for your comments and suggestions.

We have discussed these points in details in the previous responses to RC1. Regarding the local sources in the surrounding environment, we have supplemented additional information and descriptions. Overall, there is likely little influence from the surrounding environment (few residences and no biomass burning spotted).

Regarding the regression methods for keeling plot evaluations, we have indicated that "general linear regression method is used for keeling plot calculations, while the uncertainties were evaluated based on the ordinary least squares (OLS) method".

12．Line 451 onwards – global comparison – see references below.

R: Thanks for your suggestion. We will consider the references and enrich our comparison with other relevant studies.

CONCLUSION

This paper present important new results that will be very useful in attributing China's methane emissions to specific sources. The work should certainly be published. But the paper needs some work still.

REFERENCES to consider: don't cite all but pick and choose which fit best in the text as it is revised.

SPECIFIC Oil and gas and Keeling：

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Ars, Sébastien, et al. "Using in situ measurements of δ13C in methane to investigate methane emissions from the western Canada sedimentary

basin." *Atmospheric Environment: X* 23 (2024): 100286.

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Fisher, Rebecca E., et al. "Measurement of the 13C isotopic signature of methane emissions from northern European wetlands." *Global Biogeochemical Cycles* 31.3 (2017): 605-623.

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Zazzeri, G., et al. "Plume mapping and isotopic characterisation of anthropogenic methane sources." *Atmospheric Environment* 110 (2015): 151-162.

Zazzeri, Giulia, et al. "Carbon isotopic signature of coal-derived methane emissions to the atmosphere: from coalification to alteration." *Atmospheric Chemistry and Physics* 16.21 (2016): 13669-13680.

GLOBAL budget：

Michel, Sylvia Englund, et al. "Rapid shift in methane carbon isotopes suggests microbial emissions drove record high atmospheric methane growth in 2020–2022." *Proceedings of the National Academy of Sciences* 121.44 (2024): e2411212121.

Nisbet, Euan G., et al. "Practical paths towards quantifying and mitigating agricultural methane emissions." *Proceedings A*. Vol. 481. No. 2309. The Royal Society, 2025.

Nisbet, Euan G., et al. "Atmospheric methane: Comparison between methane's record in 2006–2022 and during glacial terminations." *Global Biogeochemical Cycles* 37.8 (2023): e2023GB007875.

Nisbet, Euan G. "New hope for methane reduction." *Science* 382.6675 (2023): 1093-1093.

Saunois, Marielle, et al. "Global methane budget 2000–2020." *Earth System Science Data Discussions* 2024 (2024): 1-147.

R: Thank you for the references you provided; we will cite some of them.