Author Responses (AR) to Referee Comments (RC) and resulting revisions of manuscript egusphere-2026-411: “Distinct dual-isotopic signatures of major methane sources in South Asia” by Peng Yao, Katja Belec, Henry Holmstrand, Josh Balacky, Abdus Salam, Krishnakant Budhavant, Mohanan Remani Manoj, Khaled Shaifullah Joy, Md. Alamin Hossain, Atinderpal Singh, Anil Patel, Neeraj Rastogi, Chinmay Mallik, Kirpa Ram, Gyanesh Kumar Singh and Örjan Gustafsson.

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We sincerely thank the editor and both reviewers for thoroughly reviewing and giving constructive feedback that is helping to clarify the significance and importance of this manuscript during revision. All reviewer comments are included below in *italic font* each followed by our detailed author responses, in normal font.

Anonymous Referee #1: [*https://doi.org/10.5194/egusphere-2026-411-RC1]*

This article provides very valuable data for understanding global methane emissions. The measurements concern both carbon 13 and deuterium isotopes in methane from several anthropogenic sources in South Asia. There were almost no data of this kind available in the literature before, therefore I recommend publishing this study. However, I think the methodology and interpretation needs to be severely revised first.

Response: We appreciate the recognition that this study fills an important gap and the overall strong support. We have addressed the detailed review comments below, which have contributed to further improve the manuscript.

General comments:

- *You don’t mention background samples in the methods, though Miller-Tans was chosen over Keeling, without background-related justification. In the Keeling plots of the supplementary material, we see the lowest concentrated samples are always >2 ppm… even more in the case of ruminants; which means the hypothesis of the stability of background conditions, which is inherent to the Keeling plot approach[1], isn’t fulfilled. Your data shows background samples taken at least for ruminants and biomass burning (labeled “blank”?), why aren’t they appearing on the Keeling plots?*

Response: We understand your concerns regarding the treatment of **background** in the **application of Keeling and Miller-Tans plots**. To address this point, we have revised the manuscript to add clarifications and discussion in both the main text and the Supporting Information.

(1) We would like to emphasize that the CH_4 concentrations in our source samples are very high, and therefore largely insensitive to background influence. Both Keeling and Miller-Tans approaches are fundamentally based on isotopic mass balance:

$$\delta_{obs} = \frac{c_{bg} \times \delta_{bg} + c_{source} \times \delta_{source}}{c_{bg} + c_{source}} \qquad (S1)$$

When the source concentration is much higher than the background:

$$c_{source} \gg c_{bg} \tag{S2}$$

Eq. (S1) simplifies to:

$$\delta_{obs} \approx \delta_{source} \tag{S3}$$

This implies that when source concentrations are sufficiently high, the observed isotopic composition approaches that of the source. Therefore, high-concentration data points alone can provide a good approximation of the source isotopic signature, even without applying Keeling or Miller-Tans analyses.

(2) Why stable background conditions are often emphasized in Keeling and Miller-Tans analysis? In practice, background conditions are never strictly constant, raising the question of how much variability is acceptable. Our answer is that it depends on the relative magnitude of source and background signals. To clarify this, we briefly revisit the mathematical structure of the Keeling approach.

In the Keeling formulation, background terms are embedded in the slope:

$$\delta_{obs} = c_{bg} \times (\delta_{bg} - \delta_{source}) \times \frac{1}{c_{obs}} + \delta_{source} \tag{S4}$$

As a result, variability in background leads to variations in the slope, which can distort the linear relationship (e.g., rotation or fan-shaped distributions in the data).

In contrast, the Miller-Tans formulation places the background contribution in the intercept:

$$\delta_{obs} \cdot c_{obs} = \delta_{source} \times c_{obs} + c_{bg} \cdot (\delta_{bg} - \delta_{source}) \tag{S5}$$

This means that the slope (i.e., the isotopic source signature) is less directly affected by background variability, while background fluctuations primarily influence the intercept, potentially increasing scatter without fundamentally altering the slope.

Nevertheless, both approaches reflect the combined influence of source and background variability. The key point is that the relative importance of background depends on the magnitude of source-driven variability. When source signals (variation of c_{source}) are much stronger than background signal (variations of c_{bg}), the derived Keeling intercepts and Miller-Tans slopes remain dominated by the source signal (c_{obs} variation dominated by c_{source}), even if background variability is present. This is consistent with the case of $c_{source} \gg c_{bg}$ (Eq. S2).

Conversely, when source-induced variations are comparable to background variability, even small background fluctuations may bias the inferred source signature. This situation may occur when source signals are weak or when observations are far away from the source but close to background conditions. For example, in some atmospheric observations over rice paddies, CH_4 concentrations are close to background levels, such that background variability, although small in absolute terms, can still influence Keeling-derived source signatures.

(3) In our dataset, atmospheric background conditions are represented in the Keeling plots shown in the Supporting Information Figs. S1–S2, where low-concentration data points (around ~2 ppm) are included for ruminant and biomass burning samples, although their influence is negligible due to the dominance of high-concentration samples ($c_{source} \gg c_{bg}$). For aqueous samples (rice paddies and wastewater), background methane is expected to be very low (effectively negligible), and the observed concentrations span several orders of magnitude, further confirming the dominance of source signals over background variability.

We have added the above clarifications to the Supporting Information (Section S2) to improve transparency and facilitate interpretation (Main text Page 12, Line 252–264). We agree that this is an important methodological consideration.

"Keeling and Miller-Tans plots are two formulations of the isotopic mass balance (Eq. 4), differing primarily in their treatment of background contributions. The Keeling approach (Eq. 2) derives the source signature from the intercept but is sensitive to background variability through its effect on the slope, which can distort linearity. In contrast, the Miller-Tans formulation (Eq. 3) derives the source signature from the slope, with background variability mainly affecting the intercept and increasing scatter while largely preserving linearity. As both methods rely on linear regression, increased scatter is generally less detrimental than distortion of linearity, making the Miller-Tans approach more robust in practical applications. Both approaches are most reliable when source-driven variability dominates over background variability. In our case, some high-concentration observations approach the condition $c_{source} \gg c_{bg}$ (Eq. 5), leading to $\delta_{obs} \approx \delta_{source}$ (Eq. 6), such that the influence of atmospheric background variability becomes negligible. Further discussion of background effects is provided in the Supporting Information Section S2." (Page 12, Line 252–264)

- *I also notice the source signatures are derived from samples taken at different sites, with potentially specific environments. How different the isotopic signatures per source would be if there were average of individual signatures calculated per site?*

Response: For some individual sites, the number of samples is not sufficient to apply Keeling or Miller-Tans analyses. Therefore, we performed a combined analysis using all available data. As discussed above, a subset of our samples exhibits very high methane concentrations, for which the isotopic composition can directly serve as a good approximation of the isotopic source signature. Therefore, we calculated weighted average isotopic values, which better represent the integrated characteristics across different sites and environments.

- *not sure if this would help, but the link between δ^2H of precipitations and into CH_4 from biomass burning was studied before[2]. Your study only looks at the link with microbial CH_4.*

Response: Thank you, this is a very interesting point. We have now added this relevant reference (δ^2H in H_2 from biomass burning) and included a brief discussion in the revised manuscript (Main text Page 26, Line 567–571). However, current constraints on the isotopic signature of methane from biomass burning remain limited at the global scale. In particular, the available data of methane δ^2H are still insufficient to robustly assess potential relationships with precipitation δ^2H across different regions and environmental conditions, so we cannot move further but only raise a potential hypothesis. Therefore, in this study, we focus on the link with microbial methane, for which observational constraints are relatively better established. Revised text:

"In addition, previous studies have shown that the δ^2H of H_2 produced from biomass burning exhibits a latitudinal dependence (Röckmann et al., 2010). By analogy, the δ^2H of CH_4 from biomass burning may also be influenced by the isotopic composition of surface water and precipitation. However, as shown in Fig. 2E (Supplementary Data S2), the currently available global dataset is too limited to resolve such variability." (Main text Page 26, Line 567–571)

Specific comments:

l. 57: "methane emitters" -> "methane emitting region"

Response: Revised as suggested.

l. *92: please rephrase to avoid too many pronouns.*

Response: Revised to "A global review of methane isotopic values was further conducted to compare with South Asian sources". (Page 5, Line 94–95)

l. 103-104 & 106: can you provide more details on the "clean air" you've used? What is the composition and/or manufacturer?

Response: Revised to "The flasks were pre-conditioned with clean air (Strandmøllen, 20.9% oxygen, and 79.180% nitrogen, $C_nH_m \leq 3$ ppm, $CO_2 \leq 1$ ppm, $CO \leq 1$ ppm, $H_2O \leq 3$ ppm) to eliminate contaminants." (Page 5, Line 105–107)

l. 123: Can you specify how deep the samples were taken (it is written "mid-depth") ? I am concerned by how representative of CH_4 emissions the dissolved CH_4 is. Do you have any information on the isotopic effect (fractionation) of transport processes (through plant-mediated transport or oxidation in the water column)? If not, I would say the distance to the surface is an important parameter to take into account.

Response: We have added additional details on the sampling procedure in the revised manuscript. The samples were collected at approximately 20 cm depth, although the exact depth varied depending on the flooding conditions in individual paddy fields. Sampling at this depth is expected to capture dissolved CH_4 that integrates multiple processes occurring within the water column, including plant-mediated transport and potential oxidation. At the same time, it minimizes direct influence from air-water exchange at the surface, as well as from additional complexities associated with the underlying soil. We have clarified this information in the manuscript:

"Samples were then collected by submerging the vials to mid-depth (approximately 20 cm depth, the exact depth varied depending on the flooding conditions in individual paddy fields) for 20 seconds until bubbling ceased, followed by an additional five-second hold." (Page 6, Line 125–128)

l. 167: "mesh size ##" ?

Response: Thank you for catching this, where ## should have been replaced by mesh size number. This has now been corrected to "mesh size 60–80".

l. 190 to 196: The Miller-Tans and Keeling approaches are based on assumptions. I would appreciate a more detailed analysis of which assumptions are valid in your case, and resulting arguments for choosing one approach or another.

Response: We agree and have now provided a quite detailed discussion above and in the revised manuscript. Here, we briefly repeat and summarize the key points. The primary difference between the two approaches lies in their treatment of background contributions: in the Miller-Tans formulation, the background term is incorporated into the intercept, whereas in the Keeling approach it affects the slope, making the latter more sensitive to background variability and potential distortion of linearity. As a result, the Miller-Tans method is generally less sensitive to background fluctuations. For our dataset, characterized by high methane concentrations, both approaches yield consistent results. Under these conditions, source-driven variability clearly dominates over background variability, such that the assumptions underlying both methods are well satisfied. Indeed, high-concentration samples alone provide a close approximation of the source isotopic signature.

l. 236 to 240: Your assumptions here need to be supported by references, to provide more evidences and precisions. For example: "more sensitive" (to which parameter?); "wide range of conditions" (what type of conditions? explain with clear variables or parameters).

Response: We have revised the comparison between the Keeling and Miller-Tans methods by starting from the fundamental equations, clarifying their differences and the conditions under which they are applicable to our dataset:

"Keeling and Miller-Tans plots are two formulations of the isotopic mass balance (Eq. 4), differing primarily in their treatment of background contributions. The Keeling approach (Eq. 2) derives the source signature from the intercept but is sensitive to background variability through its effect on the slope, which can distort linearity. In contrast, the Miller-Tans formulation (Eq. 3) derives the source signature from the slope, with background variability mainly affecting the intercept and increasing scatter while largely preserving linearity. As both methods rely on linear regression, increased scatter is generally less detrimental than distortion of linearity, making the Miller-Tans approach more robust in practical applications. Both approaches are most reliable when source-driven variability dominates over background variability. In our case, some high-concentration observations approach the condition $c_{source} \gg c_{bg}$ (Eq. 5), leading to $\delta_{obs} \approx \delta_{source}$ (Eq. 6), such that the influence of atmospheric background variability becomes negligible. Further discussion of background effects is provided in Supporting Information Section S2." (Main text Page 12, Line 252–264)

l. 240: "primarily" -> "only"

Response: Revised as suggested.

l. 246 and 250: the effect of C3/C4 types of vegetation on the CH_4 isotopic composition is well known; please provide references to support what you observed.

Response: We have added references that report and discuss the isotopic source signatures of C3 and C4 biomass burning as follows:

"There appeared to be a significant $\delta^{13}C$ difference between methane emissions from C3 and C4 biomass combustion globally (Vernooij et al., 2022; Nisbet et al., 2022), presumably driven by the differing $\delta^{13}C$ content of the feedstocks (Yao et al., 2022)." (Page 13, Line 269–271)

l. 261: refer to Table 1, since the calculation to understand it are explained here.

Response: We have added a reference to Table 1 at the appropriate location in the revised manuscript:

"Using the isotopic values measured for C3 combustion in South Asia, the global mean for C4 combustion, and the regional C3/C4 ratio, we derived a C3/C4-weighted $\delta^{13}C$ value of –29.5±2.0‰ for South Asia (Table 1)." (Page 13, Line 286–288)

l. 271: "In tropical regions, …" this sentence is true on the global scale, not specifically for tropical regions. Your C3 $\delta^{13}C$ data is in agreement with global, within the uncertainties. It provides evidence *of the type of plant being the main driver of CH_4 isotopic composition variations, in the case of biomass burning.*

Response: We agree that this statement applies at the global scale rather than being specific to tropical regions and have revised to:

"The relative proportions of C3 and C4 biomass remain a key determinant of isotopic signatures globally, while geographic variations have a minor influence." (Page 14, Line 298–300)

l. 300-301: Variations in δ^2H of CH_4 is in a way influenced by diet, as it reflects the hydrogen isotopic signature in water. This sentence is strange because the causality isn't very clear; what is it from the global mean value that suggests the δ^2H of CH_4 isn't influenced by diet? By diet, if you only mean C3 or C4 plants, please clarify.

Response: Thank you for pointing this out. We agree that the original sentence was ambiguous and could be misleading. We have revised the text accordingly. The δ^2H signature of CH_4 is primarily influenced by the isotopic composition of environmental water, which varies systematically with latitude and climate, rather than directly by diet (e.g., C_3 vs. C_4 plants). In this context, our intention was to convey that the hydrogen isotopic signature ultimately reflects the water ingested by the animals, which is controlled by environmental conditions. We have clarified this point in the revised manuscript to avoid confusion:

"The δ^2H signature of methane is expected to be primarily derived from surface water, and thus may exhibit regional variability. The global mean δ^2H value (–311±46‰) likely reflects this variability, which may arise from differences in the isotopic composition of environmental water as well as variations in rumination processes." (Page 15, Line 327–330)

l. 302-311: You write the adjusted δ^{13}C in methane (–63.3±1.1‰) compares well with the global value (–67.0±3.0‰), but C3-fed ruminant data for S Asia (-68.7±0.5‰) is written to be more depleted while being closer to the global value. It isn't consistent.

Response: Our previous description may have been ambiguous. These values correspond to the δ^{13}C signatures of C3 diet ruminants and the C3/C4-weighted mean, respectively. We have revised the manuscript to clarify this distinction:

"Methane emissions from C3-fed ruminants in South Asia (–68.7±0.5‰, Fig. 3A) were more depleted in δ^{13}C than the global mean of C3-fed ruminants (–67.0±3.0‰, Fig. 3D)." (Page 15, Line 331–332)

"Using the isotopic values measured for C3 diet ruminants in South Asia, the global mean for C4 diet ruminants, and the regional C3/C4 ratio, we derived a C3/C4-weighted δ^{13}C value of –63.3±1.1‰ for South Asia, which is comparable to the global C3/C4-weighted mean (–63.8±2.4‰)." (Page 16, Line 336–339)

l. 331: you state that ruminant δ^2H in South Asia deviates from the global mean, but it doesn't fall out of the range of uncertainty. Hydrogen isotopes can present large variations, with certainly more complex drivers linked to the H_2O cycle. Your result are within these variation window.

Response: We agree that the ruminant δ^2H values in South Asia still fall within the overall global range, but they are consistently offset from the global mean. This systematic deviation suggests a regional signal rather than random variability. We have clarified this point in the revised manuscript to better reflect both aspects:

"Ruminant methane showed similar $\delta^{13}C$ source signatures globally but displayed distinct $\delta^{2}H$ values in South Asia that deviate from the global mean (still within the uncertainty)." (Page 17, Line 359–360)

l. 338: The linearity of the Keeling plot is recognized as being poor, but the causes aren't discussed. Anyway, these plots can't be interpreted because are not scientifically valid (see general comment on Keeling plots)

Response: We agree that the linearity of the Keeling plot for the rice paddy samples is not ideal, and we have clarified this point in the revised manuscript and Supporting Information. We interpret the reduced linearity as reflecting the complexity of methane production and processing in rice paddies. In particular, multi methane sources and partial oxidation within the water column can generate isotopically distinct methane at different concentrations, effectively resulting in multiple coexisting source signatures. Under such conditions, both Keeling and Miller-Tans approaches tend to be influenced by the highest-concentration (least oxidized) endmember. However, because the Keeling formulation incorporates background contributions into the slope, the presence of multiple source-like components can act analogously to variable background, leading to reduced linearity. In contrast, in the Miller-Tans formulation, background contributions are incorporated into the intercept. When concentration differences span several orders of magnitude, the slope remains largely controlled by the dominant source signal, even in the presence of multiple components. For this reason, we primarily rely on the Miller-Tans approach for the rice paddy samples and have added a discussion in the manuscript to explain the limitations of the Keeling method in this context.

We have added more description in the revised manuscript (Main text Page 17, Line 367–386; Supporting Information Section S3) for the details.

"In contrast, the Keeling plots showed reduced linearity and more enriched $\delta^{13}C$ and $\delta^{2}H$ values (Supplementary Fig. S3), reflecting the complexity of methane production and processing in rice paddies (Supplementary Section S3.2). The sample concentration range spanned several orders of magnitude and some high-concentration samples satisfied the condition $c_{source} \gg c_{bg}$ (Eq. 5), yielding $\delta_{obs} \approx \delta_{source}$ (Eq. 6), and their isotopic values still exhibited noticeable variability, indicating the coexistence of multiple methane sources and/or the influence of in situ oxidation within the water column. Both Keeling and Miller-Tans methods are fundamentally designed for single-source perturbations; in multi-source systems, they tend to be biased toward the highest-concentration source (Monte Carlo mixing simulation in Supplementary Section S3.1), while weaker sources are suppressed or even negligible when concentration differences are large. In the Keeling method, background contributions are incorporated into the slope (Eq. 2). Under multi-source conditions, lower-concentration methane sources do not represent true background, but their influence becomes effectively indistinguishable from background variability within the Keeling framework. As a result, the combined variability of background and lower-concentration sources became significant in rice paddy samples, leading to deviations from linearity and reduced robustness. In contrast, the Miller-Tans formulation incorporates background into the intercept (Eq. 3); when concentration differences spanned several orders of magnitude for rice paddy methane, the slope was primarily controlled by the highest-concentration source, resulting in a more stable and interpretable relationship." (Page 17, Line 367–386)

l. 350-351: This hypothesis explains more depleted values obtained with Miller-Tans, what are the reasons for other methods to give higher values from the same samples?

Response: In our rice paddies dataset, the Miller-Tans δ^{13}C value (–53.8 ± 0.8‰) is relatively enriched compared to previous studies, whereas the δ^{2}H value (–311 ± 6‰) is consistent with the literature. The agreement in δ^{2}H suggests that oxidation is unlikely to be the primary driver of the observed differences, as oxidation would be expected to affect both isotopes simultaneously. Both the Keeling and Miller-Tans approaches tend to be weighted toward the highest-concentration source, meaning that the Miller-Tans result primarily reflects the isotopic signature of the dominant, minimally oxidized methane source. In contrast, simple averages and concentration-weighted means integrate contributions from a broader mixture of sources and therefore more strongly reflect the influence of partially oxidized methane. In rice paddies, plant-mediated transport is the dominant emission pathway, transferring methane from subsurface anoxic layers and thus generally preserving a relatively unoxidized isotopic signature. However, due to mixing within the water column and the presence of oxic microenvironments near roots, partially oxidized methane can also be entrained and transported through plant aerenchyma. Therefore, concentration-weighted averages are not without significance; rather, they provide a complementary perspective by capturing the integrated effect of both primary emissions and oxidation processes. We have revised the description for better understanding.

"A significant linear relationship between δ^{13}C and δ^{2}H (Fig. 4C) further supports the presence of methane oxidation, consistent with isotopic enrichment associated with methanotrophic activity (Schaefer and Whiticar, 2008). In rice paddies, only 1–2% of methane is emitted via diffusion through floodwater, whereas ~90% is transported via plant-mediated pathways (aerenchyma) and 8–9% through ebullition (Cicerone and Shetter, 1981; Schütz et al., 1989; Smartt et al., 2016). Plant-mediated transport primarily transfers methane from subsurface anoxic layers and is therefore generally less affected by oxidation. However, due to mixing and circulation within the water column and the presence of oxic zones near roots, partially oxidized methane may also be entrained and transported through plant aerenchyma. Ebullition is also less affected by oxidation, while the diffusion pathway is more susceptible to isotopic enrichment through oxidation.

Previous studies have primarily relied on atmospheric sampling, whereas this study focuses on aquatic measurements, raising questions of representativeness. Key challenges include the presence of multiple sources, oxidation processes, and multiple transport pathways. These issues are discussed in detail in Supplementary Sections S3.3–S3.4. Briefly, both atmospheric and aquatic sampling may be subject to representativeness biases, as the Keeling and Miller-Tans methods are dominated by the highest-concentration source (Supplementary Section S3.1), while contributions from lower-concentration sources may be indistinguishable from background variability within the fitting framework. Nevertheless, the Miller-Tans estimates are considered to best represent the isotopic signature of the dominant, minimally oxidized methane source and are therefore adopted as the most consistent metric across sampling approaches." (Page 18, Line 390–410)

l. 384-387: Indeed, the sensitivity to this source is very high. Not only it is important to apply region-specific signatures, but also to reduce the uncertainties by doing more measurements ? (Which is what your study started to do!)

Response: Yes, agree – thank you for seeing this importance. We revised the sentence to:

"applying region-specific isotopic source signatures and reducing the uncertainties is essential for accurately constraining methane emissions in South Asia." (Page 20, Line 445–446)

l. 395-396: Can you explain what you provide the values of the "concentration gradient", and the reasons why it can be linked to minimal oxidation? Generally for the water sources, were all the samples taken at mid-depth, and what does this imply in term of oxidation?

Response: Thank you for this question. The interpretation is based on the known direction of isotopic fractionation during methane oxidation. This can be inferred from the distribution of data points in the dual-isotope space (Fig. 5C), where oxidation would produce a characteristic co-enrichment trend in both isotopes. In our wastewater dataset, such a trend is not clearly observed, suggesting that oxidation is limited.

All water samples were collected at mid-depth. Under these conditions, the differences between rice paddies and wastewater systems are mainly related to microbial processes. Rice paddies typically host active methanotrophic communities that can oxidize methane, whereas wastewater systems often undergo substantial anaerobic treatment prior to sampling, favoring methanogenic conditions. As a result, methane oxidation is expected to be less pronounced in the wastewater samples. We have clarified these points in the revised manuscript:

"Methane oxidation would be expected to produce a characteristic co-enrichment trend in both isotopes. However, no clear relationship between δ^{13}C and δ^{2}H was observed for methane in wastewater (Fig. 5C). This lack of a systematic isotopic trend suggested minimal oxidation, indicating that degradation processes prior to release were limited for wastewater methane." (Page 21, Line 453–457)

l. 408: "dispersed and irregular patterns", or wider range of values?

Response: Yes, this is a clearer and more precise description. We have revised the sentence to:

"Other sources, such as composting, biogas fermentation and other organic waste decomposition (Lu et al., 2021; Bakkaloglu et al., 2022), exhibited wider range of values." (Page 21, Line 467–468)

l. 422-429: there can be variation up to 10 ‰ in the waste methane δ^{13}C, this is quite large. Also, we know wastewater CH_4 is more enriched than from landfills, but you don't have landfill data in your study. I think your wastewater results could be representative, as you claim on l. 224, but not for landfills. Please rephrase.

Response: We agree that these data are for wastewater and not for landfills. We have scrutinized and revised the text so that this should not be misunderstood:

"However, our global review showed only minor distinctions among various waste sources, suggesting that the isotopic signatures we measured in South Asia should be representative for wastewater in the region. Further exploring other waste sources and various factors may improve our understanding of methane emissions from the waste sector." (Page 22, Line 483–487)

l. 451-453: Please explain in which way the "general oxidation level" is reflected here. These more enriched signatures show that some oxidation occurred, but if you write "level", is it that you can quantify it?

Response: We cannot quantify the oxidation fraction, but the isotopic fractionation direction is known. When comparing with the Miller-Tans results, the isotopic values are more enriched, which means oxidation happened. The manuscript text has been slightly revised to more clearly articulate this:

"More enriched production-weighted concentration-weighted means (δ^{13}C=–41.7±7.5‰ and δ^{2}H=–236±45‰) reflected the influence of oxidation." (Page 23, Line 513–514) *l. 454: "these fractionation patterns". Do you refer to oxidation or diffusion here? Perhaps using "process" rather than "pattern" is more suitable?*

Response: Yes, oxidation process is more suitable here. Manuscript text revised accordingly (Page 23, Line 515).

l. 471: "similar" to what?

Response: The surface water and microbial sources are similar in regional patterns of δ^{2}H. We have revised the sentence to clarify this:

"Global microbial methane δ^{2}H exhibited a moderate or weak correlation with surface water δ^{2}H (Fig. 7; R^2=0.549 for ruminants, 0.363 for rice paddies and wetlands, 0.217 for waste), reflecting similar regional patterns among surface water and microbial sources." (Page 24, Line 530–533)

l. 470-471: can you provide values or representation of this correlation? It isn't very clear on the maps.

Response: We added the R^2 for the δ^{2}H of three different microbial groups with the surface water, i.e., R^2=0.549 for ruminants, 0.363 for rice paddies and wetlands, 0.217 for waste (Page 24, Line 530–533).

l. 472: "Hydrogen atoms in surface water likely served as a source for microbial methane, contributing to the observed spatial similarities in isotopic signatures.". Please refer

Response: We have added relevant references to support this statement, including early foundational studies that describe the origin of hydrogen atoms in microbial methane and their relationship with environmental water:

"Hydrogen atoms in surface water likely served as a source for microbial methane (Whiticar et al., 1986; Whiticar, 1999), contributing to the observed spatial similarities in isotopic signatures." (Page 24, Line 534–536)

l. 494: "resulting in fewer studies focusing on δ^{2}H". The lack of study on hydrogen isotopes isn't because of one or "Some studies", it's mostly because of the technical challenges in the measurements. Also, other studies point at the additional constrains hydrogen gives.

Response: We agree that the limited number of studies on δ^{2}H is primarily due to technical challenges associated with its measurement. We have revised the text accordingly and added recent studies highlighting that δ^{2}H can provide additional constraints on methane source attribution:

"Compared to the extensive observations and studies of δ^{13}C (Nisbet et al., 2023), measurements and constraints based on δ^{2}H remain much more limited, largely due to technical challenges associated with its analysis. However, a growing body of recent studies suggests that δ^{2}H can provide valuable additional constraints on methane sources (Dasgupta et al., 2025; Riddell-Young et al., 2025)." (Page 25, Line 556–560)

l. 513: "Conversely, methane from rice paddies and wastewater displayed more enriched δ^{13}C values than global means.". For wastewater, please compare with the mean for wastewater as well.

Response: We agree that a comparison between South Asian wastewater emissions and global wastewater-specific values is more appropriate. The sentence has been revised accordingly:

"Conversely, methane from rice paddies displayed more enriched δ^{13}C values than global means, and wastewater methane was more enriched in δ^{13}C relative to global waste means (Fig. 8A) and also global wastewater means (Fig. 5; Table 4)." (Page 26, Line 581–584)

l. 545: "and… and…". Please rephrase.

Response: We have rephrased the sentence to:

"In South Asia, reported emissions varied substantially in both magnitude and source composition, from 37±3.7 Tg C yr^{-1} in the 2000s (Patra et al., 2013) to 50.3 Tg C yr^{-1} in more recent estimates(Ito et al., 2023), with further estimates of 52 Tg C yr^{-1} from top-down approaches (n=6) and 58 Tg C yr^{-1} from bottom-up approaches (n=27) (Saunois et al., 2025)." (Page 28, Line 612–617)

l. 546: you mention seasonality. But does it affect all the sources, and why? Why not including this consideration in your analysis for the sources that are concerned?

Response: We discussed that seasonality can affect the strength of different methane sources, although the magnitude and mechanisms of this influence may vary among sources. In this study, seasonal variability is mentioned primarily for discussion purposes, rather than being explicitly incorporated into the determination of isotopic source signatures.

Our aim is to highlight that continuous atmospheric methane isotope observations, when combined with source-specific isotopic signatures, have the potential to provide additional constraints on temporal variations in source strength. Such seasonal details are often difficult to resolve using concentration measurements alone or through conventional bottom-up and top-down approaches. Explicitly incorporating seasonal variations in source isotopic signatures would introduce substantial additional complexity and uncertainty, potentially making the system underconstrained and more difficult to interpret robustly. We revised the content to avoid misunderstanding:

"Atmospheric methane in South Asia exhibited pronounced seasonal variations in both mixing ratios and isotopic composition (Rao et al., 2008; Tiwari et al., 2020; Metya et al., 2022; Guha et al., 2018), reflecting a combination of changes in source activity, transport, and atmospheric processing that are difficult to capture using conventional models. Given these limitations, regional isotopic source signatures, together with dual-isotope top-down approaches, offer an independent and valuable framework for improving constraints on regional methane budgets." (Page 28, Line 617–623)

l. 550: … if the underlying factors of variations are well-understood.

Response: We have revised the sentences to avoid misunderstanding:

"Given these limitations, regional isotopic source signatures, together with 619 dual-isotope top-down approaches, offer an independent and valuable framework for improving 620 constraints on regional methane budgets." (Page 28, Line 621–623)

l. 570: remove "potential"

Response: Removed.

Figure 1: I think adding the countries boundary lines would improve the figure.

Response: We initially included country boundary lines in the figure. However, after discussion among co-authors, we chose to remove them because some borders in the region are subject to dispute. To avoid potential geopolitical sensitivities and keep the focus on the scientific content, we present the figure without national boundaries.

Figure 2:

- *"isotopic characteristics" isn't the right phrasing; rather use "isotopic source signatures" or "isotopic composition"*
- *(D) and (E): I suggest to add a comparison with averages of this study.*

Response: We have revised "isotopic characteristics" to "isotopic source signatures" throughout Figures 2–5, which is indeed a more accurate description.

Regarding panels D and E of Figure 2, we have already provided a detailed comparison of South Asian and global values in Table 1. The South Asian values are displayed in Figure 2A–2B, while Figure 2D–2E show global statistics, which are clearly distinguished from the regional data. Including South Asian values in Figure 2D–2E would overly complicate the figure and could make it harder for readers to interpret. This separation was chosen to maintain clarity and avoid potential confusion. Furthermore, comparisons between South Asian and global values are presented again in Figure 8 and Table 5, ensuring that the regional-to-global context is fully emphasized without adding unnecessary complexity to Figure 2.

Table 1: Please explain in the legend that you've used the global values for C4 to derive the mean for South Asia.

Response: We added one sentence accordingly.

"The global δ^{13}C value for C4 biomass burning was applied in computing the weighted mean for the South Asian WM δ^{13}C of C3/C4." (Page 33, Line 670–671)

Figure 3, (D) and (E): I suggest to add a comparison with averages of this study.

Response: We appreciate the suggestion, but a detailed comparison of South Asian and global values is provided in Table 2. Figure 3A–B shows the South Asian values, while Figure 3D–E present global statistics, distinguished from the regional data. Including South Asian values in Figure 3D–E would overly complicate the figure and reduce readability. This separation maintains clarity, and regional-to-global comparisons are further emphasized in Figure 8 and Table 5, avoiding unnecessary figure complexity in Figure 3.

Figure 6: can you indicate the region boundaries for the averages we see on the figure?

Response: Some country borders in this region are subject to political dispute. To avoid potential geopolitical sensitivities and maintain focus on the scientific analysis, we will not display explicit political boundaries.

Figure S4: why 2 different color scales for the same variables? Also, the concentration units are in ppm on the x-axis, but should be L/nmol, considering the color scale and that there were water samples.

Response: We have now standardized the color scales across all three panels and corrected the x-axis labels with the proper units (L nmol^{-1}).

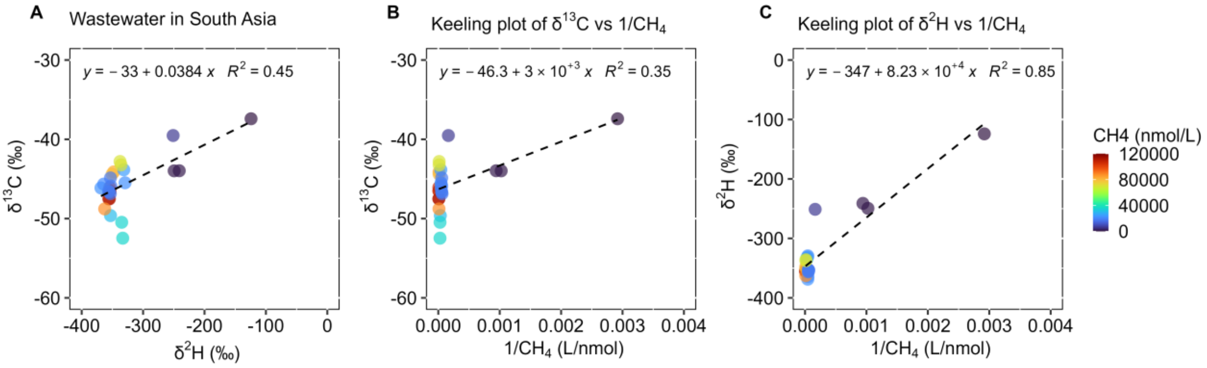

Fig. S4. Mixing ratios and isotopic characteristics of CH_4 from wastewater in South Asia. (A) Synchronous variations in $\delta^{13}C$ and $\delta^{2}H$ of CH_4. **(B)** Keeling plot for $\delta^{13}C$. **(C)** Keeling plot for $\delta^{2}H$.

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