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Distinct dual-isotopic signatures of major methane sources in South Asia

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Abstract

Methane is a powerful greenhouse gas contributing significantly to global warming. South Asia is a major methane emission region, yet source-diagnostic isotopic signatures remain poorly constrained, limiting top-down source attribution. To address this gap, we conducted extensive sampling and isotopic analyses of major methane sources in South Asia. Our results reveal substantial deviations of South Asian methane source fingerprints from global means. Methane from C3 biomass burning is more depleted in $\delta^{13}C$ (–30.9±2.2‰) but more enriched in $\delta^{2}H$ (–201±18‰), while ruminant methane (C3) is strongly depleted in both $\delta^{13}C$ (–68.7±0.5‰) and $\delta^{2}H$ (–343±6‰). In contrast, rice paddy methane is more enriched in $\delta^{13}C$ (–53.8±0.8‰) and $\delta^{2}H$ (–311±6‰), with their ratios signaling pre-emission oxidation. Wastewater methane shows enriched $\delta^{13}C$ (–45.0±2.4‰) and depleted $\delta^{2}H$ (–350±10‰) relative to global means, with minimal oxidation or spatial variation. These pronounced regional differences highlight the importance of using regionally constrained source fingerprints in isotope-based source apportionment. A global synthesis further shows that $\delta^{13}C$ signatures of biomass burning and ruminant methane are primarily controlled by C3/C4 feedstocks, whereas $\delta^{2}H$ is relatively insensitive to substrate type. Methane from rice paddies and wetlands exhibits strong latitudinal gradients worldwide. Combining emission inventories with source-specific isotope fingerprints reveals a mismatch with atmospheric methane in South Asia, suggesting an overestimation of rice paddy emissions and/or an underestimation of other microbial sources. These findings demonstrate the utility of top-down dual-isotope constraints to refine regional methane budgets and mitigation strategies.

Keywords: biomass burning, ruminant, rice paddy, wastewater

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1. Introduction

Mitigating methane emissions is critical for achieving the Paris Agreement 2°C target (e.g., (Rogelj et al., 2016)). Methane (CH_4) is a potent greenhouse gas (GHG) with a 20-year global warming potential 84 times that of an equal mass of CO_2, contributing ~20% to total global warming (Naik et al., 2023). Despite its significance, the drivers of recent methane increases remain uncertain (Nisbet et al., 2023; Schaeffer et al., 2025), highlighting the need for precise monitoring and effective mitigation strategies. Anthropogenic emissions are major contributors (Bousquet et al., 2006; Zhang et al., 2022; Saunois et al., 2025) and understanding methane sources and sinks is essential for targeted reduction efforts. The tropics, particularly South Asia, account for an estimated ~60% of global methane emissions (Jackson et al., 2020; Feng et al., 2022; Saunois et al., 2025). South Asia is one of the largest and fastest-growing methane emitters, with contributions believed to be primarily from anthropogenic sources (Stavert et al., 2022). The region experiences extensive biomass burning (Kirschke et al., 2013), hosts the world's largest ruminant population (Ganesan et al., 2017), is a major rice producer (Singh et al., 2021) and has substantial waste emissions from the dense population (Chakraborty et al., 2011). However, methane source apportionment and quantification in this region is limited and remain highly uncertain.

Methane sources are broadly classified as microbial, combustion and thermogenic (Whiticar, 1999). Microbial sources include e.g., wetlands, rice paddies, ruminants, landfills and wastewater (Masson-Delmotte et al., 2021). Combustion sources of methane is believed to be dominated by biomass burning emissions but also include coal combustion, traffic emissions and other combustion processes (Saunois et al., 2025). Thermogenic methane originates from fugitive emissions during fossil fuel extraction, transport and processing (Sherwood et al., 2017; Menoud

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et al., 2022). The spatial and temporal variability of these sources, coupled with the atmosphere's open system, introduces substantial uncertainties in methane estimates (Saunois et al., 2025). Bottom-up estimates of methane emissions remain uncertain due to varying methodologies and biases across different source sectors (Zavala-Araiza et al., 2015; Hristov et al., 2017). Recent satellite-based top-down observations have helped to improve some estimates (Lauvaux et al., 2022; Shen et al., 2023; Cusworth et al., 2024), yet are challenged by dispersed sources such as from ruminants and waste that are distributed through the landscape.

Estimates of methane emissions based on isotopic constraints are promising for fingerprinting the relative source contributions in an intercepted receptor setting, yet remain limited by uncertainties in both source-specific isotopic signatures and in atmospheric sinks. Moreover, large-scale top-down isotopic observations are lacking. Nevertheless, isotopic analysis can be a powerful tool for not only the source attribution but also for quantification of their reaction sinks (Fischer et al., 2008; Bock et al., 2017; Dyonisius et al., 2020; Nisbet et al., 2023). However, methane isotopic studies in South Asia remain highly limited (Rao et al., 2008; Metya et al., 2022), with isotopic source signatures nearly completely lacking (Metya et al., 2022). Establishing regional isotopic source signatures is critical for achieving source apportionment and reducing uncertainties in estimates of methane emissions.

In this study, we analyzed $\delta^{13}C$ and $\delta^{2}H$ signatures from four key methane emitting sources in South Asia, namely biomass burning, ruminants, rice paddies and wastewater. By evaluating isotopic variability across emission processes, sampling techniques and geographic regions, this work aims to constrain methane isotopic source signatures and thereby facilitate subsequent top-down isotope-based source apportionment to reduce uncertainties in methane emissions. A global

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review of methane isotopic values was further conducted to compare with those of these herewith constrained South Asian sources.

2. Materials and Methods

2.1. Gaseous and aqueous methane source sampling

Cattle ruminant samples in South Asia were collected using a custom-built sampling instrument. Sample air was passed through magnesium perchlorate (CAS# 10034−81−8, Alfa Aesar) to remove moisture, into an electrically-powered membrane pump (KNF Neuberger N86), and out into two cylindrical 1000 mL borosilicate 3.3 glass flasks (Normag, Germany) with axial inlet and outlet, connected in series. The inlet and outlet of each flask were sealed with a Normag needle valve with high-diffusion-minimized sealing. Tubing was made of PTFE and Synflex(R) and connections were Swagelok(R) and UltraTorr (TM). The flasks were pre-conditioned with clean air to eliminate contaminants. Before sampling, the flasks were conditioned in a 4-step protocol: Evacuated at high vacuum at 50°C for 12h, purged with nitrogen at 50°C for 2h, again evacuated at high vacuum at 50°C, for 3h, and finally filled with pre-conditioned clean air to a pressure of 1.3 bar (absolute). Sampling was conducted by positioning a funnel 2–5 cm from the cattle's mouths to capture their breath. The sample air was pumped through the flasks for 5 min, then closing the outlet valve and letting pressure build up to 1.7 bar (absolute), after which the flask valves were closed. Finally, flask in- and outlets were sealed with parafilm to prevent contamination from dust etc.

For combustion sources, we collected exhaust samples from agricultural crop residue burning in South Asian fields using the same custom-built instrument. Sampling was performed 3–15 cm

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from the burning rice paddies. A 0.45 μm inline gas filter was placed between the PTFE tubing and the metal tubing to remove aerosols. Each sampling session lasted 5 minutes, with the final flask pressure reaching 1.2 bar (absolute).

Samples were also collected to constrain the isotope fingerprints of aqueous microbial sources in South Asia, including rice paddies and wastewater. Rice paddy sampling involved dividing each paddy into four quadrants and taking one to three replicate samples from the center of each quadrant, totaling 4-12 samples per paddy field. For wastewater, three replicate samples were collected from sewage at each location. Before sampling, glass vials (VMR) were rinsed thrice with 125 mL of either rice paddy water or wastewater. Samples were then collected by submerging the vials to mid-depth for 20 seconds until bubbling ceased, followed by an additional five-second hold. The vials were then sealed with a bromine butyl rubber stopper (Apodan Nordic) attached to a string. After sampling, 0.5 mL of saturated $ZnCl_2$ solution was added as a preservative, and the vials were crimp-sealed, labeled, and stored at 4 °C in the dark before and after being shipped to Stockholm University for further analysis.

Thus, we collected a substantial number of methane samples from the four sources: ruminants, biomass burning, rice paddies and wastewater (see Supplementary Data S1 for details of each sample). Among them, ruminants and biomass burning represent two major sources of gaseous methane, while rice paddies and wastewater are significant atmospheric sources of aqueous, dissolved methane. The ruminant samples were obtained from 6 farms across South Asia, totaling 40 samples. For biomass burning, we conducted 4 sampling campaigns in different regions, collecting a total of 17 samples. Rice paddy samples were collected from 18 different rice-growing areas, amounting to 185 samples. Wastewater samples were gathered from 13 sewage treatment plants, totaling 38 samples. The sample distribution is illustrated in Fig. 1, with gaseous methane

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samples from biomass burning and ruminants primarily collected in Bangladesh, while aqueous methane samples from rice paddies and wastewater are distributed across Bangladesh and several densely populated regions of India. The background color of Fig. 1 represents total methane fluxes in 2023, sourced from EDGAR (Crippa et al., 2021), indicating significant methane emissions in South Asia.

2.2. Analysis of methane mixing ratios and isotopic composition

Methane mixing ratios were measured using gas chromatography with flame ionization detection (GC-FID, Agilent Technologies 7890A). For gaseous source samples, methane was extracted from a glass flask using a syringe and injected directly into the instrument. For aqueous source samples, a portion of the liquid was extracted, and helium (He) was introduced. After equilibration, a syringe was used to collect the headspace mixture of helium, methane and other dissolved gases for analysis. Three methane standards with methane concentrations of 1.6 ppm, 80.3 ppm and 250 ppm in synthetic air were used for calibration.

The equilibrium between the gaseous and aqueous phases was evaluated using Henry's Law (equation 1):

$$c = k \times P \quad (1)$$

where c is the concentration of dissolved methane (nmol L^{-1}), k is Henry's law constant, and P is the partial pressure of methane. For the calculations: the water volume was 40 mL, the headspace volume was 10 mL, the headspace pressure was 1 atm, the equilibration temperature was 25°C,

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the gas constant R was 0.08025 atm·L mol^{-1}·K^{-1}, and Henry's Law constant k for methane at 25°C was 0.0014 mol L^{-1} atm^{-1}.

Once the methane mixing ratios were determined, gaseous and aqueous source samples were analyzed for $\delta^{13}C$ and $\delta^{2}H$ using gas chromatography isotope ratio mass spectrometry (GC-IRMS; Delta V Plus, Thermo Fisher). Due to variable methane mixing ratios in source samples, two methods were used: pre-concentration (Precon) for diluted samples (Rice et al., 2001) and direct injection, using the GC injector, for concentrated samples. The Precon system was modified with custom-built components to improve isotopic analysis. In this configuration, only liquid nitrogen was used as the cryogen for all traps. CO_2 and water vapor were first removed with chemical absorbents, followed by Trap 1 for additional purification. Trap 2 (a 1/8" stainless steel tube, 20 cm in length, packed with HayeSep D, mesh size ##) was then employed, with sufficient venting through the Precon six-port valve to remove most of the residual oxygen that could interfere with $\delta^{2}H$ measurements. The sample was subsequently transferred to Trap 3 (a PoraPLOT capillary, 0.32 mm internal diameter), and final separation was performed on a 5 m × 0.32 mm PoraPLOT column at –78 °C (dry ice). This procedure ensured effective resolution of the methane peak from any remaining oxygen before conversion in the high-temperature reactor. Any interference by krypton (Kr) in the $\delta^{13}C$ analysis was eliminated by post-column GC separation from the methane-derived carbon dioxide peak (PoraPLOT 7 m × 0.32 mm; (Schmitt et al., 2013)). To match the relatively narrow detection range of the IRMS, syringe dilutions with He were applied. Isotopic values were corrected for instrumental drift and calibrated using standards.

Isotope values are reported in δ notation, representing the relative deviation of isotope abundance in a sample compared to international standards: Vienna Pee Dee Belemnite (V-PDB) for $\delta^{13}C$ and Vienna Standard Mean Ocean Water (V-SMOW) for $\delta^{2}H$. For diluted samples, the two standards

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used were both 1.85 ppm, with $\delta^{13}C$ values of −48.4±0.3‰ and −68.6±0.3‰, and $\delta^{2}H$ values of −63±5‰ and −240±5‰. For concentrated samples, $\delta^{13}C$ was measured directly using a 100-ppm standard with a $\delta^{13}C$ value of −43.8‰, while $\delta^{2}H$ was measured after pre-dilution and corrected using the same approach as for diluted samples. Analytical uncertainties of the reported isotopic composition are 0.09‰ for $\delta^{13}C$ and 2.1‰ for $\delta^{2}H$. The here constrained isotopic data of the major methane sources in South Asia are summarized in Supplementary Data S1.

2.3. Determination of isotopic source signatures

To determine the isotopic values of the sources, we analyzed the isotopic data for all samples using the Keeling (Keeling, 1958; Pataki et al., 2003) and Miller-Tans (Miller and Tans, 2003) methods. These approaches follow the equations 2 and 3:

$$\delta^{13}C_{obs} = c_{bg} \times \left(\delta^{13}C_{bg} - \delta^{13}C_{source}\right) \times \frac{1}{c_{obs}} + \delta^{13}C_{source} \quad (2)$$

$$\delta^{13}C_{obs} \cdot c_{obs} - \delta^{13}C_{bg} \cdot c_{bg} = \delta^{13}C_{source} \cdot \left(c_{obs} - c_{bg}\right) \quad (3)$$

where *c* represents the CH_4 mixing ratio, and the subscripts *obs*, *bg*, and *source* denote atmospheric observations, background levels, and source contributions, respectively. The Miller–Tans approach, which yielded narrow uncertainties, was used in the main text, while the Keeling plots are provided as additional information in Supplementary Figs. S1–S4.

We employed Kriging interpolation using the *gstat* package in R to evaluate the spatial distribution of isotopic values. This geostatistical method estimates values at unsampled locations based on the spatial autocorrelation of observed data, modeled through a fitted variogram. We applied this

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approach to interpolate δ^2H values of global surface water and representative microbial methane sources (ruminants, wetlands and rice paddies, and waste) for comparative spatial analysis.

To calculate methane isotopic source signatures and integrate contributions from multiple sources, we used a combination of statistical approaches. Uncertainty propagation was quantified using Monte Carlo simulations (10,000 iterations), accounting for variability in both isotopic measurements and source fractions.

2.4. Literature review of isotopic signatures of global methane sources

A comprehensive literature review was conducted to compile isotopic source signatures, which were further assessed for major global and regional methane sources (Supplementary Data S2). The review was carefully curated to minimize the influence of individual studies by selecting only a single representative value per region from each publication. Source-specific mathematical approaches were applied, as detailed in the following sections.

In the final section, we integrated the synthesized isotopic signatures with a range of top-down and bottom-up estimates to evaluate the discrepancies between current emission inventories and isotopic source constraints. Global data were compiled from our extensive literature review (Data S2). Isotopic values for microbial sources were calculated using Monte Carlo simulations, integrating our findings with estimates from Saunois et al. (2025) (Saunois et al., 2025), Ito et al. (2023) (Ito et al., 2023), and the IPCC (Masson-Delmotte et al., 2021) assessment. For South Asia, we incorporated isotopic signatures of rice paddy methane, while for natural wetlands, we retained tropical region values from the global review, as there is no evidence indicating significant methane oxidation in South Asian wetlands. Thermogenic methane isotopic values were sourced

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from extensive global (Sherwood et al., 2017) and European (Menoud et al., 2022) databases. The South Asian dataset focuses on methane sources across Afghanistan, Bangladesh, India and Pakistan. Thermogenic methane primarily originates from natural gas, coalbed methane, shale gas and other methane emissions associated with fossil fuel extraction, transportation and processing. This thermogenic category also includes minor contributions from biogenic methane present in various mineral deposits, incorporated to facilitate the source analysis of atmospheric methane.

3. Results and discussion

3.1. Methane from agricultural biomass burning

The isotopic source signatures of methane from agricultural biomass burning in South Asia was constrained and compared to measurements elsewhere (Fig. 2, Table 1, Supplementary Data S1–S2) to establish robust and representative source end-member values. The $\delta^{13}C$ and $\delta^{2}H$ values derived from Miller-Tans plots (Fig. 2A–2B) were –30.9±2.2‰ and –201±18‰, respectively. Keeling plots yielded comparable $\delta^{13}C$ values but slightly more enriched $\delta^{2}H$ values (Supplementary Fig. S1). The Keeling plot is mathematically rigorous, whereas the Miller–Tans method is empirical. The Keeling plot requires a more pronounced perturbation against a stable background, while the Miller–Tans method is more sensitive and provides more reliable estimates under weaker perturbations. In this study, we applied the more sensitive Miller–Tans method to account for a wide range of conditions. In these sampling campaigns, methane primarily originated from agricultural crop residue burning of C3 biomass. The linear relationship between $\delta^{2}H$ versus $\delta^{13}C$ showed that the isotopic composition was influenced by atmospheric methane, with a gradient reflecting the transition from source to atmospheric background values (Fig. 2C).

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To minimize bias from overrepresented datasets in specific regions, our global review consolidated data from each study and region into a single representative value (Fig. 2D and Supplementary Data S2). There appeared to be a significant $\delta^{13}C$ difference between methane emissions from C3 and C4 biomass combustion globally, presumably driven by the differing ^{13}C content of the feedstocks. By weighting $\delta^{13}C$ values according to the global proportions of C3 and C4 vegetation (77% and 23%) (Still et al., 2003), we derived a global biomass-type-weighted mean $\delta^{13}C$ value of –25.0±2.1‰. In contrast, the $\delta^{2}H$ values of methane from C3 vs C4 biomass burning did not exhibit a clear distinction (Fig. 2E), suggesting that $\delta^{2}H$ was not strongly influenced by biomass type. The mean $\delta^{2}H$ value for global biomass burning methane was –222±39‰.

Given that $\delta^{13}C$ variability in methane from biomass burning was influenced by the relative contributions of C3 and C4 biomass, these factors must be carefully considered when characterizing atmospheric-receptor isotopic signatures in specific regions. Based on our previous isotopic source apportionment of elemental carbon (EC) in South Asian atmospheric aerosols, C3 and C4 biomass combustion accounted for 90% and 10% of EC, respectively (Dasari et al., 2020). Since EC and methane are co-emitted during combustion, a first approximation is that they have the same proportional contributions. 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. In contrast, $\delta^{2}H$ was not influenced by C3/C4 composition and does not require such adjustment. Overall, methane from biomass burning in South Asia was more depleted in $\delta^{13}C$ and more enriched in $\delta^{2}H$ than the global mean (–25.0±2.1‰).

Global wildfire-related methane emissions may be underestimated due to undetected small fires (Zhao et al., 2025), highlighting the need for top-down constraints of biomass burning emissions.

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Estimate of methane emission from tropical biomass burning spanned a wide range of 14–34 Tg yr^{-1} (Kirschke et al., 2013), making alternative approaches to methane assessments in South Asia particularly important. A recent study reported $\delta^{13}C$ values of CH_4 from tropical biomass burning, ranging from –12‰ to –16‰ for grassland fires and –16‰ to –28‰ for farmland fires (Nisbet et al., 2022), which align with global estimates. In tropical regions, the relative proportions of C3 and C4 biomass remain a key determinant of isotopic signatures, while geographic variations have a minor influence. Additionally, combustion conditions and fuel moisture content can influence isotopic signatures, necessitating additional research to refine isotopic source characterization (Vernooij et al., 2022).

In South Asia, biomass burning is dominated by agricultural residue combustion and other fire types, such as wildfires and forest fires, and are expected to have similar methane isotopic signatures. Other combustion sources, such as traffic and coal combustion, contribute modestly to methane emissions but exhibit $\delta^{13}C$ signatures of their raw materials similar to C3 biomass (Yao et al., 2022). Improved isotopic characterization of these sources can enhance source attribution. In South Asia, biomass burning emissions displayed more depleted $\delta^{13}C$ and enriched $\delta^{2}H$ values than global means reported from elsewhere, reflecting regional variations in fuel type and C3/C4 biomass composition. Region-specific isotopic endmembers are therefore critical for accurate source apportionment.

3.2. Methane from ruminants

The isotopic source signatures of ruminant methane from South Asia were constrained and compared with such measurements globally (Fig. 3, Table 2, Supplementary Data S1–S2). The

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289 $\delta^{13}C$ and $\delta^{2}H$ values derived from Miller-Tans plots (Fig. 3A–3B), yielded –68.7±0.5‰ (primarily
290 reflecting C3 biomass) and –343±6‰, respectively. Keeling plots yielded comparable $\delta^{13}C$ and
291 $\delta^{2}H$ values (Supplementary Fig. S2). The relationship between $\delta^{2}H$ and $\delta^{13}C$ showed a clear
292 gradient as the isotopic composition transitions from the source to the atmospheric background
293 (Fig. 3C).

294 Methane isotopic values from global ruminant sources were summarized from the literature (Fig.
295 3D), revealing a notable $\delta^{13}C$ difference between C3 and C4 diets, driven by the distinct ^{13}C
296 content of these feedstocks. By weighting $\delta^{13}C$ values according to the global proportions of C3
297 and C4 diets (70% and 30%) from a recent database study (Chang et al., 2019), we calculated a
298 global C3/C4 biomass-weighted mean $\delta^{13}C$ value of –63.8±2.4‰. In contrast, the $\delta^{2}H$ values for
299 methane from ruminants globally showed no clear differentiation between C3 and C4 diets (Fig.
300 3E). The global mean $\delta^{2}H$ value was –311±46‰, suggesting that $\delta^{2}H$ in methane emissions was
301 not strongly influenced by diet composition or rumination processes.

302 Methane emissions from C3-fed ruminants in South Asia (–68.7±0.5‰, Fig. 3A) were more
303 depleted in $\delta^{13}C$ than the global mean (–67.0±3.0‰, Fig. 3D). However, regional variability in
304 C3/C4 feed composition was an equally important factor that must be considered when
305 determining the representative isotopic signature for South Asian ruminants. Based on a database
306 study (Chang et al., 2019), ruminant diets in South Asia consisted of approximately 65% C3 and
307 35% C4 plants. Using the isotopic values measured for C3 diet ruminants in South Asia, the global
308 mean for C4 diet ruminants, and the regional C3/C4 ratio, we calculated a C3/C4-weighted $\delta^{13}C$
309 value of –63.3±1.1‰ for South Asia. In contrast, $\delta^{2}H$ was not significantly influenced by C3/C4
310 dietary composition and does not require adjustment. After accounting for the C3/C4 feed ratio,
311 the $\delta^{13}C$ signatures of ruminant methane in South Asia were comparable to the global mean. In

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312 contrast, δ^2H signatures showed a substantial discrepancy, with depletion exceeding by 32‰ in
313 South Asia compared to the global mean, underscoring the importance of determining and using
314 regionally-constrained source fingerprints in isotope-based source apportionment studies.

315 Recent studies have indicated that biogenic methane emissions have increased in the tropics, with
316 considerable emissions from agricultural activities such as ruminant livestock farming and rice
317 cultivation (Schaefer et al., 2016). South Asia, home to the world's largest ruminant stock, is
318 potentially one of the major contributors to these emissions (Ganesan et al., 2017). Isotopic source
319 fingerprinting to characterize ruminant methane emissions in the tropics and South Asia offers a
320 promising approach to place quantitative constraints on the importance of ruminant and other
321 sources. Isotopic source signatures must be carefully adjusted based on regional dietary
322 compositions and environmental conditions, as the prevalence of C4 vegetation in tropical regions
323 results in more enriched $\delta^{13}C$ values in some areas, such as −57‰ in Kenya (Nisbet et al., 2022),
324 −52 to −57‰ in Zimbabwe (Brownlow et al., 2017), −60 to −63‰ in Australia (Lu et al., 2021),
325 and −65‰ in sub-Saharan Africa (Chang et al., 2019). Additionally, methane from ruminants is
326 primarily produced in the rumen through enteric fermentation and then exhaled (Hook et al., 2010),
327 but cattle are not the only ruminants contributing to methane emissions. Other species, such as
328 buffalo, sheep, and goats also play a significant role. Incorporating these additional ruminant
329 sources may help develop a more comprehensive isotopic characterization. Ruminant methane
330 showed similar $\delta^{13}C$ source signatures globally but displayed distinct δ^2H values in South Asia
331 that deviate from the global mean. Taken together, also for the ruminant releases, isotope-based
332 source apportionment of atmospheric methane should employ region-specific endmember values.

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3.3. Methane from rice paddies

The isotopic signatures of methane from South Asian rice paddies were quantified and compared with global values (Fig. 4, Table 3, Supplementary Data S1–S2). The $\delta^{13}C$ and $\delta^{2}H$ derived from Miller–Tans plots were –53.8±0.8‰ and –311±6‰, respectively (Figs. 4A–B). While Keeling plots exhibited a poor linearity and may be less reliable, they yielded even more enriched $\delta^{13}C$ and $\delta^{2}H$ values (Supplementary Fig. S3). Alternative statistical approaches, quantiles, arithmetic means, and concentration-weighted means, also produced more enriched signatures than the Miller–Tans method (Figs. 4D–E). Among them, the concentration-weighted mean ($\delta^{13}C$=–45.3±12.3‰, $\delta^{2}H$=–250±71‰) likely reflected methane dissolved in floodwater. A significant linear relationship between $\delta^{13}C$ and $\delta^{2}H$ in rice paddy water (Fig. 4C) was consistent with isotopic enrichment from methane oxidation by methanotrophic bacteria (Schaefer and Whiticar, 2008). However, diffusion through floodwater accounts for only 1–2% of total methane emissions from rice paddies, 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). Emissions via plant-mediated transport and ebullition are minimally affected by pre-emission oxidation, while the diffusion pathway is more susceptible to isotopic enrichment through oxidation. Therefore, the Miller–Tans values likely best represented the unoxidized, source-specific isotopic signature of rice paddy methane.

The global compilation of $\delta^{13}C$ and $\delta^{2}H$ values of methane emissions from rice paddies and wetlands revealed similar isotopic signatures of these two aqueous sources (Figs. 4F–4G). The global mean $\delta^{13}C$ and $\delta^{2}H$ values for rice paddies were –59.8±5.3‰ and –324±18‰, respectively, while these for wetlands were –60.0±7.6‰ and –309±49‰. Both sources exhibited clear latitudinal trends, with more enriched isotopic signatures in tropical regions and more depleted

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values in boreal zones. These patterns were consistent with previous observations, which attributed the depletion in boreal wetland $\delta^{13}C$ to reduced oxidation and the absence of C4 vegetation (France et al., 2022; Brownlow et al., 2017; Tyler et al., 1988; Fisher et al., 2017; Ganesan et al., 2018). In tropical and temperate zones, $\delta^{13}C$ values for rice paddies and wetlands were nearly identical. However, due to the absence of rice paddies in boreal regions, the global mean $\delta^{13}C$ value for rice paddy methane appeared slightly more enriched compared to that from wetlands. Conversely, global mean $\delta^{2}H$ value was slightly more depleted, potentially reflecting data availability biases, as boreal wetlands exhibited the most depleted $\delta^{2}H$ values. Methane from South Asian rice paddies (Miller-Tans values) was notably more enriched in $\delta^{13}C$ compared to the global mean, while $\delta^{2}H$ values slightly enriched than global mean. This enrichment was consistent with previous regional measurements (e.g., $\delta^{13}C = -54.3‰$ and $-57.2‰$; (Rao et al., 2008)) and might reflect enhanced pre-emission oxidation under South Asian field conditions.

Methane formation in rice paddies and wetlands primarily occurs via acetoclastic (acetate fermentation) and hydrogenotrophic (CO_2 reduction with H_2) pathways. The hydrogenotrophic pathway typically yields methane with more depleted $\delta^{13}C$ values, whereas acetoclastic methanogenesis produces methane with relatively enriched $\delta^{13}C$ values (Whiticar et al., 1986). The dominant pathway varies with substrate availability, temperature, and redox conditions across wetland and lake types. In wetlands, methane is also emitted through plant-mediated transport (~30%–90%; more than 90% in some studies), ebullition (up to ~60%; more than 90% in non-plant systems), and diffusion (up to ~30%) (Van Der Nat and Middelburg, 1998; Ding et al., 2002; Jeffrey et al., 2019; Villa et al., 2020; Ma et al., 2017), similar to rice paddies but with varying pathway contributions. Both methane source pathways and oxidation processes influence the isotopic composition of these aquatic emissions, although the extent of these effects remains

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uncertain and requires further study. Given the broad spatial coverage of our dataset, the Miller–Tans values for rice paddy methane reflected minimally oxidized isotopic signatures and were considered regionally representative. In contrast, isotopic values for wetland methane require further evaluation; currently, literature-based values from tropical regions are recommended. Given that approximately half of global methane emissions originate from aquatic ecosystems (Rosentreter et al., 2021) and South Asia accounts for ~20% of global rice production (Ganesan et al., 2017), applying region-specific isotopic source signatures is essential for accurately constraining methane emissions in South Asia.

3.4. Methane from wastewater

The isotopic source signatures of methane were constrained from South Asian wastewater and compared with global wastewater sources (Fig. 5, Table 4, Supplementary Data S1–S2). The $\delta^{13}C$ and $\delta^{2}H$ values derived from Miller-Tans plots (Figs. 5A–5B), yielded -46.4 ± 1.2‰ and -355 ± 5‰, respectively. Although Keeling plots exhibited poor linearity and may be less reliable, they yielded similar $\delta^{13}C$ and enriched $\delta^{2}H$ values (Supplementary Fig. S4). There was no clear relationship between $\delta^{13}C$ and $\delta^{2}H$ for methane in wastewater (Fig. 5C). The concentration gradient suggested minimal oxidation, indicating that degradation processes prior to release were limited for wastewater methane. The methane isotopic signatures were compared for isotopic quantiles, arithmetic means and concentration-weighted means (Figs 5D–5E). The median- and concentration-weighted means aligned closely with the values obtained from Miller-Tans plots, further supporting their reliability.

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A global review of $\delta^{13}C$ and $\delta^{2}H$ values was conducted for methane emissions from waste sources (Fig. 5F–5G), i.e., wastewater, landfills and other sources. The results indicated minor differences, suggesting that $\delta^{13}C$ and $\delta^{2}H$ signatures were not significantly distinct among various waste sources. Methane from global waste sources had mean $\delta^{13}C$ and $\delta^{2}H$ values of –54.0±5.4‰ and –295±18‰, respectively. Slight differences existed between methane emissions from wastewater and landfills, with wastewater showing more enriched $\delta^{13}C$ and slightly more depleted $\delta^{2}H$ values. Other sources, such as composting, biogas fermentation and other organic waste decomposition (Lu et al., 2021; Bakkaloglu et al., 2022), exhibited more dispersed and irregular patterns. Nonetheless, our findings showed that methane isotopic signatures from waste sources were consistent globally, which facilitated isotopic source apportionment. This similarity may be attributed to similar methane production mechanisms across these sources. Additionally, the narrow range of $\delta^{13}C$ values for global waste methane suggested minimal latitudinal variation, making further differentiation unnecessary. However, in South Asia, methane from wastewater was more enriched in $\delta^{13}C$ and depleted in $\delta^{2}H$ compared to the global mean values.

Methane emissions from waste sources were estimated to contribute approximately 12% of global anthropogenic emissions (Saunois et al., 2025). In South Asia, landfill methane emissions were particularly significant (Chakraborty et al., 2011), and atmospheric data also suggested that the waste sector played a key role in regional methane emissions, as supported by $\delta^{13}C$ constraints (Metya et al., 2022). Emissions from waste sources were also influenced by a range of factors, including microbial communities, temperature, pH, the CH_4/O_2 ratio, nutrient levels and inhibitory chemicals (Polag et al., 2015). Additionally, studies indicated that the operational status of landfills (active or closed) can influence the carbon isotopic signature (Bakkaloglu et al., 2022). However, our global review showed only minor distinctions among various waste sources, suggesting that

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the isotopic signatures we measured in South Asia should be representative for the region. Further exploring various factors may improve our understanding of methane emissions from the waste sector. Although isotopic signatures of methane from waste sources showed limited variability globally, values in South Asia deviated significantly from the global mean. This highlights the need for region-specific isotopic endmembers also for wastewater in methane source apportionment studies.

3.5. Geographical distribution

There are geographic variations in methane isotopic compositions across the globe for any source class due to a combination of environmental factors and source materials. The isotopic signatures of microbial methane vary across regions due to multiple factors, including differences in raw materials, methanogenic pathways (Whiticar et al., 1986; Conrad, 2005), and the methane oxidation by methanotrophic bacteria. These factors are essential to consider and suggests that region-specific and sometimes system-specific isotope source fingerprinting are necessary to facilitate accurate isotope-based source apportionment. Previous studies identified correlations between methane isotopic values and regional environmental factors (Sherwood et al., 2017; Douglas et al., 2021). Building on our isotopic data and a comprehensive literature review, we investigated the geographic distribution of the isotopic signals of microbial methane in South Asia and worldwide.

The geographical distribution of methane isotopic signatures in South Asia was assessed for two microbial sources: rice paddies and wastewater (Fig. 6). Regional Miller–Tans-derived values for rice paddy methane showed substantial variability (Fig. 6A), with similar signatures in western

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India, the Indo-Gangetic Plain (IGP), and Bangladesh, but more depleted values in southern and eastern India. The enrichment in both $\delta^{13}C$ and $\delta^{2}H$ (Fig. 4C) suggested that pre-emission oxidation was the dominant driver of this spatial variation. Given that rice cultivation was concentrated in the IGP and Bangladesh (Gumma, 2011), the production-weighted means of Miller–Tans values ($\delta^{13}C$=–45.5±2.5‰ and $\delta^{2}H$=–266±17‰) represented pre-oxidation signatures of floodwater methane, though partial oxidation and associated fractionation may still be present. More enriched production-weighted concentration-weighted means ($\delta^{13}C$=–41.7±7.5‰ and $\delta^{2}H$=–236±45‰) reflected the general oxidation level. Although diffusion contributes only ~1–2% of rice paddy methane emissions, these fractionation patterns may offer insights for wetlands, where diffusion accounts for a larger share (5–30%). Nevertheless, the overall Miller–Tans values ($\delta^{13}C$=–53.8±0.8‰ and $\delta^{2}H$=–311±6‰; Fig. 4A) were minimally influenced by oxidation and best represented the unaltered, source-specific isotopic signature of rice paddy methane.

Wastewater methane isotopic signatures exhibited minimal regional variation, with India and Bangladesh showing similar $\delta^{13}C$ values (Fig. 6B). Pre-emission oxidation of wastewater methane was negligible (Fig. 5C). To better represent regional emissions, we applied population-weighted averaging, assuming similar per capita methane production across areas, yielding $\delta^{13}C$=–45.0±2.4‰ and $\delta^{2}H$=–350±10‰.

Our global synthesis revealed pronounced latitudinal variations in the isotopic signatures of methane from wetlands and rice paddies (Figs. 4F–4G). Beyond the effects of oxidation and vegetation type, regional water conditions may also influence the hydrogen isotopic composition of microbial methane. To investigate this, we compared the global distributions of $\delta^{2}H$ in surface water (H_2O) and microbial methane (Fig. 7). Surface water isotopic data were sourced from the literature (Nan et al., 2019; Global Network of Isotopes in Precipitation (GNIP); Halder et al.,

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2015), and microbial methane δ^2H values were derived from our dataset and the global review. Global microbial methane δ^2H exhibited a moderate correlation with surface water δ^2H (Fig. 7), reflecting similar regional patterns. This correlation was particularly pronounced in North America. Hydrogen atoms in surface water likely served as a source for microbial methane, contributing to the observed spatial similarities in isotopic signatures. Among microbial sources, δ^2H values varied by source category: ruminants exhibited the most depleted isotopic values, followed by waste, while rice paddies and wetlands were relatively more enriched in isotopic composition. In tropical regions, microbial methane δ^2H values were more depleted than global mean values, potentially indicating unique microbial and/or environmental processes that require further investigation. Variations across microbial sources mainly stem from differences in methanogenesis, with each source maintaining internal consistency.

Latitudinal variations in aquatic methane δ^2H (from rice paddies and wetlands) appeared to be influenced by both water isotopic composition and pre-emission oxidation. In South Asia, $\delta^{13}C$ and δ^2H enrichment in rice paddies methane (Fig. 4C) provided clear evidence of oxidation. Additionally, the latitudinal patterns of aquatic methane δ^2H closely mirrored those of surface water δ^2H (Fig. 7C; Figs. 4F–G), suggesting both factors may contribute. Similarly, ruminant methane exhibited parallel δ^2H trends with surface water across latitudes but showed minimal oxidation, as reflected by depleted δ^2H values (Fig. 7B) and a narrow δ^2H range globally (Fig. 4G), likely due to direct atmospheric release. In contrast, waste sources showed minimal δ^2H enrichment (Fig. 7D) and narrow $\delta^{13}C$ and δ^2H distributions globally (Figs. 5F–5G), suggesting limited impacts from water sources and oxidation. In comparison, biomass burning methane exhibited a consistently narrow global δ^2H range (Fig. 2E), as it was minimally influenced by surface water and was emitted directly into the atmosphere without oxidation.

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Data scarcity in many regions limited the development of a comprehensive global distribution map (Fig. 7). Some studies supposed that δ^2H values provided weaker constraints on methane isotopic sources compared to $\delta^{13}C$ values (Nisbet et al., 2023), resulting in fewer studies focusing on δ^2H. Nevertheless, other research indicated correlations between the δ^2H of surface water (and precipitation) and the δ^2H of aquatic methane sources in certain regions (Douglas et al., 2021). Our results indicated that δ^2H followed predictable trends shaped by surface water isotopic composition and microbial processes. The correlation remained valid on a global scale (Fig. 7), though it was weaker, as numerous factors collectively influenced the isotopic signatures of each microbial source. Therefore, incorporating δ^2H into isotopic source apportionment can enhance our understanding of the factors driving the rapid rise in global methane concentrations. Despite progress, studies on methane isotopic source signatures remain incomplete, with significant data gaps across many regions. This study alleviated some of these gaps for South Asia, contributing to the required source fingerprint data for isotope-based source apportionment of airshed-receptor methane.

4. Summary of methane isotopic signatures in South Asia and globally

The extensive new source-isotope datasets were combined with earlier studies to yield updated dual-isotope endmember databases for South Asia and the globe (Fig. 8 and Table 5). Methane isotopic signatures for several sources differed in South Asia relative to their global means. Biomass burning and ruminant emissions in South Asia, both primarily associated with C3 biomass, exhibited more depleted $\delta^{13}C$ values than global means (Fig. 8A). Conversely, methane from rice paddies and wastewater displayed more enriched $\delta^{13}C$ values than global means. For δ^2H, methane from biomass burning and thermogenic sources in South Asia was more enriched

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than global means (Fig. 8B). Among microbial sources, ruminants and wastewater were more depleted in δ^2H, while rice paddies were more enriched than global values. The δ^2H versus $\delta^{13}C$ comparisons between South Asian and global methane sources provided a two-dimensional perspective (Fig. 9). While South Asian sources generally aligned with global categories, they exhibited distinct deviations. South Asian isotopic signatures showed a narrower distribution, whereas global isotopic signatures displayed greater variability. Among microbial sources, South Asian isotopic signatures appeared tighter constrained than their global counterparts.

Based on previous bottom-up and top-down studies, emissions-weighted microbial methane isotopic signatures in South Asia ranged from $\delta^{13}C$=–54.6±1.2‰ and δ^2H=–323±8‰ (Ito et al., 2023) to $\delta^{13}C$=–57.1±1.8‰ and δ^2H=–329±11‰ (Saunois et al., 2025). These $\delta^{13}C$ values are notably more enriched than the global compiled one ($\delta^{13}C$=–60.2±4.8 and δ^2H=–308±32) (Masson-Delmotte et al., 2021; Saunois et al., 2025), largely due to substantial rice paddy and waste contributions. Considering the $\delta^{13}C$ of atmospheric methane in South Asia (e.g., −47.41±0.94‰ in India (Metya et al., 2022)) and accounting for isotopic fractionation during OH oxidation (approximately 6–7‰ in $\delta^{13}C$ (Whiticar and Schaefer, 2007; Fischer et al., 2008; Schwietzke et al., 2016)), the inferred isotopic values of the total source approach or even fall below the microbial estimates. This discrepancy suggests biases in current emission inventories, likely overestimating rice paddy emissions and underrepresenting other microbial sources. These uncertainties highlight the need for dual-isotope measurements at receptor sites to better constrain methane budgets in South Asia.

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5. Concluding discussion

Methane emission estimates remain considerably uncertain. Some studies attributed recent atmospheric methane increases primarily to microbial sources, including tropical wetlands (Saunois et al., 2025), waste and agriculture (Peng et al., 2022; Michel et al., 2024), while others highlighted contributions from thermogenic and ruminant sources (Chandra et al., 2021). Biomass burning was also proposed as a significant contributor (Zhao et al., 2025). Bottom-up inventories showed large discrepancies, with estimates differing by severalfold (Stavert et al., 2022). 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 more recent values of 50.3 Tg C yr^{-1} (Ito et al., 2023), and 52 Tg C yr^{-1} for top-down (n=6) and 58 Tg C yr^{-1} for bottom-up (n=27) (Saunois et al., 2025). Methane emissions 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 dynamic shifts in source activity that are difficult to capture using conventional models. Given these uncertainties, dual-isotope top-down approaches offer an independent and valuable tool for constraining regional methane budgets.

Comparisons of methane isotopic signatures between South Asian and global means revealed significant distinction (Figs. 8–9), underscoring the need for region-specific isotopic data to ensure accurate source apportionment. $\delta^{13}C$ signatures reflected feedstock characteristics, distinguishing sources such as biomass burning and ruminants based on C3/C4 biomass ratios. Similarly, aquatic methane $\delta^{13}C$ was influenced by organic precursors, with South Asian sources showing enriched values compared to other regions. Globally, $\delta^{2}H$ in methane appeared linked to surface water and organic interactions, but highly depleted $\delta^{2}H$ observed in South Asia suggests different microbial processes requiring further investigation. Additionally, pre-emission oxidation significantly

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affected methane from rice paddy water in South Asia, warranting more research to better understand this process and similar processes in other aqueous sources.

The availability and accuracy of isotopic source signatures was critical for constraining methane sources (Schwietzke et al., 2016). At present, isotopic measurements of tropical methane sources remain scarce, particularly for δ^2H, still limiting their use in atmospheric top-down source constraints. While $\delta^{13}C$-based constraints are growing in applications globally (Nisbet et al., 2023), δ^2H constraints have been underutilized due to data limitations and unclear geographical distribution. Our study enhances the isotopic source fingerprint database, especially by adding δ^2H data for sources in South Asia.

While isotopic source signatures of major methane sources in South Asia are now improved, estimating the isotopic composition of well-mixed atmospheric methane remains challenging due to potential fractionation during oxidation (e.g., OH and Cl radicals). Existing models applied fixed isotopic fractionation factors, yet these vary considerably across studies (Whiticar and Schaefer, 2007; Fischer et al., 2008; Rice et al., 2016; Schwietzke et al., 2016; Schaefer et al., 2016; Bock et al., 2017; Sherwood et al., 2017; Douglas et al., 2021; Nisbet et al., 2023; Michel et al., 2024; Thanwerdas et al., 2024; Fujita et al., 2025). Despite these uncertainties, background methane mixing ratios and isotopic compositions in South Asia and globally remain relatively stable, indicating that a steady-state approach, incorporating region-specific isotopic fingerprints, may help reconcile inconsistencies in current methane budget estimates.

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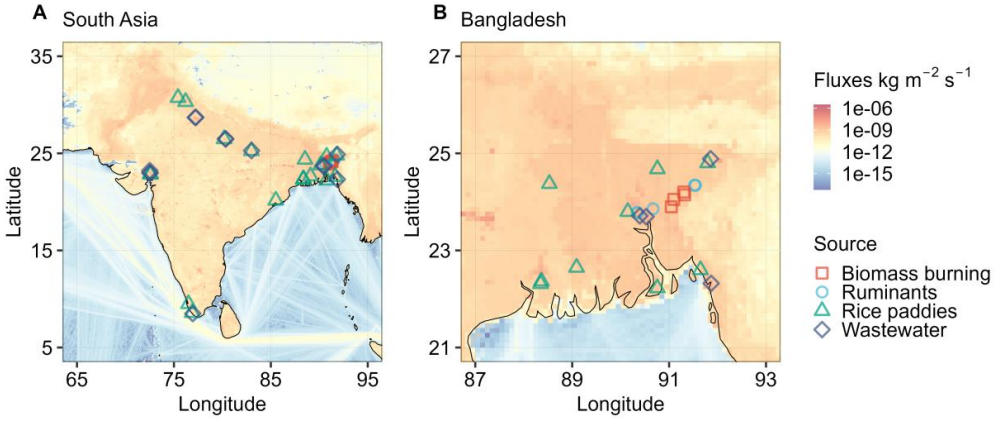

579

580 **Fig. 1. Map of collected methane source samples for (A) entire South Asia and (B) a close-up**
581 **for Bangladesh.** The background color represents total methane fluxes in 2023, sourced from
582 EDGAR (Crippa et al., 2021).

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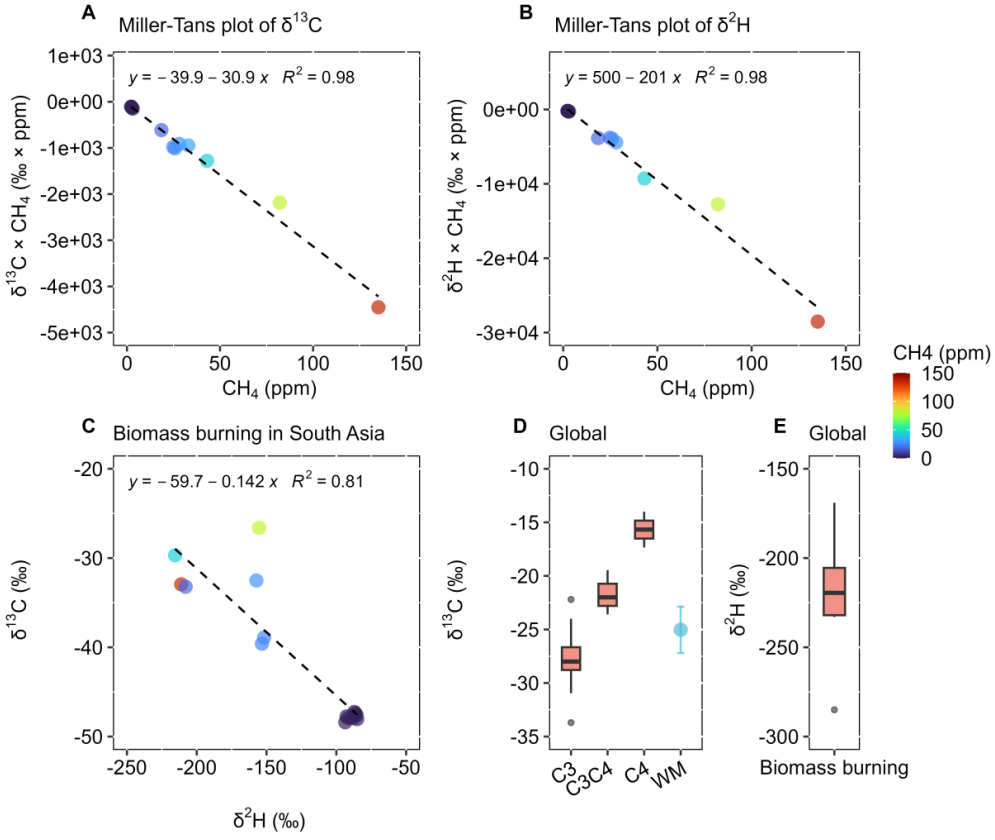

584

585 **Fig. 2. Isotopic characteristics of methane from biomass burning in South Asia and globally.**
586 **(A)** Miller-Tans plot of $\delta^{13}C$-CH_4 for South Asia crop residue burning. **(B)** Miller-Tans plot of
587 $\delta^{2}H$-CH_4 for South Asia crop residue burning. **(C)** Coupled variation in $\delta^{13}C$ and $\delta^{2}H$. **(D)** Global
588 $\delta^{13}C$ values of biomass burning methane (C3 vs. C4 biomass, WM=weighted mean of C3 and C4
589 biomass). **(E)** Global $\delta^{2}H$ values of biomass burning methane. Biomass burning in South Asia
590 primarily here refer to agricultural wheat crop residue burning. Global review in Supplementary
591 Data S2.

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Table 1. Isotopic signatures of CH_4 from biomass burning in South Asia and globally, as determined by various analytical and statistical methods.

	Region	Type	$\delta^{13}C$ (‰)	$\delta^{2}H$ (‰)	Data/Ref.
Keeling	South Asia	C3	–31.6±2.7	–186±19	Data S1
Miller-Tans	South Asia	C3	–30.9±2.2	**–201±18**	Data S1
	South Asia	WM of C3/C4*	**–29.5±2.0**		Data S1
Review	Global	C3	–27.8±2.7		Data S2
	Global	C4	–15.7±2.4		Data S2
	Global	C3&C4	–21.7±2.1		Data S2
	Global	WM of C3/C4	**–25.0±2.2**		Data S2
	Global	Mean		**–222±39**	Data S2

*The weighted mean (WM) δ-values for biomass burning methane in South Asia are based on a C3:C4 ratio of 0.9:0.1, derived from an EC isotopic source apportionment study (Dasari et al., 2020). For the global biomass burning methane, the WM is calculated using a C3:C4 ratio of 0.77:0.23, based on the global distribution of C3 and C4 vegetation (Still et al., 2003).

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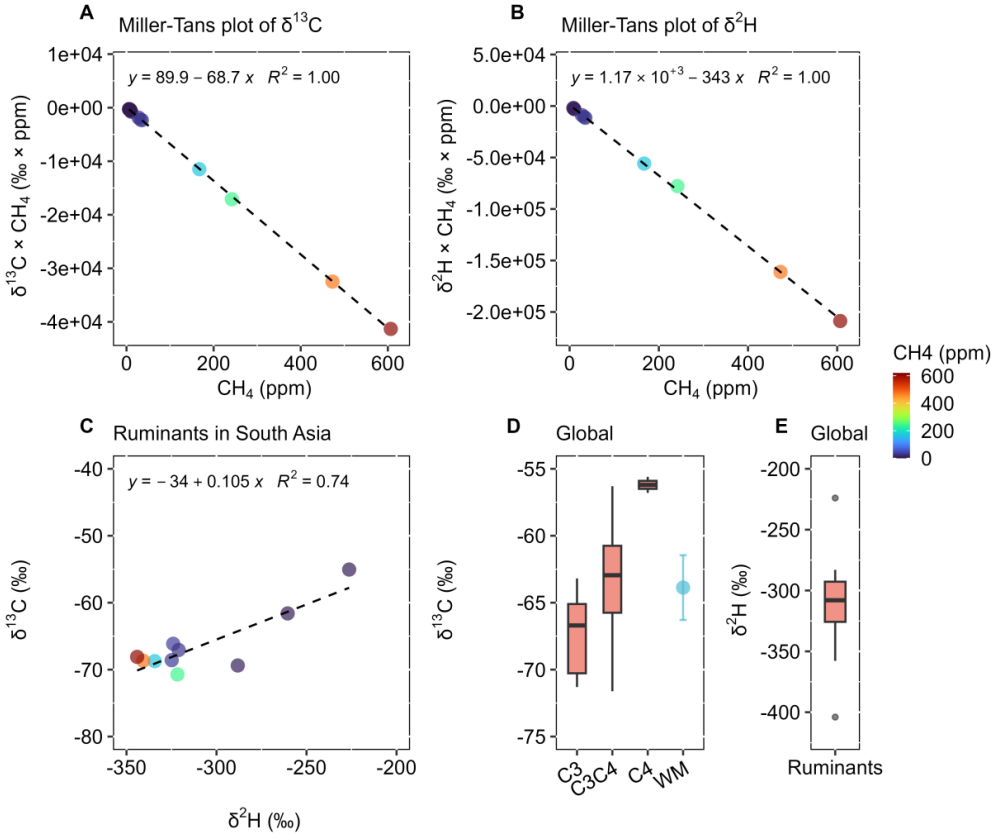

600

601 **Fig. 3. Isotopic characteristics of methane from ruminants in South Asia and globally. (A)**
602 Miller-Tans plot of $\delta^{13}C$-CH_4 for South Asia ruminants. **(B)** Miller-Tans plot of $\delta^{2}H$-CH_4 for South
603 Asia ruminants. **(C)** Coupled variation in $\delta^{13}C$ and $\delta^{2}H$. **(D)** Global $\delta^{13}C$ values of ruminant
604 methane (C3 vs. C4 diets; WM=weighted mean of C3 and C4 diets). **(E)** Global $\delta^{2}H$ values of
605 ruminant methane. Global review in Supplementary Data S2.

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Table 2. Isotopic signatures of CH_4 from ruminants in South Asia and globally, as determined by various analytical and statistical methods.

	Region	Type	$\delta^{13}C$ (‰)	$\delta^{2}H$ (‰)	Data/Ref.
Keeling	South Asia	C3	–71.0±3.8	–342±13	Data S1
Miller-Tans	South Asia	C3	–68.7±0.5	**–343±6**	Data S1
	South Asia	WM of C3/C4*	**–63.3±1.1**		Data S1
Review	Global	C3	–67.0±3.0		Data S2
	Global	C4	–53.2±3.1		Data S2
	Global	C3&C4	–61.3±6.4		Data S2
	Global	WM of C3/C4	**–63.8±2.4**		Data S2
	Global	Mean		**–311±46**	Data S2

*The weighted mean (WM) δ-values for ruminant methane in South Asia are based on a C3:C4 dietary of 0.65:0.35, reflecting the regional distribution of ruminant feed (Chang et al., 2019). For the global ruminant methane, the WM is calculated using a C3:C4 ratio of 0.7:0.3, based on the global mean feed composition (Chang et al., 2019).

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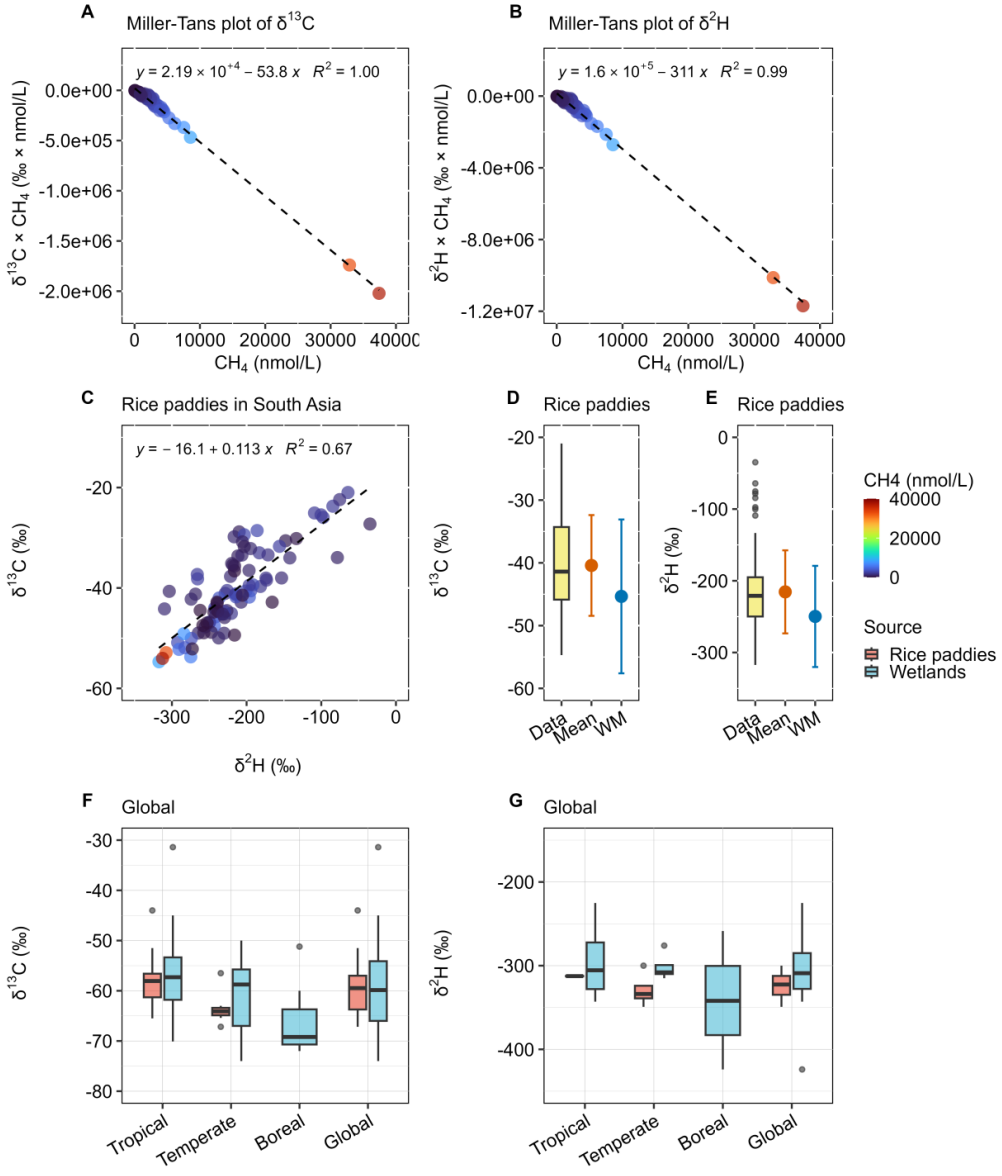

614

615 **Fig. 4. Isotopic characteristics of methane from rice paddies South Asia and globally. (A)**
616 Miller-Tans plot of δ^{13}C-CH_4 for South Asia rice paddies. **(B)** Miller-Tans plot of δ^{2}H-CH_4 for
617 South Asia rice paddies. **(C)** Coupled variation in δ^{13}C and δ^{2}H. **(D)** Quantiles, arithmetic mean,
618 and concentration-weighted mean of δ^{13}C-CH_4 for South Asia rice paddies. **(E)** Quantiles,

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619 arithmetic mean, and concentration-weighted mean of δ^2H-CH_4 for South Asia rice paddies. **(F)**
620 Global δ^{13}C values of methane from rice paddies and for comparison also from wetlands. **(G)**
621 Global δ^2H values of methane from rice paddies and for comparison also from wetlands. Global
622 review in Supplementary Data S2.

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Table 3. Isotopic signatures of CH_4 from rice paddies and wetlands in South Asia and globally, as determined by various analytical and statistical methods.

	Region	Type	$\delta^{13}C$ (‰)	$\delta^{2}H$ (‰)	Data/Ref.
Keeling	South Asia		–39.7±1.7	–212±14	Data S1
Miller-Tans	South Asia		**–53.8±0.8**	**–311±6**	Data S1
Data analysis	South Asia	Median	–41.4	–221	Data S1
Rice paddies	South Asia	Mean	–40.4±8.0	–215±58	Data S1
	South Asia	WM of conc*	–45.3±12.3	–250±71	Data S1
	South Asia	WM of geo conc	–41.7±7.5	–236±45	Data S1
	South Asia	WM of geo MT	–45.5±2.5	–266±17	Data S1
Review	Global	Mean	–59.8±5.3	–324±18	Data S2
Rice paddies	Tropical	Mean	–58.0±5.2	–313±1	Data S2
	Temperate	Mean	–63.5±3.4	–329±21	Data S2
Review	Global	Mean	–60.0±7.6	–309±49	Data S2
Wetlands	Tropical	Mean	–57.1±7.0	–295±52	Data S2
	Temperate	Mean	–60.5±6.9	–302±15	Data S2
	Boreal	Mean	–66.6±5.4	–342±83	Data S2
Review	Global	Mean	–60.0±7.2	–314±42	Data S2
All	Tropical	Mean	–57.3±6.6 (m=47)	–301±41 (m=4)	Data S2
	Temperate	Mean	–61.1±6.4	–314±22	Data S2
	Boreal	Mean	–66.6±5.4	–342±83	Data S2

*"WM of conc" refers to the concentration-weighted mean δ-values of rice paddy methane in South Asia. "WM of geo conc" represents the geographically weighted mean, where each region's contribution is based on its concentration-weighted mean. "WM of geo MT" denotes the geographically weighted mean derived from Miller–Tans method results for each region.

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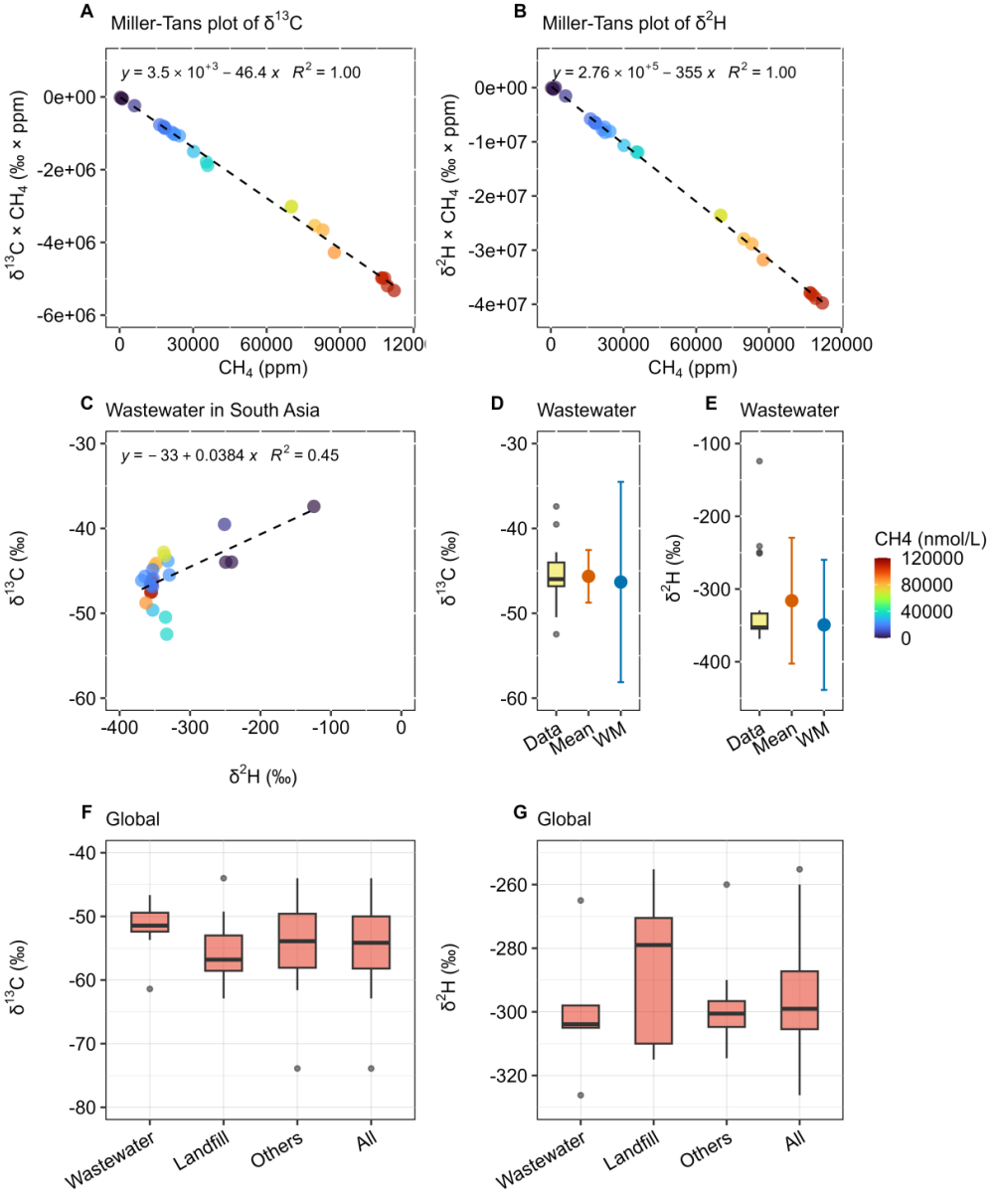

631

632 **Fig. 5. Isotopic characteristics of methane from South Asian wastewater and global waste**
633 **sources. (A)** Miller-Tans plot of $\delta^{13}C$-CH_4 for South Asia wastewater. **(B)** Miller-Tans plot of
634 $\delta^{2}H$-CH_4 for South Asia wastewater. **(C)** Coupled variation in $\delta^{13}C$ and $\delta^{2}H$. **(D)** Quantiles,

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635 arithmetic mean, and concentration-weighted mean of $\delta^{13}C\text{-}CH_4$ for South Asia wastewater. **(E)**
636 Quantiles, arithmetic mean, and concentration-weighted mean of $\delta^{2}H\text{-}CH_4$ for South Asia
637 wastewater. **(F)** Global $\delta^{13}C$ values of methane from waste sources. **(G)** Global $\delta^{2}H$ values of
638 methane from waste sources. Global review in Supplementary Data S2.

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Table 4. Isotopic signatures of CH_4 from waste sources in South Asia and globally, as determined by various analytical and statistical methods.

	Region	Type	$\delta^{13}C$ (‰)	$\delta^{2}H$ (‰)	Data/Ref.
Keeling	South Asia	C3	–46.3±1.1	–338±29	Data S1
Miller-Tans	South Asia	C3	–46.4±1.2	–355±5	Data S1
Data analysis	South Asia	Median	–46.0	–353	Data S1
Wastewater	South Asia	Mean	–45.6±3.1	–316±87	Data S1
	South Asia	WM of conc*	–46.3±11.8	–349±89	Data S1
	South Asia	WM of pop MT	**–45.0±2.4**	**–350±10**	Data S1
Review	Global	Mean	–54.0±5.4	–295±18	Data S2
	Wastewater	Mean	–51.5±3.8	–300±22	Data S2
	Landfills	Mean	–55.7±4.3	–286±22	Data S2
	Others	Mean	–53.7±6.3	–299±13	Data S2

*"WM of conc" refers to the concentration-weighted mean δ-values of wastewater methane in South Asia. "WM of pop MT" denotes the population-weighted mean (weighted with the population of each province), calculated from the Miller–Tans method results for each region.

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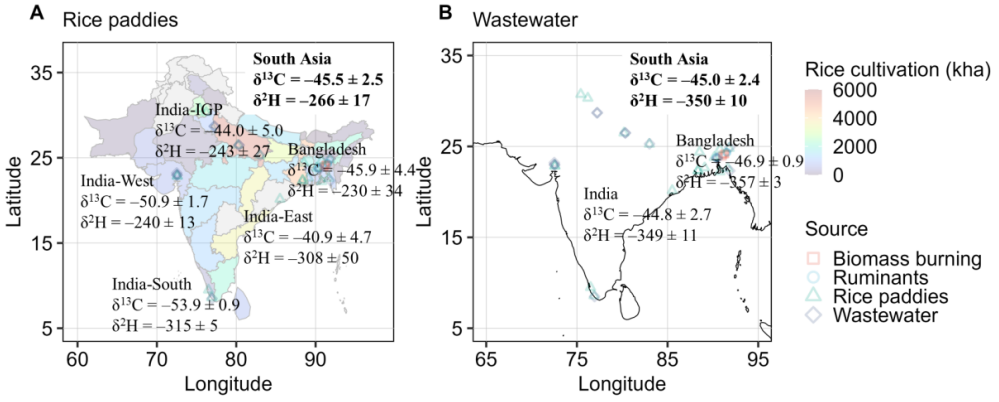

646

647 **Fig. 6. Geographical distribution of methane isotopic signatures from two microbial sources**
648 **in South Asia. (A)** Rice paddies. **(B)** Wastewater. Rice cultivation data is derived from MODIS
649 multitemporal data (Gumma, 2011). The isotopic signatures for rice paddies represent cultivation-
650 weighted means, while those for wastewater are population-weighted means.

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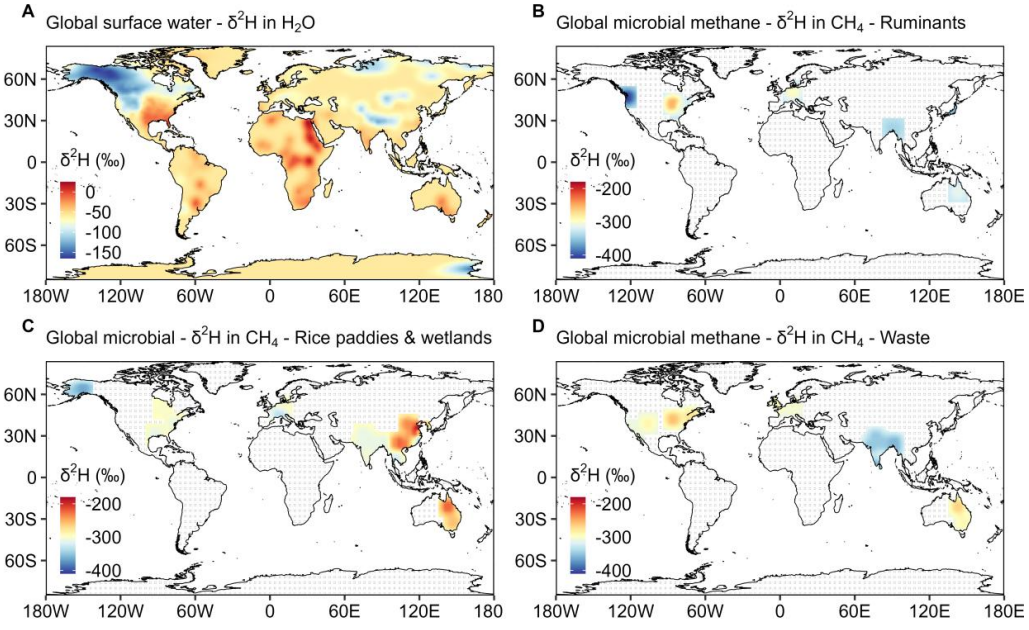

652

653 **Fig. 7. Global distribution of δ²H in surface water and in microbial methane. (A)** δ^2H
654 distribution in surface water systems, based on isotopic data from the literature (Nan et al., 2019;
655 Global Network of Isotopes in Precipitation (GNIP); Halder et al., 2015). **(B)** δ^2H distribution in
656 microbial methane from ruminants. **(C)** δ^2H distribution in microbial methane from rice paddies
657 and wetlands. **(D)** δ^2H distribution in microbial methane from waste. The isotopic and geographic
658 data of microbial methane are compiled from this study (South Asia) and the literature - global
659 (Sherwood et al., 2017) and European (Menoud et al., 2022). Grid cells without any observation
660 are marked with diagonal lines to indicate interpolation-only areas.

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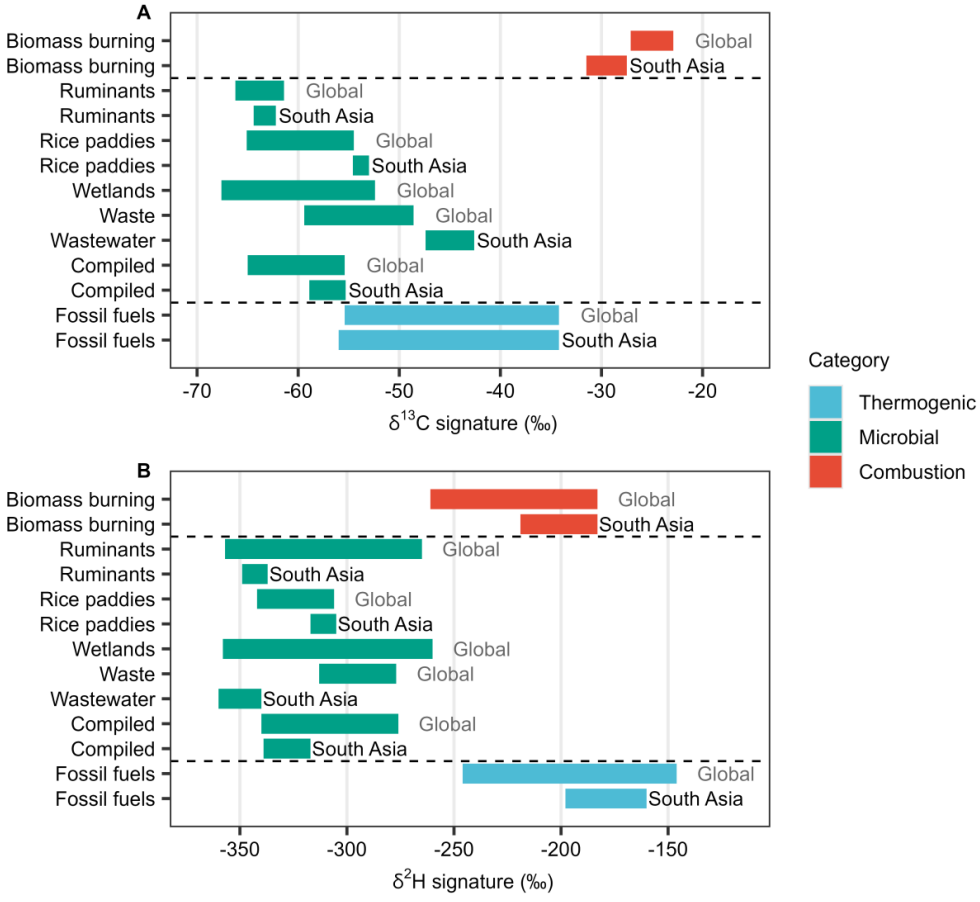

662

663 **Fig. 8. Isotopic signatures of major methane sources in South Asia and globally. (A)** $\delta^{13}C$

664 signatures. **(B)** $\delta^{2}H$ signatures.

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Table. 5. Isotopic signatures of major methane sources in South Asia and globally. n represents to the number of samples analyzed in this study, while **m** indicates the number of literature sources summarized, where isotopic data from a specific region in a single study are compiled as a single entry. **x** refers to the number of isotopic data from the literature. Raw data, literature review and corresponding references are provided in Supplementary Data S1–S2.

Category	Source	Region	$\delta^{13}C$ (‰)	$\delta^{2}H$ (‰)
Combustion	Biomass burning	South Asia	–30.9±2.2 (n=17; 100% C3) –29.5±2.0 (90% C3)	–201±18 (n=15)
	Biomass burning	Global	–25.0±2.2 (m=19; 77% C3)	–222±39 (m=6)
Microbial	Ruminants	South Asia	–68.7±0.5 (n=37; 100% C3) –63.3±1.1 (65% C3)	–343±6 (n=11)
	Ruminants	Global	–63.8±2.4 (m=36; 70% C3)	–311±46 (m=11)
	Rice paddies	South Asia	–53.8±0.8 (n=90)	–311±6 (n=90)
	Rice paddies	Global	–59.8±5.3 (m=20)	–324±18 (m=6)
	Wetlands	Tropical	–57.3±6.6 (m=47)	–301±41 (m=4)
	Wetlands	Global	–60.0±7.6 (m=94)	–309±49 (m=12)
	Wastewater	South Asia	–45.0±2.4 (n=27)	–350±10 (n=27)
	Waste	Global	–54.0±5.4 (m=69)	–295±18 (m=29)
	Compiled	South Asia	–57.1±1.8	–328±11
	Compiled	Global	–60.2±4.8	–308±32
Thermogenic (mainly)	Fossil fuels	South Asia	–45.1±10.9 (x=83)	–179±19 (x=28)
	Fossil fuels	Global	–44.8±10.6 (x=8128)	–196±50 (x=2878)

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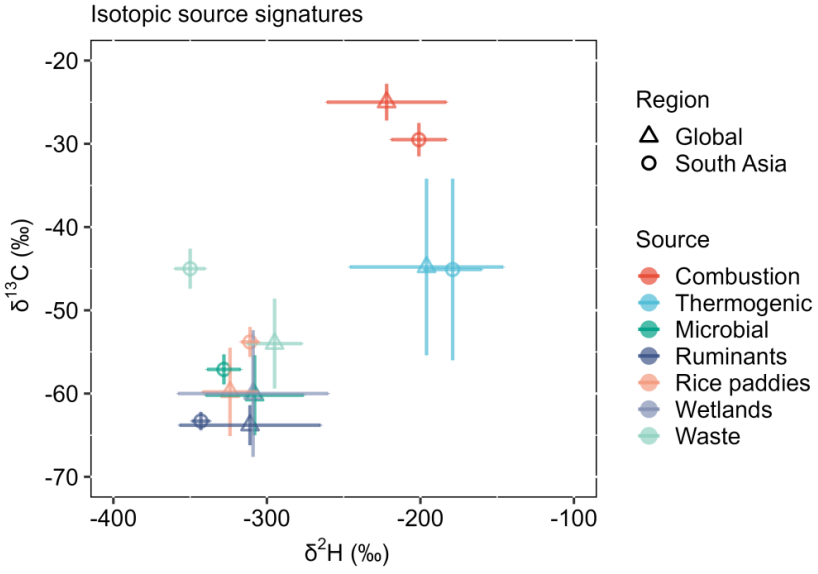

673

674 **Fig. 9. Comparison of South Asian and global isotopic signatures of methane sources.** δ^2H
675 versus $\delta^{13}C$. Microbial source refers to the compiled values of ruminants, rice paddies, wetlands,
676 waste, etc.

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Data availability

The dataset will be hosted and maintained by a database management at the Bolin Centre for Climate Research at Stockholm University. The dataset is accessible at the Bolin Centre Database (https://bolin.su.se/data/draft?id=14278&token=f0770bfd-210f-4a51-a9bd-a355467058a4).

Supporting Information

Supporting Information for this study is available.

Author Contribution

ÖG acquired funding and conceived the study. ÖG, HH, A. Salam, K. Budhavant, MRM, KSJ, MAH, A. Singh, AP, NR, CM, KR, and GKS. designed and conducted the field campaigns. PY, K. Belec, JB, and HH performed the isotope analyses. PY, K. Belec, HH, and ÖG conducted the data analysis and interpretation. PY, HH, and ÖG prepared the manuscript with contributions from all co-authors. All authors reviewed and edited the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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