Response to Reviewer 1

"A global dataset of $\delta^{13}C$-CH_4 source signatures and associated uncertainties (1998–2022), with a sensitivity analysis to support isotopic inversions"

By Tapin et al. (essd-2025-668)

Submitted to Earth System Science Data (ESSD)

Legend:

Reviewer comments are reproduced in dark blue.

Author responses are in black.

Manuscript changes are shown in purple italics, with line numbers referring to the revised manuscript.

General response

We sincerely thank Reviewer 1 for the careful and constructive evaluation of our manuscript, and for the strong support expressed for publication. We are grateful for the detailed list of references provided, several of which we had not yet incorporated, and for the helpful suggestions concerning latitudinal variability, the freshwater and geological sources, the chlorine and stratospheric sinks, and tropical measurement programs. We have addressed all comments individually below. The reviewer's suggestions have led to substantive improvements, in particular to the discussion of latitudinal C_3/C_4 gradients, the explicit acknowledgement of large uncertainties in freshwater and geological emission estimates, the expanded treatment of the chlorine and stratospheric sinks, and the inclusion of recent tropical measurement campaigns in our reference list. All line numbers in our responses refer to the revised manuscript.

Response to general comments

G1. Lack of tropical data and latitudinal variation (C_3/C_4 plants in agriculture, wetlands, and biomass burning)

Reviewer comment: "There are many factors driving strong latitudinal gradients. In particular, C_4 plants, which pre-concentrate and thus accept more 13-C in photosynthesis, are dominant in the tropical grasslands and wetlands (e.g. papyrus), while C_3 rules in the boreal realm. Also in the sinks, in the Brewer-Dobson circulation the polar vortex delivers low mixing ratio, highly 13-C rich methane, while in the tropical troposphere the upward loss to stratosphere is of high mixing ratio 12-C rich methane."

Response: We fully agree that latitudinal gradients arising from the C_3/C_4 plant distribution and from stratospheric transport processes deserve more explicit discussion. We have substantially expanded our treatment of these issues in three places in the revised manuscript.

First, in Section 2.3 (uncertainty assessment) we now explicitly state that propagated uncertainties carry a latitudinal dimension driven by the C_3/C_4 distribution and by stratospheric transport. Second, in Section 4.1.1 (spatial patterns) we expanded the discussion of all three affected sectors (AGW, BB, WET) to highlight the role of C_3 versus C_4 vegetation, citing the additional references suggested by the reviewer. Third, in Section 4.1.3 we added a dedicated paragraph on the latitudinal structure of propagated uncertainties.

Manuscript change (Section 4.1.1, AGW bullet, L 416–419): *"Livestock signatures show notable regional variability: tropical regions tend to be more enriched (heavier than −60‰) due to C_4-dominated forage, while temperate extensive grazing systems on C_3 grasslands yield more depleted values. However, intensive dairy systems in the northern hemisphere, fed largely on C_4 maize silage, can also produce relatively enriched signatures (Chang et al., 2019)."*

Manuscript change (Section 4.1.1, BB bullet, L 420–424): *"Isotopic gradients are primarily latitudinal, driven by the distribution of C_3 and C_4 plants. Tropical and subtropical regions dominated by C_4 vegetation exhibit more enriched values, while boreal regions show more depleted signatures (Still et al., 2003; Randerson et al., 2012; Lan et al., 2021a). Within the tropics, grass fires burning C_4 vegetation tend to produce more enriched $\delta^{13}C$-CH_4 than bush and tree fires, which involve predominantly C_3 biomass (Barker et al., 2020; France et al., 2022)."*

Manuscript change (Section 4.1.1, WET bullet, L 425–427): *"Generally depleted values (−50 to −70‰) are observed, with more depleted signatures in high-latitude and boreal wetlands, and relatively enriched ones in tropical wetlands, where C_4 aquatic vegetation such as papyrus contribute to isotopically heavier signatures (Ganesan et al., 2018; Oh et al., 2022; Nisbet et al., 2021; France et al., 2022)."*

Manuscript change (Section 4.1.3, latitudinal structure paragraph, L 540–545): *"Beyond sectoral aggregation, the propagated uncertainties also exhibit a latitudinal structure. The distribution of C_3 and C_4 plants drives systematic gradients in isotopic signatures across the WET, BB, and AGW sectors: tropical regions, dominated by C_4 vegetation, tend to produce more enriched $\delta^{13}C$–CH_4 signatures, while boreal and temperate regions, where C_3 plants prevail, yield more depleted values (Still et al., 2003; Chang et al., 2019; France et al., 2022; Nisbet et al., 2021)."*

Regarding the lack of tropical isotopic measurements, we now emphasise this issue more strongly in Sections 4.1.3 and 4.1.4 and refer explicitly to the recent MOYA and ZWAMPS campaigns (France et al., 2022; Nisbet et al., 2021; Shaw et al., 2022; Barker et al., 2020). The tropical wetland source signatures of -59.0 ± 1.3‰ (Bolivian Amazonia, France et al., 2022) and -59.3 ± 2.0‰ (Zambia/Bolivia composite, Nisbet et al., 2021) are now discussed in Section 4.1.4, where we note their consistency with the tropical wetland values used in our maps and acknowledge that these recent airborne measurements have not yet been assimilated into process-based wetland isotope models.

G2. Weighting given to freshwater sources

Reviewer comment: "Freshwater systems. I'm very sceptical here, but I realise this is a long-running puzzle and in this paper I think the best approach is simply to say briefly that fluxes are very uncertain. The re-scaling by one-third maybe should get a further note here, in addition to the reference to Saunois et al 2025. Note for example the Shaw et al. paper, which is one of the very few to study the Congo freshwater and wetlands."

Response: We agree with the reviewer's scepticism and adopt the suggested approach. The aim of our work is not to defend a particular freshwater flux estimate, but rather to be consistent with the Global Methane Budget framework while clearly flagging that this estimate is highly uncertain. We have expanded the description of freshwater fluxes in Section 3.1.2 to explicitly state the large uncertainties surrounding these estimates and the limitations of the one-third rescaling, and we now cite Shaw et al. (2022), which we had inadvertently overlooked.

Manuscript change (Section 3.1.2, L 277–282): *"Freshwater systems: Emissions from lakes and reservoirs are based on the CH_4 flux maps by Stavert et al. (2022). Initial global emissions are estimated at 95 Tg yr^{-1}, reduced to 73 Tg yr^{-1} after ice-cover corrections. Following Martinez et al. (2024), emissions are further rescaled by a factor of one-third, resulting in a global total of 53 Tg yr^{-1} (Saunois et al., 2025). We acknowledge that freshwater flux estimates remain subject to large uncertainties, arising from poorly mapped inland water extent, complex emission pathways, and spatial variability in methanotrophy (e.g., Van Bergen et al., 2019; Lauerwald et al., 2023; Shaw et al., 2022). These uncertainties are further discussed in Sect. 4.2.2."*

We therefore retain the GMB freshwater value as a working assumption (rather than an established constraint) and emphasise in Section 4.3.2 that omitting freshwaters entirely would bias regional attribution, while including them as a separately optimised category remains premature given the available observational constraints.

G3. Weighting given to geological sources

Reviewer comment: "Petrenko et al have presented convincing evidence that the Etiope et al estimate of geological emissions is much too high, and I suspect even the downsized 21.1 Tg guess in Table 1 is still high. (...) I strongly suspect this 23 Tg/yr figure is too high."

Response: We agree with the reviewer that the geological emission estimate of ~21–23 Tg yr^{-1} is contested by the ice-core constraints from Petrenko et al. (2017) and other related work. In our framework, we adopted the geological flux from the Global Methane Budget (Martinez et al., 2024) for consistency across our reference simulations, and we did not perform a sensitivity test on this value. To address the reviewer's concern, we have added an explicit caveat in Section 3.1.2 stating that the current value follows the GMB framework but that ice-core-based constraints suggest substantially lower pre-industrial geological emissions. We note that since this value is held constant in our reference simulations and contributes only a small fraction of the FFG sector flux, the impact on the modeled $\delta^{13}C\text{-}CH_4$ signal is limited; however, the reviewer's point underscores a structural uncertainty that should be addressed in future inversion studies.

Manuscript change (Section 3.1.2, L 285–290): *"Geological methane: Geological emissions use the gridded climatology from Etiope et al. (2019), rescaled to a total global flux of 21 Tg CH_4 yr^{-1} by Martinez et al. (2024). Offshore geological emissions, including marine seepage, are excluded to avoid double counting. We note that geological emission estimates remain uncertain; ice core-based constraints suggest that pre-industrial geological emissions may have been substantially lower (Petrenko et al., 2017), though the current value follows the Global Methane Budget framework (Martinez et al., 2024) for consistency with the broader modeling setup."*

G4. The chlorine sink

Reviewer comment: "The chlorine sink is small for methane in Tg but disproportionately large in its leverage on $\delta^{13}C$… this is an important factor as the sink is still poorly quantified. It probably needs a somewhat longer discussion. Maybe also refer to Allen et al. 2007."

Response: We agree that the Cl sink, while small in mass terms, has disproportionate isotopic leverage and merits a more substantial discussion. We have added a dedicated paragraph in Section 4.2.3 covering: (i) the magnitude of the tropospheric Cl sink based on the most recent estimates (~1–3% of total CH_4 oxidation; Hossaini et al., 2016; Sherwen et al., 2016; Gromov et al., 2018; Wang et al., 2021; Saunois et al., 2025), with explicit reference to Allan et al. (2007) for context on earlier estimates; (ii) the strong KIE of the Cl reaction (≈1.066), more than an order of magnitude larger than that of OH; and (iii) a quantitative summary of the Thanwerdas et al. (2022b) sensitivity analysis, which used the same CIF–LMDz–SACS framework as ours and reported a near-linear sensitivity of +11.7 Tg CH_4 yr^{-1} and −1.0‰ in the globally averaged source signature per 1000 molec. cm^{-3} increase in mean tropospheric Cl, with stratospheric Cl alone contributing a ~0.30‰ surface enrichment via stratosphere–troposphere exchange.

We explain that we did not repeat a Cl sensitivity experiment because it would essentially duplicate Thanwerdas et al. (2022b) within the same model framework, but we acknowledge that uncertainties in Cl concentrations remain a leading factor limiting isotope-based source partitioning, in agreement with Basu et al. (2022) and Röckmann et al. (2024b). We also added a note in Section 4.4 on emerging evidence for previously unaccounted-for Cl sources, notably photocatalytic release from mineral dust–sea spray aerosol (van Herpen et al., 2023; Röckmann et al., 2024a), as a relevant target for future inversion setups.

Manuscript change (Section 4.2.3, chemistry paragraph, L 858–873): *"The Cl sink accounts for a small fraction of total CH4 oxidation. Recent estimates converge on a tropospheric contribution of ~1–3% of the total chemical sink (Hossaini et al., 2016; Sherwen et al., 2016; Gromov et al., 2018; Wang et al., 2021), with the latest Global Methane Budget reporting a climatological tropospheric Cl sink of 6 [1–13] Tg CH4 yr−1 (Saunois et al.,2025), substantially smaller and better constrained than earlier estimates (Allan et al., 2007). Despite this small magnitude, the Cl reaction carries an exceptionally large kinetic isotope effect (KIE ≈ 1.066 at 298 K; Saueressig et al., 1995), more than and order of magnitude larger than that of OH, so even modest uncertainties in Cl concentrations translate into substantial shifts in modeled δ13C-CH4 (see Table 2). Basu et al. (2022) further identified the combined uncertainty in fractionation (OH-KIE and Cl contribution) as the single most*

important factor limiting isotope-based source partitioning at the global scale (Röckmann et al., 2024b). Thanwerdas et al. (2022b) quantified this influence within the same CIF-LMDz-SACS framework used here, and reported a near-linear sensitivity of +11.7 Tg CH4 yr−1 and −1.0 ‰ in the globally averaged source signature per 1000 molec. cm−3 increase in mean tropospheric Cl, with stratospheric Cl alone contributing a ~ 0.30 ‰ surface enrichment via stratosphere–troposphere exchange and modifying the δ13C-CH4 seasonal cycle amplitude by up to 10–20% depending on latitude. Because our configuration adopts the Cl field from Wang et al. (2021), consistent with the most recent tropospheric chlorine chemistry, the Cl-related uncertainty in our simulations is bounded by the ranges quantified in Thanwerdas et al. (2022b), which are of the same order of magnitude as the OH-KIE sensitivity reported in Table 6. A dedicated Cl sensitivity experiment was therefore not repeated here to avoid duplicating a recent and comprehensive analysis with the same model."

G5. The stratospheric sink

Reviewer comment: "I am concerned by two problems – 1) the Cl sink with its large isotopic leverage and 2) the stratospheric sink. Both of these seem poorly understood and with high uncertainty. (...) Table 6 – Chemistry – mention the stratospheric sink?"

Response: We agree that the stratospheric sink merits more explicit discussion. In our model, methane oxidation in the stratosphere is represented by reactions with OH, $O(^1D)$ and Cl, with 3-D oxidant fields prescribed (Section 3.1.4, Table 2). The stratospheric component of the Cl sink is therefore implicitly included in the simulations. To make this clearer, we have added a footnote to Table 6 stating that the Cl sink is not perturbed independently in the sensitivity ensemble, with reference to the Thanwerdas et al. (2022b) results that quantify both tropospheric and stratospheric Cl contributions in the same framework. We have also expanded the discussion in Section 4.2.3 (see also G4 above) to explicitly address the stratospheric Cl contribution and its impact on the surface $\delta^{13}C–CH_4$ signal via stratosphere–troposphere exchange.

Manuscript change (Section 4.4, transport paragraph, L 1029–1038): *"A second priority concerns atmospheric transport and chemistry, which were not perturbed in our sensitivity ensemble. All simulations were performed with LMDz at a single resolution (Sect. 3.1.1), and the TransCom-CH_4 intercomparison (Patra et al., 2011) showed that modeled CH_4 budgets are sensitive to troposphere–stratosphere exchange rates and to vertical grid structure, with CH_4 lifetimes spanning 9.50–10.27 yr across 12 CTMs using identical OH fields. For $\delta^{13}C$-CH_4, vertical transport additionally controls the rate at which ^{13}C-enriched stratospheric air re-enters the troposphere via the Brewer–Dobson circulation, as well as the vertical distribution of the Cl sink and its strong fractionation (Butchart, 2014; Thanwerdas et al., 2022b)."*

Response to specific comments

L23 – Updated atmospheric CH_4 mole fraction

Reviewer comment: "1930 in 2024 – maybe cite NOAA (Lan et al. 2026) for end 2025."

Response: We have updated the value to reflect the most recent NOAA observations and now cite Lan et al. (2026).

Manuscript change - L21: *"…to reach 1946 ppb in November 2025 (Forster et al., 2023b; Lan et al., 2026)."*

L25 – Atmospheric lifetime

Reviewer comment: "Maybe distinguish perturbation life (12 yr, as cited in IPCC) from burden/flux life of 9 yr."

Response: Thank you for this useful clarification. We have rephrased the sentence to explicitly distinguish the two timescales.

Manuscript change - L24-25: *"Its relatively short atmospheric lifetime (about 9 years for the burden; Prather et al., 2012, and approximately 12 years for the perturbation lifetime, Forster et al. 2023b)…"*

L33 – Long reference list

Reviewer comment: "Maybe add Ciais et al. 2026, Fujita et al. 2025, Riddell-Young et al. 2025, and switch Nisbet 2019 to 2023."

Response: We have added Ciais et al. (2026), Fujita et al. (2025b), and Riddell-Young et al. (2025) to the reference list at this location, and we have updated the citation of Nisbet et al. to the 2023 version. The full reference list at L33 in the revised manuscript now includes the original references plus these additions.

L36 – Biomass burning

Reviewer comment: "Add biomass burning (both natural and anthropogenic)."

Response: We have updated the sentence to explicitly mention biomass and biofuel burning as both natural and anthropogenic sources.

Manuscript change - L37-39: *"Methane emissions have both natural (around 200 Tg CH_4 yr^{-1}, e.g., wetlands, freshwaters, geological sources, natural wildfires) and anthropogenic origins (around 320 Tg CH_4 yr^{-1}, e.g., agriculture, fossil fuel, waste, and anthropogenic biomass and biofuel burning)."*

L64–66 / L271 – Geological emissions

Reviewer comment: "See also Line 270, and Table 1 FFG segment… Petrenko et al have presented convincing evidence that the Etiope et al estimate of geological emissions is much too high, and I suspect even the downsized 21.1 Tg guess in Table 1 is still high."

Response: Addressed in the response to general comment G3 above. We have added a caveat in Section 3.1.2 referring to Petrenko et al. (2017) and acknowledging that the GMB-aligned value may overestimate geological emissions.

L82 – Reference to Fujita et al. 2025

Reviewer comment: "Also see Fujita et al 2025."

Response: We have added a citation to Fujita et al. (2025b) in the discussion of multi-isotopic constraints on fossil methane emissions l33.

L227 – Sub-sector variability and latitudinal uncertainty

Reviewer comment: "Sub sector uncertainty and aggregation. This is interesting, but perhaps there should also be a brief discussion of latitudinal uncertainty… there are many factors driving strong latitudinal gradients."

Response: We have addressed this point in detail (see G1). In addition to the spatial-pattern paragraphs in Section 4.1.1, we added a dedicated paragraph in Section 4.1.3 on the latitudinal structure of propagated uncertainties, and a closing sentence in Section 2.3 that anticipates this discussion.

Manuscript change - L236-239: *"In summary, this framework provides consistent tools to estimate sectoral $\delta^{13}C\text{-}CH_4$ uncertainties, combining sub-sector variability and aggregation effects. Beyond sectoral aggregation, isotopic uncertainties also carry a latitudinal dimension, driven notably by the distribution of C_3 and C_4 plants across source sectors and by stratospheric transport processes; these spatial gradients are discussed further in Sect. 4.1.1."*

L265 – Freshwater systems

Reviewer comment: See general comment G2.

Response: Addressed in the response to general comment G2 above. We have expanded the discussion of freshwater uncertainties in Section 3.1.2 and added a reference to Shaw et al. (2022).

L285 – Hydroxyl

Reviewer comment: "See Morgenstern et al. 2025, and Ciais et al. 2026."

Response: We have added these references in Section 4.2.3, where we discuss the temporal evolution of tropospheric oxidative capacity and its implications for recent CH_4 trends.

Manuscript change - L825-829: *"Recent studies further suggest that tropospheric oxidative capacity is itself evolving over time: Morgenstern et al. (2025) infer an increasing global OH abundance from radiocarbon monoxide (^{14}CO) observations, with implications for the interpretation of recent CH_4 trends (Ciais et al., 2026; Nisbet and Manning, 2026)."*

L292 / Table 2 – Chlorine sink

Reviewer comment: See general comment G4.

Response: Addressed in the response to general comment G4 above. We have expanded the Cl-sink discussion in Section 4.2.3 and added Allan et al. (2007) to the reference list.

L394–397 / Fig. 1 – Latitudinal gradient of C_3 and C_4 plants

Reviewer comment: "Ganesan et al. is mentioned elsewhere (L482) in the paper but could be cited here too. Also see France et al. 2022, and Nisbet et al (MOYA) 2022. The C_3/C_4 gradient affects: 1) wetland emissions – equatorial wetlands are papyrus rich, while outer tropics have more reeds (C_3/C_4) and boreal wetlands have almost no C_4 plants. 2) agricultural emissions – ruminant grazers eat grasses in the tropics that are mostly C_4, but in the drier outer tropics cattle and antelope are often browsers, on C_3 trees. 3) biomass burns – tropical grass fires are C_4 rich but tropical bushes and tree fires are C_3, so methane given off by incomplete combustion is somewhat less heavy. Boreal forest fires are C_3. See Barker et al. 2020."

Response: We thank the reviewer for this very detailed comment, which has helped substantially improve the spatial pattern discussion. We have integrated all three points (wetland, agricultural, biomass burning) into the bullet list of Section 4.1.1, with the appropriate citations. The corresponding manuscript changes are shown in our response to general comment G1 (AGW, BB and WET bullets).

L436 – AGW signature regional variability

Reviewer comment: "AGW signature is regionally very variable. In the tropics, heavier than −60‰. In the populated northern hemisphere, breath of grassland animals (mostly beef) is probably lighter than −60‰ but intensive livestock (especially dairy) are fed C_4 maize and their breath can be quite heavy."

Response: We added a sentence to the AGW bullet in Section 4.1.1 reflecting this variability (see manuscript change in our response to G1, AGW bullet, L 416–419).

L482 / L501 – Lack of tropical measurements

Reviewer comment: "Emphasise the lack of tropical measurement?"

Response: We have strengthened this point in two places. In Section 4.1.3 (propagated uncertainty paragraph for WET), we now explicitly state that "Tropical wetlands in particular remain under-sampled, despite representing a dominant fraction of global wetland CH_4 emissions (France et al., 2021, 2022)", and similarly for BB ("tropical fire-dominated regions being particularly under-represented in isotopic measurement databases", citing Nisbet et al., 2021). In Section 4.1.4, we list "Uneven geographic coverage" as the first systematic bias inherited from observational databases, and explicitly note that recent tropical measurement programs (France et al., 2022; Nisbet et al., 2021; Shaw et al., 2022) have not yet been integrated into the maps.

L523 – Aggregation uncertainty for WET

Reviewer comment: "No aggregation uncertainty stated, but actually the aggregation involves aggregating different components of low latitude and high latitude results."

Response: We agree that this point deserves clarification. Although our aggregation uncertainty σ_agg, defined as the variability across emission inventories used for flux-weighting, is null for WET because we use a single wetland flux dataset, the latitudinal contrast between tropical and boreal wetlands generates substantial isotopic variability that is captured by the propagated uncertainty σ_prop. We have added an explicit comment in Section 4.1.3 to clarify this.

Manuscript change - L570-577: *"On the contrary, for WET, no aggregation uncertainty is reported because this sector relies on a single wetland flux dataset for weighting, precluding a cross-inventory sensitivity assessment. We note, however, that the latitudinal contrast between tropical (more enriched, ~−50‰) and boreal (more depleted, ~−70‰) wetlands contributes substantially to the propagated uncertainty σ_prop already reported for this sector (0.4–8.2‰)."*

Table 5 – Tropical wetland measurements

Reviewer comment: "Maybe mention the France et al (2022) and Nisbet et al. (2022) tropical wetland results?? These were from both S. America and Africa…"

Response: We have added a sentence to Section 4.1.4 explicitly reporting the recent tropical airborne measurement values and comparing them with the values used in our maps.

Manuscript change - L632-637: *"Recent tropical airborne measurement programs report wetland source signatures of −59.0 ± 1.3‰ in Bolivian Amazonia (France et al., 2022) and −59.3 ± 2.0‰ for a Zambia–Bolivia composite (Nisbet et al., 2021), consistent with the tropical wetland values used in our maps from Oh et al. (2022). The slightly more enriched values produced by our maps over African papyrus-dominated wetlands likely reflect that these recent airborne measurements have not yet been assimilated into process-based wetland isotope models, contributing to the systematic biases on tropical wetlands discussed below."*

L560 – China and India coal emissions

Reviewer comment: "China is dominant – but India is also a big coal emitter."

Response: We have updated this sentence to explicitly mention India alongside China as a major coal-emitting region with relatively enriched signatures.

Manuscript change: - L608 *"Our estimate is derived using flux-weighted averaging that emphasizes high-emitting coal regions, such as China and India, where emissions tend to be less depleted than the global average…"*

Table 6 – Stratospheric sink

Reviewer comment: "Chemistry – mention the stratospheric sink?"

Response: We have added a footnote to Table 6 explicitly noting that the Cl sink (which contributes both tropospheric and stratospheric components) is not perturbed independently in our sensitivity ensemble, with reference to the comprehensive analysis by Thanwerdas et al. (2022b) using the same model framework.

Manuscript change: *Footnote added to Table 6: "The Cl sink is not perturbed independently in this sensitivity ensemble. Its impact within the same CIF–LMDz–SACS framework has been*

comprehensively quantified by Thanwerdas et al. (2022b), who report that stratospheric Cl alone contributes a ~0.30‰ surface enrichment in $\delta^{13}C–CH_4$ via stratosphere–troposphere exchange, and modifies the seasonal cycle amplitude by 10–20% depending on latitude (see Sect. 4.2.3).”

L633 – Ethiopian cattle diet

Reviewer comment: “See Brychkova et al – Ethiopian cattle eat C_3 above about 2500m but around 2000m many forage grasses are C_4.”

Response: We thank the reviewer for this useful reference. We have incorporated Brychkova et al. (2022) into Section 4.2.1, in the discussion of regional AGW variability in Ethiopia, and again in Section 4.3.1 in the Indo-Gangetic Plain case study where we use Ethiopia as a finer-scale illustration.

Manuscript change - L718-719: *“For instance, in Ethiopia, the balance between C_3 and C_4 forage grasses varies with altitude, leading to distinct isotopic signatures within the same country (Brychkova et al., 2022).”*

Manuscript change - L949-951: *“The Ethiopian highlands provide a finer-scale illustration of this need: the C3/C4 forage balance varies with altitude, producing sub-national contrasts in livestock signatures (Brychkova et al., 2022) that current global inventories cannot resolve. ”*

L671 – Landfills

Reviewer comment: “Maybe Nisbet et al 2020.”

Response: We have added Nisbet et al. (2020) to the reference list in the discussion of waste management practices in Section 4.2.2 (L776).

L687 – Caspian Sea classification

Reviewer comment: “This isn’t clear – is the Caspian here treated as a Freshwater source? I think it’s about 12‰ salinity… Also it has methane/mud volcanoes and there are huge petrochemical methane sources nearby – gasfields and oilfields.”

Response: The reviewer raises a fair point. The Caspian Sea is indeed a brackish endorheic lake, and our classification of its emissions as freshwater is a default consequence of using the Stavert et al. (2022) lake-and-reservoir dataset, which does not separate brackish endorheic systems. We have clarified this explicitly in both Section 4.2.2 and Section 4.3.1, acknowledging the classification ambiguity, the brackish nature of the lake, and the proximity of major oil-and-gas infrastructure (Turkmenistan, Azerbaijan), which complicates regional source attribution.

Manuscript change - L794-796: *“The Caspian Sea is a large endorheic saline lake whose emissions are treated as freshwater in our framework given the absence of a dedicated dataset; its brackish nature and proximity to major oil and gas infrastructure introduce additional uncertainty in this region.” (Section 4.2.2). And in Section 4.3.1 - L960-963: “The Caspian Sea is a large endorheic brackish lake, treated here as a freshwater source given the absence of a dedicated dataset for endorheic systems; this classification ambiguity, combined with proximity to major oil and gas infrastructure in Turkmenistan and Azerbaijan, makes regional source attribution particularly challenging.”*

L703 – Atmospheric chemistry concerns (Cl sink and stratospheric sink)

Reviewer comment: "I am concerned by two problems – 1) the Cl sink with its large isotopic leverage and 2) the stratospheric sink. Both of these seem poorly understood and with high uncertainty. Also note that OH in the tropical troposphere is changing (Morgenstern et al 2025) and likely has significant longitudinal variation around the globe."

Response: Addressed in the responses to general comments G4 and G5 above. We have substantially expanded the chemistry discussion in Section 4.2.3, including a new paragraph on the Cl sink and its KIE leverage with reference to Allan et al. (2007), Wang et al. (2021), Saunois et al. (2025), Thanwerdas et al. (2022b) (L858-L873), and a discussion of the temporal and longitudinal variability of tropospheric OH following Morgenstern et al. (2025), Zhao et al. (2019) and Penn et al. (2025) (L823-829).

Manuscript change - L823-829: *"These regions also display marked longitudinal contrasts in tropospheric OH, driven by zonal asymmetries in convection, lightning NOx, and biomass burning, which are not consistently captured across global chemistry models (Zhao et al., 2019; Morgenstern et al., 2025)."*

L715 – Oxidative capacity

Reviewer comment: "Could mention Ciais et al. 2026 and Nisbet & Manning 2026."

Response: We have added both references to the discussion of OH-related uncertainties in Section 4.2.3.

Manuscript change - L832: *"This confirms that the oxidative sink is a dominant factor controlling methane concentrations and that its uncertainties propagate broadly rather than being confined to specific regions (Zhao et al., 2019; Ciais et al., 2026; Belikov et al., 2026; Nisbet and Manning, 2026; Skeie et al., 2023)."*

L721 – Update older references

Reviewer comment: "References here are a bit elderly. ? Penn et al 2025, Skeie et al 2023?"

Response: We thank the reviewer for suggesting these references. We have added Skeie et al. (2023) alongside the existing citations on OH-driven CH_4 trends, as it directly examines how anthropogenic influence on OH through NO_x and CO has modified the CH_4 growth rate over recent decades (L833). We have also added Penn et al. (2025) in the context of OH uncertainty characterization, as this study explores satellite-based constraints on tropospheric OH concentrations using GOSAT and AIRS observations, representing a promising avenue to reduce the OH-related uncertainties discussed in this section.

Manuscript change - L829: *"Satellite-based observations have also been explored as a means to better characterize tropospheric OH distributions and reduce these uncertainties (Penn et al., 2025)."*

L918 – Targeted campaigns

Reviewer comment: "See MOYA – double volume of Phil Trans Royal Soc. 2021-2022."

Response: We have explicitly cited the MOYA and ZWAMPS campaigns in Section 4.4 (Pathways for improvement) as examples of the type of coordinated airborne and ground-based campaigns needed to fill observational gaps in tropical regions.

Manuscript change - L1042-1044: *"Coordinated airborne and ground-based campaigns, exemplified by MOYA and ZWAMPS in tropical wetlands, rice fields, and biomass burning regions (Nisbet et al., 2021; France et al., 2022), provide a model for the type of effort needed to fill these observational gaps."*

Closing remarks

We thank Reviewer 1 once again for the very thorough and constructive review, and especially for the extensive list of references provided, which has substantially enriched the manuscript. We believe that the revisions, in particular the expanded treatment of latitudinal C_3/C_4 gradients, the explicit acknowledgement of large freshwater and geological flux uncertainties, the strengthened discussion of the chlorine and stratospheric sinks, and the integration of recent tropical measurement programs, address all the concerns raised. We hope the revised manuscript will meet the expectations of the reviewer and the editor.

Summary of new references added

In response to this review, the following references have been added to the manuscript:

Allan, W., et al. (2007). Methane carbon isotope effects caused by atomic chlorine in the marine boundary layer. J. Geophys. Res.: Atmospheres, 112(D4).

Barker, P. A. et al. (2020). Airborne measurements of fire emission factors for African biomass burning sampled during the MOYA campaign. Atmos. Chem. Phys., 20, 15443–15459.

Brychkova, G., et al. (2022). Climate change and land-use change impacts on future availability of forage grass species for Ethiopian dairy systems. Sci. Rep., 12, 20512.

Ciais, P., et al. (2026). Why methane surged in the atmosphere during the early 2020s. Science, 391, eadx8262.

France, J. L., et al. (2022). $\delta^{13}C$ methane source signatures from tropical wetland and rice field emissions. Phil. Trans. R. Soc. A, 380, 2215.

Fujita, R., et al. (2025b). Global fossil methane emissions constrained by multi-isotopic atmospheric methane histories. J. Geophys. Res.: Atmos., 130, e2024JD041266.

Lan, X., et al. (2026). Trends in globally averaged CH_4 (Updated NOAA GML data, end-2025).

Morgenstern, O., et al. (2025). Radiocarbon monoxide indicates increasing atmospheric oxidizing capacity. Nature Communications, 16, 249.

Nisbet, E. G., & Manning, M. R. (2026). What is causing the methane surge? Science, 391, 556–557.

Nisbet, E. G., et al. (2021/2022). Isotopic signatures of methane emissions from tropical fires, agriculture and wetlands: the MOYA and ZWAMPS flights. Phil. Trans. R. Soc. A, 380, 2215.

Nisbet, E. G., et al. (2020). Methane mitigation: methods to reduce emissions, on the path to the Paris Agreement. Reviews of Geophysics, 58, e2019RG000675.

Nisbet, E. G., et al. (2023). Atmospheric methane: comparison between methane's record in 2006–2022 and during glacial terminations.

Penn, E., et al. (2025). What can we learn about tropospheric OH from satellite observations of methane? Atmos. Chem. Phys., 25, 2947–2965.

Petrenko, V. V., et al. (2017). Minimal geological methane emissions during the Younger Dryas–Preboreal abrupt warming event. Nature, 548, 443–446.

Riddell-Young, B., et al. (2025). Microbial driver of 2006–2023 CH_4 growth indicated by trends in atmospheric $\delta D–CH_4$ and $\delta^{13}C–CH_4$. PNAS, 122, e2516543122.

Shaw, J. T., et al. (2022). Large methane emission fluxes observed from tropical wetlands in Zambia. Global Biogeochem. Cycles, 36, e2021GB007261.

Skeie, R. B., et al. (2023). Trends in atmospheric methane concentrations since 1990 were driven and modified by anthropogenic emissions. Comms. Earth & Envir., 4, 317.