### The Global Methane Budget: 2000-2020

Saunois et al., ESSD, 2024

### **Detailed Response to Anonymous Referee #1**

We acknowledge the referee for the time spent on reading and commenting on the paper. We thank Anonymous Referee#1 for the useful corrections and suggestions provided on the paper, which have helped clarify and improve the manuscript. Below are the responses (in black) to the comments (in italics, blue). Changes in the text follow each response in bold font.

This is a most important paper that makes a major contribution not just in terms of scientific discovery, but more generally to the wider effort of protecting the well-being of our planet. The work is extremely well documented and carefully presented.

*I strongly recommend publication.* 

That said, I do have a number of comments that the authors may wish to take into account for minor revisions prior to final publication.

We thank warmly the anonymous reviewer #1 for the compliment and the consideration of the importance of this work for the community.

### **General comments**

Quantifying the rapidly changing global methane budget is of the highest importance in tracking climate forcing, and of major societal value, particularly in the Global Methane Pledge, now signed by 155 nations, and in the UN Framework Convention on Climate Change, signed by all nations.

This paper is the most recent update in a series of authoritative and very influential assessments of the global methane budget, that have been produced by the Global Carbon Project over the past decade. Throughout the series of papers, the work of this team has been detailed, very welldocumented and of high academic quality.

The work in the paper is comprehensive and generally appears accurate, with thoughtful evaluations of uncertainties and gaps in knowledge. Once published, the budget assessment will be immediately valuable both to the very active scientific methane community, and in the wider climate debate.

We thank the reviewer again for this valuable comment. Indeed, the last year of the budget, 2020, is the reference year of the Global Methane Pledge (GMP) objective. The values provided by the Global Methane Budget could be further used to assess the success of the GMP.

However, I do have a number of specific questions that the authors may wish to consider.

In particular, it would be useful to add at least a comment on the need for isotopic balance. There is enough <sup>13</sup>C isotopic information available to provide an independent test of the budget, and thereby to help to constrain uncertainties.

Indeed we acknowledge that the budget provided here does not include any isotopic constraints, while the isotopic signal is quite clear in the atmosphere. In the past we tested the consistency of the budgets (BU and TD) with the isotopic signal (Saunois et al (2017) for the 2000-2012 periodand we also did in Zhang et al (2022), an exhaustive assessment for the 2000-2017 period. A follow-up study is planned to address this issue based on the GMB estimates. Though it would have been better to integrate this in the main budget, we could not for the sake of time. However, isotopic studies are not as straightforward and face challenges. Indeed, the uncertainties and variabilities (regional mostly but also temporal) of the isotopic signatures remain a difficulty when it comes to attributing emissions to sectors, (especially anthropogenic versus natural emissions from microbial sources). We were probably a bit too optimistic in the 2020 paper in the projection that more inversions would include isotopes capabilities. To date only 2–3 groups have published global analysis with 3D atmospheric inversions using isotopic constraints over several years (Thanwerdas et al., 2024; Basu et al., 2022). Box models may be used to try to constrain the budget with the isotopic signal but that probably would not give the same results as a full 3D inversions assimilating both CH<sub>4</sub> and delta <sup>13</sup>C-CH<sub>4</sub> data. We still have a goal to obtain more runs and hope to be able to report an ensemble of inversion results in the next release of GCP-CH4.

A sentence at the end of the 3rd paragraph of the introduction has been added: "Another difficulty of the CH<sub>4</sub> budget lies in the necessity to also match the isotopic balance and in particular reflect the decreasing methane isotopic signal <sup>13</sup>C (Nisbet et al., 2016; 2019). The previous budgets were tested against the isotopic observations (Saunois et al., 2017) and follow an exhaustive assessment (Zhang et al., 2022). To date only a couple of atmospheric inverse systems are able to assimilate both CH4 mixing ratios and stable isotopic signal to retrieve fluxes at the global scale (Thanwerdas et al., 2024; Basu et al.,2022), but these systems still need improvements in terms of configuration set-up and computing time resources, in addition to characterization of source signatures and chemical kinetic effect (Chandra et al., 2024). We hope to be able to report isotopic constrained budgets in the coming years, or at least test the budget against the isotopic balance."

# In the concluding sections it might also be interesting to compare the findings to the official UNFCCC declarations and other important assessments (e.g. IEA), and perhaps to say a little more about the remarkable changes that have taken place in the 2020s.

1. Regarding the official UNFCCC declarations, and other assessments (such as IEA for fossil emissions), not all countries regularly report their methane emissions to UNFCCC (yet). Few studies have investigated and compared the UNFCCC reporting against bottom-up (including IEA) and top-down approaches (for example Deng et al. (2022, 2024), Tibrewal et al. (2024)). Such comparison requires thorough investigation and are stand-alone studies that are done following the publication of the Global Methane Budget. We have added the following sentences near the end of the conclusion: "Further investigation is needed in follow-up studies to (1) compare these results to the official UNFCCC declarations and to important assessments such as those of IEA) as done previously for example in Deng et al. (2022; 2024) or more specifically for fossil fuel emissions in Tibrewal et al. (2024) and (2) further discuss the trend and interannual variability of CH<sub>4</sub> sources and sinks at sectoral and regional scales

as in Jackson et al. (2020, 2024), Stavert et al. (2021) or RECCAP-2 related publications (e.g., Petrescu et al., 2021; 2023; Lauerwald et al., 2023b), and discuss the compatibility of the budget against the atmospheric isotopic signal such as in Saunois et al. (2017). The next budgets will be critical to assess whether the Global Methane Pledge is successful and assess methane mitigation efforts."

2. Regarding the recent changes in the early 2020s, the budget does not cover the period. However, we already had a Section (Section 6) addressing some of the recent changes, providing insights of studies that have investigated this.

As a matter of presentation, it would be a lot easier on the tired-eyed reader to break up some of these enormously long paragraphs! Also, as is inevitable in such a comprehensive paper, some sections seem a bity elderly and could do with updating and a few more recent references.

Indeed some paragraphs were not updated as much as others. Using the useful comments provided by the three reviewers, we have addressed this issue, hoping that will satisfy the reviewer.

### Conclusion

This is a major paper of great importance to society as a whole. It is a credit to the authorship team and should be published after minor revision. The biggest weakness is the absence of any effort to use the isotopic constraints. Perhaps that should wait until the next update in a few years' time, but it should be considered or at least mentioned here, if only in a brief section.

The isotopic section has been removed since the budget published in 2020 as we do not (yet) use these data directly to constrain the budget. Nevertheless, and as mentioned earlier, we added a few relevant sentences to the introduction and in the last results section (Section 6)

In the introduction, we have added a few sentences at the end of the third paragraph (see above).

in section 6, we have added the following text: "The very high records of CH<sub>4</sub> growth rate over 2020-2022 have also been accompanied by a sharp decline in the stable isotopic signal,  $\delta^{13}C_{CH4}$ , driven by increased emissions from microbial sources such as those found in wetlands and freshwater, agriculture, and waste systems (Michel et al., 2024)."

### **Specific Comments**

Abstract: Line 155. The abstract will be widely cited, and many will look at it but not read the paper in detail (sadly). Thus it would help if the abstract could include more source-specific numbers detailing inputs like fossil fuels, agriculture, biomass burning and also sink estimates. If that makes the abstract too long, then maybe move lines 130-135 (which are very important, but not necessarily needed in the abstract) to the introduction?

Following this advice, we have removed former L148-153 (about the latitudinal distribution). We have added the global decadal estimate by category after the estimates of the tropospheric OH sink and total sink: "The tropospheric loss of methane, as the main contributor to methane lifetime, has been estimated at 563 [510-663] Tg CH<sub>4</sub> yr<sup>-1</sup> based on chemistry climate models. These values are slightly larger than for 2000-2009 due to the impact of the rise in atmospheric methane, and remaining large uncertainty (~25%). The total sink of CH<sub>4</sub> is estimated at 633 [507-796] Tg CH<sub>4</sub> yr<sup>-1</sup> by the bottom-up approaches and

at 554 [550-567] Tg CH<sub>4</sub> yr<sup>-1</sup> by top-down approaches. Though, most of the top-down models use the same OH distribution, which introduces less uncertainty to the global budget than is likely justified.

For 2010-2019, agriculture and waste contributed an estimated 228 [213-242] Tg CH<sub>4</sub> yr<sup>-1</sup> in the top-down budget and 211 [195-231] Tg CH<sub>4</sub> yr<sup>-1</sup> in the bottom-up budget. Fossil fuel emissions contributed 115 [100-124] Tg CH<sub>4</sub> yr<sup>-1</sup> in the top-down budget and 120 [117-125] Tg CH<sub>4</sub> yr<sup>-1</sup> in the bottom-up budget. Biomass and biofuel burning contributed 27 [26-27] Tg CH<sub>4</sub> yr<sup>-1</sup> in the top-down budget and 28 [21-39] Tg CH<sub>4</sub> yr<sup>-1</sup> in the bottom-up budget."

### L190 – GMP now has 155 signatures.

It is true that this number changes over time. This has been updated – 158 as of October 2024

L208 – maybe somewhere mention Anderson, D. C., et al. (2024). Trends and interannual variability of the hydroxyl radical in the remote tropics during boreal autumn inferred from satellite proxy data. Geophysical Research Letters, 51,

This citation has been included in Section 7, complementing the reference to their technical note published in 2023. "Recently, numerous efforts based on satellite data have been made to constrain OH distribution, variability and trends (e.g, Anderson, 2023; 2024 ; Pimlott et al. 2022 ; Zhao et al., 2023; Zhu et al., 2022). "

L251 – perhaps mention Yu, X, et al. (2023) A high-resolution satellite-based map of global methane emissions reveals missing wetland, fossil fuel, and monsoon sources. Atmospheric Chemistry and Physics 23): 3325-3346.

This citation will not be at the right place here. Here we discuss the accuracy of the satellite measurements. The paper suggested by the referee uses TROPOMI to correct prior estimates of fluxes. This correction highly depends on the prior used and also on the constraint observations.

Though the paper has been cited as an example of the use of TROPOMI in Section 7.

L306 – EDGAR v6? – See also L424.v7, and now v8. I guess in a big synthesis exercise like this it is hard to keep up with changes to inventories. Maybe mention new work by one of the co-authors. Crippa, M., et al. (2023) Insights on the spatial distribution of global, national and sub-national GHG emissions in EDGARv8. 0. Earth System Science Data Discussions 2023: 1-28.

Indeed, since the start of the study, the EDGAR team has released updates of their inventories. We started to use EDGARv6 as prior for the inversions. We then included EDGARv7 ending in 2021 for the BU budget - which was enough as the study spanned 2000-2020. EDGARv8 and now EDGAR\_2024\_GHG were released later on and provided data up to 2022 and 2023 respectively. This is used in Section 6 where we discuss the post 2020 period. We included the following statement in section 2.2 after mentioning the use of EDGARv6 as prior for the inversions: "Though EDGARv7 (EDGAR, 2022; Crippa et al., 2023) spanning until 2021 was then released, and was used for the bottom-up budget. EDGARv8 (EDGAR, 2023; Crippa et al., 2023) spanning until 2022 and released in 2024, was used in Section 6 to discuss the post 2020 methane budget. "

### L327 – good to have N2O and CH4 regions comparable.

### This is a comment.

## L348 – maybe could add a few general words on how to assess heavily human-dominated floodplain deltas like the Nile and Ganges and the Yangtse and Mekong (Tonlé Sap) floodplains.

This assessment is indeed quite challenging, and separating natural from anthropogenic in those regions is challenging. We have added the following comment to acknowledge this issue: "Separating natural from anthropogenic sources could be quite challenging, especially over regions where sources overlap, as over heavily human-dominated floodplain deltas for example"

### L371 – soil uptake – major uncertainty.

We consider this only as a comment; the uncertainty on the soil sink will be further discussed in its specific subsection (Sect. 3.3.4).

L424 – see earlier comment on EDGAR v 6,7,8. (L306).

This has been addressed above.

### L439 – abatement coefficient - how much uncertainty does this introduce?

Unfortunately, emission inventories come rarely with uncertainty estimates; and in particular do not estimate the uncertainties introduced by each input variable, in particular activity data that have often no alternative dataset. However, comparing different bottom-up inventories and looking at the range of their estimates could inform on the impact brought by the methodology used to build each inventory. This abatement coefficient is very sector and region specific and looking into this would require deep analysis of the inventories at national scale for example, which is out of the scope of this study.

L507 – Methane mitigation challenge – maybe mention Nisbet et al. 2020 Rev. Geophys.

Indeed Nisbet et al. 2020 as well as Shindell et al. 2024 are good references here. They have been included.

L510 – 'well within the range of scenarios' — the paper seems to suggest the recent climb is OK and expected, so we can be complacent. But the past few years have been much more like 5-8.5. Scary! See also fig. 6 in Nisbet et al Phil Trans Roy Soc A 379 (2021): 20200457.

Sure this is not the message we wanted to draw. This has been rephrased to: "observations of global CH<sub>4</sub> concentrations fall well within the range of scenarios in absolute values but their trend over the past few years is closest to those of scenario SSP5-8.5 "

L530 – FF 34% - maybe a comment on how this is changing?

This contribution is a bit increasing over time - considering the mean estimate of the bottomup approches. This study shows that FF contributed a bit less than 32% (31,5%) on average in 2000-2009, a bit less than 34% (33,5%) and a bit more than 34% (34.4%) for 2020. We have modified the text as follows: **"The sector accounts on average for 34% (range 31-42%) of total global anthropogenic emissions in 2010-2019. This contribution has slightly increased from 32% on average in 2000-2009."** 

### L563 – underground fires – real guesswork.

This is true. This value seems to be the same for years in EDGAR inventories across any version of EDGAR, despite the increase in coal exploitation in China for example. This estimate is largely unknown but we lack information to better discuss this. Though we have added a comment on this value: "An additional assumed very small source corresponds to fossil fuel fires, which are mostly underground coal fires. This source is estimated at around 0.15 Tg yr<sup>-1</sup> in EDGARv7, though this value remains the same across EDGAR versions and all years despite the changes in coal production, which could influence this estimate. However, to date, insufficient data is available to better estimate this largely unknown source."

L592-594 Low fracking losses - My gut feeling is to concur with this, but it might be a bit higher. The reference list is pretty old here - maybe cite more recent studies both by the EDF supported and other teams. Examples include Zhang, Y, et al. Science advances 6 (2020): eaaz5120. Or Li, et al. Science of The Total Environment 912 (2024): 169645. Or Williams, et al. EGUsphere 2024 (2024): 1-31. Shen, Lu, et al. Nature Communications 14 (2023): 4948.

We are thankful to the reviewer for the list of recent studies to include here.

The former lines 592-594 addressed the emission factors of the Oil and gas facilities. Zhang et al., 2020 suggest that emissions represent 3.7% of the gross gas extracted from the Permian. Li et al. (2024) do not estimate an emission factor but reported more on the underestimation of emissions Alberta in the inventories in Alberta. Williams et al. (2024) discuss the loss rate depending on the production gross. Finally, Shen et al. (2023) compared the estimate of UNFCCC and other BU inventories to TROPOMI based atmospheric inversions over 24 countries and found large underestimation, though they do not specify emission factors. The suggested studies looking at the underestimation of the emissions have been used later in this section.

Looking at the literature and considering the key point we wanted to highlight the sentence (former lines 592-594) has been rephrased as follows: "The latest studies tend to infer emission factors from the oil gas production chain of about 1% to 2% (e.g., Schneising et al., 2020; Varon et al., 2023; Zhang et al., 2020), but loss rate could be has high as more than 10% in low producing well sites (Omara et al., 2022, Williams et al., 2024)."

### L611 – 613 – again, elderly references in a fast moving field.....but Lavaux ref is good.

This has been modified to: "Most recent studies (e.g., Zhang et al., 2020; Shen et al., 2023; Li et al.; 2024, Tibrewal et al., 2023; Sherwin et al., 2024) still suggest that the methane emissions from oil and gas industry are underestimated by inventories, industries, and agencies, including the USEPA and UNFCCC reporting. "

L624 – extra 8 Tg....perhaps – but ultra-emitters tend to be short lived.

This is true. It would be interesting to analyze the trend of the emissions from those superemitters, but we do not know of any study that looked into this.

### L640-642 90/10 ratio of eructation vs flatulence – give a reference for this? Or Johnson?

We found a similar value in Hill et al. (2016), and included this reference.

### L669 – enteric 114-124 Tg – my instinct is that the uncertainty is wider than this?

Indeed, this range is the min-max from the estimates and does not include the uncertainty of the individual estimate, which would be larger than this. We have added the following sentence : "It is worth recalling here that the ranges provided in this study correspond to the minimum-maximum of the existing estimates and do not include the uncertainty of the individual estimate; these uncertainties could be larger than the range proposed here."

L737 and L769 – maybe cite some direct measurements? For example Barker, P., et al. (2023) Airborne measurements of fire emission factors for African biomass burning sampled during the MOYA campaign. ACP 20s 15443-15459.

We have modified the text as follows: "Small fires associated with agricultural activity, such as field burning and agricultural waste burning, are often not detected by moderate resolution remote sensing methods and are instead estimated based on cultivated area <u>or</u> <u>through in-situ measurements such as dedicated airborne campaigns (e.g., Barker et al.,</u> <u>2023).</u>"

L834 - 842 – 853 There's a problem in depending on land surface models. Note the bad failure of land surface models in tropical African and Bolivian wetlands – see especially Fig 8 and discussion in Shaw J.T. et al. (2022) Large methane emission fluxes observed from tropical wetlands in Zambia. Global Biogeochemical Cycles 36: e2021GB007261. Also see France et al. France, J. L., et al. (2022) Very large fluxes of methane measured above Bolivian seasonal wetlands. PNAS 119 e2206345119. Also note models in Zhang, Z. et al. Nat. Clim. Change https://doi.org/10.1038/s41558-023-01629-0 (2023) and comment by Nisbet 2023 Nature Climate Change 13 421-422.

Indeed LSMs still suffer from many uncertainties. To acknowledge this, we have included the following sentences, citing the suggested references: "However, large uncertainty remains in both spatial and temporal emission distributions, especially over tropical wetlands where data are lacking to evaluate the models but are nevertheless a key region for climate feedbacks (Nisbet et al., 2023; Zhang et al., 2023). Direct measurement campaigns and remote sensing are providing key insights for where to improve the land surface models (e.g., France et al., 2022; Shaw et al., 2022)."

L957 – note that Shaw et al (section 3.1.1) found low emissions over the wide shallow lake Bangweulu, even though the lake is shallow, warm and there is a lot of organic input – instead, the high emissions were dominated by the wetlands beside the lake. Although I have often observed warm afternoon ebullition events (sometimes dramatic) in lakes and open water bodies, as a general but intuitive impression (i.e. personal observation not backed up by quantitative study) my 'feel' is that where water is more than about 3 to 4m deep and where plant stems (e.g. reeds,

### papyrus) do not project to entrain methane up from mud to air in the sap, then lake emissions are fairly low. Any bubbles are taken up in the aerobic conditions in oxic upper water.

The estimates produced here are from studies that have exhaustively collected data from the literature, or produced data using a process-based model trained on such literature data, more than a general intuitive impression (these process-based models include an explicit representation of gas transport and bubble dissolution). The general consensus in the oceanographic literature is you need a depth of >100 meters for the methane in a bubble to equilibrate with surrounding waters. The total and ebullitive CH4 fluxes reported here for global lakes are thus based on the best empirical evidence and scientific knowledge currently available. Of course, significant uncertainties remain (due to limited observations and incomplete understanding) and we have conveyed this by expanding on lines 995-996 of the original manuscript (see next comment).

L975 – see comment on lakes – these numbers are pretty high and based on few observations - may be higher than reality.

We agree that the number of observations remains limited. Yet it is important to note that if our assessment is based on a few studies/models, these efforts try to use all available observations. We nevertheless recognize that the observation ensemble might not fully capture the wide diversity of processes and environmental drivers impacting lake and reservoir CH4 emissions and have thus have expanded on uncertainties and directions for future research (line 995-96 of original ms.): "Finally, for all inland water systems a greater scrutiny of the limiting factors (including the impact of ice-cover, stratification of the water column and seasonality) CH4 production, consumption and transport pathways is needed. In addition, a better understanding of the climatic, environmental and geomorphological controls on key CH4 processes (e.g., sedimentary diffusive and ebullitive production, bubble dissolution, CH4 oxidation) on the large-scale remains critically needed. For instance, the consistently lower global emissions determined by the process-based model of Zhuang et al. (2023) compared to observations, suggest that current datasets are too limited to fully capture the spatio-temporal variability in CH4 dynamics and their key control factors, possibly leading to biased-high estimates. "

### L1020 – run-off of agricultural Nitrogen fertilizer and manure to nearly wetlands?

The sentence has been modified: "It is also clear that the cultural eutrophication of natural lakes <u>driven by run-off of agricultural nitrogen fertilizer and manure</u> is augmenting CH4 emissions (DelSontro et al., 2018; Li et al., 2021), with shallow lakes particularly likely to experience eutrophication (Qin et al., 2020)."

#### L1068. Interesting discussion of this vexed problem. Good.

Thank you.

### L1084 - may still be too high? Very much guesswork!

We agree that the total natural wetland (vegetated+inland water) methane emissions flux is still likely too high. The bottom-up method maintains fidelity to each of the emitting sectors, and the issue of double counting (leading to the high numbers) is gradually being reduced with each assessment. The lessons learned are carried forward each time, with this budget providing better distinction for wetlands and inland waters, and including a lateral flux correction term. The wetlands team will soon be looking at lessons learned from this budget to provide improved estimates for the next CH<sub>4</sub> budget. The inland water team will need to continue expanding their observational database. Furthermore, these observations should help inform a wider range of (newly-developed) models resolving the spatial and temporal variability in inland water fluxes on the global scale.

L1116 – The Petrenko group's evidence is compelling – theirs are very high quality measurements. Maybe also cite Dyonisius, Michael N., et al. (2020) Science 367(2020): 907-910. I'm not convinced by the Thornton et al (2021) arguments.

There are still contradictory estimates of geological emissions of methane. Dyonisius et al. (2020) also suggests low geological (14C- free CH4 emissions) over the time period spanned by ice-core record (between 0 and about 15 Tg/year). This paper has been added in the discussion: "(Dyonisius et al. (2020) also suggest a low range of geological emissions over the last deglaciation period and for the late Holocene (0-10 Tg CH<sub>4</sub>yr<sup>-1</sup>)".

We believe we tried to remain impartial. No improvement or reconciliation has been made since Saunois et al. (2020). Thornton et al. (2021) is still the latest paper comparing and discussing the issue. Different individual and independent studies suggest significant geological emissions at regional scale. We have further acknowledged this in the following sentence: "This discrepancy highlights another main unresolved uncertainty in the methane budget and calls for further investigations to reconcile the different estimates and reduce the uncertainty on geological emissions. "

L1127 – typo – Schwietzke.

### This has been corrected

L1133 – Termites – interesting how little work has been done since Pat Zimmerman's work in the 1980s. The 10 Tg estimate looks like a placeholder! Maybe cite Chiri, E., et al (2020). Termite mounds contain soil-derived methanotroph communities kinetically adapted to elevated methane concentrations. The ISME journal, 14, 2715-2731. And Chiri, E, et al. Termite gas emissions select for hydrogenotrophic microbial communities in termite mounds. PNAS118 (2021): e2102625118.

Indeed, the scaling up of termite emissions still lacks a sufficient number of studies to cover extensively all the uncertainty areas underlined in the introductory section on termites in this paper. Most advances concern aspects that were investigated with laboratory analyses, like the increase of data on CH4 emissions factors in vitro and also most recent interesting studies of biochemistry/genetic like the ones proposed by the reviewer by Chiri et al., which provided new insight on the complexity of the mound environment and termite- microorganisms connections. This surely is a frontier aspect that is very challenging. However a fundamental big gap of knowledge on the basics of termites ecology remains, related to the variability of termites groups, nests typologies, biodiversity distribution of the different populations (and hence trophic modes and nest types), among different ecosystems and across continents, as well as our ability to a more precisely estimate the total termite biomass of an ecosystem. This task would require a challenging field monitoring approach at large scale, with significant international collaboration over continents among groups of entomologists, ecologists, experts of flux emissions and more.

We have included the citations in the following text : " The species of each family and subfamily of the two major groups of lower and higher termites, listed by Sugimoto et al. (1998) were associated with EF values based on emissions from in-vitro experiments as reported by Sanderson (1996) and Eggleton et al. (1999), to which a correction factor ( $cf_{MOUND}$ ) of 0.5 (Nauer et al., 2018; <u>Chiri et al., 2020; 2021</u>) was applied in order to take into account the mound effect on the CH<sub>4</sub> produced by termites, once inside the nest. "

L1205 – I strongly suspect this 2 Tg number is a serious underestimate. Even in cattle monocultures there are dense populations of small nocturnal antelope (in Africa) and small deer (in EU and N America), in bush and scrub forest, etc as well as larger tree clumps and forests. For Africa see for example Hempson, G.P., Archibald, S. and Bond, W.J., 2017. The consequences of replacing wildlife with livestock in Africa. Scientific reports, 7, p.17196.

This value could seem underestimated, though not many global studies exist. We have modified the text as follows, "However, the estimate of 1-3 Tg CH<sub>4</sub> yr<sup>-1</sup> seems underestimated when considering that Hempson et al. (2017) found actual CH<sub>4</sub> emissions from African wild life alone to be around 9 Tg CH<sub>4</sub> yr<sup>-1</sup> but without discussing the uncertainty of this value. As a result, high uncertainty remains and recalls the need for further investigation of this natural source of CH<sub>4</sub>.

Based on these findings and waiting for further global estimates, the range adopted in this updated CH<sub>4</sub> budget is 2 [1-3] Tg CH<sub>4</sub> yr<sup>-1</sup> (Table 3)."

L1216 – gas plumes – see Fig 1 in Westbrook, Graham K., et al. (2009) Escape of methane gas from the seabed along the West Spitsbergen continental margin. Geophysical Research Letters 36. Plumes disappear quickly and even very large plumes don't manage to rise through more than about 250m of water.

Some studies show evidence of gas bubble plumes searching the surface, even at high depth, depending on the intensity of the events. We have added the reference (and other) and modified the sentence : " <u>Gas bubble plumes generally reach the atmosphere in relatively shallow waters (<400 m) of continental shelves depending on the intensity of the events (e.g., Westbrook et al. 2009); however, massive deep-water seepage events could contribute significant amount of CH4 to the atmosphere even from depths > 1000m (e.g., e.g., Schmale et al., 2005; Greinert et al., 2006, Solomon et al., 2009)."</u>

## L1275 – the in-situ oceanic source from phosphonates is real. It may be small, but it's essentially unknown. This 5 Tg number looks a bit like the 'placeholder' in Cicerone and Oremland 1988.

The 5 Tg is mostly based on recent studies: Rosentreter et al. (2023) and Weber et al. (2019). For such sources, the uncertainty remains large with at least +/- 50%, as shown with the 3-10 Tg yr<sup>-1</sup> range. We have added a reference to Resplandy et al. (2024) that discusses the possible contribution of sedimentary methane vs. direct aerobic in situ production in the surface waters (including the methylphosphonate pathway). Further studies are needed to better estimate the small coastal and oceanic emissions and their source contributions at the global scale as well as reduce uncertainties. The sentence has been revised as follows: "Overall, these marine biogenic emissions are sustained by a mixture of sedimentary production and

in-situ production in the sea-surface layers <u>(including the methylphosphonate pathway)</u> (e.g., Karl et al., 2008; and Repeta et al., 2016; <u>Resplandy et al., 2024</u>). "

### L1314 – see also L 1116 – seems too high given the Petrenko team's results.

Indeed, this is related to the above estimate. However, no other studies provided detailed estimates of geological emissions by pathways/locations. To acknowledge this, we have included the following sentence: "While we use the estimate by Etiope and Schwietzke (2019) , we acknowledge that high uncertainty remains and others studies suggest lower ranges of emissions based on radiocarbon (<sup>14</sup>C-CH<sub>4</sub>) data in ice cores (e.g., Hmiel et al., 2020). The suggested estimate may overestimate this source and be part of the top-down bottom-up discrepancy as discussed in Section 5.1.2."

### L1374 – does this also include features like the Yamal blowout collapse structures? Bogoyavlensky, Vasily, et al. (2021) Permanent gas emission from the Seyakha Crater of gas blowout, Yamal Peninsula, Russian Arctic. Energies 14: 5345.

This section reports permafrost direct emissions, which are difficult to estimate as stated in the text and in Hugelius et al. (2023). Individual formation exists as for the Yamal one but emission estimates are rare. Actually, we did not find quantification of the emissions of the Yamal structure in Bogoyavlensly et al (2021). Further investigation would be needed to provide a reporting of the existing thermokarst structures due to permafrost thaw associated with their emission. However, such inventory will probably be quite difficult to create.

L1381 – Nisbet et al 2009 demonstrated transport of methane via sap – i.e. plant methane comes from underlying **an**aerobic soil methanogens, not in situ aerobic processes in the leaf. In other words the plants were acting as 'straws' (see Line1385). UV-caused emissions are tiny (Bloom et al.). Note the important work of Gauci et al and Pangala et al. (see Line 1388): are their emissions encapsulated in the other flux categories (Line 1401).

To acknowledge this we have added Nisbet et al. 2009 in the sentence in former line 1385: "Second, and of clearer significance, plant<u>stems</u> act as "straws", drawing up and releasing microbially produced CH<sub>4</sub> from anoxic soils (Cicerone and Shetter, 1981; Rice et al., 2010; <u>Nisbet, et al. 2009</u>). "

In order to update this section, we have added the following sentence to mention that trees could also act as methane sink: "Recently, field-work suggested that trees may also act as a CH<sub>4</sub> sink (Machacova et al., 2021 ; Gorgolewski et al., 2023 ; Gauci et al., 2024)".

L1433 some of this OH section is a bit elderly. Maybe add some new references? E.g. (see also above L208) Anderson, D. C., et al. (2024); and Stevenson, David S., et al. (2020) Trends in global tropospheric hydroxyl radical and methane lifetime since 1850 from AerChemMIP. *Atmospheric Chemistry and Physics* 20 12905-12920. Or Naus, S., et al. (2021). A three-dimensional-model inversion of methyl chloroform to constrain the atmospheric oxidative capacity. *Atmospheric Chemistry and Physics*, 21, 4809-4824.

It is true that some old citations have been kept. We have significantly updated this section and the discussion to highlight the most recent studies on the OH issue. Other small parts of

the text have been marginally changed. In the following, we include the most important changes made to the text: "OH concentrations and their changes can be sensitive to climate variability (e.g., Nicely et al., 2018; Anderson et al., 2020), biomass burning (e.g., Anderson et al., 2024), and anthropogenic emissions of precursors (Peng et al., 2022; Stevenson et al., 2020). OH distributions calculated by chemistry climate models show large regional differences and various vertical profiles (Zhao et al., 2019). OH changes present also regional differences over the long term (Stevenson et al., 2020). Despite large regional changes, the global mean OH concentration was suggested to have changed only slightly from 1850 to 1980, but followed by strong (9 %) increases up to the present day (Stevenson et al., 2020). This increase simulated by models over 2000-2015 are however not in agreement with observation-based approaches (Thompson et al., 2024; Patra et al., 2020; Nicely et al., 2018; Rigby et al., 2017; Turner et al., 2017) where OH decreases or remain constant over the period. CCMI and CMIP6 models show OH interannual variability ranging from 0.9% to 1.8% over 2000-2010 (Table S4), in agreement with the values of IAV derived from some observationally constrained studies (e.g., Thompson et al., 2024; Montzka et al., 2011) but lower than value deduced from methyl chloroform measurements (Patra et al., 2021; Naus et al., 2021)."

### L1459-1461 and 1465-1466 – maybe these numbers should be in the abstract?

As suggested we have added the value of the tropospheric loss in the abstract with the following sentences: "The tropospheric loss of methane, as the main contributor to methane lifetime, has been estimated at 563 [510-663] TgCH4 yr-1 based on chemistry climate models. These values are slightly larger than for 2000-2009 due to the impact of the rise in atmospheric methane, and still present large uncertainty (~25%). "

## L1504 – interesting this number is low – the Cl sink has a strong isotopic leverage, so if the low number is correct that leverage is small.

Indeed, recent studies tend to lower tropospheric concentrations of chlorine, leading to a smaller impact on the total methane sink than previously thought. However, the isotopic leverage of chlorine in the troposphere is still meaningful for methane isotopic modeling as discussed in Strode et al., 2020 and Thanwerdas et al., 2022b (See previous line 1500-1502).

L1507 soil uptake. This is a major uncertainty in the budget because there are so few measurements from tropical seasonal rainfall woodlands and savannas. It's a huge unknown, linked to the termite unknown. (see Chiri papers L1133). I suspect the already wide uncertainty range on L1535 and L1543 is low!

The authors fully agree with this comment. Studies are rare: modelling ones may miss some processes and not enough observations are available to validate and calibrate them. We also acknowledge that this section has been marginally modified due to a lack of novel global estimates. We have added the following two sentences near the end of the section: "Indeed, Murgia-Flores et al. (2021) estimated that the global soil-uptake doubled between 1900 and 2015 and could further increase due to enhanced diffusion of CH<sub>4</sub> into soil as a result of increases in atmospheric CH<sub>4</sub> mole fraction. Further investigation of the soil uptake is required to better constrain this process at the global scale while it is highly dependent on

local scale microbial activity and environmental conditions (e.g., D'Imperio et al., 2023; Fest et al., 2017). "

L1546 – lifetime definition – burden/sink - maybe make clear that this is not the same as the 12 yr perturbation lifetime cited in some IPCC chapters. Media people get very confused between the two sorts of lifetime. Maybe cite Prather here (as well as on L1619)

The text at former line 1619 has been removed, first to avoid confusion from another reviewer and second because it did not fit the discussion we intend to have. Regarding the comment for L1546 - start of the section on methane lifetime- We have included a clear sentence on the difference between lifetime and perturbation time: "This value is different from what is called perturbation lifetime. Perturbation lifetime is used to determine how a one-time pulse emission may decay as a function of time as needed for the calculation of Global Warming Potentials (GWPs), and as a result is related to a theoretical concept. For CH<sub>4</sub>, the corresponding perturbation lifetime that should be used in the GWP calculation is  $11.8 \pm 1.8$ years (Forster et al., 2021). In this section, we discuss the global atmospheric lifetime (also called 'burden lifetime' or 'turnover lifetime') that characterizes the time required to turn over the global atmospheric burden and is defined by the burden divided by the removal flux. "

L1683 608 Tg – see also L 1836. maybe this number should be in the abstract also? It's ~10% higher than 2010's 554 Tg. See also Nisbet et al 2023 for discussion of post 2020 changes.

We have added the following sentence to the abstract: "The 2020 emission rate is the highest of the period and reaches 608 [581-627] Tg CH<sub>4</sub> yr<sup>-1</sup>, which is 12% higher than the average emissions in the 2000s."

The article from Nisbet et al. (2023) was already cited in Section 6 discussing post 2020 changes.

### L1824 – I suspect this 3 Tg number is a major underestimate!

Indeed, we recall this here as follows: "Wild animal and permafrost maps do not exist and are missing from the calculation, leading to a<u>t least</u> 3 Tg CH<sub>4</sub> yr<sup>-1</sup> of discrepancy. <u>However</u>, as aforementioned (Sections 3.2.5 and 3.2.7) this 3 Tg CH<sub>4</sub> yr<sup>-1</sup> estimate is probably <u>underestimated in the bottom-up budget.</u>"

L1883 regional budgets – maybe mention how these numbers contrast with recent studies like Yu, Xueying, et al. (2023) A high-resolution satellite-based map of global methane emissions reveals missing wetland, fossil fuel, and monsoon sources. Atmospheric Chemistry and Physics 23: 3325-3346.

Comparing different studies requires covering similar periods and regional scale. Yu et al. (2023) for example provide emission estimates over 2018-2019, the year with the highest emissions from our decadal period 2010-2019, and provide national estimates while we present values for a mix of large countries (Russia, the USA, Brazil, China..) and regions (South-East Asia, ..). For example, for China we found 57 [37-72] Tg CH<sub>4</sub> yr<sup>-1</sup> for 2010-2019, while they found 60 Tg CH<sub>4</sub> yr<sup>-1</sup> for 2018-2019. For the USA, we found 38 [32-46] Tg CH<sub>4</sub> yr<sup>-1</sup> for 2010-2019, while they found 44 Tg CH<sub>4</sub> yr<sup>-1</sup> for 2018-2019. The conclusion would be that we found similar estimates, considering the range of uncertainty that is larger in the Global Methane

Budget as we use an ensemble of top-down systems and not a single one; even if the study includes sensitivity tests.

We prefer providing the ensemble estimates and let further studies investigate the differences, if any, between studies.

### L1983 ? Mention the current d13C(CH4) isotopic plunge – and concerns (e.g. Nisbet et al 2023).

The following sentence has been added: "The very high values of CH<sub>4</sub> growth rate over 2020-2022 have also been accompanied by a sharp decline in the stable isotopic signal,  $\delta^{13}C_{CH4}$ , which suggests that this recent increase of growth rate is at least partly explained by increased emissions from microbial sources such as wetlands, inland waters, agriculture and waste (Nisbet et al., 2023; Michel et al., 2024). "

L2076 – FINALLY!!! A mention of isotopes!!! That's the big missing factor in the whole study!!!! The isotopes must balance – if they don't the budget needs to be re-examined. Obviously it is too late to add a full isotopic analysis to this paper, but the need for isotopic constraint surely should be mentioned. I would suggest that a short section on isotopes should be added here at this stage in the paper, including maybe a brief one-paragraph attempt at balance, and a promise to do something about it next time.

We fully agree that the budget must agree with the isotopic signal. This statement is also true for any top-down studies constrained by CH<sub>4</sub> only (even if based on TROPOMI as in Xueying et al. (2023) mentioned earlier by the reviewer). We have included the following sentence in the introduction: " Another difficulty of the CH<sub>4</sub> budget lies in the necessity to also match the isotopic signal and in particular reflect the decreasing methane isotopic signal <sup>13</sup>C (Nisbet et al., 2016; 2019). The previous budgets were tested against the isotopic observations (Saunois et al., 2017) and followed an exhaustive assessment (Zhang et al., 2022). To date only a couple of atmospheric inverse systems are able to assimilate both CH4 mixing ratios and stable isotopic signal to retrieve fluxes at the global scale (Thanwerdas et al., 2024; Basu et al., 2022), but these systems still need improvements in terms of configuration set-up and computing time resources, in addition to characterization of source signatures and chemical kinetic effect (Chandra et al., 2024).. We hope to be able to report isotopic constrained budgets in the coming years."

We are working on examining the 2024 version budget against the isotopic signal through 3D transport forward modelling. The results are too preliminary to be included here but will be part of a coming paper, hopefully next year. This paper will also aim at establishing a protocol to evaluate the top-down (and bottom-up budget) against the atmospheric signal of CH4 and  $\delta^{13}C_{CH4}$ .

To recall that most top-down studies are only based on CH4 constraints only we have included the following sentence in Section 6 - formerly L1986: "However, it is worth noting that almost all published top-down studies aforementioned include constraints only on CH<sub>4</sub>, and do not discuss the consistency with the atmospheric isotopic signal."

In the conclusion section we had the following sentences (see previous comment): "Further investigation is needed in follow-up studies to (1) compare these results to the official UNFCCC declarations and to important assessment (as those of IEA) as done for example in Deng et al. (2022; 2024) or more specifically for fossil fuel emissions in Tibrewal et al. (2024)

and (2) further discuss the trend and interannual variability of CH<sub>4</sub> sources and sinks at sectoral and regional scales as in Jackson et al. (2020, 2024), Stavert et al. (2021) or RECCAP-2 related publications (e.g., Petrescu et al., 2021; 2023; Lauerwald et al., 2023b), and discuss the compatibility of the budget against the atmospheric isotopic signal such as in Saunois et al. (2017). The next budgets will be critical to assess whether the Global Methane Pledge is successful and assess methane mitigation efforts."

### L2190 – maybe mention Nisbet et al 2023 here and/or in Line 2194?

The citation has been added at the two mentioned lines (former L2190 et L2194).

TABLES – these are extremely valuable and will be very heavily used. The Table 'callouts' could maybe be better integrated in the text and perhaps need to have more mentions: they'll be very prominent on publication.

As suggested, we have included more call-out to the Tables along the text.

FIGURES – these are excellent and again the 'callouts' maybe need some more prominence in the text. For Fig 2 see comparison with Nisbet et al 2021 Phil Trans. In Fig 3 wetlands is heavily biassed by the Congo peatlands – but somewhat misses/downplays the upper Congo wetlands and also downplays big emitters like the Pantanal and Mamore R. and the Sudd, despite lots of recent evidence for big emissions. I accept these reagions are very difficult to study by remote sensing, as cloud cover is so pervasive in wet season. I've flown over many and seen only cloud! Also I'm very sceptical of the 'lowish' biomass burn in India where crop waste burning is horrendous (or is crop waste burning classed under 'waste'?) Also India has a huge coal mining industry and giant landfills. Fig 5 'Geological' seems improbable that central Asia is less prominent than Romania (e.g. see MEMO2 / ROMEO measurements).

As suggested, we have included more call-out to the figures along the text. Further investigation would be needed to discuss the regional distribution of the sources and sinks, and their changes over time. We feel this is out of the scope of this study.

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