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Peer Review File

Methane trend over the last decades driven and modified by anthropogenic emissions

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28th Mar 23

Dear Dr Skeie,

Your manuscript titled "Methane trend over the last decades driven and modified by anthropogenic emissions" has now been seen by 3 reviewers, and I include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

A thorough examination of the drivers of change in the global methane budget is presented using atmospheric chemical transport models, box models and emission inventories. The study is very thorough and presents some useful results. I recommend it for publication in Communications Earth and Environment, provided that several relatively minor issues are addressed.

General comments

While the authors cite a commendable amount of literature, a more complete discussion of their findings in relation to some publications is needed in parts. For example:
- I felt that the work of Nicely et al. (2017; 2018) should be cited and discussed as it describes very similar model experiments but reaches somewhat different conclusions.
- The findings of this work appear to have some similarities to the inverse modelling-based findings of Rigby et al. (2017) and Turner et al. (2017), but these studies are only cited as part of a general introduction on previous methods.
- Studies such as Lan et al. (2022) discuss 13CH4 trends using box models, reaching somewhat different conclusions to those presented here.

While I certainly sympathise that the literature is vast and growing rapidly, I felt that some of the advances claimed here have been discussed elsewhere (at least in part), and so a slight broadening of the discussion is warranted.

Specific comments

L11 and throughout: the term “temporal development” is used extensively in this manuscript. While I’m not sure whether it’s technically incorrect, it is an extremely uncommon way of describing a

trend, or temporal variability. I suggest replacing the use of this term with “trend”, “temporal variability”, “evolution”, “changes”, or similar, depending on the appropriate context.

L19 and elsewhere: the use of “1/3” here sounds too precise. I’d suggest “approximately one third”

L22: It is ambiguous and potentially misleading to describe methane as the “second strongest” GHG. E.g., this could refer to its GWP, in which case the statement would be wrong. Re-word to clarify that this is in reference to its contribution to anthropogenic radiative forcing of climate.

L23: Presumably “global mean concentration”? Furthermore, I don’t believe that we know this number to 0.1 ppb, or the percentage increase to the nearest 1%.

L25: Delete “levels of”: “The current atmospheric concentrations are…”. However, it’s not clear what “higher than in scenarios consistent with limited global warming” means. I suggest deleting this part of the sentence.

L26: “point to”, rather than “point at”

L29 – 30: I have two problems with this sentence: a) you can’t evaluate future projections, b) I don’t know what “reaching the goal in the Paris Agreement” means. Which goal? I think you can delete this sentence.

L32: I don’t think the semicolon is needed in this sentence.

L34: suggest rewording to: “Between 2000 and 2007, atmospheric methane concentrations were relatively stable, but from 2007 it started rising again, with a further acceleration in 2014”.

L34: I suggest citing Dlugokencky et al. (2009) and Rigby et al. (2008) for the description of the renewed methane growth.

L39: “consists”, rather than “consist”

L44: “exceed”, rather than “exceeds”

L50: Delete “Also”

L56: What is the role of CO2 here? Is this via its influence on temperature?

L58: “influences”, rather than “influence”

L68: Other methods, or species have been used to estimate [OH], e.g., Wolfe et al. (2019), Montzka et al (2011) (i.e., it’s not just CTMs or methyl chloroform)

L70: “… THE OH trend IS large…”

L83: this needs rewording. Perhaps “as a consequence of COVID-19 lockdowns…”

L85: “investigated”, rather than “presented”?

L98: I assume the NOAA data needs to be acknowledged with an appropriate literature citation, rather than a URL?

L118: Somewhere here, say where the observed trends are taken from.

L126: “are not able to reproduce the observed atmospheric trend…” This needs to be softened. No model will ever be able to fully reproduce the trend, but there are some features in common.

L137: “also seen as stable anthropogenic emissions in the 1980s continuing up to year 2000”. This needs rewording, as I don’t know what it means.

L141: “gives AN atmospheric trend…”

L146: I suggest “after the stabilization…”

L173: “and up to 1980-90 of 3%”. It’s not clear what this means. Reword.

L176: some commas would help here: “and longer, respectively, as in 1850. From 1990, OH…”

L183: “declines”, rather than “decline”

L204: “results, also indicating a role for the emissions”, rather than “results, that also indicate”. However, you also need to say which emissions here (CO, NOx, CH4?)

L205: what is meant by “OH development”?

L206: When translating the 3D CTM results to the box model, how do you account for the influence of the varying CH4 emissions in the 3D model? Wouldn’t there be a non-linear response on the methane lifetime from these methane emissions variations?

L214 - 215: “Changes in the OH sink have likely contributed to the stabilization…”. Similar conclusions were reached (albeit with very large uncertainty) by Turner et al. (2017) and Rigby et al. (2017). It seems these papers should be cited here and/or in the discussion.

L217: The caption and legend for Figure 1 suggests that only the CEDS21+COVID simulation was used, but here it seems to suggest that CEDS17 was used prior to 2000? Apologies if I’ve misread. In any case, why was a change in simulation needed? What would be the effect of this change?

L220: Should this be -3.2 ppb/yr? The use of “negative” in this sentence confuses matters.

L222: “COVID emission reductions”: need to say for which species here, as I don’t think you mean for CH4 (rather, NOx/CO)?

L222: “contributing”, rather than “contributed”?

L236: “show” rather than “span”?

L242: perhaps it's the relatively low-resolution version in the document that I have, but it's not clear to me where the boxes overlap. I'm therefore not sure why the caption says this.

L262: "were", rather than "was"?

L265: "results to the", rather than "as the"

L266: "does" rather than "do"

L272: What are the fluxes from non-inundated areas?

L275: something like: "… datasets to drive the model ARE SHOWN"? Also, it should be "indicates", rather than "indicate".

L288: "is seen", rather than "can be expected"?

L293: Again, what are the emissions from non-inundated areas?

L305: Fig 5b, rather than 6b?

L325: "negative anomaly", rather than "spike"?

L326: "growth rate", rather than "trend" (which is non-specific)

L340: Is "Solazzo" reference to a publication, or personal communication, or something else?

L344: "The COVID lockdown did not affect methane the most compared to other emission components". I don't know what this means. It's a very strong statement though. Needs rewording.

L355: emissions of which species?

L355: "OH variability", rather than "OH development"?

L354 – 356: I don't really understand what is being claimed here. Why does this indicate that the emission inventory is important, since the previous sentence just states that the AerChem and Oslo models had similar OH trends?

L367: A reference is needed here

L372: "After the methane stabilization…"

L383: "… less than a factor of two…"

L388: "anomaly", rather than "spike"?

L402: "Can an increase in OH from the 1980s to early 2000s using CEDS-2017 emissions…", rather than "Can an increase in OH from the 1980s to early 2000s as the models show using CEDS-2017 emissions…"? This sentence should be reworded in any case, ideally not having the figure reference

in the middle.

L406 – 415: I think that Rigby et al. (2017) show similar conclusions (e.g., Figure S6 in their paper).

L446: as above, “1/3” seems too precise. “approximately one-third”?
L449: What is meant be “components” here? Do you mean “atmospheric constituents”? Or perhaps just “factors”

L450: I suggest deleting this sentence. It’s just one factor that could influence CH4 lifetime, and it seems very out of context as a concluding line here.

L465: sets, rather than “set”.

L579: Isn’t [OH] a bigger source of uncertainty in the methane budget?

Additional references:

Dlugokencky, E. J., Bruhwiler, L., White, J. W. C., Emmons, L. K., Novelli, P. C., Montzka, S. A., Masarie, K. A., Lang, P. M., Crotwell, A. M., Miller, J. B., and Gatti, L. V.: Observational constraints on recent increases in the atmospheric CH4 burden, Geophysical Research Letters, 36, L18803, https://doi.org/10.1029/2009GL039780, 2009.

Lan, X., Basu, S., Schwietzke, S., Bruhwiler, L. M. P., Dlugokencky, E. J., Michel, S. E., Sherwood, O. A., Tans, P. P., Thoning, K., Etiope, G., Zhuang, Q., Liu, L., Oh, Y., Miller, J. B., Pétron, G., Vaughn, B. H., and Crippa, M.: Improved Constraints on Global Methane Emissions and Sinks Using δ 13 C-CH 4, Global Biogeochemical Cycles, 35, https://doi.org/10.1029/2021GB007000, 2021.

Nicely, J. M., Salawitch, R. J., Canty, T., Anderson, D. C., Arnold, S. R., Chipperfield, M. P., Emmons, L. K., Flemming, J., Huijnen, V., Kinnison, D. E., Lamarque, J.-F., Mao, J., Monks, S. A., Steenrod, S. D., Tilmes, S., and Turquety, S.: Quantifying the causes of differences in tropospheric OH within global models: Quantifying Global Model OH Differences, Journal of Geophysical Research: Atmospheres, 122, 1983–2007, https://doi.org/10.1002/2016JD026239, 2017.

Nicely, J. M., Canty, T. P., Manyin, M., Oman, L. D., Salawitch, R. J., Steenrod, S. D., Strahan, S. E., and Strode, S. A.: Changes in Global Tropospheric OH Expected as a Result of Climate Change Over the Last Several Decades, Journal of Geophysical Research: Atmospheres, 123, 10,774-10,795, https://doi.org/10.1029/2018JD028388, 2018.

Rigby, M., Prinn, R. G., Fraser, P. J., Simmonds, P. G., Langenfelds, R. L., Huang, J., Cunnold, D. M., Steele, L. P., Krummel, P. B., Weiss, R. F., O’Doherty, S., Salameh, P. K., Wang, H. J., Harth, C. M., Mühle, J., and Porter, L. W.: Renewed growth of atmospheric methane, Geophysical Research Letters, 35, L22805, https://doi.org/10.1029/2008GL036037, 2008.

Reviewer #2 (Remarks to the Author):

<B>Review of “Methane trend over the last decades driven and modified by anthropogenic

emissions" by Skeie et al.</B>

This manuscript explores the drivers of the trends and shorter-term variability of atmospheric methane (CH4) over recent decades through evaluation of some of its major sources and sinks over that time period. The main conclusion is that anthropogenic emissions dominate both the long-term trends of methane and much of the variation in these trends, with smaller impact from natural sources. The anthropogenic contribution to CH4 variation is both direct, through emission of CH4 into the atmosphere, and indirect – changing the methane lifetime due to human-influenced variations in other species in the atmosphere. Finally, and most interestingly, the authors investigate the possibility that delayed influence of increased atmospheric hydroxyl (OH) concentrations might have influenced the declining isotopic ratio of atmospheric CH4 in the past decade.

Generally, the work is very well-written with clear and appropriate figures, whilst the methods are suitable and the authors have combined a range of datasets to investigate their hypotheses. I did not come across many major issues regarding the clarity, accuracy and appropriateness of the work that is demonstrated here.

However, my main issue is that many of the early findings of the paper are not convincingly novel. Previous groups have shown that anthropogenic emissions have been responsible for much of the rise in CH4 over previous decades (e.g. McNorton et al. (2018); Turner et al. (2019); Stavert et al. (2021); Zhang et al. (2021)), whilst others have proposed and assessed the effect of increased OH concentrations during the slowdown period for CH4. (e.g. McNorton et al. (2016); Turner et al. (2017); Rigby et al. (2017)). There is mixed evidence for stable or changing global wetland and/or natural emissions. (e.g. Melton et al., (2013); Saunois et al. (2020); Oh et al., (2022)). Others have quantified the effect of methane lifetime on the growth rate during the 2020 Covid pandemic. (e.g. Peng et al. (2020)) The strength of this work relative to these others is perhaps in bringing these strands together within one model, and covering a relatively long time period. However, in my opinion, to be considered for publication the authors need to explain much more clearly why their results and/or methods are innovative, and not simply an update on or confirmation of the findings of others.

One respect in which I did find their results to be novel, but maybe a little underdeveloped, is in the idea that the prolonged effects of OH variations on the isotopic ratio of 13CH4/12CH4 in the atmosphere can explain at least some of the recent observed variations in this metric. The results shown in Figure 7 are interesting, but the discussion is quite brief. The authors can explain some 30% of the observed decrease in ∂13C_{CH4} due to this effect. However, the variation shown by the model is much smaller and slower than the observations, so some discussion on why that might be is appropriate. The following questions could be answered:
• If there is indeed no trend in natural emissions, what might account for the rest of the decrease, particularly since the observed changes are much more sudden?
• Is it possible to explore this question further with the box model, including and perturbing the isotopic signatures for different emissions?
• If not, or if results are inconclusive, what data is needed for further exploration of this isotopic variation? What changes in sources or sinks would be needed to produce changes in ∂13C_{CH4} of the magnitude observed?

If this aspect of the paper was further developed in this way and the novelty of earlier results was emphasised, then I think that the paper could be considered for publication here.

<B>References:</B>

McNorton, J., Chipperfield, M. P., Gloor, M., Wilson, C., Feng, W., Hayman, G. D., Rigby, M., Krummel, P. B., O'Doherty, S., Prinn, R. G., Weiss, R. F., Young, D., Dlugokencky, E., and Montzka, S. A.: Role of OH variability in the stalling of the global atmospheric CH4 growth rate from 1999 to 2006, Atmos. Chem. Phys., 16, 7943–7956, https://doi.org/10.5194/acp-16-7943-2016, 2016.

McNorton, J., Wilson, C., Gloor, M., Parker, R. J., Boesch, H., Feng, W., Hossaini, R., and Chipperfield, M. P.: Attribution of recent increases in atmospheric methane through 3-D inverse modelling, Atmos. Chem. Phys., 18, 18149–18168, https://doi.org/10.5194/acp-18-18149-2018, 2018.

Melton, J. R., Wania, R., Hodson, E. L., Poulter, B., Ringeval, B., Spahni, R., Bohn, T., Avis, C. A., Beerling, D. J., Chen, G., Eliseev, A. V., Denisov, S. N., Hopcroft, P. O., Lettenmaier, D. P., Riley, W. J., Singarayer, J. S., Subin, Z. M., Tian, H., Zürcher, S., Brovkin, V., van Bodegom, P. M., Kleinen, T., Yu, Z. C., and Kaplan, J. O.: Present state of global wetland extent and wetland methane modelling: conclusions from a model inter-comparison project (WETCHIMP), Biogeosciences, 10, 753–788, https://doi.org/10.5194/bg-10-753-2013, 2013.

Oh, Y., Zhuang, Q., Welp, L.R. et al. Improved global wetland carbon isotopic signatures support post-2006 microbial methane emission increase. Commun Earth Environ 3, 159 (2022). https://doi.org/10.1038/s43247-022-00488-5

Peng, S., Lin, X., Thompson, R.L. et al. Wetland emission and atmospheric sink changes explain methane growth in 2020. Nature 612, 477–482 (2022). https://doi.org/10.1038/s41586-022-05447-w

Rigby, M., Montzka, S.A., Prinn, R.G., White, J.W.C., Young, D., O'Doherty, S., Lunt, M.F., Ganesan, A.L., Manning, A.J., Simmonds, P.G., Salameh, P.K., Harth, C.M., Mühle, J., Weiss, R.F., Fraser, P.J., Steele, L.P., Krummel, P.B., McCulloch, A., Park, S., 2017. Role of atmospheric oxidation in recent methane growth. Proc Natl Acad Sci USA 114, 5373–5377. https://doi.org/10.1073/pnas.1616426114

Saunois, M., Stavert, A. R., Poulter, B., Bousquet, P., Canadell, J. G., Jackson, R. B., Raymond, P. A., Dlugokencky, E. J., Houweling, S., Patra, P. K., Ciais, P., Arora, V. K., Bastviken, D., Bergamaschi, P., Blake, D. R., Brailsford, G., Bruhwiler, L., Carlson, K. M., Carrol, M., Castaldi, S., Chandra, N., Crevoisier, C., Crill, P. M., Covey, K., Curry, C. L., Etiope, G., Frankenberg, C., Gedney, N., Hegglin, M. I., Höglund-Isaksson, L., Hugelius, G., Ishizawa, M., Ito, A., Janssens-Maenhout, G., Jensen, K. M., Joos, F., Kleinen, T., Krummel, P. B., Langenfelds, R. L., Laruelle, G. G., Liu, L., Machida, T., Maksyutov, S., McDonald, K. C., McNorton, J., Miller, P. A., Melton, J. R., Morino, I., Müller, J., Murguia-Flores, F., Naik, V., Niwa, Y., Noce, S., O'Doherty, S., Parker, R. J., Peng, C., Peng, S., Peters, G. P., Prigent, C., Prinn, R., Ramonet, M., Regnier, P., Riley, W. J., Rosentreter, J. A., Segers, A., Simpson, I. J., Shi, H., Smith, S. J., Steele, L. P., Thornton, B. F., Tian, H., Tohjima, Y., Tubiello, F. N., Tsuruta, A., Viovy, N., Voulgarakis, A., Weber, T. S., van Weele, M., van der Werf, G. R., Weiss, R. F., Worthy, D., Wunch, D., Yin, Y., Yoshida, Y., Zhang, W., Zhang, Z., Zhao, Y., Zheng, B., Zhu, Q., Zhu, Q., and Zhuang, Q.: The Global Methane Budget 2000–2017, Earth Syst. Sci. Data, 12, 1561–1623, https://doi.org/10.5194/essd-12-1561-2020, 2020.

Stavert, A. R., Saunois, M., Canadell, J. G., Poulter, B., Jackson, R. B., Regnier, P., Lauerwald, R., Raymond, P. A., Allen, G. H., Patra, P. K., Bergamaschi, P., Bousquet, P., Chandra, N., Ciais, P., Gustafson, A., Ishizawa, M., Ito, A., Kleinen, T., Maksyutov, S., … Zhuang, Q. (2021). Regional trends and drivers of the global methane budget. Global Change Biology, 28, 182– 200. https://doi.org/10.1111/gcb.15901

Turner, A. J., Frankenberg, C., Wennberg., P. O., and Jacob, D. J.: Ambiguity in the causes for decadal trends in atmospheric methane and hydroxyl, Proc. Natl. Acad. Sci., 114 (21) 5367-5372, (2017), https://doi.org/10.1073/pnas.161602011

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Zhen Zhang, Benjamin Poulter, Sara Knox, Ann Stavert, Gavin McNicol, Etienne Fluet-Chouinard, Aryeh Feinberg, Yuanhong Zhao, Philippe Bousquet, Josep G Canadell, Anita Ganesan, Gustaf Hugelius, George Hurtt, Robert B Jackson, Prabir K Patra, Marielle Saunois, Lena Höglund-Isaksson, Chunlin Huang, Abhishek Chatterjee, Xin Li, Anthropogenic emission is the main contributor to the rise of atmospheric methane during 1993–2017, National Science Review, Volume 9, Issue 5, May 2022, nwab200, https://doi.org/10.1093/nsr/nwab200

Reviewer #3 (Remarks to the Author):

This manuscript presents an analysis of the temporal development of atmospheric methane concentration from 1970 to 2020 (this temporal window sometimes changes), with a focus on the impacts of anthropogenic methane emissions, OH, and wetland emissions using a box model. Although the discussion is comprehensive, the manuscript lacks consistency and clarity in both the methodology and structure. Many aspects influencing the atmospheric methane concentration are discussed, however, the discussion is somewhat scattered. It is not clear what the novel contribution of this study is or how it compares to existing synthesis studies. Therefore, I recommend that the authors improve the logical flow and organization of the contents.

The methodology lacks consistency. The paper makes use of existing simulations (which is great); however, in the way it is currently presented, it lacks a cohesive framework. Simulations often cover different temporal periods making the interpretation very challenging. It may be helpful to focus on the same time period and remove some unnecessary distractions.

For the impacts of anthropogenic methane emissions on the year-to-year methane growth rate (termed as methane trend in the paper, but it is a year-to-year concentration increase), an ensemble of emission inventories is used with a one-box model that has a fixed emission to mixing ratio conversion factor over the five decades. The key quantity being discussed in this paper is the methane growth rate, which is strongly influenced by atmospheric transport, and hence a box model with fixed emission to mixing ratio conversion under the hypothesis of fully mixing is problematic. Please refer to Pandey et al., 2019 in GRL for more discussion. For instance, the 3D simulations for OH using dynamic meteorology result in much larger interannual variations. I consider this a very important limitation of the study. At least, sensitivity tests using 3D chemistry transport models are

needed to illustrate the impact of dynamics.

For the impacts of changes in OH and methane lifetime, the authors used a chemistry transport model (OsloCTM3) to simulate changes in OH concentration covering the period from (a) 1850-2020 with fixed meteorology and (b) variable meteorology from 1997-2017. Then, a scaling factor is applied to the methane lifetime in the box model to simulate its impacts on the methane growth rate.

For the impacts of wetland emissions, CLM5.0 is used using different meteorological drivers and different parameters for sensitivity tests. Here, the simulations cover the period of 2008 to 2017 (Fig. 4). And yet, in Figure 5, the temporal coverage is from 1990 to 2020. Similarly, the expected influences on methane growth rate is simulated using the box model. All factors are then added together to estimate the overall impact on the growth rate.

The modeled changes in δ13CCH4 due to changes in OH shown in the discussion are interesting. I think the paper could be significantly improved if it identified clearer objectives and structure the discussion around these key points.

Technical notes:
Line 13: define “recent”
Line 50: define “recent” (it is often confusing what “recent” means in the paper as they appear to refer to different periods)
Line 77-87: while the authors lay out the current understanding of the methane issue, it is not apparent what new contributions this paper is going to bring.
Figure 1. It is more appropriate to term it as “growth rate” since a year-to-year increase rate is analyzed here
Line 305: Fig. 6b?

Below is our point-by-point response to the reviewers. The original reviewer's comments are in black and our response in green.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

A thorough examination of the drivers of change in the global methane budget is presented using atmospheric chemical transport models, box models and emission inventories. The study is very thorough and presents some useful results. I recommend it for publication in Communications Earth and Environment, provided that several relatively minor issues are addressed.

We would like to thank the reviewer for the thorough and careful review of the manuscript.

General comments

While the authors cite a commendable amount of literature, a more complete discussion of their findings in relation to some publications is needed in parts. For example:
- I felt that the work of Nicely et al. (2017; 2018) should be cited and discussed as it describes very similar model experiments but reaches somewhat different conclusions.

The results of the work by Nicely is now discussed in the Discussion: "Nicely et al 2018 considered various factors affecting tropospheric OH and found the oxidizing capacity to be steady since 1980 but note that the effect of NOx was based on model results using the RCP emission inventory."

- The findings of this work appear to have some similarities to the inverse modelling-based findings of Rigby et al. (2017) and Turner et al. (2017), but these studies are only cited as part of a general introduction on previous methods.

These studies are now referred to in the Discussion: "The model results using the CEDS-2017 emission inventory points at an important contribution of changes in OH via anthropogenic NOx and CO emissions to the methane stabilization period. Several previous studies have indicated an important role of an increased methane sink for the methane stabilization using observationally derived OH concentration (Turner et al., 2017;McNorton et al., 2016;Rigby et al., 2017) without presenting a physical explanation of the OH trend."

- Studies such as Lan et al. (2022) discuss 13CH4 trends using box models, reaching somewhat different conclusions to those presented here.

We have added a couple of sentences on the Lan et al. study in the Discussion section:
"On the other hand, Lan et al. (2021) did not find an influence of OH on the $\delta^{13}C_{CH4}$ decrease, but they only explored a scenario with negative OH trend after 2006.
...
The comprehensive analysis of modelled versus observed $\delta^{13}C_{CH4}$ by Lan et al. (2021) identified a few scenarios that could explain the renewed methane growth after 2006, and these involved different combinations of increased wetland emission, more moderately increasing fossil fuel emissions, decreases in biomass burning emissions and/or a significant decrease in soil sink."

While I certainly sympathise that the literature is vast and growing rapidly, I felt that some of the advances claimed here have been discussed elsewhere (at least in part), and so a slight broadening of the discussion is warranted.

The discussion section is broadened, and the results presented here are put in context to previous published studies as the ones listed above.

Specific comments

L11 and throughout: the term "temporal development" is used extensively in this manuscript. While I'm not sure whether it's technically incorrect, it is an extremely uncommon way of describing a trend, or temporal variability. I suggest replacing the use of this term with "trend", "temporal variability", "evolution", "changes", or similar, depending on the appropriate context.

The term "temporal development" is replaced by different terms in the text.

L19 and elsewhere: the use of "1/3" here sounds too precise. I'd suggest "approximately one third"

Suggestion accepted. The box model results are however corrected, as previously the increase in annual mean methane concentrations was calculated, and not the growth over the year. The effect due to COVID is then enhanced to "approximately two third". The growth rate from the NOAA network is also slightly revised downwards.

L22: It is ambiguous and potentially misleading to describe methane as the "second strongest" GHG. E.g., this could refer to its GWP, in which case the statement would be wrong. Re-word to clarify that this is in reference to its contribution to anthropogenic radiative forcing of climate.

We have replaced the sentence by the following:

"Methane is the second strongest contributor to anthropogenic greenhouse gas radiative forcing after CO_2 (Forster et al., 2021)"

L23: Presumably "global mean concentration"? Furthermore, I don't believe that we know this number to 0.1 ppb, or the percentage increase to the nearest 1%.

We have replaced the sentence by the following, adding more up to date concentration values from WMO: "The global mean concentration reached 1908 parts-per-billion (ppb) in 2021(WMO/GAW, 2022), more than 160% higher than the pre-industrial level (year 1750) of methane and this increase is largely driven by anthropogenic activities (Canadell et al., 2021)."

L25: Delete "levels of": "The current atmospheric concentrations are…". However, it's not clear what "higher than in scenarios consistent with limited global warming" means. I suggest deleting this part of the sentence.

We have deleted "levels of" and rephrased: "The current atmospheric concentrations are higher than in scenarios consistent with limiting global warming to 1.5 and 2 degrees (Nisbet et al., 2019), and several studies point to the importance of reducing atmospheric methane to meet the temperature goals of the Paris Agreement(Ganesan et al., 2019;Collins et al., 2018;Fletcher and Schaefer, 2019;Nisbet et al., 2019)."

L26: "point to", rather than "point at"

Corrected.

L29 – 30: I have two problems with this sentence: a) you can't evaluate future projections, b) I

don't know what "reaching the goal in the Paris Agreement" means. Which goal? I think you can delete this sentence.

We have deleted future projections and added the "temperature" to the goal in the Paris Agreement: "To better evaluate mitigation efforts in view of reaching the temperature goals in the Paris Agreement(UNFCCC, 2015), an improved understanding of the past methane trends is crucial (Ganesan et al., 2019)."

L32: I don't think the semicolon is needed in this sentence.

Deleted.

L34: suggest rewording to: "Between 2000 and 2007, atmospheric methane concentrations were relatively stable, but from 2007 it started rising again, with a further acceleration in 2014".

Replaced with the suggestion.

L34: I suggest citing Dlugokencky et al. (2009) and Rigby et al. (2008) for the description of the renewed methane growth.

References added.

L39: "consists", rather than "consist"

Corrected.

L44: "exceed", rather than "exceeds"

Corrected.

L50: Delete "Also"

Deleted.

L56: What is the role of CO2 here? Is this via its influence on temperature?

The role of CO2 here is the increase in net primary productivity and heterotrophic respiration. We have added to the text: "and atmospheric CO_2 that consequently increase net primary productivity and heterotrophic respiration (Arora et al., 2018)"

In addition, we have added this here: "A strengthening of the wetland methane feedback, an increase in wetland emissions driven by temperature and precipitation increases, is of concern as the climate is changing (Nisbet et al., 2019). Using one wetland model Zhang et al. (2023) found that wetland emissions had increased over the last two decades driven by climate change." These results are also added to the box-models.

L58: "influences", rather than "influence"

Corrected.

L68: Other methods, or species have been used to estimate [OH], e.g., Wolfe et al. (2019), Montzka et al (2011) (i.e., it's not just CTMs or methyl chloroform)

Added the following:
"inverse methods using methyl chloroform or other components (Bousquet et al., 2005;Montzka et al., 2011;Rigby et al., 2017;Turner et al., 2017;Patra et al., 2021;Thompson et al., 2018;Manning et al., 2005)" The focus here is on trends, and the method used by Wolfe et al 2019 is not used to estimate the trend, and is hence not added.

L70: "… THE OH trend IS large…"

Corrected.

L83: this needs rewording. Perhaps "as a consequence of COVID-19 lockdowns…"

We have rewritten:
"The possible influence on the high growth rate of atmospheric methane in 2020 due to emission reductions of NOx and CO as a consequence of COVID-19 lockdowns are assessed."

L85: "investigated", rather than "presented"?

Suggestion accepted.

L98: I assume the NOAA data needs to be acknowledged with an appropriate literature citation, rather than a URL?

A citation is added rather than the URL.

L118: Somewhere here, say where the observed trends are taken from.

Added reference at L127.

L126: "are not able to reproduce the observed atmospheric trend…" This needs to be softened. No model will ever be able to fully reproduce the trend, but there are some features in common.

This is rewritten as: "The box model calculations using the CEDS-2017 emission inventory and a SSP scenario (CEDS-2017 in Fig. 1a, see Method) give an increase in methane that is too small prior to 1990 and too strong post 2005 compared to the observed growth rate."

Further down we have now written:
"A single anthropogenic emission inventory alone will never be able to reproduce the observed trend in atmospheric methane."

L137: "also seen as stable anthropogenic emissions in the 1980s continuing up to year 2000". This needs rewording, as I don't know what it means.

Replaced by:
"and this can also be seen directly from the relatively stable CEDS-2021 emissions in the 1980s and 1990s (Fig. 2)."

L141: "gives AN atmospheric trend…"

Corrected.

L146: I suggest “after the stabilization…”

Suggestion accepted.

L173: “and up to 1980-90 of 3%”. It’s not clear what this means. Reword.

We have replaced 1980-90 with 1990.

L176: some commas would help here: “and longer, respectively, as in 1850. From 1990, OH…”

Commas added.

L183: “declines”, rather than “decline”

Corrected.

L204: “results, also indicating a role for the emissions”, rather than “results, that also indicate”. However, you also need to say which emissions here (CO, NOx, CH4?)

Added specifications of emission components at L200 “emissions of chemically reactive components”. Suggestion for L204 accepted.

L205: what is meant by “OH development”?

Replaced by “OH time evolution”.

L206: When translating the 3D CTM results to the box model, how do you account for the influence of the varying CH4 emissions in the 3D model? Wouldn’t there be a non-linear response on the methane lifetime from these methane emissions variations?

In the method section it is written: “The CTM is run with prescribed surface concentrations of methane (Søvde et al., 2012) scaled to the historical global mean methane concentrations (Meinshausen et al., 2017) for the year simulated.” So the influence of the historical change in methane concentration is accounted for in the OH and methane lifetime results. Also, the multi-model initiatives are driven by prescribed surface concentration of methane.

L214 - 215: “Changes in the OH sink have likely contributed to the stabilization…”. Similar conclusions were reached (albeit with very large uncertainty) by Turner et al. (2017) and Rigby et al. (2017). It seems these papers should be cited here and/or in the discussion.

As stated above we have included a discussion of our results in relation to these results in the discussion.

L217: The caption and legend for Figure 1 suggests that only the CEDS21+COVID simulation was used, but here it seems to suggest that CEDS17 was used prior to 2000? Apologies if I’ve misread. In any case, why was a change in simulation needed? What would be the effect of this change?

For the results highlighted in Fig. 1 (CEDS21+COVID), the time evolution of methane lifetime had to be extended back in time as the OsloCTM3 simulation with CEDS21 emissions was only performed from year 2000. The trend in NOx/CO is similar in CEDS21 and CEDS17 prior to year 2000. In the methods section it was written: “The OsloCTM3 CEDS21 is extended back in time from 2000 using the CEDS17 results”

In the result section it is now written "After year 2000 the lifetime is adjusted based on the CEDS21+COVID simulation and before year 2000 the lifetime is adjusted based on the CEDS17 simulations, as the trend in the emissions used in these simulations are similar before year 2000 (Fig. S8)."

L220: Should this be -3.2 ppb/yr? The use of "negative" in this sentence confuses matters.

The sentence is removed to focus on the latter part of the period and the COVID response here.

L222: "COVID emission reductions": need to say for which species here, as I don't think you mean for CH4 (rather, NOx/CO)?

Added "of NOx and CO" after reduction.

L222: "contributing", rather than "contributed"?

Corrected.

L236: "show" rather than "span"?

Corrected.

L242: perhaps it's the relatively low-resolution version in the document that I have, but it's not clear to me where the boxes overlap. I'm therefore not sure why the caption says this.

In the first column, the boxes that are plotted are next to each other with no white space in between. The colors are not transparent, so you are not able to see that they overlap. That is why the sentence in the caption is added. We have rephrased the sentence to: "Note that some of the coloured boxes overlap."

L262: "were", rather than "was"?

Replaced "wetland areas" with "wetland area" and kept was.

L265: "results to the", rather than "as the"

Corrected.

L266: "does" rather than "do"

Corrected.

L272: What are the fluxes from non-inundated areas?

Fluxes from non-inundated areas, are methane fluxes that are calculated in the CLM model from the area of the gridbox that are not specified as inundated area (wetland area).

We have rewritten: "while the net emissions that in addition to wetland emissions, defined as emission fluxes in the inundated fraction of the grid cell in the CLM, include fluxes from the non-inundated areas and soil sink,"

L275: something like: "… datasets to drive the model ARE SHOWN"? Also, it should be "indicates", rather than "indicate".

Corrected.

L288: “is seen”, rather than “can be expected”?

Suggestion accepted.

L293: Again, what are the emissions from non-inundated areas?

This is now clarified. See response above.

L305: Fig 5b, rather than 6b?

Corrected. Also on L300.

L325: “negative anomaly”, rather than “spike”?

Suggestion accepted.

L326: “growth rate”, rather than “trend” (which is non-specific)

Replaced trend with growth rate

L340: Is “Solazzo” reference to a publication, or personal communication, or something else?

This is now rewritten. The range was provided to me by Solazzo, as the range was not explicitly written in the publication. The range is explicitly written out in Minx et al. (2021) where Solazzo is a co-author, so now we just refer to Solazzo and add the uncertainty range provided by Minx et al as well:

“to be is -33% to +46% (at 2σ) for total anthropogenic methane emissions (Solazzo et al., 2021), and based on best value judgement Minx et al. (2021) narrowed the uncertainties to ±30 % (90% confidence interval), corresponding to 263 to 489 Tg (for 2018 in EDGARv7, Fig. 2).”

We have added this uncertainty range to Figure 2 as well.

As EDGARv7 is available, EDGARv6 is now replaced by EDGARv7, which is an extension of EDGARv6 from 2018 up to 2021.

L344: “The COVID lockdown did not affect methane the most compared to other emission components”. I don’t know what this means. It’s a very strong statement though. Needs rewording.

As EDGARv7 is included, this section is rewritten:
“Only the EDGARv7 inventory include emission in year 2020, when observed methane growth rate hits a new record (Fig. 1). Driven by the agricultural and waste sector, the EDGARv7 emissions increased by 1% from 2019 to 2020, while other estimate a weak decrease in anthropogenic methane emissions due to COVID lockdowns (Forster et al., 2020). McNorton et al. (2022) found an emission increase in 2020…”

L355: emissions of which species?

Added “…emission inventory of chemically reactive gases”

L355: “OH variability”, rather than “OH development”?

Replaced.

L354 – 356: I don’t really understand what is being claimed here. Why does this indicate that the emission inventory is important, since the previous sentence just states that the AerChem and Oslo models had similar OH trends?

AerChemMIP models and OsloCTM were driven by the same emission inventory and had similar OH trends.

Rewritten as: “As these models were driven by the same emission inventory of chemically reactive gases, this indicates that the emission inventory used are important for the OH variability, as also pointed out by Stevenson et al. (2020) for AerChemMIP.”

L367: A reference is needed here

The reference to Dalsøren, et al. is in the sentence before this (but separated by a figure), and we have moved these sentences together.

L372: “After the methane stabilization…”

Suggestion accepted.

L383: “… less than a factor of two…”

Corrected.

L388: “anomaly”, rather than “spike”?

Suggestion accepted.

L402: “Can an increase in OH from the 1980s to early 2000s using CEDS-2017 emissions…”, rather than “Can an increase in OH from the 1980s to early 2000s as the models show using CEDS-2017 emissions…”? This sentence should be reworded in any case, ideally not having the figure reference in the middle.

This part has been rewritten.

L406 – 415: I think that Rigby et al. (2017) show similar conclusions (e.g., Figure S6 in their paper).

Thank you for making us aware of the result from Rigby et al. We have now added the following sentence to the Discussion section:
“Rigby et al. (2017) also used a box model to show that evolving OH concentrations, inferred from methyl chloroform (CH_3CCl_3) observations by inversion, could explain some of the decline in $\delta^{13}C_{CH4}$ (their Fig. S6), but their inferred OH concentration had both an increase until around 2005 and a decrease thereafter.”

L446: as above, “1/3” seems too precise. “approximately one-third”?

Rewritten.

L449: What is meant be “components” here? Do you mean “atmospheric constituents”? Or perhaps just “factors”

Rewritten: “and mitigate atmospheric constituents that influence methane indirectly by affecting the methane lifetime such as CO.”

L450: I suggest deleting this sentence. It’s just one factor that could influence CH4 lifetime, and it seems very out of context as a concluding line here.

Sentence deleted.

L465: sets, rather than “set”.

Corrected.

L579: Isn’t [OH] a bigger source of uncertainty in the methane budget?

Rephrased: “and an important source of uncertainty”

Additional references:

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Reviewer #2 (Remarks to the Author):

Review of "Methane trend over the last decades driven and modified by anthropogenic emissions" by Skeie et al.

This manuscript explores the drivers of the trends and shorter-term variability of atmospheric methane (CH4) over recent decades through evaluation of some of its major sources and sinks over that time period. The main conclusion is that anthropogenic emissions dominate both the long-term trends of methane and much of the variation in these trends, with smaller impact from natural sources. The anthropogenic contribution to CH4 variation is both direct, through emission of CH4 into the atmosphere, and indirect – changing the methane lifetime due to human-influenced variations in other species in the atmosphere. Finally, and most interestingly, the authors investigate the possibility that delayed influence of increased atmospheric hydroxyl (OH) concentrations might have influenced the declining isotopic ratio of atmospheric CH4 in the past decade.

Generally, the work is very well-written with clear and appropriate figures, whilst the methods are suitable and the authors have combined a range of datasets to investigate their hypotheses. I did not come across many major issues regarding the clarity, accuracy and appropriateness of the work that is demonstrated here.

However, my main issue is that many of the early findings of the paper are not convincingly novel. Previous groups have shown that anthropogenic emissions have been responsible for much of the rise in CH4 over previous decades (e.g. McNorton et al. (2018); Turner et al. (2019); Stavert et al. (2021); Zhang et al. (2021)), whilst others have proposed and assessed the effect of increased OH concentrations during the slowdown period for CH4. (e.g. McNorton et al. (2016); Turner et al. (2017); Rigby et al. (2017)). There is mixed evidence for stable or changing global wetland and/or natural emissions. (e.g. Melton et al., (2013); Saunois et al. (2020); Oh et al., (2022)). Others have quantified the effect of methane lifetime on the growth rate during the 2020 Covid pandemic. (e.g. Peng et al. (2020)) The strength of this work relative to these others is perhaps in bringing these strands together within one model, and covering a relatively long time period. However, in my opinion, to be considered for publication the authors need to explain much more clearly why their results and/or methods are innovative, and not simply an update on or confirmation of the findings of others.

Thank you for your positive and constructive review. At the end of the introduction section as well as in the discussion section, we have tried to explain more clearly the innovative aspects of our work. In the discussion section we have put our results in context to previous work. For example, the previous studies on the role of OH on the slowdown of methane growth in the 1990s and early 2000 did not give any physical explanation for the changes in OH. We also highlight the uncertainties in the anthropogenic NOx and CO emission inventories driving the OH changes in the models, as well as the uncertainties in anthropogenic methane emissions. We have also added wetland model results from Zhang et al 2023, who find increased methane emissions over the last two decades to the box models. Bringing all the three main components in the methane budget together, from a bottom-up perspective, is a strength of the paper as you mentioned.

One respect in which I did find their results to be novel, but maybe a little underdeveloped, is in the idea that the prolonged effects of OH variations on the isotopic ratio of 13CH4/12CH4 in the atmosphere can explain at least some of the recent observed variations in this metric. The results shown in Figure 7 are interesting, but the discussion is quite brief. The authors can explain some 30% of the observed decrease in ∂13CCH4 due to this effect. However, the variation shown by the model is much smaller and slower than the observations, so some discussion on why that might be is appropriate. The following questions could be answered:

• If there is indeed no trend in natural emissions, what might account for the rest of the decrease, particularly since the observed changes are much more sudden?
• Is it possible to explore this question further with the box model, including and perturbing the isotopic signatures for different emissions?
• If not, or if results are inconclusive, what data is needed for further exploration of this isotopic variation? What changes in sources or sinks would be needed to produce changes in ∂13CCH4 of the magnitude observed?

If this aspect of the paper was further developed in this way and the novelty of earlier results was emphasised, then I think that the paper could be considered for publication here.

This part of the paper is further developed. We have moved the isotopic box-model results to a separate section at the end of the Results section of the paper. In addition to the effect on the isotopic ratio of OH, the response of wetland and four different anthropogenic emission sectors on the isotopic ratio are included. Sensitivity simulations have also been performed to explore the effects of uncertainty in the isotopic signature of each emission sector. The results are shown in Fig. 6 (previously Fig. 7) and the associated discussion has been expanded.

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Reviewer #3 (Remarks to the Author):

This manuscript presents an analysis of the temporal development of atmospheric methane concentration from 1970 to 2020 (this temporal window sometimes changes), with a focus on the impacts of anthropogenic methane emissions, OH, and wetland emissions using a box model. Although the discussion is comprehensive, the manuscript lacks consistency and clarity in both the methodology and structure. Many aspects influencing the atmospheric methane concentration are discussed, however, the discussion is somewhat scattered. It is not clear what the novel contribution of this study is or how it compares to existing synthesis studies. Therefore, I recommend that the authors improve the logical flow and organization of the contents.

The methodology lacks consistency. The paper makes use of existing simulations (which is great); however, in the way it is currently presented, it lacks a cohesive framework. Simulations often cover different temporal periods making the interpretation very challenging. It may be helpful to focus on the same time period and remove some unnecessary distractions.

Thank you for your review comments. A strength of this study is to combine a range of results for three of the main components in the methane budget. As emission inventories and input data needed for model simulations do not all cover the same time period, the results presented here

will cover different time periods. However, in the box model, the different results are combined and the same time period is considered. In the method section it is clearly written how the anthropogenic emissions are combined and extended, as well the adjustments of the methane lifetime. This is now clarified for the anomalies in natural emissions.

We have also restructured the manuscript, so that the results from the isotopic box model have a separate section in the results. The last part of the introduction is slightly modified and, in the discussion, the results are more discussed in relation to previous studies to highlight the novelty of this study (see also response to the other reviewers).

For the impacts of anthropogenic methane emissions on the year-to-year methane growth rate (termed as methane trend in the paper, but it is a year-to-year concentration increase), an ensemble of emission inventories is used with a one-box model that has a fixed emission to mixing ratio conversion factor over the five decades. The key quantity being discussed in this paper is the methane growth rate, which is strongly influenced by atmospheric transport, and hence a box model with fixed emission to mixing ratio conversion under the hypothesis of fully mixing is problematic. Please refer to Pandey et al., 2019 in GRL for more discussion. For instance, the 3D simulations for OH using dynamic meteorology result in much larger interannual variations. I consider this a very important limitation of the study. At least, sensitivity tests using 3D chemistry transport models are needed to illustrate the impact of dynamics.

Some modifications to the text replacing trend with growth rate or year-to-year concentration increase are done.

The study by Pandey et al, highlights especially the importance of transport when looking at interhemispheric differences in methane growth rate. In this study we use a one-box model, and we have added a section on box-model limitations in the method section:

"The box model is used for illustration of the effect on year-to-year increase in atmospheric methane and the relative role of anthropogenic emissions, changes in oxidation capacity of the atmosphere and wetland emissions and does not take into account complex interaction of chemistry, dynamics and spatial and temporal variability in emissions."

Limitations in the simple box models for isotopes were also added to the method and discussion section, respectively:

"The limitations mentioned above for the methane box model apply also here for the isotopic box model. In addition, an important limitation is that one global mean value for the isotopic signature is used for each emission sector, while in reality these values differ in space and partly also in time."

"It should be noted that box models have important limitations. Our isotopic box model is based on global and annual means and does not account for the highly variable OH concentrations in both time and space, nor the inhomogeneous distribution of the isotopic ratio and emission signatures. Further studies are needed using 3-D models to better resolve and understand the influence of OH and its temporal evolution on the δ13CCH4 evolution."

For the impacts of changes in OH and methane lifetime, the authors used a chemistry transport model (OsloCTM3) to simulate changes in OH concentration covering the period from (a) 1850-2020 with fixed meteorology and (b) variable meteorology from 1997-2017. Then, a scaling factor is applied to the methane lifetime in the box model to simulate its impacts on the methane growth rate.

For the impacts of wetland emissions, CLM5.0 is used using different meteorological drivers and different parameters for sensitivity tests. Here, the simulations cover the period of 2008 to 2017 (Fig. 4). And yet, in Figure 5, the temporal coverage is from 1990 to 2020. Similarly, the expected influences on methane growth rate is simulated using the box model. All factors are then added together to estimate the overall impact on the growth rate.

For clarification, in Fig. 4 we have moved the text line describing the error bar. In the caption it is now written: "Coloured boxes show the annual mean values from the CLM simulations for the period 2008 to 2017 (or the end year of the simulation, see Method) compared to the range of reported studies in The Global Methane Budget (GMB) for the 2008-2017 decade from top-down inversions models and bottom-up models represented by the error bars." The reason why a different time period was plotted in Fig 4 compared to Fig 5, was to compare the emission with emission ranges from GMB. Hope this make it clearer.

The modeled changes in δ13CCH4 due to changes in OH shown in the discussion are interesting. I think the paper could be significantly improved if it identified clearer objectives and structure the discussion around these key points.

To highlight the results of δ13CCH4 due to changes in OH, this is moved to a separate section at the end of the Results section.

Technical notes:
Line 13: define "recent"

Line 50: define "recent" (it is often confusing what "recent" means in the paper as they appear to refer to different periods)

Throughout the manuscript we have tried to clarify or rewrite when "recent" was used. Eg. On line 50 it is rewritten as "For the increase in atmospheric methane since 2007".

Line 77-87: while the authors lay out the current understanding of the methane issue, it is not apparent what new contributions this paper is going to bring.

In this part we have added the time dependent isotopic response of OH as well as highlighted that we consistently assess all of the three largest terms in the methane budget.

Figure 1. It is more appropriate to term it as "growth rate" since a year-to-year increase rate is analyzed here

We have replaced the y-label with "Methane growth rate", as well as included "growth rate" and "annual methane mole fraction increase" in the figure legend.

Line 305: Fig. 6b?

This is corrected.

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23rd Jun 23

Dear Dr Skeie,

Your manuscript titled "Methane trend over the last decades driven and modified by anthropogenic emissions" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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If you have any questions or concerns about any of our requests, please do not hesitate to contact me.

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Joshua Dean, PhD
Editorial Board Member
Communications Earth & Environment

Clare Davis, PhD
Senior Editor
Communications Earth & Environment

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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have done a very good job of addressing the reviewer's comments. I am satisfied that the manuscript is suitable for publication, following some minor additional suggestions:

- The newly added line: "Only the EDGARv7 inventory include emission in year 2020..." should be "includes", not "include"

- The new results section on the influence on 13CH4 is a good addition. However, I'd consider renaming it to be more specific. E.g., "Influence on 13C-CH4 trends".

- Thank you for including references to Lan et al and Rigby et al in the discussion of the 13C-CH4 trends. I feel that both of these studies should appear somewhere in the newly added results section. Furthermore, given that these studies were also both essentially box model studies, isn't it too strong a statement to say that "We make a first attempt to investigate this by using an isotopic box model (see Method)"?

Reviewer #2 (Remarks to the Author):

Review of updated manuscript for “Methane trend over the last decades driven and modified by anthropogenic emissions” by Skeie et al.

This manuscript has been significantly updated and improved since the previous submission. The authors did well in incorporating updates based on my previous comments, emphasising and explaining the novelty of their work. They have also expanded the section that investigated observed the isotopic signature of 13δCH4 and its relationship with various terms in the methane budget, particularly loss through reaction with hydroxyl radicals.

Based on these improvements, I can recommend this work for publication if the following minor and technical corrections are made.

Specific comments

L19 and throughout: change “two third” to “two thirds”.

L21: Change to “Methane (CH₄) is the…”

L22: Change to “…carbon dioxide (CO₂).”

L29: Change to “understanding of past methane trends”

L30: “reasons for the methane trend” should be changed to something like “reasons for the variations in the observed methane trend”

L34-35: Change to something like “Large annual increases equivalent to those observed in 2020 and 2021 (15.2 and 17.8 ppb, respectively) have not been seen since the 1980s”.

L40: wetland -> wetlands

L41: Change to “The main sink of methane is through chemical reactions…”

L62: to represent -> in representing

L76: “important role for the OH trend”

L85: What does “consistently” mean here? Consider deleting?

L94: “Jointly changes…” -> “Joint changes…”

L110 (Figure caption 1): “In the supplementary” -> “In the supplementary material…”

L126 (Table caption 1): Insert space in SupplementaryTable

L194: occur -> occurs

L197: change to “prior to the year 2000.”

L199 – 201: I took me a long time to follow this. I now understand that 2020 emissions were produced by scaling the emissions that were available for 2019, but as written this is not clear. Please try to clarify this sentence.
L235: “In line with…” – I don’t think that a fraction of two thirds is “in line” with one of “up to one half” from Stevenson et al., or even really with the fraction found by Peng et al.’s study. Personally, I would find “larger than” more appropriate than “in line with”.

L257: “GMB” should be defined in the main text when first used, and not only in the figure caption.

L260 – L262: This sentence is difficult to follow.

L296: Clarify the sentence that starts “The general pattern in the time development is…” as its meaning is currently unclear to me. In particular, remove use of “time development”.

L364: extra ‘.’ before Stell et al.

L379: Change to “whether from wetlands, ruminants, waste or all…”.

L435: Change to “whilst others estimate”

L448: Change to “methane, CO and other compounds”

L451: inventory -> inventories

L515: Change to: “There is no trend”

L519: Change to “wetland emissions”

Reviewer #3 (Remarks to the Author):

I am very pleased to see that the manuscript has greatly improved in terms of readability and clarity. The revised version effectively articulates its main findings in relation to recent literature, examining key factors influencing the changes in methane growth rate and the 13CH4, including anthropogenic emissions (from various sectors), wetland emissions, and OH sink. The commentary on the legacy effect of OH change on the isotopic ratio is interesting. I only have a few minor suggestions.

Line 1: “Last decades” is rather vague. It will be helpful to specify which period this paper is addressing. If not in the title, it should at least be stated in the abstract (line 12).

Line 64: some global inversions also suggest an increase in tropical wetland emissions, see Yin et al., 2021, ACP.

Line 98: I suggest replacing "trend" with "growth rate" in this context (and in similar instances). I recommend using "growth rate" for annual changes and "trend" for consistent changes over several years.

Fig. 1b To avoid confusion, consider using dots and dashed lines for the OsloCTM3 CEDS21+COVID

time-slice runs (as in Fig. 3). It is not clear from the figure caption whether the shaded area represents different OsloCTM3 runs that participate in CCMI and AerChemMIP? Or are they model ensembles? From the text, it seems to be the model ensemble. It will be good to clarify.

Fig. 5 It's not entirely clear what is included in the "net emissions". Could you clarify the relationship between (a) and (b)?

Line 305: “The interannual variability in the wetland emissions are smaller than for the net emissions” I find this statement quite confusing. Which one has a smaller IAV? It seems to be the wetland emissions, not the net, correct?

Line 319: What are the differences in the settings of Zhang et al. that result in different trends in recent years compared to other wetland models? A discussion regarding the key factors either here or later in the manuscript would be helpful.

We would again like to thank the reviewers for their constructive reviews. Below is our point-by-point response to the reviewer's remaining comments. The original reviewer's comments are in black and our response in green.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have done a very good job of addressing the reviewer's comments. I am satisfied that the manuscript is suitable for publication, following some minor additional suggestions:

- The newly added line: "Only the EDGARv7 inventory include emission in year 2020..." should be "includes", not "include"

Corrected in the manuscript text.

- The new results section on the influence on 13CH4 is a good addition. However, I'd consider renaming it to be more specific. E.g., "Influence on 13C-CH4 trends".

Suggestion accepted. It is renamed to: Influence on $\delta^{13}C_{CH4}$ trends.

- Thank you for including references to Lan et al and Rigby et al in the discussion of the 13C-CH4 trends. I feel that both of these studies should appear somewhere in the newly added results section. Furthermore, given that these studies were also both essentially box model studies, isn't it too strong a statement to say that "We make a first attempt to investigate this by using an isotopic box model (see Method)"?

Based on your comment and that the journal does not want the word "first" to be used, we have rephrased the sentence:
"We investigate the isotopic effect of the strong increase in OH prior to the stabilization period by using an isotopic box model (see Method)."
As Lan et al. did not investigate this scenario and Rigby et al did inverse modelling, we do not include a reference to these studies in the results section. With the rephrased sentence I hope this is ok for you.

Reviewer #2 (Remarks to the Author):

Review of updated manuscript for "Methane trend over the last decades driven and modified by anthropogenic emissions" by Skeie et al.

This manuscript has been significantly updated and improved since the previous submission. The authors did well in incorporating updates based on my previous comments, emphasising and explaining the novelty of their work. They have also expanded the section that investigated observed the isotopic signature of 13δCH4 and its relationship with various terms in the methane budget, particularly loss through reaction with hydroxyl radicals.

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Corrected the three occurrences.

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Done.

L22: Change to “…carbon dioxide (CO2).”

Changed to “carbon dioxide.” as it is not used elsewhere in the text.

L29: Change to “understanding of past methane trends”

Done.

L30: “reasons for the methane trend” should be changed to something like “reasons for the variations in the observed methane trend”

Done.

L34-35: Change to something like “Large annual increases equivalent to those observed in 2020 and 2021 (15.2 and 17.8 ppb, respectively) have not been seen since the 1980s”.

Suggestion accepted.

L40: wetland -> wetlands

Done.

L41: Change to “The main sink of methane is through chemical reactions…”

Done.

L62: to represent -> in representing

Done.

L76: “important role for the OH trend”

Done.

L85: What does “consistently” mean here? Consider deleting?

We have deleted the word.

L94: “Jointly changes…” -> “Joint changes…”

Done.

L110 (Figure caption 1): “In the supplementary” -> “In the supplementary material…”

Replaced with a reference to the Supplementary Figures 2-4.

L126 (Table caption 1): Insert space in SupplementaryTable

Done.

L194: occur -> occurs

Done.

L197: change to “prior to the year 2000.”

Done.

L199 – 201: I took me a long time to follow this. I now understand that 2020 emissions were produced by scaling the emissions that were available for 2019, but as written this is not clear. Please try to clarify this sentence.

This is rewritten as: “For the year 2020, the CEDS21 emission for year 2019 is scaled by estimated emission reduction due to policies put in place to combat the COVID19 pandemic (see Method).”

L235: “In line with…” – I don’t think that a fraction of two thirds is “in line” with one of “up to one half” from Stevenson et al., or even really with the fraction found by Peng et al.’s study. Personally, I would find “larger than” more appropriate than “in line with”.

“In line with” replaced by “larger than”.

L257: “GMB” should be defined in the main text when first used, and not only in the figure caption.

Done.

L260 – L262: This sentence is difficult to follow.

This is rewritten: “*Supplementary Figure 6 show the net emissions that in addition to wetland emissions, defined in the CLM as emission fluxes in the inundated fraction of the grid cell, include fluxes from the non-inundated areas and soil sink. The different meteorological data used to drive the model give slightly different wetland fluxes (Fig. 4) and span a larger range for net emissions (Supplementary Figure 6).”*

L296: Clarify the sentence that starts “The general pattern in the time development is…” as its meaning is currently unclear to me. In particular, remove use of “time development”.

The two sentences are now combined: “The general pattern in the time evolution is similar for the CLM model driven by different meteorological datasets, where the global net-emission increased from 1992 to 2000, then increased again from the end of the stabilization period up to 2011 and decreased thereafter (Fig 5a).”

L364: extra ‘.’ before Stell et al.

Corrected.

L379: Change to “whether from wetlands, ruminants, waste or all…”.
Done.

L435: Change to “whilst others estimate”

Done.

L448: Change to “methane, CO and other compounds”

Done.

L451: inventory -> inventories

Done.

L515: Change to: “There is no trend”

Done.

L519: Change to “wetland emissions”

Changed to wetland area.

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I am very pleased to see that the manuscript has greatly improved in terms of readability and clarity. The revised version effectively articulates its main findings in relation to recent literature, examining key factors influencing the changes in methane growth rate and the 13CH4, including anthropogenic emissions (from various sectors), wetland emissions, and OH sink. The commentary on the legacy effect of OH change on the isotopic ratio is interesting. I only have a few minor suggestions.

Line 1: “Last decades” is rather vague. It will be helpful to specify which period this paper is addressing. If not in the title, it should at least be stated in the abstract (line 12).

We have added in the abstract (line12): “…on the methane growth rate over the last *three* decades…”

Line 64: some global inversions also suggest an increase in tropical wetland emissions, see Yin et al., 2021, ACP.

Thank you for pointing at this study. We have added a reference to it: “recent increase in methane[25,26] *and global inversion studies find contribution from tropical wetlands*[27]*,* and regional studies do “

Line 98: I suggest replacing "trend" with "growth rate" in this context (and in similar instances). I recommend using "growth rate" for annual changes and "trend" for consistent changes over several years.

Replaced trend with growth rate in the Figure caption as well as replaced elsewhere in the text.

Fig. 1b To avoid confusion, consider using dots and dashed lines for the OsloCTM3 CEDS21+COVID time-slice runs (as in Fig. 3). It is not clear from the figure caption whether the shaded area represents different OsloCTM3 runs that participate in CCMI and AerChemMIP? Or are they model ensembles? From the text, it seems to be the model ensemble. It will be good to clarify.

The figure is kept as it is, but I have clarified this in the figure caption:

"The green line shows contribution to the growth rate allowing for changes in methane lifetime following the OsloCTM3 CEDS21+COVID, while the green shading indicates the range in contribution from allowing methane lifetime to change following the other OsloCTM3 CEDS17 simulations (with fixed and varying meteorology), CCMI and AerChemMIP results (see Box model section in Method). "

Fig. 5 It's not entirely clear what is included in the "net emissions". Could you clarify the relationship between (a) and (b)?

It is written in the figure caption: "In a) global anomalies for net methane emissions (wetland emissions, fluxes from non-inundated areas and soil sink) and in b) global anomalies for wetland emissions"

We think this is clearer now. In the main text, we have tried to clarify for Figure 4:
"*Supplementary Figure 6 show the net emissions that in addition to wetland emissions, defined in the CLM as emission fluxes in the inundated fraction of the grid cell, include fluxes from the non-inundated areas and soil sink. The different meteorological data used to drive the model give slightly different wetland fluxes (Fig. 4) and span a larger range for net emissions (Supplementary Figure 6)."*

Line 305: "The interannual variability in the wetland emissions are smaller than for the net emissions" I find this statement quite confusing. Which one has a smaller IAV? It seems to be the wetland emissions, not the net, correct?

We have tried to clarify: "The interannual variability in the wetland emissions are smaller than for the net emissions (Fig. 5), as the emissions in the non-inundated areas *included in the net emissions* are more affected by meteorological factors."

Line 319: What are the differences in the settings of Zhang et al. that result in different trends in recent years compared to other wetland models? A discussion regarding the key factors either here or later in the manuscript would be helpful.

One difference compared to the CLM and the wetland models that contributed to the GMB, is that the study of Zhang et al. used a hydroglogical model to determine the wetland area. I have highlighted this in the discussion:
"Using a hydrological model to determine the wetland area, Zhang, et al. [21] modelled an intensification of the tropical wetland"