The Global Methane Budget: 2000-2020

Saunois et al., ESSD, 2024

Detailed Response to Anonymous Referee#2

We acknowledge the referee for the time spent on reading and commenting on the paper. We thank Anonymous Referee #2 for the useful corrections and suggestions 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.

Global Methane Budget needs focus and discernment'

This manuscript represents a lot of work by a large number of co-authors. The work is massive and I admit to being unable to seriously review the entire document. It contains a lot of valuable material plus a number of errors or misguided recommendations. Given the number of authors, it would be nice if more of them spent time carefully reading/editing. I begin to wonder if the massive tomes generated so frequently by the Global Carbon Project are a benefit to the community. I suspect it is too late to raise this, but I do not think this work is ready for publication.

A number of the sections seem to be repeats of the same old ideas and references in Saunois 2020. Why is this not an update of what is new?

We acknowledge the difficulty of reviewing such manuscript due to his length and amount of information and details. Indeed, this manuscript is a living document, updated every 3-4 years to produce a new budget but keeping the foundation of the main text. Such a regular budget is highly expected by the community. The first paper (Kirschke et al., 2013) has been cited 2264 times, the second (Saunois et al., 2016) has been cited 1157 times and the last update (Saunois et al., 2020) has been more cited than the 2016 paper: 1871 times.

Though some parts have been less updated than others – in terms of reference, this budget brings novel insights and latest estimates of methane sources and sinks, and uncertainties that still remain for global emissions and at the individual sector scale. These uncertainties are often overlooked in many individual studies providing a single number for methane emissions. The "repeats" from the previous budget are intentional when the information has not changed much. We believe this helps the reader not to have to read the older budgets to keep up with the background and material in the current budget. However, we agree that some old references remained in the submitted version of the text. Thanks to the constructive reviews received (mainly from reviewer #1) we improved this weakness by providing a more up-to-date review of the current knowledge for most of the concerned paragraph. Other parts may not have been updated enough but were kept for the sake of completeness of the budget.

Section 7 appears to be a personal wish list of the authors' personal research goals, rather than a critical review. Many of the references are to a single paper, often involving one or more of the authors.

Section 7 is not meant to be a critical review of our synthesis. Also, it has been constructed by many of the authors and reflects at least partly, priorities from the methane community. We included papers and studies known by the authors. We improved this part by adding literature and studies that were not cited in the current version.

Abstract:

L115: "maintaining CH4 as…" is incorrect. Even if CH4 emissions remained constant, it would maintain CH4 as the 2nd GHG for a long while!

Indeed. Original sentence : "*Emissions and atmospheric concentrations of CH4 continue to increase, maintaining CH4 as the second most important human-influenced greenhouse gas in terms of climate forcing after carbon dioxide (CO2)"*. This has been rephrased to: "**CH4 is the second most important human-influenced greenhouse gas in terms of climate forcing after carbon dioxide (CO2) and both emissions and atmospheric concentrations of CH4 continue to increase since 2007**"

L117: "importance of CH4 EMISSIONS compared to THOSE OF CO2…" The atmospheric abundance is what it is, and is observed! Here I think you want to emphasize the emissions, not the abundance.

Indeed. Thank you for your comment. This has been corrected.

L119: "in reducing uncertainties in the factors.." is very contorted, try: "in quantifying the factors responsible for the observed atmospheric.." we do not really need 'well' here either.

This has been modified as suggested.

L130: this 2024 edition (help remind the reader of which edition they are reading)

This has been modified as suggested.

L133: either the double counting is "potential" in which case it MAY exist, or it is not 'potential' but an estimate of the REMAINING double accounting.

This has been rephrased to " **the potential double counting that may exist**" .

Introduction

L172ff: No, this is simply incorrect. CH4 is not a stronger absorber in the current atmosphere, there is more CO2 and hence CO2 is the stronger absorber (witness the ERF of each). Anyhow, what you are talking about is GWP>1 meaning stronger impact on climate over the next 100 years per kg emitted. Anyway, this paper does not have the background in radiative transfer and forcing and you should not be talking about 'absorbers', stick to climate forcing and GWPs.

We acknowledge that this was incorrectly written. This part has been rewritten based on climate forcing and GWP only. The revised version now includes: "An **equal mass of CH_4**

emissions has a stronger impact on climate than carbon dioxide (CO_2), which is reflected by its global warming potential (GWP) relative to CO_2 on a given time horizon"

L173ff: It is not 'climate feedbacks' that matter here, but 'climate and chemical composition changes' (not feedbacks!). Chemical feedback are included in the GWP.

The GWP indeed includes indirect chemical effects. We have removed "without considering climate feedbacks" and rephrased to: "**For a 100-yr time horizon the GWP of CH_4 emitted by fossil sources is 29.8 (GWP of CH_4 emitted by microbial sources is 27), whereas the values reach 82.5 over a 20-year horizon for CH_4 emitted by fossil sources and 79.7 for CH_4 emitted by microbial sources (Forster et al., 2021)**"

L174ff: Since you are insisting on quoting all the GWPs for methane (20 & 100 yr, fossil and non-fossil) let us get it right. OK, you are quoting GWPs from last IPCC, but it is about time we correct the maths that were done in Forster et al 2021. I know this is not the job of this CH4 budget review to fix IPCC problems, but we should avoid further propagating errors.
(1) The difference in fossil vs non-fossil GWP is 2.75 based on the molecular wt of CH4 and CO2. IPCC rounds to 2.8 – OK.
(2) This assumes that the fossil 1 kg CH4 releases immediately 2.75 kg CO2 (stoichiometric) and that the CO2 is there for the full GWP time scale.
(3) In reality the CO2 is released as the CH4 decays and thus the difference from non-fossil to fossil must be smaller than 2.75.
*(4) We agree that a tropospheric CH4 pulse decays as exp(-t/11.8) since a perturbation lifetime is 11.8 yr per IPCC Table 7.15. For 20 years, the average CH4 is about 9.63 yr or about 48% of the 20-yr period. Thus, the duration of the CO2 is 0.518*20 or only 10.37 years (i.e., add +1.4 to the GWP-20). Thus the fossil CH4 GWP-20 should be corrected to 81.1 not 82.5. This is a minor correction, but since you are quoting decimal places, we should all correct the value.*
(5) For 100 year GWP, this correction is smaller, 29.4 not 29.8.

As stated by the reviewer this is not the place to correct IPCC values. Small incorrections may have been done in Forster et al. (2021), and the uncertainties on the values are larger than the given precision. Though we acknowledge the reviewer to quote these errors, we keep the original Forster et al. (2021) values in the text.

L175: "CH4-non fossil" is very poor English it is more in the French style with the noun first. I love the French language, but here try "non-fossil CH4" and "fossil CH4"

This is not fair as a remark. This spelling is the one used in Table 7.15 of Forster et al. (2021), and it has just been reused as is. Maybe the person who did the Table was French, and none of the hundreds of co-authors and reviewers caught this? Anyway this has been rewritten in a more detailed and English way as follows: "**For a 100-yr time horizon the GWP of CH_4 emitted by fossil sources is 29.8 (GWP of CH_4 emitted by microbial sources is 27), whereas the values reach 82.5 over a 20-year horizon for CH_4 emitted by fossil sources and 79.7 for CH_4 emitted by microbial sources (Forster et al., 2021).** "

L1467FF This section on the stratosphere is remarkably out of date and misguided. The idea that strat loss could be 10 Tg/y is ridiculous (I know that models may calculate this, but why

report absurd values). This gives a strat lifetime of 500 years!! Then it would be well mixed to the mesosphere. Scaling to N2O and other known species it should be ~120 yr (100-200?) = ~40 Tg, and it is uncertain to what level, maybe 30-50, but not 10-70. The large percentages for O(1D) can Cl loss must refer to regions of the stratosphere and not to total stratospheric loss? If so, drop them because they are misleading and it does not matter what drives CH4 loss at 48 km.

Actually, we did not find recent updates on the estimation of stratospheric loss of methane on the top of the reference or simulations (e.g. CMIP6 & the latest chemistry climate estimates) used in the paper. Providing literature on the matter would be valuable to improve the manuscript. Though the first lines are very introductive, **recent references have been added (Zhang et al., 2023) and Morgenstern et al. (2018).** We acknowledge that the lower range of loss (10 Tg/yr) is not very likely, noting that average estimates are more at 30-40 Tg/yr. We propose to exclude the single model with this low estimate from the ensemble. The text has been modified as follows: "**In this study, six chemistry climate models that contributed to CMIP6 modelling activities (Table S5) provided estimates of CH_4 chemical loss, including reactions with OH, $O(^1D)$, and Cl; CH_4 photolysis is also included but occurs only above the stratosphere. Considering a 200 hPa tropopause height, these six CMIP6 simulations suggest an estimate of 34 [10-51] Tg CH_4 yr^{-1} for the CH_4 stratospheric sink for the 2000-2009 decade (Table S5), similar to the value derived from the previous CCMI activity reported in Saunois et al. (2020) (31 [12-41] Tg CH_4 yr^{-1}). The lowest estimate provided by a model (10 Tg CH_4 yr^{-1}) is quite unrealistic and would yield a methane stratospheric lifetime of several hundreds of years. As a result this outlier is excluded and we prefer to report a mean of 39 Tg CH_4 yr^{-1} associated with a min-max range of [27-51] for 2000-2009.**
For 2010-2019, we report here a climatological range of 28-43 Tg CH_4 yr^{-1} associated with a mean value of 37 Tg CH_4 yr^{-1} based on five models that contributed to CMIP6 runs (historic followed by SSP3-7.0 projections starting in 2015; Table S5)."

Also, it does matter what drives methane loss in the stratosphere and if models get it right or wrong in terms of sinks and then in terms of concentrations. This is especially true when it comes to using total columns measured from satellites within atmospheric inversions framework to infer surface fluxes. A wrong stratospheric gradient induced overestimating the total column in the chemistry transport model not because of the fluxes at the surface (which will be wrongly corrected).

L1617ff: This text looks like the previous 2020 edition that made the mistake of assuming that the fluctuations in CH4 abundance caused by emissions bumps would follow the budget lifetime rather than the perturbation lifetime (12 yr). Thus, the excellence of your fit to 9 yr should give you pause and admit that the variability is a mix of time scales for varying emissions and OH. The IPCC clearly discusses the different lifetimes and prominently displays the perturbation lifetime (11.8 yr) in the Table 7.15 from which you took the GWPs.

Indeed the sentence regarding the fit made using NOAA observation has been kept as in the previous version, and corresponds to a calculation made in Dlugockenky et al. (2003) to partly discuss the plateau in the early 2000s. We decided to remove that part of the text to stick to

describing observations as done for the period after the plateau and provide the same discussion for the different periods of atmospheric methane changes.

Nonetheless, we would like to highlight that this sentence was correct. The perturbation lifetime is theoretical and does not account for the impact of fast chemistry compared to the lifetime of methane (involving NOx for example, see Nicely et al. (2018))

L1690: "In addition, most of the top-down models use the same OH distribution from the TRANSCOM experiment (Patra et al., 2011),.." If this is so, then the top-down model mean of 575 Tg/y is really fixed by the assumed OH values. It is hardly a chemistry model, since the chemistry is fixed. This makes all the top-down numbers useless as they are fixed by Patra's choice of OH in 2011. Can you explain?

The top-down community faces difficulties in handling OH amount and variations, two key parameters for the methane budget, that are not well-constrained. Most of the systems still use prescribed OH concentrations, some tend to try to optimize that quantity along with the fluxes (though the reviewer tends to criticize the use of satellite data for this in a following comment). Using the same OH quantity definitely constrained the overall budget and range across the simulations, though different transport and simulated methane distribution lead to slightly different loss even with the same prescribed OH. As a result the loss is not fixed at 575 Tg CH_4/yr. This is acknowledged in the second part of this sentence: *"…which introduces less variability to the global budget than is likely justified, and so contributes to the rather low range (10%) compared to bottom-up estimates (see below*)." The values are not useless as we recognized the limitations. Part of the simulations were done including OH changes over time – low IAV <2% and no significant trend based on Patra et al. (2021) and in agreement with Thompson et al. (2024). Differences between the two ensembles (constant versus varying OH) are much lower than the range across the inverse systems. Also two inversions used different OH fields and their results fall into the same range. If these values are useless then all the top-down inverse studies published so far could be discarded as none is really doing a better job.

Though to acknowledge such limitation we recall Zhao et al.(2020) main message in the result section 5.1.1 after stating the range from the top-down studies: "**We recall here that Zhao et al. (2020) found an uncertainty of about 17% in global methane emissions (518 to 611 Tg CH_4 yr^{-1} for the early 2000s) due to changes in OH burden and distribution (OH ranging from 10.3 to 12.6 10^5 molec. cm^{-3})**"

L2052: I think we need to improve the chemistry-transport models and chemistry-climate models using full chemistry rather than try to replace interactive chemistry with a ML approximation so you can use tracer-transport models. (There is a confusion here that CTMs are tracer models with fixed OH, they are not.) It is more important to find out why the CCMs and CTMs (with interactive chemistry) may be wrong. That should be a higher recommendation than the Zhao approach that the lead author here is recommending.

Improving the chemistry in chemistry transport model (CTM) and chemistry climate models (CCM) has been a needed path for years. Atmospheric inversions do not use interactive chemistry as it could be too costly and we would need the adjoint of the chemistry solver. This is why we use pre-calculated OH fields from CTM or CCM. However, over the last three rounds (10 years) of this exercise of methane budget, we have not noticed any improvement in the

matter: the ranges of OH quantity, trends and IAV are still of the same order of magnitude, and CTM and CCM are not converging. Biases of those models against observations are still existing. Correcting the existing OH fields based on observations could be efficient awaiting a benchmarking of OH among CTM and CCM. Zhao et al. (2024) proposed a recipe to evaluate OH based on interactive chemistry in CCM/CTMs and a box model that could be useful to improve the convergence of OH fields among CCMs if more groups were adopting it. However, we acknowledge that the recommendation spectrum was too narrow. Thus, the details provided on Zhao et al. (2023) study have been removed and other references have been added. The text is as follows:

"**These results emphasise the need to first assess, and then improve, atmospheric transport and chemistry models, especially vertically, and to integrate robust representation of OH fields in atmospheric models. Recently, numerous efforts based on satellite data have been made to constrained OH distribution, variability and trends (e.g, Anderson et al. 2023 ;2024 ; Pimlott et al. 2022 ; Zhao et al., 2023; Zhu et al., 2022).**"
For the steps forward:
"**- Developping benchmarking of CTM and CCM regarding simulated OH distribution and variability (as in Zhao et al. (2019) for example) to increase efforts to assess biases and improve atmospheric chemical schemes in CTM and CCM.**
- Developping methods to better constrain OH, numerous have been proposed: satellite CH_4 observations (Zhang et al., 2018; Anderson et al., 2023; 2024) could afford this but strategy is needed (see Duncan et al. preprint 2024 and references therein);... "

L2063: If you recommend studying the reactivity of air parcels, then you really should reference the major effort on that done by the NASA ATom mission (e.g., 2023 ESSD, doi: 10.5194/essd-15-3299-2023).

This reference has been added to the text.

L2067: You can recommend satellite-constrained OH and that is a very attractive area for some, but it lacks the vertical resolution needed for CH4 studies and it seems to be overhyped here.

Indeed, this limitation is very real. We acknowledge this, we completed the text as follows:
"**- - Developping methods to better constrain OH. Numerous have been proposed: satellite CH_4 observations (Zhang et al., 2018; Anderson et al., 2023; 2024) could afford this but strategy is needed (see Duncan et al. preprint 2024 and references therein); using halogenated compounds beyond methyl chloroform (MCF), such as done in box models (Thompson et al., 2024) to derive a 3D dynamical OH. Such methods should be able to reach very low uncertainty for OH burden and trends (<2%) in order to really better constrain the CH_4 budget. Duncan et al. (preprint) discuss the existing satellite-based methods and propose a strategy to constrain OH from space-based approaches.**"

L2153: Again, this whole list of recommended directions seems like the co-authors personal research agenda and hardly a critical review of where advances might come form. The idea of porting transport models to GPUs to reach sub-one-degree will not improve the overall transport characteristics of these models. Many CTM/CCMs are already running one-degree

simulations. Much of the error is in the large-scale flows. Further, who is going to generate and store the petabytes needed to run the tracer models? Already we have full up CCMs running refined mesh at 3 km scales embedded in ESMs. If you want hi-res, that is where you must go. We already have CTM/CCMs with hybrid coordinates, what is so special here (L2155). The choice of odd grids will come from the dynamics of the underlying climate/forecast model rather than being designed by the tracer-transport operator.

This paragraph and directions concern the atmospheric inversion approaches. Most of them, at the time of the submission of this paper, still run at around 2-3 degrees resolutions for the sake of computer resources and time to produce an inversion over 20 years. Aiming at finer resolution allows finer resolution for the flux retrievals, better synoptic resolution of the variations of concentrations at surface stations (especially coastal and mountain sites), and also a better vertical transport (regarding convection in particular and troposphere-stratosphere exchange). Indeed, the transport model errors consider here also the error made against the observational constraint used for the optimizations. Also, running atmospheric inversions at finer resolution on CPU as usual will take a few months which is unrealistic, porting the atmospheric inversions system to GPU will overcome (has overcome) this issue. Indeed, some atmospheric inversion systems rely on dynamical cores that now could run on odd grids, but inversion systems need to adapt to this and this is far from straightforward. Odd grids could allow better representation of the high latitudes regions, which are, considering the Arctic, important regions regarding greenhouse gas emissions and climate feedback. Finally, although we agree that some recommendations may be unadapted (e.g. hybrid coordinates (**This has been removed**)), the recommendations made in the paper come from years of analysis of the global methane budget from 80 scientists worldwide and several workshops organized by various groups among which the global carbon project. We think they mean more than personal thoughts of the authors.
The sentence has been modified as follows : "**In the long run, developments within the dynamical core of the atmospheric transport models through the implementation of hexagonal-icosaedric grid with finer resolution (Dubos et al., 2015; Niwa et al., 2017, 2022; Lloret et al., 2023), and improvements in the simulated boundary layer dynamics or troposphere-stratosphere exchanges are promising to reduce atmospheric transport errors.** "
Finally, the question of the computing resources and storage for these simulations is a common question for the whole modeling community whatever the subject, and not specific to atmospheric inversions. Though it is worth noting that many institutions are already considering the environmental impact of their research.

References:

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