

# The Global Methane Budget: 2000-2017

Saunois et al., ESSDD, 2019

## Detailed Response to Michael Prather (Referee #2)

We acknowledge the work from Michael Prather for the time spent on reading and commenting on the paper. We thank him for his useful corrections and suggestions on the paper, which have helped clarifying and improving the manuscript. Below are the responses to his comments (in *italics, blue*). Changes in the text follow each response in **bold font**. Following both reviewers advices, the text has been shortened by 5 pages (8%), from 61 to 56 pages in the ESSDD format.

*This is a very well written and thorough review of the methane budget. The abstract and introduction read easily and layout the scope of the work. The remaining 2000 lines are a bit harder to get through, but this paper is meant as a reference work and not a "beach read." For a paper on methane, it is great to see Ehhalt's original work as one of the prime references. I have two major suggestions and then several minor/editorial comments on my read-through.*

We thank M. Prather for the compliment.

*#1 As a reference work, it would be very useful to have a Table of Contents up front. The TOC should be as detailed as possible (e.g., add another level: 3.3.2.1. Lakes and Ponds) and possibly include a list of key words for each section. This would greatly help those looking for specific discussions.*

A table of content has been added. Unfortunately, ESSD allows only 3 levels for the sectioning. We will add as much as possible details in the TOC, that fit ESSD style requirement.

*#2 The Section 3.3.5 on CH4 Lifetime contains some serious errors in understanding. I am not sure how to fix it, but the authors should think about the purpose and what they want to get across. See details below.*

Section 3.3.5 has been revised accordingly to the detailed comments. We have kept only the calculation of lifetime based on CCMI models and using the "budget" definitions: Total\_burden/Tropo\_loss for the tropospheric lifetime and Total\_burden/total\_loss for the "total lifetime". The other definitions were removed from this section to avoid confusion.

*L217: The Prather 10% is only 68% confidence interval, perhaps if the Saunois 15% is 90% confidence, they are consistent.*

The Saunois et al. (2016) uncertainty is based on a min-max range using the different ACCMIP models. Some of the models have difficulties in well representing methane loss due to uncomplete chemical scheme. The uncertainty is larger but its conservative and includes all models...

*L225: Probably more important here is not 'credible scenarios,' but credible mitigation strategies.*

Yes. Mitigation strategies are more important. Though the future climate scenario should help in defining mitigation strategies... We have replaced "credible future climate scenario" by "**appropriate mitigation strategies**".

*L231: do you want to mention the Paris Accord's "stocktaking"?*

This has been added in the sentence as: "**In order to verify such reductions, for example to help conducting Paris Agreement's stocktake,...**"

*L243: This is a slight mis-direct. It is not the lack of some direct observations, but by my calculation, the number of OH "measurements" needed to integrate the loss of methane (1 km x 100 m parcels, every 1/2 hour (clouds)) is of order 2x10<sup>14</sup> per yr. I would put this as an impossible task without some modeling and other tricks.*

Yes, several ideas were mixed there. This part has been reformulated to: **“The spatial and temporal distributions of OH are highly variable. Although OH can be measured locally, calculating CH<sub>4</sub> loss through OH measurements would require OH measurements every half on hour (to integrate cloud cover), in each small parcel (below 1km spatially to consider OH high reactivity and heterogeneity) of the atmosphere. As a result, such a calculation is currently possible only through modelling. However, simulated OH concentrations from chemistry climate models still show uncertain spatio-temporal distribution at regional to global scales (Zhao et al., 2019).”**

*L290: Here and later, you cannot reference a work in prep as a real reference. You can describe ongoing work by Staever and colleagues that will follow up this paper and come up with a shorthand notation for this work.*

This paper is in preparation and should be submitted before the final proof-reading of our paper. In case it will not be submitted, we will change the phrasing.

*L318-324: If you are being precise, please define where the edges of the 1-degree grid fall, one the 0.0, or 0.5, or? Also does what you describe mean that coastal boxes (<100% Land and <100% Ocean) have no emissions – you should definitely want to warn people as it will look funny when plotted. Also what do you do about large lakes (resolved at 1 degree)?*

We have added information on the edges: **“The edges of the 1°x1° grid fall in 0.0°.”** The land/sea (or lake) mask is an issue when estimating regional budget but not the global one. The reallocation of land emissions is based on the percentage of sea/land in the initial and final grid cells. Coastal boxes have then non-zero fluxes. The same treatment is reserved to the big lakes.

*L383: This discussion of natural vs. anthropogenic intrigued me. We have a long history of trying to break emissions cleanly in these two camps. With natural often being ignored in the scenarios (RCP, SRES), such that in the IPCC SAR & TAR, I had to make up the natural sources to ensure a consistent budget and initial trajectory for the future scenarios. The other problem is that attribution is still not an exact science and thus declaring something like wildfires or wetland loss as "anthropogenic" is not trivial.*

This issue has been raised by referee#1, and also by several co-authors, which would like specific treatment for some sources at the frontier between these two categories.

*Let me propose that the sources be split into "pre-agricultural" and new "anthropogenic" types of sources (such as ag or industry). This allows us to work our best science on how wetland methane emissions have changed, without trying to ascribe cause. The changes in the "pre-ag" sources probably can be attributed in many cases to direct human intervention.*

Thank you very much for this suggestion. This fits the definition the lead author has in mind, and the response provided to referee#1. Such a definition allows to sit permafrost emissions in the “natural” sources, while for sure, thawing permafrost will release more in the future due to climate warming, being an indirect anthropogenic source. The text in the Methodology section has been changed to:

**“In the following, we present the different methane sources classified from anthropogenic or natural origin. “Natural sources” refer to pre-agricultural emissions even if they are perturbed by anthropogenic climate change, and “anthropogenic sources” are caused by direct human activities since pre-industrial/pre-agricultural time (3000-2000 BP, Nakasawa et al. (1993)) including agriculture, waste management and fossil fuel related activities. Natural emissions are split between “wetland” and “other natural” emissions (e.g., non-wetland inland waters, wild animals, termites, land geological sources, oceanic geological and biogenic sources, and terrestrial permafrost). Anthropogenic emissions contain: “agriculture and waste emissions”, “fossil fuel emissions”, “biomass and biofuel burning emissions”, assuming that all types of fires cause anthropogenic sources, although they are partly of natural origin (Fig. 6, see also Table 3 and 6).**

**Our definition of natural/anthropogenic sources does not correspond exactly to the definition used by UNFCCC following the IPCC guidelines (IPCC, 2006), where, for pragmatic reasons, all emissions from managed land are reported as anthropogenic, which is not the case here. For instance, we consider all wetlands as natural emissions, despite some wetlands being managed and their emissions being partly reported in UNFCCC national communications. The human induced perturbation of climate, atmospheric CO<sub>2</sub>, and nitrogen and sulfur deposition may cause changes in**

the sources we classified as natural. Following our definition, emissions from wetlands, inland water or thawing permafrost will be accountable in “natural” emissions, even though, we acknowledge that climate change – a human perturbation – may cause increasing emissions from these sources. Methane emissions from reservoirs are considered as natural even though reservoirs are human man-made, and since the 2019 refinement to the IPCC guidelines (IPCC, 2006; IPCC, 2019) emissions from reservoirs and other flooded lands are considered anthropogenic by UNFCCC.”

*L529: can you make this simpler to read: "perhaps because AMAP analysed data from a wider range of inventories and projections, plus it was referenced to one year only (2005) rather than averaged over a decade, as done here."*

Thank you very much for the suggestion. This has been corrected accordingly.

*L552: "realized" in what? the scenarios or real life?*

In the real life... This has been changed to: "... **current emissions appear to follow the higher-emission trajectories over the next decade**".

*L660ff: In terms of shale gas emissions, is the DeCarlo work on the Marcellus Shale consistent with these? Goetz, J. D., et al., Analysis of local-scale background concentrations of methane and other gas-phase species in the Marcellus Shale, Elementa Science of the Anthropocene, 1–20, doi:10.1525/journal.elementa 182, 2017.*

The “oil and gas” section has been revised accordingly to Referee#1’s comments. The discussion focuses on the budget of oil and gas emissions. Shale gas emissions are included in the “oil and gas” estimate from inventories. Few sentences have been kept to discuss the uncertainty regarding shale gas emissions – mainly through their different emissions factors. The authors looked at the Goetz et al. paper, they did not specifically discuss methane emission to compare to inventories such as EPA.

*L1026: Is this really true? If we have all positive values but some far outliers, then you would reject a positive flux because the std dev exceeded the mean?*

Here we use a simple diagnostic to roughly estimate the area over which land surface models agree on the emissions. Indeed, we did not do the calculation using a bootstrapping method (considering all but one model and looping over the models). This would indeed identify outlier model and provide an average area. However, we use the method as in the previous budget for consistency.

*L1174: I am unsure what the "up scaling issues" means.*

This includes the spatial and temporal uncertainty of the flux density used for a single class of ecosystems for example. This paragraph has been removed, belonging now to Section 6, following Referee#1’s comment.

*L1205ff: This sentence on the Petrenko work is a bit perjorative and full of inuendo ("If it is correct", "which is questionable"). There is nothing obviously wrong with the work, unless the 14C community is worried about it, and if so state why. The sentence on L1208 about the discrepancy is fine.*

Indeed, we agree the wording was judgmental, and useless for the budget. The sentence has been removed.

*L1233: I am not sure what 'termite' as a unit is?*

We understand why the referee as difficulty with the unit. The density flux is given per biomass of termite, that is per ‘gram of termite’, which should be “(g termite)<sup>-1</sup>” instead of “g<sup>-1</sup> termite”  
The unit has been corrected.

*L1500-1507: I am a little worried about this section, but have no simple fix to recommend.*

*(1) the effect of CH<sub>4</sub> oxidation on HO<sub>x</sub> depends on the pathway for H<sub>2</sub>CO, if it photolyzes on one path you get 2 HO<sub>x</sub>, otherwise, if OH is high, then OH-destruction it yields 0 net HO<sub>x</sub>. NO<sub>x</sub> plays a role in this, but HO<sub>x</sub> levels are also very important.*

*(2)The short lifetime of OH should go back to Levy or Logan, not a 2004 reference.*

*(3) "estimate"? One can estimate OH from models without observations.*

(4) Many, many OH measurements are made in the free troposphere by Brune and others. I am beginning to think it best to stop at 'compounds.' on L1501 and jump to L1507 'Following. . .'

Indeed, we agree that a long review would be needed to discuss OH. And this is not the point of the budget paper. This part has been removed as suggested.

L1511: I think you have to recognize here that mass weighted OH concentrations are NOT a good measure of the methane loss. Since this is a methane paper, you should focus on the average methane loss rate from these models. I know this is a hobby of mine, but please look at the table in my 1990 paper (Prather & Spivakovsky, *Tropospheric OH and the lifetimes of HCFCs*, JGR: 95, 18723-18729, 1990), also reproduced by Mark Lawrence in 2000, and one can see that the average OH varies by 20-40% depending on how it is weighted. What is relevant here is when OH is weighted by mass and  $\exp(-1800/T)$ . In fact, the OH values do not tell you the methane lifetime unless you know the temperature weighting.

It is true that the mass weighted OH concentrations do not represent a measure of the methane loss directly. Even though, the discussion should focus on ethane loss directly, some explanations on OH range in concentrations and horizontal and vertical distributions are needed to explain – partly – the range in methane loss. The following text has been added prior the discussion on OH:

**“Mass-weighted OH tropospheric concentrations do not directly represent methane loss, as the spatial and vertical distributions of OH affect methane loss, through, in particular, the reaction temperature dependency and the distribution of methane. However, estimating OH concentrations and spatial and vertical distributions is a key step in estimating methane loss through OH. »**

L1527-1560: The Holmes et al paper (2013, *Future methane, hydroxyl, and their uncertainties: key climate and emission parameters for future predictions*, *Atmos. Chem. Phys.*, 13, 285–302, doi:10.5194/acp-13-285-2013) really addresses recent OH variability and should be included with this discussion. (Sorry to push my own papers again, but it is a balanced survey of OH variations from cause [all those listed] to methylchloroform-derived variability.)

The Holmes et al. (2013) paper has been included as a reference in this discussion.

L1579: It is unclear if your 60 Tg is irreversible mixing (i.e., loss in strat) or the cross tropopause flux. The amount of CH<sub>4</sub> entering the stratosphere is 10x or more larger than this number, most of which crosses into the lower stratosphere in the sub-tropics and is then transported into the troposphere with little chemistry. I would drop this whole sentence as it does not say much.

This part has been removed, in agreement with referee#1 comment.

L1590: We have a much more accurate measure of the stratospheric loss from the Plumb & Ko relationship and the observed N<sub>2</sub>O-CH<sub>4</sub> tracer slopes in the lower stratosphere. I do not know when this was last revisited, however.

The authors have searched for updated estimates using Plumb and Ko relationship, but without success, to the best of their knowledge...

L1654ff: This section 3.3.5 on CH<sub>4</sub> Lifetime has some major problems. Excuse my didactic diversion here. "Lifetime" is a budgetary number since the standards and notation in the 1995/96 IPCC SAR through AR5. It is the burden divided any loss rate. Note that the total burden in the system must be used to take advantage of adding inverse lifetimes. Hence you will see the "lifetime of CH<sub>4</sub> against trop OH loss" is the T<sub>g</sub> (including stratosphere) divided by the T<sub>g</sub>/yr lost to OH in the troposphere. If you use this definition carefully then inverse lifetimes can added and we can think of the lifetime due to stratospheric loss being about 160 yr as is recommended here. The OH lifetime should be noted and taken from the MIPs, it should be about 11 yr.

"Perturbation time, response time, e-fold time" are used to define the decay of a perturbation about any atmospheric state (again, steady-state does not matter for these definitions). Since 1994, for CH<sub>4</sub> we have known that this time scale is about 1.4 times the total lifetime because of chemical feedbacks whereby CH<sub>4</sub> suppresses OH. This is well known, consistently modeled and increases all the integrated impacts by this factor. With a budget lifetime of 9.1 yr, the time scale for CH<sub>4</sub> perturbations is about 12 yr. Any perturbation to a chemically reactive species will excite a large number of chemical modes – each with its own pattern of species and its own decay time. Hence CO is an indirect greenhouse gas because it generates a CH<sub>4</sub> perturbation that decays with a 12-yr e-fold. "Steady state" is not required

*for these numbers, but the steady-state lifetime does have some magic properties. It is the effective average over all the different chemical modes (their amplitudes and time scales) excited by a perturbation.*

*The rest of the lifetime numbers from the recent MIPS look to be OK, but make sure that these are calculated using the full burden.*

We removed the sentence mentioning “Perturbation time, response time, e-fold time” from this section (bottom-up) and keep only the definition of lifetime as burden over loss.

The lifetimes were calculated using the total burden of methane from CCMI models – associated to the tropospheric methane loss to calculate lifetime with respect to OH removal. As a result, the numbers were correct. In this Section (bottom-up) we have removed the couples of sentences discussing lifetime from observation – considered as top-down estimates.

*L1708: The references to Rigby 2017 and Turner 2017 really should include also the accompanying Prather and Holmes paper (Over-explaining or under-explaining methane’s role in climate change, PNAS 114(21) 5324-5326, doi: 10.1073/pnas.1704884114, 2017) that points out the fundamental error in modeling CH<sub>4</sub> perturbations for both papers (i.e., they did not include the chemical feedbacks).*

This part of the text has been removed from the revised manuscript, following referee#1 comments – discussing with non-used data set in this budget...

*L1750. Based on notes above, I might expect the time scale here to be 12 yr rather than 9 yr, but I think it is probably close enough for this simple analysis.*

The commented approach is based on atmospheric methane concentrations recorded before and during the stabilization period. In this approach, we assumed no trend in emissions and in lifetime (so no trend in sinks). Such assumption can be criticized in regard with some literature explaining the stabilization period by decreasing emissions associated to increasing sink. We acknowledge that there are uncertainties associated with this calculation. Nevertheless, the result seems consistent with lifetime values from the literature. We have changed the text as follows:

**“When a constant atmospheric lifetime is assumed, the decreasing growth rate from 1983 through 2006 may imply that atmospheric CH<sub>4</sub> was approaching steady state, with no trend in emissions. The NOAA global mean CH<sub>4</sub> concentration was fitted with a function that describes the approach to a first-order steady state (*ss* index):  $[CH_4](t) = [CH_4]_{ss} - ([CH_4]_{ss} - [CH_4]_0)e^{-t/\tau}$ ; solving for the lifetime,  $\tau$ , gives 9.3 years, which is very close to current literature values (e.g., Prather et al., 2012,  $9.1 \pm 0.9$  years). Such an approach includes uncertainties, especially due to the strong assumption of no trend in emissions and sinks, which does not agree with some study explaining the stabilization period by decreasing emissions associated to increasing sink (e.g. Bousquet et al., 2006). However, this value seems consistent albeit higher than the chemistry climate estimates (8.2 years, see Sect. 3.3.5)”**

*L1795ff: This is a good discussion of results, but I wonder how much these inversions depend on the a priori’s for lifetime, etc. I do not see how you can cover this here, but can you comment? OK, see L1988. L1988: Yes, if everyone uses the same trop OH, then the budget total is pretty well fixed (except for T biases, and maybe ITCZ gradients).*

Indeed, this is one of the main caveats of the top-down budget.

*L2154ff & L2200ff: Agreed. We continue to produce models with the same range in CH<sub>4</sub> OH-lifetime and no means of differentiating them. I would not emphasize the vertical as our model failings include horizontal as well. We need to develop some key observational tests that reflect the reactivity of the air parcels. To start, we need to run the MIPS with some more focused diagnostics that tell us why the models CH<sub>4</sub> lifetimes are so different. Even more interesting is that the historical trends and the scenario projections often go in different directions across the model ensemble. And we should not forget that our goal is accurately simulate CH<sub>4</sub> loss, not just get OH correct.*

We thank Michael Prather for his comments and point of view of the future directions to better estimate methane loss. This comment has been used to feed Section 6.

*L2273: This is where you might want to look at the Holmes (2013 ACP) effort to model OH variability and match it to CH<sub>3</sub>CCL<sub>3</sub> observations and CH<sub>4</sub> lifetimes. It is multi-model and more physically based than these inversions.*

We thank the reviewer for the reference. This has been included and we will look at this study carefully for the further study on methane change.