

# *Interactive comment on* "The Global Methane Budget: 2000–2012" *by* Marielle Saunois et al.

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# General answer to both referees

We acknowledge both referees for the time spent on our paper, for their comments and helpful suggestions to improve the manuscript. Our aim is to present the global methane budget on annual to decadal time steps, and not to focus on methane budget changes (variability and trends), although this is of course a very interesting topic. Reviewers have made some comments for more discussion on budget changes, but as it is already long, we think that this is beyond the scope of this paper. Indeed we are preparing a second "GCP methane paper" more focused on methane variations over the 2000-2012 period. As a result, and to avoid misleading interpretation of this paper, we have sometimes deleted or moved some parts of the initial text mentioning budget changes to the perspective section, where we open the discussion and introduce

C1

the second paper to be submitted soon. These changes can be followed in the track change version of the manuscript. This is one of the significant changes of the text. The second significant change is a specific sub section dedicated to methane isotopes as proposed by referee Euan Nisbet.

During the review process, a graphic using interactive data visualization techniques has been produced. It is now proposed as a complementary way of viewing the methane budget and is advertised at the beginning of subsection 5.2.2.

While reviewing the text and figures of the initial manuscript, we also discovered some misleading sentences in the text, an error in Figure 1 (corrected with an author comment during the opening phase) and an issue in Figure 2 (RCP data used for Figure 2 were not the recommend ones). The text and figures have been corrected in the revised version of the manuscript. The revised manuscript has been sent to the co-authors and some minor corrections (not requested) have been made throughout the text (typos and english mainly...).

We answer below (normal text) to all comments (italic text) of both referees. Changes in the manuscript are highlighted in bold text.

## **Detailed Response to Anonymous Referee 2**

We acknowledge referee 2 for his time spent on reading and commenting on the paper, providing comments and helpful suggestions to improve the manuscript.

The summaries of the existing estimates of anthropogenic emissions: fossil fuels, waste, agriculture etc. was very interesting but it could be noted that underlying all these different approaches is the IPCC guidelines (IPCC 2006) (except Shale Gas which was not significant when the IPCC guidelines were written) so, despite their

#### different assumptions and data used they are not completely independent.

Indeed, these data sets are not completely independent as they follow some recommendations of the IPCC guidelines; we have added this piece of information in the text (Sect. 3.1.1, formerly page 9 line 21). Instead of "There are major differences between these three inventories"; we rephrased in: "These datasets differ in their assumptions and the data used for the calculation, however they are not completely independent as they follow the IPCC guidelines (IPCC, 2006)."

The over-estimate of coal methane from China in the EDGAR estimates highlights why the IPCC guidelines recommend national factors for significant contributors to total emissions, simply applying "Tier 1" factors without checking their appropriateness can lead to significant errors.

We have emphasized the recommendation of IPCC in this paragraph on coal emissions (formerly page 12 line 23) and added: "[...] twice too high in China. This highlights that significant errors on emission estimates may result from inappropriate use of some emission factor and that applying "Tier1" for coal mine emission is not accurate enough as stated by the IPCC guidelines."

The paper notes that overall the T-D estimates are greater than the B-U estimates BUT this is mainly due to inland water, wetlands and geological leaks. For those sources that counties report nationally to the UNFCCC (with a probable increase in reporting under the Paris Agreement) it would be useful to stress the good agreement between B-U and T-D estimates thus providing additional credibility to these reports in the policy arena.

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The reviewer probably means that B-U estimates are larger for B-U than for T-D, mostly due to natural sources. Indeed, anthropogenic emissions are in better agreement as stated by the reviewer. We think this point was clear in the initial text but tried to make it even clearer in the revised version (page 39, line 31): "[...] This overestimation likely results from errors in the estimation of natural sources and sinks: extrapolation or double counting of some natural sources (e.g. wetlands, inland waters), or estimation of atmospheric sink terms. The anthropogenic sources are much more consistent between B-U and T-D approaches (Sect. 5.1.2)."

The paper gives values as mean and +/- 25%ranges of the estimates presented in the different sources. Some discussion of the uncertainties in the individual estimates (especially as they are not completely independent) would be helpful to inform the reader about the overall accuracy of these budgets.

In the paper (text and tables) we give mean, minimum and maximum values and not +/- 25% ranges. We estimated that the number of studies for the different estimates provided in the paper is generally too low (down to 2-3) to provide more classical statistics such 1-sigma values. This is stated in the first paragraph in the Methodology section. However we acknowledge that these minimum and maximum values do no take into account the uncertainties of each estimate, which is poorly documented in the inventories. Thus the full uncertainty range, provided by the mean of the different studies gathered here, may be greater than the given range. While the bottom-up approaches may be better in estimating regional or per source emissions than TD, top down approaches are better in constraining the global total source. Indeed, the range reported here at the regional and/or per source scales is larger for TD than for BU. Also we have acknowledged the uncertainty in the global budget and stated that the 3 digit values (formerly page 6, line 15) are given to close the budget and should not be taken as is. We think that the accuracy of the budget for the different sources can

be read through the [min-max] range given in the BU approaches, though the reader should keep in mind that the studies providing the range are not fully independent, so that the range of uncertainty may be larger.

To make this clearer, we have rephrased a bit the sentence in the Methodology section (formerly page 6, line 13): "Following Kirschke et al. (2013) and considering the relatively small and variable number of studies generally available for individual numbers, uncertainties are reported as minimum and maximum values of the gathered studies in brackets. Doing so, we acknowledge that we do not take into account all the uncertainty of the individual estimates (when provided). This means that the full uncertainty range may be greater than the range provided here. "

The paper states that emissions are characterized according to their anthropogenic or natural origin. This should be qualified that these are not the definitions of anthropogenic and natural used by the UNFCCC or in the IPCC Guidelines where, for pragmatic reasons, all emissions from managed land are reported as anthropogenic.

Indeed, choosing a way to characterize the sources and categorize them can lead to long discussions for some sources to partition anthropogenic and natural emissions. As suggested, we have added the following sentence at the end of the header paragraph of Section 3 (formerly page 8, line 23).

"In the following, we choose to present the different methane sources depending on their anthropogenic or natural origin, which seems more relevant for planning climate mitigation activities. However this choice does not correspond exactly to the definition of anthropogenic and natural used by UNFCCC and IPCC guidelines, where, for pragmatic reasons, all emissions from managed land are reported as anthropogenic, which is not the case here. For instance, we con-

C5

sider all wetlands in the natural emissions whereas there are managed wetlands"

In the discussion about coal mine methane it should be added that (page 12 lines 15-17) that the geological history (basin uplift) is also an important determinant of the coal emission factor.

Thank you for pointing the missing important detail. In the coal emission paragraph, we have added this in the following sentence (formerly page 12, lines15-17): "Coal mining emission factors depend strongly on the type of coal extraction (underground mining emitting up to 10 times more than surface mining), the geological underground structure (very region-specific) and history (basin uplift), and the quality of the coal (brown coal emitting more than hard coal)."

page 43 line 16 " central North America" - should this be "boreal North America"?

At this line, we discuss the regions for which agriculture and waste are dominant sources, which is the case for central North America, but not boreal North America. Our sentence seems correct so.

## **Response to Euan Nisbet**

We acknowledge referee Euan Nisbet for his time spent on reading and commenting on the paper. He did a thorough and helpful review. He has highlighted many points that needed or would need more attention in the future review of methane. His useful corrections and suggestions on the paper have helped clarifying and improving the manuscript. [...] Overall, the paper, which will be very highly cited, is publishable with only the most minor amendments. But it would be nice at least to give isotopes their own sub-section!

In the first draft we wrote, almost two years ago, there was a dedicated sub-section for isotopes! Then the paper grown and evolved and isotopes were included in a "other observations" section. We acknowledge the benefit from isotopes the methane budget can gain. In the revised version, we dedicate a specific sub-section to isotopes and enlarge the discussion around isotopic studies. This is the main change in the revised manuscript, as already stated in the general answer to both referees.: The subsection 4.1.3 is now on Isotopes and subsection 4.1.4 on other observations.

# Specific Comments

1. Introduction: This section provides a very useful general review. It is a telling comment that uncertainties in emissions reach 40-60% of how to achieve optimal use of satellite retrievals, which have in the past led to some perhaps misleading inferences.

We added a sentence in the abstract about RCP8.5. : "Since 2010, the bottom-up global emission inventories are closer to methane emissions in the most carbon intensive Representative Concentrations Pathway (RCP8.5) and higher than all other RCP scenarios." We also added a sentence about active space mission, which is a way to limit systematic errors and get closer to the optimal usage of satellite data: (page 5, line 17) : "[...] The development of low-bias observations system from space, such as active LIDAR technics, is promising to overcome these issues

C7

# (Kiemle et al., 2014)."

2. Methodology. The paper broadly follows the established methodology of Kirschke et al. (2013). This is good. However for future updates it might be worth considering an attempt to split agricultural emissions into ruminants, rice, and agricultural waste/silage/manure and biomass burning categories, and to address urban waste fully independently from agricultural waste/silage/manure.

We acknowledge the suggestion for future update of the paper. That would be feasible for most of the bottom-up approaches, which have part of the suggested individual categories. However, for example, GAINS provides gridded data for the following sectors only: energy, industry, solvent use, transport, domestic combustion, agriculture, open burning of agricultural waste, waste treatment. Also, such a detailed budget is, at present, only suitable for process-based approaches (B-U) but not for T-D inversions as most of the systems resolve net methane emissions for the moment, and few resolve 3-4 categories only. If multi-tracers inversions were to further developed (using isotopes, ethane, ...), one could think of going further in the TD category estimates, but it will probably never reach the detail of B-U inventories.

3. Methane sources and sinks. The discussion of inventories is excellent and very helpful as a general summary. The recent sharp increase in bottom-up source inventories, close to RCP8.5, is commented on. What is only lightly discussed (nor later in Sect. 4) is the isotopic implication and the clash with observations. Also, the ethane results (3.1.3) may merit a little more comment.

The aim scope of the paper is a decadal (and last year) budget and we do not aim to discuss the inter-annual variations of the methane budget here, neither the trend.

We show Figure 1 with RCP scenarios to illustrate that today none of the RCP scenarios match the different inventory emissions. However, a second paper will specifically address inter-annual variations. Therefore in the revised version we moved all discussions on methane emission variations to the perspectives part at the end of the paper.

The discussion of China's coal emissions is valuable. Perhaps R. Thompson et al's (JGR 2015) findings that methane emissions from China increased by 3% annually from 2000 to 2011 is worth citing?

Indeed, we missed this recent reference. They use a top-down regional system assimilating both methane and C13-CH4 data to constraint different sources. They found lower emissions in their posterior compared to their prior (in agreement with our analysis), but explain this difference with rice emissions more than coal emissions. In fact they do not found significantly different coal emissions than from their prior (EDGARv42) and even a bit higher (32 compared to 30 Tg yr-1 in 2010-2011) We have added the following sentence (formerly page 12, line 21): "Also, assimilating also 13CH4 data, Thompson et al. (2015) showed that their prior (based on EDGARv42) overestimated the Chinese methane emissions by 30%, however they found no significant difference in the coal sector estimates between prior and posterior."

Shale gas / Fracking: Zavala-Araiza et al probably need more discussion as this is a very interesting paper and TD and BU estimates were close. One point is that the emissions were dominated by a few high-emitters, and that these came largely from the 'conventional' part of the extraction complex – it's not the fracking, but what happens after the fracking. Indeed, the paper could lead to an optimistic inference that these high emitters (which must surely be expensive) can easily be found and

C9

controlled. The increasing cost-consciousness brought on by falling prices may be driving leak reduction as much as regulatory controls. There is emerging evidence (e.g. Peischl et al.) that nowadays there isn't much difference between conventional and unconventional gas, at least as far as methane is concerned.

To address part of this comment, we have added the following sentence on Zavala-Araiza paper (formerly Page 14 line 10): "This study also showed that emissions were dominated by a few high emitters, neglected in the inventories. Moreover these high emitting points, located on the conventional part of the facility, could be avoided through better operating conditions and repair of malfunctions. It also suggest that emission factor of conventional and non-conventional gas facilities might not be as different as originally thought (Howarth et al., 2011) " Peisch et al. paper has been adequately cited and surely this sub-section will be updated in the next release.

Livestock. Assessing emissions is difficult: Africa cattle eat trees and are often water-limited. Indian cattle have experienced poor monsoons. Parts of S. America had severe droughts in this period. But in 2001-2012 Chinese cattle increased (as did melamine consumption).

Indeed, to highlight the uncertainty in the assessment of these emissions, we have added the following sentence (formerly page 15, line 2): "Methane emissions from enteric fermentation are also variable from one country to another as cattle experience water-limited conditions that highly vary spatially and temporally (especially in the tropics)"

Waste. Urban waste in the Middle East and parts of Africa and rapidly urbanising Asia

has had little attention. Our own work in Kuwait indicates it may be a significant source.

Our global analysis does not reached that level of refinement about Koweit , which will deserve more specific attention in the future. We add a sentence about global urban development (page 16, line 2) : "The large and fast urban development worldwide, and especially in Asia, could enhance methane emissions from waste if adequate policies are not designed and implemented rapidly".

Rice. The changes in rice area in China, and perhaps growth in non-conventional locations like Australia, will need attention in future.

As China is the dominant emitter for rice, we added a sentence in the revised version. More will be addressed in future or more regional studies. (page 17, line 5) : "The decrease of CH4 emissions from rice cultivation over the past decades is confirmed in most inventories, because of the decrease in rice cultivation area, the change in agricultural practices, and a northward shift of rice cultivation since 1970s (e.g. Chen et al., 2013).

Biomass burning. This is a major topic – it is possible that in the isotopic balance, a decline in biomass burning has masked a rise in fossil fuel emissions. The discussion in the paper mostly addresses forest biomass burning, which is very important in SE Asia and S. America. However, my own anecdotal experience from 45 years of travelling and flying annually across the length of Africa is that the bulk of biomass burning in Africa is in C4 grasslands. Similarly, even in forest, peat burning merits more attention. Also, a significant part of the burning may be of seasonal grasses in clearings. It might be worth mentioning the CO record as it pertains to biomass burning

C11

The first part of this comment is about trends, which is beyond the scope of the paper as explained before. As stated in the introduction of our answer, **we have deleted the paragraph on the biomass burning variability (formerly page 19, line 13 to 18)**. Indeed, as more detailed in the second paper in preparation, a decline in biomass burning (enriched in isotope) has likely masked a rise in fossil fuel emissions, and this important result deserves more dedicated discussion in future works.

Concerning components of biomass burning, we agree that only forest and savannahs were mentioned. We have changed the sentence (formerly page 17, line, 28) to: "Anthropogenic fires are concentrated in the tropics and subtropics, where forests, savannahs and C4 grasslands are burned to [...]"

Carbon monoxide is a good tracer for biomass burning emission. On (formerly) page 17-line18, we have added: "Among the species emitting during biomass burning, carbon monoxide is a pertinent tracer for biomass burning emissions (Pechony et al., 2013 ; Yi et al., 2015)"

# Biofuel – this is a placeholder really.

This is true, that we do not provide much information on biofuel burning. We did not find many studies synthetizing this part of the budget. Hopefully, this will be completed in the future updates. We acknowledge this in the revised version adding a sentence (page 19, line 28): "Although more than 2 billions [...] (Andre et al., 2014), methane emissions from biofuel combustion have not yet received the attention it should have to estimate its magnitude."

Natural Sources Wetlands, lakes, ponds and streams. Saunois et al point to the

discrepancy between bottom-up and top-down estimates of wetland and freshwater emissions. This tallies with my personal anecdotal experience that freshwater bodies deeper than a couple of metres emit little methane. Ebullition is dissolved on rising, or is captured by methanotrophy. There is much need for better studies of freshwater emissions from open lakes and streams. To some extent, the scientific funding system may be a problem here: funding bodies do not like null results and there is always an incentive to claim bigger and more impressive methane emissions from whatever source is being investigated. "My burp is bigger than theirs – give me a grant!").

Land surface models do not in general currently differentiate isotopically between C3 wetland systems (as in boreal muskeg) and C4 wetland vegetation (e.g. papyrus, some C3/C4 phragmites). The uncertainties are huge and need attention.

We already stressed the fact that more studies are needed about methane emissions from freshwaters in section 6 (first issue raised). In this part, indeed, the isotopic point of view was a bit missing in the perspective section. To overcome this and address this comment, we have added in Section 6 (formerly page 45, line 3) the following sentence: "More measurements of the isotopic atmospheric composition of the various ecosystems (bogs/swamps, C3/C4 vegetation, ...) would also help better constraining methane fluxes as well as its isotopic signature in the wetland models."

This poor knowledge of freshwater sources is arguably the largest single barrier to a proper understanding of the global methane budget. In particular the fluxes from lakes, ponds and streams need sharp critical evaluation.

Indeed this poor knowledge has been highlighted in the first paragraph of our Section 6 Futur developments, missing elements and remaining uncertainties.

C13

A significant puzzle is that in some El Nino events, methane emission often seems to rise, while land surface models usually find that it should plummet. Part of the reason may be temperature: Q10 in land surface models is not well constrained and could be a major source of uncertainty. Also, hydrology can be important in large tropical wetlands. In the start of an El Nino event, ground water may be well-charged from the previous season. Thus even a small run off can flood wetlands, albeit not to the extent in a 'normal' season. Later, after evaporation and transpiration, the wetland groundwater level becomes depleted. Thus the fall-off in methane emission should show a hysteresis – it should lag the El Nino.

Again, this is a very interesting comment that may explain part of the differences between TD and BU phases of wetland and freshwater emissions. It would require a complete analysis of the impact of El Niño events on methane emissions, which seems beyond the scope of this paper focused on global decadal budgets.

Geological Sources. Here too there may have been a tendency to aggrandisment of fluxes. Locally, large seeps etc may occur, but how significant are they? Our own group's work in Kuwait suggests the use of mobile CRDS instruments may help constrain regional seepage sources, for example around oil and gas fields.

Indeed, in the perspective section (Section 6), we did not mention the improvement needed in the assessment of the geological sources. To address this, we have rephrased the first point of Section 6:

See section 6.

First point title (formerly page 44): "Annual to decadal CH4 emissions from major natural sources (wetlands, fresh water, geological) are highly uncertain"

Formerly page 45, line 13, we added: "Similarly more local measurements using continuous laser based techniques would allow refining the estimation of geological methane emissions." Then the following sentence has been changed (formerly page 45, line 13): "Further efforts are needed: 1) extending the monitoring of the methane emissions from the different natural sources (wetlands, freshwaters and geological) complemented with key environmental variables to allow proper interpretation (e.g. soil type, temperature and moisture [...]"

Termites: these insect cows channel emissions from their 'gardens' on the water table via the chimneys of termite mounds, so that egress can bypass methanotrophy. The Saunois paper depends heavily on one study by Sanderson. Perhaps it would also be worth going back to some of the earlier work by Pat Zimmerman and Stan Tyler?

As the topic is complex and the paper we are submitting is already so full of information, we found more suitable to refer to Sanderson and Kirschke works, rather than mentioning again detailed literature, which the reader can indeed find in the two above-mentioned papers. Indeed, both are literature review and their estimates derived from available literature. The present estimate, calculated from the average CH4 emission per unit of termite mass by spatialized average termite biomass per ecosystem type, takes into account data collected in the field either directly from mounds or from soil (Kirschke et al. 2013). The data used for upscaling are based on both Sanderson et al., 1996 and Kirschke et al. 2013. Analysed papers are mentioned in the supplementary materials of Kirschke et al. 2013. .

So somehow we have tried to provide an average value, which considers main mechanisms, which lead to net CH4 emissions (direct chimney loss or mediated loss from soil). Even chimneys seem to host however methanotrophic communities on their walls, so that even in this case we could speak of net emissions. To be honest the water table issue has not been dealt with, but we think that in the majority of the areas

C15

where termites are widespread superficial watertables might be rare.

Wild animals. These are perhaps a larger factor than estimated. There are enormous numbers of deer still in hiding – SE Asian forest has large populations of small deer, as does North America. How do reindeer fit in? – semi domestic. Camels? Incidentally as sources to amuse, we have found that elephant dung is not significant, but maybe in Venezuela one might consider the hoatzin (an avian ruminant, the stinkbird, the last ruminant of the dinosaur clade).

Indeed wild animals do not report! We have added a sentence highlighting the uncertainties and difficulties to estimate these emissions. Formerly on page 25, line 32: "However, as suspected, numerous and various wild animals live partly hidden in the forests, savannahs, etc., challenging the assessment of these emissions."

Oceanic – This is a very useful revision of the Cicerone and Oremland 'placeholder' flux that has survived in inventories for nearly 30 years. Maybe cite Westbrook et al (GRL 2009) – methane plumes do not reach surface.

Hydrates – maybe cite Fisher et al 2011, showing Arctic hydrate emissions are small.

We have added a citation of Westbrook et al., (2009) and Fisher et al., 2011 in Section 3.2.6 (formerly page 26, line 18): "[...] more uncertain is the flux of oceanic methane reaching the atmosphere. For example, bubble plumes of CH4 from the seabed have been observed in the water column, but not detected in the Arctic atmosphere (Westbrook et al., 2009; Fisher et al., 2011)."

Vegetation. Plants are powerful channels for methane escaping past the jaws of hungry methanotrophs. In wetlands, cotton grass seems to do this; in the tropics tree transpiration brings methane up from anaerobic soil methanogens. Pangala's work is appropriate here. What is interesting as an aside is that SCIAMACHY was used to bolster the plant methane story – it illustrates the risk of simplistic interpretation of retrievals, especially from some regions with near 100% thick wet season cumulo-nimbus cloud cover in daytime.

Since the first submission, we integrated a new co-author working (Dr. Covey) on methane emissions from the vegetation and we reviewed the original paragraph, still acknowledging that this source needs more studies to be more quantitatively estimated. The paragraph has been replaced with the following:

"A series of recent studies define three distinct pathways for the production and emission of methane by living vegetation. First, plants produce methane through an abiotic photochemical process induced by stress (Keppler et al., 2006). This pathway was criticized (e.g. Dueck et al. 2007; Nisbet et al. 2009), and although numerous studies have since confirmed aerobic emissions from plants and better resolved its physical drivers (Fraser et al. 2015), global estimates still vary by two orders of magnitude (Liu et al. 2015) meaning any potential implication for the global methane budget remains highly uncertain. Second, plants act as "straws", drawing methane produced by microbes in anoxic soils (Rice et al., 2010). Third, the stems of living trees commonly provide an environment suitable for microbial methanogenesis (Covey et al., 2012). Static chambers demonstrate locally significant through-bark flux from both soil- (Pangala et al., 2013, 2015), and tree stem-based methanogens (Wang et al., 2016). These studies indicate trees are a significant factor regulating ecosystem flux, however, estimates of biogenic plant-mediated methane emissions at broad

C17

scales are complicated by overlap with methane consumption in upland soil and production in wetlands. Integrating plant-mediated emissions in the global methane budget will require untangling these processes to better define the mechanisms, spatio-temporal patterns, and magnitude of these pathways."

OH oxidation. This is among the very largest unknowns in the global budget. The discussion is appropriate: perhaps it could be expanded a little, given the significance of the uncertainty.

Indeed some points were missing in this sub-section. Though, we are sure that we could say even more. We have adding discussion on the following points: NH/SH ratio, OH variation buffered by chemistry-transport connections in models and the impact of OH distribution on inferred methane budget. Formerly page 30, line 15: "Observations are generally carried out within the boundary layer, while the global OH distribution and variability are more influence by the free troposphere (Lelieveld et al., 2016)"

Formerly page 30, line 20: "However, it is worth noting that, in the ACCMIP estimations, the differences in global OH are larger between models than between pre-industrial, present and future emission scenarios simulations. Indeed Lelieveld et al. (2016) suggest that tropospheric OH is buffered against potential perturbations from emissions, mostly due to chemistry and transport connections in the free troposphere, through transport of oxidant such as ozone. Besides the uncertainty on global OH concentrations, the OH distribution is highly discussed. Models are often high biased in the northern hemisphere leading to a NH/SH OH ratio greater than 1 (Naik et al., 2013). A methane inversion using a NH/SH OH ratio higher than 1 infers higher methane emissions in the northern hemisphere and lower in the tropics and in the southern hemisphere (Patra et al., 2014). However, there are recent evidence for a parity in inter-hemispheric OH concentrations (Patra et al., 2014)."

Stratospheric loss. Again, this is a large factor and may be changing with the changing incidence of tropical clouds pushing up the tropical tropopause as global warming expands the tropics. The Brewer-Dobson circulation could be mentioned, and the impact of the polar vortex in bringing down depleted isotopically heavy air to the Arctic?

There are not many studies on methane stratospheric loss, leading to a short paragraph. However this issue probably needs to be more addressed in methane chemistry modeling, as mentioned in section 6. We now mention the Brewer Dobson circulation and the polar vortex (formerly on page 31, line 23): "[...] (Reeburgh, 2007). Stratospheric CH4 distribution is highly correlated to the changes in the Brewer Dobson circulation (Holton, 1986) and may impact Arctic air through subsidences of isotopically heavy air depending on the polar vortex location (Röckmann et al., 2011)"

Soil methanotrophy. The work cited is old and derivative. Maybe in the next update some of the more recent boreal/Arctic findings could be included.

We agree to integrate the recent findings in boreal regions in the next release of the budget as suggested by the referee.

Lifetime. (includes soil and Cl as well as OH). Maybe recapitulate on the difference between different definitions of "lifetime". Dlugokencky's point about the 9.3 yr equilibration time is powerful (mentioned in 4.1.1).

The definition of global atmospheric lifetime is attributed to a gas in steady state and

C19

corresponds to the global atmospheric burden (Tg) of this gas divided by its mean global sink (Tg/yr). In a case of a gas whose local lifetime is constant in space and time, the atmospheric lifetime equal the decay time (e-fold) of a perturbation. This is not true for gases whose lifetime is shorter than tropospheric mixing. Methane lifetime is longer than tropospheric mixing, and its lifetime is supposed not to be significantly affected by the location of the sources. Also methane is not in a steady state, this is why when calculating methane lifetime using atmospheric measurements we need to fit with a function that approaches steady state (Sect. 4.1.1). Indeed for methane, two lifetimes are generally defined (Stevenson et al. (2006) or Naik et al. (2013):

- its tropospheric lifetime corresponding to the burden divided by the loss from OH oxidation in the troposphere, sometimes called "chemical lifetime"

- its total lifetime corresponding to the burden divided by the total loss, (tropospheric loss from OH oxidation + stratospheric loss + soil sink)

The results from the steady-state calculation from atmospheric observations are consistent with the model results of total methane lifetime.

We have detailed the definition of lifetime in Section 3.3.5 and modified the text as follows:

"The global atmospheric lifetime is defined for a gas in steady state as the global atmospheric burden (Tg) of this gas divided by its mean global sink (Tg/yr). In a case of a gas whose local lifetime is constant in space and time, the atmospheric lifetime equal the decay time (e-fold) of a perturbation. As methane is not in a steady state, we need to fit with a function that approaches steady state when calculating methane lifetime using atmospheric measurements (Sect. 4.1.1). For modeled methane, two lifetimes are generally defined and computed as in Naik et al. (2013). First, its tropospheric lifetime corresponding to the burden divided by the loss from OH oxidation in the troposphere, sometimes called "chemical lifetime". Second, its total lifetime corresponding to the burden divided by the total loss: tropospheric loss from OH oxidation, the stratospheric loss and the soil sink. The tropospheric methane lifetime is of 9.3 years (range

[7.1-10.6], Voulgarakis et al. (2013)); K13) and the total methane lifetime of 8.2  $\pm$  0.8 years (for year 2000, range [6.4-9.2], Voulgarakis et al. (2013)). The model results of total methane lifetime are consistent with, though smaller than, the value reported in Table 6.8 of the IPCC AR5 of 9.1  $\pm$  0.9 years (which was the observationally constrained estimate of Prather et al. (2012)) most commonly used in the literature (Ciais et al., 2013) and the steady-state calculation from atmospheric observations (9.3 yr, Sect. 4.1.1)."

4. Observations Satellites. This is a very helpful discussion. The visually very impressive maps and sweeping conclusions from satellite studies have perhaps exerted an influence on the appreciation of the global budget, that glosses over the problems of bias, clouds and aerosols, and the problems of Arctic cover. Satellites are extremely important and powerful in their inputs, but the retrievals need to be evaluated in light of their uncertainties and inherent biases. That said, satellite results are vital in understanding the lightly-monitored tropics. The point that 'satellite' based inversions include from-the-ground priors is usefully made.

Other atmospheric observations. The IASI and TCCON discussions are good. Perhaps more could be said about the incoming use of mobile CRDS and also drones for low-altitude work. This is a major area of advance.

Satellite limitations are acknowledged in the paper (formerly) page 36 lines 20-26. We agree that new technologies using CRDS offer great opportunities for the near future. We add a few sentences in (formerly) Section 4.1.3 (formerly page 36, line 10): "New technologies have also developed systems based on cavity ring down spectroscopy (CRDS), opening a large ensemble of new activities to estimate methane emissions such as drone measurements (light version of CRDS), as land-based vehicles for real-time, mobile monitoring over oil and gas facilities,

C21

## as well as ponds, landfills, livestock etc..."

First, I must admit bias, but the isotopic section squeezed into the bottom of 4.1.3 is surely the weakest part of the paper. The discussion is OK but too brief. Isotopes are very powerful as source discriminators, and indeed for the insight they give into sinks too. Methane measurements provide four sources of information: mole fraction, *C*-isotopic ratio, D/H, and back trajectory of the air mass. Just using mole fraction and trajectory is a 2D view: in fact the full 3D picture is now becoming adequate to support inverse modelling, and hopefully 4D information will soon be available. The global budget will not be solved until the full range of isotopic information is used. Thus this treatment is very inadequate and would benefit from a significant upgrade in the next generation of the GCP work, 2-3 years from now. C-14: Lassey's 2007a paper is important, and is a major insight, but it only goes to 2000, and perhaps the weight placed on it in the conclusion is too substantial.

We agree that the isotopes deserve their own sub-section: this was our first plan, but the length of the paper pushed us to include them into the "other observations" section. Based on the recommendation of the referee, we went back to our first plan and created a specific sub section for isotopes before the "other observations section". The discussion on the contribution of biogenic versus thermogenic (and pyrogenic) sources is there, but short. In the future release, we may focus on the uncertainties of the isotopic signatures (not done here), which bring also uncertainties in modeling results. We agree that more modeling results using isotopes would be needed in the future to better constrain the methane budget. As a result, for the present submission, the text in this isotope subsection has been only enriched with relevant recent studies and reformulated. However we are willing to further develop this section it in future papers, including the impact of emerging continuous 13C concentrations based on laser technics, the integration of atmospheric inversions

#### 4.1.3 Methane isotope observations

The processes emitting methane discriminate differently its isotopologues (isotopes). The two main stable isotopes of CH4 are 13CH4 and CH3D, and there is also the radioactive carbon isotope 14C-CH4. Isotopic signatures are conventionally given by the deviation of the sample mole ratio (for example, R=13CH4/12CH4 or CH3D/CH4) relative to a given standard (Rstd) relative to a reference ratio, given in per mil as in equation 3.

$$\delta^{13}CH_4 \text{ or } \delta D(CH_4) = (\frac{R}{R_{std}} - 1) * 1000$$

For the 13CH4 isotope, the conventional reference standard is known as Vienna Pee Dee Belemnite (VPDB), with Rpdb=0.0112372. The same definition applies to CH3D, with the Vienna Standard Mean Ocean Water (VSMOW) RSMOW=0.00015575. The isotopic composition of atmospheric methane is measured at a subset of surface stations (Quay et al., 1991; 1999; Lowe et al., 1994; Miller et al., 2002; Morimoto et al., 2006; Tyler et al., 2007). The mean atmospheric values are about -47% for  $\delta$ 13CH4 and 86-96% for  $\delta$ D(CH4).  $\delta$ 13CH4 measurements are made mainly on flask air samples analysed with gas-chromatograph isotope ratio spectrometry for which an accuracy of 0.05%. for  $\delta$ 13CH4 and 1.5‰ for  $\delta$ D(CH4) can be achieved (Rice et al., 2001; Miller et al., 2002). These isotopic measurements based on air flask sampling have relatively low spatial and temporal resolutions. Laser-based absorption spectrometers and isotope ratio mass spectrometry techniques have recently been developed to increase sampling frequency and allow in situ operation (McManus et al., 2010; Santoni et al., 2012). Measurements of  $\delta$ 13CH4 can help to partition the different methanogenic processes of methane: biogenic (-70% to -55%,

C23

thermogenic (-55‰ to -25‰ or pyrogenic (-25‰ to -15‰ sources (Quay et al., 1991; Miller et al., 2002; Fisher et al., 2011) or even the methanogenic pathway (McCalley et al., 2014).  $\delta D$ (CH4) provides valuable information on the oxidation by the OH radicals (Röckmann et al., 2011) due to a fractionation of about 300‰Ålso emissions also show substantial differences in  $\delta D$ (CH4) isotopic signatures: -200‰ for biomass burning sources versus -360 to -250‰ for biogenic sources (Melton et al., 2012; Quay et al., 1999).

14C-CH4 measurements (Quay et al., 1991; 1999; Lowe et al., 1988) may also help to partition for fossil fuel contribution (radiocarbon free source). For example, Lassey et al. (2007a) used more than 200 measurements of radioactive 14C-CH4 (with a balanced weight between Northern and Southern hemispheres) to further constrain the fossil fuel contribution to the global methane source emissions to  $30\pm2\%$  for the period 1986-2000.

Integrating isotopic information is important to improve our understanding of the methane budget. Some studies have simulated such isotopic observations (Neef et al., 2010; Monteil et al., 2011) or used them as additional constraints feeding inverse systems (Mikaloff Fletcher et al., 2004; Hein et al., 1997; Bousquet et al., 2006; Neef et al., 2010; Thompson et al., 2015). Using pseudo-observations, Rigby et al. (2012) found that Quantum Cascade Laserbased isotopic observations would reduce the uncertainty in four major source categories by about 10% at the global scale (microbial, biomass burning, landfill and fossil fuel) and by up to 50% at the local scale. Although all source types cannot be separated using 13C, D and 14C isotopes, such data bring valuable information to constrain groups of sources in atmospheric inversions, if the isotopic signatures of the various sources can be precisely assessed (Bousquet et al., 2006, supplementary material).

4.1.4 Other atmospheric observations (With TCCON, Aircore, balloon, aircraft, drones, ...)"

The following text (on recent changes and trends) has been moved to the conclusion of the paper:

"Since 2007, the average annual methane growth rate amounts to 5.5pm0.6 ppb yr-1. Scenarios of increasing fossil and microbial sources have been proposed to explain the sustained increased growth rate since 2007 (Bousquet et al., 2011; Bergamaschi et al., 2013; Nisbet et al., 2014). Whereas the decreasing trend in  $\delta$ 13C in CH4 suggests a significant, if not dominant, contribution from increasing emissions by microbial CH4 sources (Schaefer et al., 2016; Nisbet et al., 2014), concurrent ethane and methane column measurements suggest a significant role (likely at least 39%) for oil and gas production (Hausmann et al., 2016), which could be consistent when assuming a concomitant decrease in biomass burning emissions (heavy source for 13C), as suggested by the GFED database (Giglio et al., 2013)."

Inversions The point that inversions use B-U or T-D priors is well made, as is the problem of the large corrections placed on satellite CH4 results (which are, strictly, not 'data' but interpretations). Perhaps there could be a little more discussions of the weaknesses of chemical transport models.

We use data and not observations as indeed satellites provide radiances, interpreted then into methane column through complex inverse modeling of radiative transfert. We think "data" is appropriate as also exists "model data". The word retrieval is another solution. Some of the weaknesses of the chemistry transport models used in the inversion are discussed later and are put in perspectives in Section 6. However, we have added in Sect 4.2.2 a few sentences on the uncertainties due to model weaknesses (former page 39, line 4).

C25

"This approach is appropriate for our purpose of flux assessment, but not necessarily for model inter-comparison. We did not require posterior uncertainty from the different participating groups, which may be done for the next release of the budget. Indeed chemistry transport models have some limitations that impact on the inferred methane budget, such as discrepancies in inter-hemispheric transport, stratospheric methane profiles, OH distribution. We consider here an ensemble of inversions gathering a large range of chemistry transport models, through their differences in vertical and horizontal resolutions, meteorological forcings, advection and convection schemes, boundary layer mixing; We assume that this model range is sufficient to cover the range of transport model errors in the estimate of methane fluxes. "

5. Methane budget The study points at the biggest hole in the budget: inland water emission (i.e. open water that is either free-running or more than, say, 1m deep and say a 10 m2 in area). Bottom-up, natural methane is 50% of sources, Top-down, natural sources are about 60%. That's a big difference relevant to the actions of policy- makers. It needs to be investigated. Also, the year-to-year variability of tropical wetland emissions has been studied by various authors – variability is not discussed much in this paper but perhaps variability deserves better, and could be given more attention in a later GCP report? Geological emissions are also a parameter needing re-study. As noted above, the 14C insight mentioned is pre-2000. Interestingly, TD and BU inventories do agree reasonably for anthropogenic emissions. But as the study indicates, that probably needs caution.

About variability in the methane budget. As presented (only decadal budget) the paper is long enough and adding discussion on variability would have made it too dense. As questioned by the referee, variability of the methane budget will be discussed in a second paper that was not ready when the ESSD review was submitted. Some of the missing points of this paper will be found in the second one. The need to better assess natural emissions (inland water and geological) is highlighted in the discussion section (Sect 6).

Regional Budgets. A very useful part of the study is the emphasis on emissions from the tropics – Africa, S. America and tropical Asia. These regions are poorly instrumented and in the wet season are covered by dense thick clouds, hard to see through. The focus on these regions is an important and valuable part of the study.

The study shows that there is a major discrepancy between N. American B-U and T-D Inventories – perhaps that's in part an example of the bias of science funding regimes encouraging 'discovery' of larger sources. and discouraging contrary studies that set out to test hypotheses of low emissions.

The result that China's emissions may have been overstated is enormously interesting. Possibly this is linked to the poor quality coal that is being burnt in some cases – high silicate content, so tonnages may be exaggerated as a lot of the mass may be shale. Thompson et al (JGR 2015) is relevant to China's emissions.

Chinese emissions are overestimated in EGDAR inventory. Peng et al. 2016 and our results suggest that this due to an overestimations in coal emissions. Yet Thompson et al. 2015) point toward rice emissions mainly. We have added this discussion in the revised version of the manuscript. Formerly page 44, line 13. "Thompson et al. (2015) showed that their prior (based on EDGARv42) overestimated the Chinese methane emissions by 30%, however they found no significant difference in the coal sector estimates between prior and posterior and attribute the difference to rice emissions."

C27

The finding that southern African wetlands are important is interesting. Hitherto the enormous upper Congo wetlands have been largely neglected, but the Chambesi and Luapula swamps are enormous, as are those in the upper Zambesi, and parts of Angola and DRC. The Mweru and Bangweulu wetlands in particular could benefit from study: as someone who has been near the source of the Chambesi, a personal comment is that Northern Hemisphere scientists tend to forget this region.

Thanks for this precise comment. Indeed more regional studies on wetland emissions are needed for tropical regions, onsite but also using satellite if progresses are made on systematic errors as mentioned in the paper.

6. Future developments. The focus is clear – we need better information on freshwater emissions, biomass burning, and better information in the tropics. Personally I would add better isotopic coverage to this list: they seem much neglected, almost forgotten, in this GCP study but perhaps that's a matter for the future: the next update in a couple of years' time could address them.

Again we acknowledge the lack of isotope discussion. We have partly addressed this missing point through the previous comments, and added some more discussion on isotopic future studies in Section 6. For sure, this will be further developed in the next release.

7. Conclusions These are sensible. The study usefully points to the latitudinal source breakdown - 2/3 tropical, 1/3 temperate, minor Arctic, and the need for better in situ tropical measurement. The importance of studying the variability of tropical wetland emissions is perhaps in need of more emphasis. Inventories suffer in general from assumptions that year-on-year changes are small, and that seasonal changes can be

ignored. Both these weaknesses need to be addressed.

In this release variability and trends have not been discussed in this paper but in a second paper, which will be submitted soon. However a better knowledge on emission variations would definitely help improving the methane budget understanding. Such conclusion will definitely be part of the second paper. Here, we put one short paragraph on variability at the end to announce this future paper.

Overall, this is an immensely valuable study, of major importance, that should be published with the most minor of changes.

We thank again Euan Nisbet for his careful and detailed reading of our paper.

Minor Comments

1. Kirschke et al. 2013 is referred to as K13 in reference call-outs. This is unsettling: it would be better to give the full name.

- 2. Page 9 has a typo in line 4 Artic.
- 3. Page 45 has a typo in line 9 resultas.
- 4. In the acknowledgements, spell out P. Bous.
- 5. Page 67 UNFCCC typo ational.

All these minors typo errors have been corrected in the revised version of the manuscript.

Interactive comment on Earth Syst. Sci. Data Discuss., doi:10.5194/essd-2016-25, 2016.

C29