

Detailed Response to Anonymous Referee #2

We acknowledge anonymous referee #2 for his/her time spent on reading and commenting on the paper, providing comments and helpful suggestions to improve the manuscript, in particular the English and phrasing of some sentences, and citations that were missing.

This is a timely, thoughtful, thorough, and important work from the scientific community involved in the Global Carbon Project. This effort focuses on the sub-decadal variability in an effort to the apparent, vexing shifts in the atmospheric growth rate of methane (CH₄). It takes a measured approach to attributing the cause of CH₄ variability in terms of natural (although perhaps perturbed by climate change) and direct anthropogenic sources. It also nicely consolidates the top-down inversions and the bottom-up emission inventories. It does not deal with the possible changes in atmospheric sinks (OH), although the evidence for large variability in the sink are proposed in some recent papers, but remain entirely obscure. This paper takes a balanced approach and could be published as is, or with some minor revisions suggested below. My apologies for the delay in reading/reviewing this manuscript. Request: Can we please all go to continuous line numbering so that it is easy to read sections and refer to them without finding the page number?

P4L18. The refs for OH chemistry models are fine as an overview, but the Holmes et al (2013, you have it later in the paper) is a good example of multi-model assessment of the interannual variability in the OH sink and its possible causes that is not found in the ACCMIP studies.

We thank the reviewer for the additional **reference** that has been **added** at this place of the text.

P5L15. The cryo-ethane history has proven useful in evaluating fossil fuel emissions and inferring ff-CH₄ sources (Aydin et al., Nature 2011, 476:198-201, 2011; Nicewonger, et al. GRL 43:214–221, 2015), and these are more relevant here than the LA-basin study of Wennberg.

We thank the reviewer for these two relevant papers, both using cryo-observations of the atmospheric composition. **These two papers** have been **added** on top of the Wennberg et al., which used in-situ present atmospheric observations.

P6L16-17. I think you mean "the first GCP global methane budget :"

Indeed, it would not be fair to pretend that other methane reviews do not exist. This has been modified.

P6L22ff. I think that this phrase is close but could be better "as most of the inversions used here assume constant OH concentrations over years, generally only optimizing its mean global concentration against methyl chloroform observations (e.g. Montzka et al. (2011))." What the models assume is not constant OH but rather constant CH₄ loss frequency (with respect to OH). These are not the same, since if temperature changes then the constant OH will result in different CH₄ loss. Moreover, the methyl chloroform decay records a mean loss frequency and not a mean OH as is frequently used. I suggest we move on to more accurate statements like: "as most of the inversions used here assume constant methane loss to OH over the time period, consistent with the observed decay of methyl chloroform (e.g. Montzka et al. (2011); Holmes et al, 2013)."

We thank the reviewer for this very interesting comment. Actually, there is probably a misunderstanding in the way MCF and CH₄ inversions are done. In the chemistry transport models used in inverse modeling, the chemical loss of a compound through OH is calculated at each time step using OH (prescribed), the compound concentrations and the reaction constant (driven by the temperature 3D field generally nudged to ECMWF inter-annual reanalyses). For MCF inversions a scaling factor is optimized for the loss, but then it is used to get inter annually varying OH (few inversion) or a seasonally-varying climatological OH

(most inversions), that is then prescribed to CH₄ inversions. Therefore, CH₄ inversions do not prescribe the pre-optimized loss but the MCF-derived OH fields, which may introduce some inconsistency about the impact of temperature changes on the loss. This effect is absorbed in OH variations as it is done currently. It remains probably small when looking only to 1-2 decades but could be significant during large climate events such as El Niño.

This part has been re-written as follows: “ **However, we do not address the contribution of the methane sinks during this period. Indeed, for most of the models, the soil sink is from climatological estimates and the oxidant concentration fields (OH, Cl, O₁D) are assumed constant over the years. The global mean of OH concentrations was generally optimized against methyl chloroform observations (e.g. Montzka et al. (2011)), but no inter annual variability is applied.**”

P9L11-15. You really need to note that if these models used their own OH & T fields that the CH₄ budget would vary by 30% or more. It is because they use an accepted OH-lifetime for methane (e.g., Prather GRL 39:L09803, 2012) that top-down agrees so closely.

This is true that the agreement between the inversions is linked to the use of similar OH concentrations and temperature fields. We have added the following sentence: “**It is to be noted that this rather good agreement between these estimates is linked with the associated rather small range of global sinks. Indeed, most inversions use similar MCF-constrained OH fields and temperature fields.**”

P14L2. typo: constrained to constrain.

This has been corrected

P14L8. While I tend to believe the Bruhwiler paper and not trust the cherry-picked satellite data over the US, you might consider referencing these two papers (Turner, Jacob, et al. GRL, 43:2218–2224, 2016; Schneising et al. Earth’s Future 2:548–558, 2014).

Following your comment and reviewer#1 comment on this paragraph. The paragraph as been changed as follows: “**Also, temperate North America does not contribute significantly to the emission changes. Contrary to a large increase in the US emissions suggested by Turner et al. (2016), none of the inversions detect, at least prior to 2013, an increase in methane emissions possible due to increasing shale gas exploitation in the U.S. Bruhwiler et al. (2017) highlight the difficulty of deriving trends on relatively short term due to in particular inter annual variability in transport.**”

P14L27. the phrase "are assumed not to contribute" is awkward. At first it sound like this paper assumes this, but what you mean is "are assumed in these model studies not to .."?

This has been rephrased as suggested.

P15L17. awkward end: "emissions occur partly over the same areas:

This has been rephrased to: “**emissions may both occur in the same or neighboring model pixels.**”

P15L18. drop the 'of' to make it a sentence.

This has been corrected.

p15L23. Now you jump from fluxes (Tg/y) in the above to trends/accelerations (Tg/yr²). How about using this line to transition to translate this difference in emissions to a trend: " and 2008-2012, i.e., a trend of about +1.7 Tg CH₄ yr⁻²).

Thank you for pointing this. The suggested change has been done as follows: “**For China, bottom-up approaches suggest a +10 [2-20] Tg CH₄ yr⁻¹ emission increase between 2002-2006 and 2008-2012, i.e. a trend of about 1.7 Tg CH₄ yr⁻² (considering a 10 Tg yr⁻¹ increase over 2004-2010), which is much larger than the top-down estimates.**”

P16L2. ?? "change, and this result holds similarly for : : :"

Thank you for the rewriting, this has been modified.

P17L11. typo: "..that the increase in methane emissions between: : :"

This has been corrected.

P17L18ff. Please revise this sentence and make more, shorter ones. I was totally lost at the "although". " The sectorial partitioning from inversions is in agreement (within the uncertainty) with bottom-up inventories (noting that inversions are not independent from inventories), though the top-down ensemble significantly decreases the methane emission change from fossil fuel production and use compared to the bottom-up inventories, although the estimate of the latter should decrease with the upcoming revised version of the EDGAR inventory (see Sect. 3.2.4)."

Indeed... This has been changed to: **"The sectorial partitioning from inversions is in agreement (within the uncertainty) with bottom-up inventories (noting that inversions are not independent from inventories). However the top-down ensemble significantly decreases the methane emission change from fossil fuel production and use compared to the bottom-up inventories. In the coming years, the revised version of the EDGAR inventory (see Sect. 3.2.4) should decrease the estimated change by bottom-up inventories, reducing the difference between bottom-up and top-down estimates."**

P17L31. " the spread of land surface models" Please pick a better word than "spread": these models do not grow like forests:

This has been rephrased to: **"The range of the methane emissions estimated by land surface models driven with the same flooded area extent shows that [...]"**

P18L8. Fix up: " wetland emissions per Meter Square." and put the Poulter ref at the end of the sentence if possible.

The sentence has been rephrased as follows: **"However, no significant trend in tropical surface temperature is inferred over 2000-2012 that could explain an increase in tropical wetland emissions (Poulter et al., in review)."**

P18L16. I think you do not want 'incorrectly' in this sentence, the following clause says it all: "Even though top-down approaches may incorrectly attribute: : :"

'incorrectly' has been removed from the sentence as follows: **"Even though top-down approaches may attribute the emissions increase between 2002-2006 and 2008-2012 to tropical regions (and hence partly to wetland emitting areas) due to a lack of observational constraints, it is not possible, with the evidence provided in this study, to rule out a potential positive contribution of wetland emissions in the increase of global methane emissions at the global scale."**

P19L5. easier to read as: "..changes leads, as expected, to unrealistically: : :"

This has been corrected accordingly.

P19L19. put a comma between the two independent clauses: " than constraints, and other :"

This has been added in the sentence.

P19L21-35. Here is maybe where it is worth looking at the firn-air record showing ethane decreases (Aydin & Nicewonger refs above).

Nicewonger et al. results span only to 1918 and could not provide any insight in this discussion on the recent change. The Aydin study used firn air data and discussed fossil fuel emissions change from 1900 to 2010. This is not completely compatible with the period discussed here so we added a sentence about these historical papers in the introduction:

“The historical record of atmospheric ethane suggests an increase of ethane sources until the 1980s and then a decrease driven by fossil fuel related emissions until the early 2000s (Aydin et al., 2011).”

P20L3-6. This sentence does not really belong in the "Ethane" discussion? " Besides, the recent bottom-up study of Höglund-Isaksson (2017) shows relatively stable methane emissions from oil and gas after 2007: : :."

The Höglund-Isaksson study does not use ethane measurement, however they show constant emission from the oil and gas sector. This result disagrees with the ethane-based study and is worth noting in this context. As a result, we decided to change the paragraph title to **“Oil and gas emissions, and ethane constraint”**

P20L17-29. This OH section is bothersome. I think you mean that models assume constant methane loss frequency – OR if they fix the 3D OH distribution, then the interannual temperature variations will drive changes in methane loss. I think they do the former and hence the correct wording would be "assume constant OH-lifetime for methane" or "assume constant methane loss frequency." These cannot just assume a uniform OH-loss because then they miss the seasonal and latitudinal gradients.

As explained above this is the second statement that occurs in practice in current inversions. Indeed, inverse modellers prescribed climatological OH (with seasonal variations) and compute the loss using varying CH₄ and temperature. This might not be fully consistent but we clarified the method in the text.

I also recommend that the authors also look at the trends in methane's OH-lifetime from the Holmes et al 2013 paper. Several models show no trends from 2006 to 2010. If anything all the models show a decreasing methane OH-lifetime from a high in 2004 to a low in 2010, an 'OH' increase of about 3%. Moreover, one model running both with GEOS MERRA vs. GEOS6 shows different trends. The Dalsøren 2016 paper is very interesting, but it is only one model – further, this Oslo CTM3 shows different trends than the same model in the Holmes paper. I am not sure which is the better result, but some caution is due. Interestingly, all the models get the big increase in OH across the 1997-89 ENSO year.

We acknowledge the caution suggested by the reviewer. **And modified the text accordingly in the last paragraph of the paper.**

P20L26. "However, decreasing OH concentrations since 2008 would require smaller emission changes to explain the observed atmospheric methane increase, also possibly implying .." This is confusing since both the Dalsoren and Holmes papers show a decrease in lifetime (2% possibly) and hence an increase in OH after 2008.

Figure 1 of Holmes 2013 and figure 15 of Dalsoren 2016 are consistent until 2007. Holmes stops in 2009 but Dalsoren shows stabilizing OH after 2007. **We rephrased this paragraph to better show the remaining uncertainties on OH variations.**

P21L27-30. Again, please check that the models kept the methane OH-lifetime (effectively the inverse loss frequency) fixed and did not freeze OH concentrations, allowing the rate coefficient to vary with temperature as it should, because then the temperature fluctuations could drive %-level variability. Also I think you have the Dalsoren paper backwards: their Fig 15 (&18) shows a steadily increasing methane loss frequency (1/lifetime, left scale) since the 1997-98 ENSO and up to 2010; the year 2008 is the only reversal of this. Their calculated change in OH does not match the CH₄ lifetime, and it is the lifetime that determines the annual loss of CH₄.

Again, as explained above, CTMs implied in inversions prescribe OH change and recomputed the loss using inter-annually varying meteorology. **The paragraph was rephrased to better reflect what was done and the remaining uncertainties.**

P21L33. "uncertainties" is odd. I am not sure we know enough to even assess the uncertainty. how about "major disagreements in OH fields simulated by the models."

We agree on this comment and have changed the sentence as suggested to: **"Estimating and optimizing OH oxidation in top-down approaches is challenging due to the major disagreements in OH fields simulated by the models."**

P22L1. It is the fact that we stopped using MCF and it is decreasing rapidly, that makes it a good surrogate for the methane OH-lifetime. When in use the uncertainty in emissions made it difficult to get better than 10-20% accuracy and variability.

We agree and have rephrased this part of the conclusion: **"Although beneficial for the recovery of the stratospheric ozone, methyl-chloroform, which is used as a proxy to derive OH variations, is decreasing rapidly in the atmosphere. MCF is therefore less sensitive to uncertain and larger emissions as in the 1980s and 1990s (e.g. Kroll et al., 2003; Prinn et al., 2001), but within years, will also be less useful to derive OH changes as its atmospheric concentrations are getting as small as the precision and accuracy of the measurements. "**

P22L5. I am not sure that this comparison with CO2 is useful or accurate. There are many thorny problems left with the CO2 budget and climate feedbacks. Stop at "understood."

The sentence has been stopped at "understood" as suggested.