Reply to Interactive comment on "Variations in global methane sources and sinks during 1910–2010" by A. Ghosh et al.

Anonymous Referee #2

Thank you very much for appreciating the work and helping us with helpful comments and suggestions for further improving the clarity of the paper. In addition to incorporating changes as per your comments, we have made following major/notable changes; (1) Figure 1 now uses new dating of the air Firn air bubbles in NEEM ice core, (2) Figure 5 is considerably remade by using new air aged NEEM data, and includes the inter-polar differences for individual data points (in addition to corrected observation, calculated from spline fitted line).

This paper presents a reconstruction of the methane emissions from 1910 to 2010 using 1/ an ensemble of methane atmospheric concentrations reconstructed data from polar archives (ice cores and firn) or directly observed in the atmosphere (since 1979), 2/ a chemistry-transport model, 3/ an initial scenario of emissions, and 4/ a massbalance procedure to optimize global emission incrementally from the initial scenario. The addition of 13CH4 data (also from ice cores, firn and direct observations) allows the authors to propose a partition of the increment in methane emissions between a light source (identified as biogenic) and a heavy source (identified as biomass burning).

General comments
The paper treats the important aspect to better understand past methane emissions in order to improve 1/ our present understanding of the global methane cycle, and 2/ the future emission scenarios for climate projections.

The main strength and originality of the paper is to bring together 12C and 13C constraints on the global methane budget over a century. Although limited to fully separate all the components of methane emissions, the use of 13C brings interesting new constraints.

The main weakness is the lack of precision in the writing all along the text, and more specifically in the methodology part (section 2). My recommendation is to re-write the method section with more precision and sometimes more details (see the numerous specific comments below) in order that the reader does not have to guess what is done behind the lines later in the results section. For instance the model description is not clear, the procedure of optimization is unclear, the construction of emission scenario also, only in the result section is clearly mentioned that only Antarctica data only are used for the optimization, a two-box model appear in the middle of the result section, …

A lot of assumptions are made in such an integrated system. I recommend that, whenever it is possible, the authors mention the impact of choices made on their results:
initial conditions, OH field used (constant, NH/SH ratios ?), isotopic signatures chosen, … See the different specific comments about this point below.
Ans. We agree with on the assessment of the weaknesses and appreciate pointing this to us. We are making modifications to better clarify the presentation of the work.

Overall, I recommend publication after addressing all issues raised in my general and specific comments

Specific comments
Abstract : Replace during the 2000s by in 2010 Replace the causes of the CH4 increase by something like Â´n the detailed causes Â¨z as we know that emission have increased, although not in detail which category increase when A suggestion : During 1910–2010, the global total CH4 emission doubled from 290Tg/yr to 580Tg/yr.
Ans. These corrections have been made

P4:
L10: Replace The present-day concentration of CH4... by the 2010 concentration of …
L19: resumed strong growth again starting in 2007 (Rigby et al., 2008; Dlugokencky et al., 2009) : please give the average growth rate since 2007.
L22: in relation with : : : and climate change : also mention tropospheric oxidant changes
Ans. Modifications made following above suggestions

P5:
L11: They are not tested for : : : : They did not address the latest: : :
L17: can be extended from today back to the mid-20th century : time scale in the two directions was not clear
Ans. Modifications made following above suggestions. The sentence in Line 17 now ends as "…to the mid-20th century from the recent decades"

P6:
L10-14: please rephrase the long sentence to make it more clear.
Ans. Sorry for this long sentence, which contained 4 long acronyms. The sentence sounds fine to us. However, suggestion for modification is welcome.

§2.1: the model description has to be rewritten as it is confusing as it is in the submitted paper. Do you use the offline model (ACTM) of a GCM (AGCM)? If yes, precise how the air mass fluxes are computed. Be more clear that you do not nudge winds in the GCM but that when doing so when analysed winds become available, the differences remain limited.
Ans. Following description is added:
"The basic physical and dynamical features of the AGCM have been described in (Hasumi et al., 2004). Advective transport of moisture and tracers is obtained from a 4th order flux-form advection scheme using a monotonic Piecewise Parabolic Method (PPM) (Colella and Woodward, 1984) and a flux-form semi-Lagrangian scheme (Lin and Rood, 1996). Subgridscale vertical fluxes of heat, moisture, and tracers are approximatedusing a non-local closure scheme in conjugation with the level 2 scheme of Mellor and Yamada (1974). The cumulus parameterizationscheme is based on Arakawa and Schubert (1974). The updraft and downdraft of tracers by cumulus convection are calculated by using the cloud mass flux estimated in the cumulus parameterization scheme. "

P7:
L16: How do you choose the different scaling factors. As the system is underdetermined, there are many solutions. Please be more precise here. L18-23: It is not clear what the different versions of EDGAR are providing (maps or only totals per sector. I suggest a table here or to clarify the text.
Ans. These scaling factors are applied to the inventory emissions so that the sectorial emissions are in general agreement with those commonly used in the recent publications, and specifically with Patra et al. (2011). We have mentioned here: (please refer to Supplementary Materials, Table S1, for annual total emissions). The data are given as supplementary file. This has been stated in the text as "Scaling factors are set to simulate the CH4 growth rate approximately for the first decade 1901--1910, and are in close agreement with Patra et al. (2011) for the period 1990--2008."

P8:
L8: Do the BBG emissions include biofuel burning? If not, this number seems a bit large if one considers today GFED's emissions.
Ans. Yes, this/GISS biomass burning product accounts for all sources. We have inserted "(including biofuels)" when biomass burning is referred for the first time.

L12-15: Please be more precise on what causes the wetland emission changes: rainfall in

which region?, does temperature also plays a role ? I suggest to describe in a few lines how the VISIT model computes wetland emissions, as it is the largest individual flux.
Ans. The VISIT ecosystem model uses historical landuse change data from Hurtt et al. (GCB, 2006). The model climate is driven by CRU gridded precipitation and temperature data. Further details may be included in the revised version.

P10:
L17: Why not adjusting on the international reference scale (NOAA04) instead of a local scale? Please precise if offsets remain between cores/firn on one side and direct measurements on the other side when comparing the common period of times.
Ans. The Tohoku University CH_4 scale has been maintained well with long-term self-consistency since Aoki et al. (1992) and is well traceable to the NOAA04 scale through the Round-Robin activity (www.esrl.noaa.gov/gmd/ccgg/wmorr/wmorr_results.php?rr=rr5&param=ch4). As seen in Figure 2, all datasets are in agreement within variability after being referenced to the Tohoku University CH_4 scale. The Tohoku University scale is probably the oldest correct scale, and for your kind information the AGAGE network reports all CH4 data in this scale. Thus it may be unfair to call this scale a 'local scale'.

P11:
L16: 2.8696 : Are all these digits really significant?
L19-20 : the reason of the 3.7
Ans. These are changed to 2.87 (our result) and 2.77 (for Fung). The value in % is approximated as 4%.

P12: _ (delta) signs are missing in equation (3)
Ans. Sorry for missing this during the proofreading. Now corrected.

Eq (4): more explanations are needed here. Do you correct the trajectory year by year? How do you allocate a global _E to the gridded Eini to produce a gridded Eopt? Without the isotopes, is it proportional to the initial source partition? I understand you need to get gridded Eopt to re-run your model and produce the concentrations at Arctic and Antarctic sites, but again this is unclear. Please be much more precise on the procedure used here to get to Eopt.
Ans. We have added this text here:
"The calculation of global total Eopt is performed for each year. Emissions at all latitude-longitude grids are multiplied by a constant scaling factor (Eopt/Eini) to prepare revised emission for running ACTM."

L9: is known: I suggest to write "is prescribed"
Ans. Suggested change is incorporated.

P13/14
L19: 12CH4 is approximated by CH4 = 12CH4 +13CH4 : Does this mean that you assume 12CH4 CH4. Please clarify?
Ans. Appropriate modifications will be made in the revised version.

L10: I suggest to add a sentence announcing what follows in order to help the reader through §2.6: Â´n in the following, we calculate by two methods the isotopic signature for global emissions _13CE. First, …. Second, _113CE can also be calculated by considering the relative fraction… Simplify lines 4 to 11 in page 14 t2 times E=Eini+DE, confusing expression E(_13CE), …
Ans. Appropriate modifications will be made in the revised version.

L16 in p14: For simplicity, ….: please rephrase : In order to remove the underdetermination, we assume, …

Ans. Suggested change is incorporated.

P15/16
The discussion on fractionations is interesting but should be in a specific paragraph (2.7) And/or announced p13 line 17 when _i are presented. Else it comes too late after the method description.
Ans. We have added a sentence here as “The choice of alpha _i used in this work and their uncertainties are addressed later in this section.”

Figure 2
Legend error (FRIN instead of FIRN)
Again there is a lack of precision here on how the transport model is sampled. What do represent Antarctic and Arctic region listed in the legend? Do you extract al model boxes at the surface above 60N for Arctic and below 60_S for Antarctic? How sensible is it considering that you gather CGO with ice cores and firn from Antarctica. Please clarify and precise things here how you sample the model outputs (in the model section §2.1).
Use of plain/dashed lines for opt/ini model output would help on this plot. Choose one colour for NH and one colour for SH. If you only apply mass balance using Antarctica, then the Arctic can act as an evaluation of the method? If yes, this should be mentioned.
§3.2 and §3.3: I suggest exchanging these two sections as the reader expects (with the present paper’s construction) emission analysis first.
Ans. Suggested changes are incorporated.
At the end of section 2.4 (useful to have the site description for this to be convincing), we have added “The annual mean concentrations of ACTM simulations are sampled at Cape Grim for the Antarctic region and Summit for the Arctic region. The direct measurements at these two sites for the decade of 2000s show good agreements (model-data mismatch less than 3 ppb, discussed later in details) with the ACTM simulation using optimized emissions, and the Cape Grim data are also homogenized with those from Law Dome measurements.”

Suggested changes for replotting Figure 2 are incorporated.
The concept of using Arctic data for evaluation of the optimised emissions using Antarctic data is interesting. However, that is only true only when the latitudinal gradient of emissions is known. We would rather say that match we find for the Arctic data with ACTM simulation using E_opt is a good sign that the latitudinal gradient of emissions prescribed by EDGAR HYDE is reasonable.
We believed simple model details should be discussed first in section 3.2, such as the loss, before going in to emissions so that the rest of the paper can maintain a flow of discussion relating only to emissions. However, exchanging section 3.2 and 3.3 can still be done if so suggested.

P17:
L24: does the mean value of [OH] is optimized using MCF obs? Please precise if so.
Ans. We have inserted a sentence here “The OH field is scaled by 0.92 for simulating the decay rate of CH3CCl3 in Earth's atmosphere (Patra et~al., 2014).”

P18:
L11: please specify the link between tropospheric air temperature and reaction rates in the text. Is the impact of temperature larger on OH reaction rate?
Ans. We have added the CH4 loss reactions and temperature dependent reaction rates in Section 2.3 in this revised version.

L19: in the bottom-up emission data.. could be replace by : in the initial scenario
L19: 380
Table 2: please provide a table S2 (supplementary/appendix material) with annual Eini, Eopt, Burden.

Ans. Suggested to change is made in L19.
We have prepared a Table S2 and cited in the Table 2 caption.

P19
L14-17: very confusing sentence because your optimisation procedure is not well explained before. To me, if you prior Eini is imbalanced, your optimized Eopt is also as you only optimize a global number. Please clarifiy after improving the optimisation procedure section
We redone the calculation of IPD and Figure 5 is now revised significantly. The revised IPD calculation stems from newly estimated 'age of air' for the NEEM data, which is done by Dr. Cathy Trudinger of CSIRO (now a coauthor). The discussion in this paragraph is modified accordingly. Hope the revised Figure 5 and related text now reads better.

L19-22: The irruption of a 2) box model is a bit strange? How did you get the estimates given for NH and SH? Please detail this (possibly in the supplementary/appendix) or remove.

L20: NH-SH emissions gradient is evoked, but what about the NH/SH gradient in OH concentrations, a topic recently published by the same group? What is the NH-SH distribution of the OH fields used here. A short discussion on OH here might be useful.
Ans. We removed the 2-box model results as the revised IPD-observed is in quite good agreement with the ACTM simulation using Eopt. In addition, some changes in OH NH/SH ratio could have taken place in the past century, and that is not addressed in this work (as mentioned in section 3.3).

P20:
L20-25 are a bit redundant with lines 15-20, please rephrase and shorten the paragraph (-60 and -21.8 explanation)
Ans. Changes will be incorporated in the revised version.

P21
L5: (and also p22 lines 15-26) decreasing trend after 1990 for the heavy delta could also be explained by fossil fuel reduction linked to former USRR collapse. Did you try to replace BBG by fossil fuel and redo the analysis still with 2 unknowns? Putting all changes on BBG is a strong assumption. The slower decreasing trend in BBG from Kirschk et et al may indicate indeed a fossil fuel contribution as well.
Ans. We have found that the isotope modeling system produce unrealistic results if we run the model with full freedom. However, we can put small degrees of freedom on each of the emissions and come up with a 'positive' solution, which again depends on the individual modeler's choice. Instead we attempted here to constrain only the biomass burning (and a mysterious wetland/animal type of source). In our forward simulation setup the biomass burning emission was kept constant because no formal estimation of trends in this category of CH4 emission exists. A more sophisticated modeling system should be tested first for the recent years, for a period with measurements from ~100 of sites, to separate multiple source categories and then it may be possible to extrapolate that information backward in time over the past 100 years when measurements are available only at the two polar regions, albeit at larger uncertainties.

L20: how sensible is the result on BBG emissions to the initial conditions taken here. You start with a high 49Tg/y constant for BBG. What if your start with lower values?
Ans. Same as the previous reply.

L28: similar trends: you wrote before (page 20 l21) that trends were different. Please make this consistent.
Ans. Lines 20-25 in page 20 are now deleted following your earlier suggestion.

L16: would have suppressed the growth of CH4 emissions from human activities: you mean

for BBG emissions? Please precise it.
Ans. Yes, we wanted to refer to BBG due to human activity. Now revised as “the growth of \chem{CH_4} emissions due to biomass burning caused by human activities”