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

Anonymous Referee #2

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Review of the paper by Gosh et al entitled: Variations in global methane sources and sinks during 1910–2010

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 mass-balance 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.

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 Âž as we know that emission have
increased, although not in detail which category increased when A suggestion: During 1910–2010, the global total CH4 emission doubled from $290 Tgyr - 1$ to $580 Tgyr - 1$.

P4:

I10: Replace The present-day concentration of CH4... by the 2010 concentration of... I19: 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

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

P6:

L10-14: please rephrase the long sentence to make it more clear.

§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.

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.
P8: L8: Do the BBG emissions include biofuel burning? If not, this number seems a bit large if one considers today GFED’s emissions.

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.

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.

P11:

L16: 2.8696: Are all these digits really significant?

L19-20: the reason of the 3.7

P12: Δ (delta) signs are missing in equation (3)

Eq (4): more explanations are needed here. Do you correct the trajectory year by year? How do you allocate a global ΔE 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.

L9: is known: I suggest to write “is prescribed”

P13/14

L19: 12CH4 is approximated by CH4 = 12CH4 +13CH4: Does this mean that you assume 12CH4 CH4. Please clarify?
L10: I suggest to add a sentence announcing what follows in order to help the reader through §2.6: ‘And in the following, we calculate by two methods the isotopic signature for global emissions $\delta^{13}CE$. First, ... Second, $\delta^{11}CE$ 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($\delta^{13}CE$), ...

L16 in p14: For simplicity, ... please rephrase: ‘In order to remove the under-determination, we assume, ...

P15/16

The discussion on fractionations is interesting but should be in a specific paragraph (2.7) And/or announced p13 line 17 when $\alpha_i$ are presented. Else it comes too late after the method description.

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 all model boxes at the surface above 60N for Arctic and below 60$^\circ$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.
P17:
L24: does the mean value of [OH] is optimized using MCF obs? Please precise if so.
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?
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.
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 clarify after improving the optimisation procedure section
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.
P20:
L20-25 are a bit redundant with lines 15-20, please rephrase and shorten the paragraph (-60 and -21.8 explanation)
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 al may indicate indeed a fossil fuel contribution as well.

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?

L28: similar trends: you wrote before (page 20 l21) that trends were different. Please make this consistent.

L16: would have suppressed the growth of CH4 emissions from human activities: you mean for BBG emissions? please precise it

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 27619, 2014.