Biogeosciences Discuss.,
https://doi.org/10.5194/bg-2018-107-RC2, 2018

Biogeosciences Discussions

Interactive comment on "Bipolar carbon and hydrogen isotope constraints of the Holocene methane budget" *by* Jonas Beck et al.

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Received and published: 3 May 2018

Review of "Bipolar carbon and hydrogen isotope constraints of the Holocene methane budget" by Jonas Beck et al.

The study presents new measurements of Holocene methane data from ice cores and combines them with literature data to provide novel constraints on the methane budget at different stages during the Holocene. The analysis is based on the combined interpretation of methane mixing ratios, stable carbon and hydrogen isotope ratios, as well as interpolar differences of the three parameters. Interpreted using a 2-box model, the combination of data sets allows to quantify the changing magnitude of hemispheric methane sources as well as their isotopic signatures. This is a highly anticipated ac-

complishment. The findings are important to understand the past methane cycle and are relevant to anticipate future changes. The authors have put a lot of thought and effort into all aspects of the analyses, particularly with regards to poorly known environmental parameters. The manuscript is very well written and structured. This is a very valuable contribution to the field.

I have two points to make. The first is the difference between polar and hemispheric data. The ice core records from Greenland are higher in mixing ratios and more depleted in the rare isotope than the northern hemispheric average, while the reverse is true for Antarctic data and the southern hemisphere. For present day conditions, this can be seen in data products like CarbonTracker-CH4 provided by NOAA-ESRL for mixing ratios and data sets maintained by INSTAAR for delta13C (for an illustration see Figure S1 of Schaefer et al., 2016). As a consequence, the emission estimates from the box model for each hemisphere will be skewed in magnitude and isotopic signature and I would expect the relative changes between different times in the Holocene to be biased as well. It is not straightforward to address this issue because past latitudinal gradients likely differ from modern ones by an unknown margin. However, an attempt should be made, e.g. by correcting the polar data for a probable offset to hemispheric means. For example, Etheridge et al. (1998) and Schaefer et al. (2016) determined the difference between polar and mean global values (for mixing ratio and carbon isotopes, respectively) as a percentage of the interpolar gradient. A similar quantification could be made for hemispheric means.

The other point is a suggestion to derive more information from the presented data. The isotopic signature of the emissions that have changed between two periods can be derived by mass balance according to E(total)*D(total)=E(initial)*D(initial) + E(change)*D(change); where E and D stand for the magnitude and isotopic signature of emissions, respectively. This would enable a more robust discussion of the changes, e.g. by comparing the isotope signatures to those of major emission types. Statements like the one on page 13, lines 37-38, can then be based on specific numbers and dis-

cussed in the context of known source isotopic signatures.

Minor comments are listed in the following.

Page 1, line 36 (1/36): these numbers don't match with Table 1 of Naik et al. (2013). Arguably, observation-based estimates like the one from Prather et al. (2012) are more reliable than model estimates.

1/37: even for current emission estimates there is a sizeable difference between bottom-up and top-down estimates (Kirschke et al., 2013; Saunois et al., 2017).

3/10-14: The sink changes between modern day and pre-industrial times as studied by Naik et al. (2013) are more relevant here (although the author's point remains valid that sink changes are unlikely to have significant impact). It cannot be ruled out that the ratio of sinks changed throughout the study period, e.g., due to different sensitivity to changing CH4 mixing ratio or environmental conditions. Such changes are likely to be minor or negligible, but the point should be made.

Table 1: it is interesting that different time series from the same lab can have different offsets to the spline. This means that the offset results are not transferable to future studies and raises questions as to the cause of the offsets. It may or may not be worth pointing that out.

Section 2.3.3: the resulting GISP2 data sets are likely of interest for future studies and I wonder if they can be made available. However, that may be problematic because the credit would have to be shared between Ed Brook as supplier of the original data and the current authors for their data analysis. Just a thought.

Section 2.3.: there should be an additional subsection to derive hemispheric averages from the polar data, as discussed above.

7/18 and following lines: the parameterization of sinks and associated fractionation is complex and without definitive solution. I see no problems with the particular choice of values by the authors, but the following points should be made clear. (i) Other

studies arrive at or use different numbers for individual parameters that are equally as valid (e.g., preindustrial tau=10.1$\pm$1.7 a, Naik et al., 2013; tropospheric expression of stratospheric fractionation = -3‰ Lassey et al., 2000). (ii) It is uncertain if modern estimates can be used for Holocene conditions. (iii) It cannot be ruled out that changes in environmental conditions and atmospheric chemistry – including CH4 mixing ratios – changed the sink and its total fractionation throughout the studied period. (iv) The exact choice of sink parameters is of minor importance as long as the focus of the interpretation are relative changes in emissions.

7/26-27: please provide a specific reference for the OH fractionation coefficients, i.e., Saueressig et al., (2001)

Section 2.4.1.: if the authors correct the input data to hemispheric averages, then the transport times derived from SF6 must be adjusted.

9/10-12: this value is likely to change when the model is run for hemispheric averages rather than high-latitude data.

9/15-17: I don't follow the description of this experiment. Is the linear change between minimum IPD at start of the run to maximum IPD at its end (or vice-versa)? Please clarify.

11/1-6: please revise these results for hemispheric averages instead of polar data.

13/37-39: it is not possible to evaluate the statement unless the authors provide quantification. This can be done by calculating the isotopic signatures of the additional emissions by isotope mass balance (as mentioned above).

13/38-41: tropical wetland emissions are more 13C-rich than other biogenic sources, so qualitatively the trend can be explained by their greater prominence. To evaluate whether an additional 13C-rich source is increasing requires the mass balance results. If a simultaneous rise of wetland and fire emissions is invoked, then the discussion should explain how increasing precipitation that enhances wetland CH4 production

also leads to more fires, which occur in dry conditions. Greater fuel supply? More smoldering fires?

14/11-13: does this statement (isotopic changes are due to changing source signatures) stand for all time periods? That would contrast with the discussion in Section 4.3.

Fig. 3: the IPDs are a crucial parameter derived in this study. I recommend showing them in the graph (or a separate graph).

Fig. 3d: the temporal changes in the three emission lines are hard to see on the presented scale, making it difficult to follow the discussion in Sections 4.1.-4.3. Consider providing an additional or alternative graph with more detail on changes in emission magnitude.

Table 5: please provide references for the various literature values. The listed value for epsilon(OH) is from an older study (Cantrell et al., 1994), a more precise estimate has been published by Saueressig et al. (2001).

Minor corrections:

5/34 “can complicate” not (“can complicates”).

6/14: should this read “residual” rather than “residuum”?

8/4: better: “in both time and magnitude”

9/41: “with a full decoupling”

10/31-32: consider rewording, e.g.: “at about the same time the d13C-CH4 signals undergo a step change from . . .”

14/8-9: “This shows the value. . .” consider rewording this sentence.

14/19: “. . .without such evidence.” Consider rewording.

Fig. 2: consider arrows for the time axes.

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