

We thank Hinrich Schaefer for this constructive and detailed review. Below we respond to each point in blue text and propose altered text in italics.

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 inter-polar 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 accomplishment. 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 inter-polar gradient. A similar quantification could be made for hemispheric means.

Yes, the values measured on polar ice cores do not represent mean hemispheric signals. And we also agree that the recommended corrections describe one possible way to address the issue. For this study, however, we decided to adapt the atmospheric transport in our deconvolution model to already account for this effect. By using high altitude (polar) SF₆ data for the calibration of the polar exchange time θ which is then used to quantify the CH₄ exchange between the boxes we inherently correct for the described effect of using polar instead of mean hemispheric values. As a result, with 1.56 years the inter-polar exchange time is larger than the literature values for the inter-hemispheric exchange time of 1.3 years (Geller et al., 1997).

In response to the review comment we tried to test the strategy recommended by Hinrich Schaefer and to use the knowledge about the modern CH₄ distribution and the past inter-polar CH₄ difference to scale the measured values. Using the CH₄ concentration data from the numerous measurement stations we calculated the latitudinal CH₄ concentration distribution over the years 1997-2017 using the monthly mean data from ftp://aftp.cmdl.noaa.gov/data/trace_gases/ch4/flask/surface/ (Dlugokencky et al., 2017). Based on the latitude weighted mean hemispheric CH₄ concentrations (with $\phi_{ITCZ} = 5^{\circ}N$) and the polar CH₄ concentrations we calculated the average correction factor for both hemisphere boxes according to the relationship

$$C_{hemisphere} = C_{polar} + \omega * IPD \text{ and got: } \omega_n = -1.77, \omega_s = 0.71$$

With the measured polar CH₄ concentration for both hemispheres (and thus the IPD) this allows us to translate the polar CH₄ data to hemispheric mean values. Assuming the same relative latitudinal distribution for $\delta D-CH_4$ and $\delta^{13}C-CH_4$ the correction can be also applied to the stable isotope time series.

However, to be consistent we also have to adapt the exchange time θ since it should now represent the

exchange time between the two hemispheres. We tried to do so by following the same SF6 calibration explained in the manuscript using mean hemispheric SF6 concentrations based on SF6 data by Levin et al. (2010). By doing so, we end up with a hemispheric exchange time of only $\theta = 1.02 \pm 0.06$ years. This value is not in line with the literature. The low value of the exchange time is most likely the result of the insufficient latitudinal resolution of the SF6 data, in particular in the low latitudes. As a consequence the calculated mean hemispheric SF6 levels of the two boxes are shifted towards each other (underestimated IPD_{SF6}) leading to the overestimation of the hemispheric air mass exchange. In summary, while the suggestion by the reviewer represents an alternative strategy, we cannot reliably calibrate our model using SF6 in this case.

Due to this difficulties we decided to stick to our method to keep the ice core time series and use the inter-polar exchange time for the deconvolution. However, the corresponding sections in the manuscript are adapted to point out this issue and our solution more explicitly. Note, that we will use the expression “inter-polar exchange time” throughout the manuscript to be consistent with our calibration approach.

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 $E_{total} \cdot D_{total} = E_{initial} \cdot D_{initial} + E_{change} \cdot D_{change}$; 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 discussed in the context of known source isotopic signatures.

So far we consciously refrained from such quantitative analysis of the inversion values due to the following reasons: (1) As we describe throughout the manuscript, the box model is based on assumptions which only allow robust conclusions on relative changes. (2) The mass balance approach proposed by the reviewer assumes constant isotopic signatures of the initial CH₄ emissions, which cannot be guaranteed. Nonetheless, we decided to make the calculation as proposed and discuss the caveats as follows:

The observed shift in δD -Es and especially $\delta^{13}C$ -Es towards heavier values in this interval cannot be explained simply by increased tropical wetland emissions in the Amazon region but requires also an enhancement of a southern hemisphere CH₄ source strongly enriched in ^{13}C . Using a simple mass balance approach ($E_{total} \cdot i\delta_{total} = E_{initial} \cdot i\delta_{initial} + E_{additional} \cdot i\delta_{additional}$) allows to determine a $\delta^{13}C$ signature of the additional CH₄ emissions of 19 Tg CH₄/a to be about -45.5 ‰ which is isotopically heavier than tropical wetland CH₄ emissions (-56.8 ‰ according to Whiticar and Schaefer (2007)). Note, however, that this quantitative approach is based on the assumption of unchanged isotope signatures of the initial CH₄ emissions and therefore needs to be taken with care.

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.

We will change the number and the citation:
... which is 9.1 ± 0.9 years (Prather et al., 2012).

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

Yes, and that is what this sentence also implies. We will clarify:

Despite the fact that the total CH₄ emissions today are well known, there is still considerable debate about the individual contributions of (in particular small) CH₄ source and their variability, e.g. evidenced by the mismatch of bottom-up and top-down estimates of the total CH₄ emissions (Crill and Thornton, 2017; Kirschke et al., 2013; Saunio 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.

We will change this paragraph as follows:

As mentioned above, changes in the sinks leave an imprint on the isotopic composition of CH₄ in the atmosphere. However, while minor changes in the relative importance of the sink processes cannot be ruled out, in general, atmospheric chemistry models point to small changes in total lifetime (Kaplan et al., 2006; Levine et al., 2011a) and we also assume the relative contribution of individual sinks to be rather constant over the Holocene when relatively stable climate conditions prevailed. Therefore, the discussion of this study centres around CH₄ source processes, hence emissions of different CH₄ source categories.

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.

Yes, the offsets are not to be transferred to any other datasets. Therefore, we tried to highlight the value of inter-polar data sets measured not only with the same measurement setup, but also during the same measurement campaign. The text in the conclusion section (14/32) will be adapted as follows: *Better IPD-CH₄ data measured on the same analytical system during the same measurement campaign in high temporal resolution, which ensures an accurate synchronisation (such as the data by Mitchell et al. (2013) for the last 2,800 years), would bring us a large step forward in further constraining the Holocene CH₄ cycle. This is especially true for the Greenland ice cores within the brittle ice zone, e.g. GISP2 records for depths between 650 and 1400 m, equivalent to the time window 2.7-8.1 ka BP where individual samples subject to modern air intrusion may compromise the Greenland CH₄ record. The new EGRIP ice core currently drilled in northeast Greenland with much lower accumulation, hence a brittle ice zone located at older ages may help to improve the CH₄ record in this time interval in the future.*

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.

The GISP2 data are published and available at <http://nsidc.org/data/nsidc-0440>. We use and cite the data in the same manner as other data used for our CH₄ data compilation.

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

See comment 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.

Regarding the choice of the lifetime we will add the following statement (7/19):

Again, using other estimates of the atmospheric lifetime (Naik et al., 2013; Prather et al., 2012) changes the absolute emissions estimates but does not affect our conclusions on relative emission changes.

Further we will clarify:

The chosen values for the atmospheric CH₄ lifetime and the strengths and the distribution of the sinks are just a best guess based on the cited literature. Hence, other studies end up with slightly different values. Note also that we assume the model parameters to remain temporally unchanged over the Holocene.

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

Yes, we will add Saueressig et al. (2001) to the references.

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

See comment above.

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

See comment above.

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.

We will clarify to:

For the second experiment (dashed lines in Figure 2), the IPD of the tropospheric isotope signatures (IPD_δD and IPD_δ13C) have been changed while the concentrations remained unchanged. By only altering the northern stable isotope signal the isotopic IPD's are changing from the minimal to the maximal observed IPD_δD and IPD_δ13C values in the splined isotope data, respectively (dashed lines in (b) and (c)).

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

See comment above.

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).

See comment 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?

We will complement as follows:

Increasing emissions from tropical and subtropical wild fires in the southern hemisphere, which are also documented in charcoal records (Daniau et al., 2012), could have become increasingly important, e.g. due to enhanced fuel production or higher CH₄ emission efficiency (more smoldering fires) both caused by wetter climatic conditions, and would readily explain the joint information

derived from inversion results on southern hemisphere CH₄ emissions and its stable isotopic signatures.

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.

We will clarify to:

Therefore, we attribute the observed long-term changes of the atmospheric CH₄ isotopes over the first half of the Holocene to shifts in the isotopic source signatures of individual CH₄ sources rather than to changes in the global CH₄ source mix.

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

We will add panels showing the IPD's in figure 3.

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.

OK, we will increase the height of figure 3 (also to get enough space for the additional IPD panels) which allows us also to stretch the axes of the inversion panels. Additionally, we will add grid lines in panel (d) to better visualise the emissions changes.

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).

We will add the following citations in the caption of table 5:

(Breninkmeijer et al., 1995; Cantrell et al., 1990; Feilberg et al., 2005; Gierczak et al., 1997; Irion et al., 1996; Tyler et al., 1994; Quay et al., 1999)

We are aware that numbers from more recent studies (e.g. Saueressig et al. (2001)) exist. The values we use have also been used for the 4 box model by Fischer et al. (2008) and Bock et al. (2017). For comparison reason we decided to not to change them.

Minor corrections:

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

Yes, we will change that.

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

Yes, we will change that.

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

Yes, we will change that.

9/41: “with a full decoupling”

Yes, we will change that.

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

We will change the text as follows:

At about the same time the IPD_δ13C changes quickly from -0.35 ‰ to -0.63 ‰, which is rather constant before and after the point of inflection.

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

We will change the text as follows:

This shows the value of our multi isotope approach in order to avoid drawing the wrong conclusions.

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

We will change the text as follows:

An alternative explanation calling upon a significant decrease of CH₄ emissions from thermokarst lakes would require compensation by other processes to fulfil the $\delta^{13}\text{C}$ constraint, e.g. a substantial decline of wild fire activity in the northern hemisphere.

Fig. 2: consider arrows for the time axes.

Principally the figure has no orientation since the shown data fulfil the deconvolution equation in both directions (time running from left to right and vice-versa). To avoid unnecessary confusion, we will apply an arrow showing the direction of the time (in the same direction as it is the case for the other figures).