Interactive comment on "Atmospheric methane control mechanisms during the early Holocene" *by* Ji-Woong Yang et al.

Anonymous Referee #2

Received and published: 23 September 2016

We thank the Referee #2 for her/his pinpoint comments and important suggestions. Below we copied the Referee comments in black, and add our responses to each comment in red italics. Further we attached the paragraphs of original discussion paper in blue, and our modifications in green.

Overall assessment

The manuscript provides new high-resolution CH4 data from the Siple Dome ice core over the time interval 8.5-11.5 kyr BP, extending previous work by Ahn et al., 2014. The data quality is generally very good (see comments below) and I commend the authors for their painstaking work to provide high-resolution data sets using discrete CH4 analyses. The data is interpreted with respect to millennial climate variations during this time interval and, based on correlation with other climate proxy data, a suggestive hypothesis about the influence of changes in the ITCZ is presented to explain the millennial variability in CH4.

Finally, the interpolar CH4 difference (IPD) is calculated using Greenland data from the literature and this difference is then analyzed using a simple three-box model. As outlined below, I have some fundamental questions about the reliability of the inferred IPD, which subsequently has also important implications for its interpretation. This prevents me from recommending the manuscript for publication in CP in its current form despite the nice data. Moreover, the quality of the manuscript in terms of the use of the English language has to be considerably improved. With a native English speaker on the author list, I see no problem that this can be achieved. In summary, after additional work I am confident that the manuscript will become suitable for publication in CP in a resubmitted version.

General comments

a) comparison of early and late Holocene CH4 variations: In the introduction the authors make the important point that only the early Holocene period allows us to study the natural CH4 variability on centennial to millennial time scales. Unfortunately, the authors do not follow up on this in the discussion. It would be interesting to compare the centennial and millennial variability in CH4 concentrations in the early Holocene (as documented in the Siple Dome, WAIS (including the continuous CH4 data by Rhodes et al., 2015) and NEEM record with the late Holocene as documented in WAIS by Mitchell et al., 2013. Are the amplitudes of this variability different and if so, is that due to an anthropogenic influence in the late Holocene or related to the changes in seasonal and geographical distribution of solar insolation (due to orbital parameter changes) between the early and late Holocene? Note that summer insolation in high northern latitudes was several tens of W/m2 higher in the early Holocene. For this analysis it may be beneficial to use the continuous WAIS data instead of the Siple Dome data to calculate a record with comparable resolution to the Mitchell data and to compare CH4 frequency spectra for the early and late Holocene.

We appreciate to Referee #2 for this important comment. We will add a dedicated chapter based on discussion below:

We compared amplitude of methane variability between the early- and late Holocene in multi centennial to millennial time scales. Figure R1 shows amplitude spectrum and root mean square (RMS) amplitude for the early Holocene and late Holocene, respectively. The amplitude of the early Holocene CH_4 change is ~10 ppb and does not change a lot except for PBO and the 8.2k event, while the late Holocene spectrum shows smaller amplitude than early Holocene for shorter-term change and larger for longer-term fluctuation. LPIH (Late Pre-Industrial Holocene) CH_4 amplitude is elevated to early Holocene level from ~0 C.E., and increases up to higher from ~1450 C.E.

The reason of low amplitude variability during 3.5 to 2.0 kyr BP, or inversely, why the early Holocene CH_4 variability is larger than this period, is probably related to different orbital configuration in both time periods. Previous studies found covariation between CH_4 amplitude and NH summer insolation change, reflecting that mean temperature of the warmest seasons is an important factor of CH_4 emission, during the interstadial conditions (Flückiger et al., 2004; Baumgartner et al., 2014). Accordingly, lower summer insolation during the late Holocene might *induce diminished CH_4 amplitude, and vice versa during the early Holocene. This evidence indicates the natural forcing in centennial- to millennial time scales is reduced in the late Holocene, given that the atmospheric CH_4 budget is similar between 3.5-2.0 kyr BP (604.9 ppb) and 9.0-7.6 kyr BP (628.6 ppb) interval, and that anthropogenic emission is larger in later Holocene than early Holocene.*

Abrupt increase of CH_4 amplitude since ~800 C.E. is likely driven by increasing anthropogenic contribution, which is consistent with anthropogenic emission scenario based on past population and agricultural activity (Mitchell et al., 2013). Also superimposed are short-term cooling events during Little Ice Age, making CH_4 variability larger.

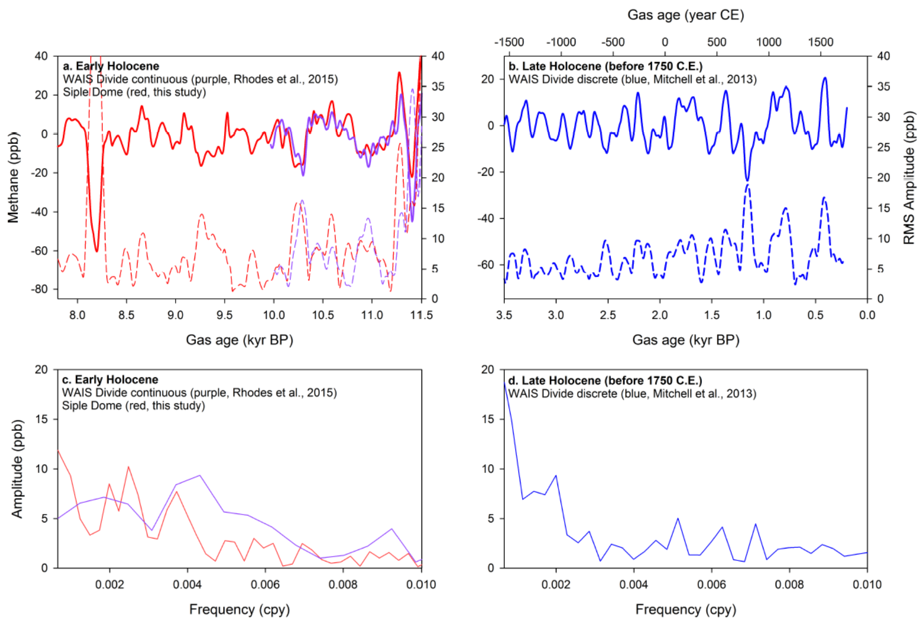

Figure R1. Upper: Detrended (75 to 1800-year band-pass filtered) CH_4 for the early (a) and late (b) Holocene from Siple Dome (red, this study), WAIS divide continuous (purple, Rhodes et al., 2015), and WAIS divide discrete (blue, Mitchell et al., 2013) data. Dashed lines are root mean square (RMS) amplitude running averaged by 75-year window. Lower: Amplitude spectrum of Early (c) and Late (d) Holocene CH_4 records. Note that CH_4 data before 1750 C.E. are used for the preindustrial late Holocene.

b) Millennial CH4 variations: The authors suggest that climate cooling in the northern hemisphere has led to a southward shift in the ITCZ, which again led to a decline in CH4 low latitude emissions due to changes in monsoon systems. The first part of this hypothesis (ITCZ shift) appears to be straightforward and has been observed in models, however, the second part (CH4 emission changes) appears not so straightforward and requires some more quantitative support. Rhodes et al. (2015) suggest a first order relationship between CH4 emissions and intense rainfall area, where from a certain point on also an increase in southern hemisphere wetland emissions is possible. Accordingly, a discussion focusing on Asian monsoon systems only, as in the manuscript by Yang, seems to be too narrow. Please explain how your hypothesis fits into this picture. Please note also the work by Bozbiyik et al., CP, 2011, performing a North Atlantic fresh water hosing experiment under interglacial conditions connected to a southward shift of the ITCZ, showing decreases in tropical precipitation and the modeling work by Zurcher et al., Biogeoscience, 2013, which shows that also boreal peatland CH4 emissions are reduced during such an experiment. Finally, the discussion of the millennial CH4 variability and the corroborating proxy evidence from other archives lacks some clarity and could be improved.

We thank again for useful comment and paper suggestion. We will consider those papers and update the sections based on the discussion and figure below:

One of main point of Rhodes et al. (2015) is that abrupt CH_4 increase occurred during Heinrich Stadial 1, 2, 4, and 5 events could be induced by the southern hemisphere emission as a result of strong southward migration of ITCZ at that time. However, mean latitudinal position of ITCZ moved northward during the Holocene climate conditions, therefore monsoon intensity in northern tropics should be strengthened while tropical rainfall in southern hemisphere decreased. This is identified from Cariaco Basin reflectance record which shows increase in rainfall during the Holocene compared to glacial period (e.g., Deplazes et al., 2013), as well as from anti-correlation *between Chinese- and Brazilian cave speleothem record (e.g., Wang et al., 2006). Further, Asian monsoon intensity during the Heinrich Stadials was weaker than during the early Holocene (e.g., Wang et al., 2008). For these reasons, we focused more on Asian/Indian monsoon variability than others, but we agree that our manuscript lacks detailed explanation on other monsoon systems. We will add more discussions and change Figure 2.*

The new Figure 2 (Figure R2 below) now shows Asian, Indian, African and Brazilian monsoon proxies, with age uncertainties indicated as black horizontal error bars. Gulf of Guinea (western tropical Africa) planktonic Ba/Ca ratio, a proxy of riverine runoff, shows decreased rainfalls at similar timings of local CH_4 minima at 8.2, 9.3 and 10.9 kyr BP, indicating that the abrupt Greenland cooling leads to hemispheric-wide hydroclimatic changes. Inverse pattern of South American rainfall (Lapa Grande Cave, eastern Brazil) supports that ITCZ was temporarily migrated southward at that time. High-resolution sediment reflectance records from Cariaco Basin and Arabian Sea clearly show that the strength of southward migration of ITCZ and its effect of precipitation change are smaller during the early Holocene than during the Heinrich Stadials (Deplazes et al., 2013; 2014).

We thank the reviewer for suggesting appropriate paper. Zurcher et al. (2013) found that abrupt cooling in Greenland and northern high latitudes by large freshwater input causes boreal peatland CH_4 emission to decrease substantially, which explains ~23% of abrupt CH_4 drop (~80 ppb) during the 8.2k event. If we assume linear scaling of model response, it implies that boreal peatland source change only accounts for ~23% of total CH_4 change during the rest of CH_4 decrease events. Given the meltwater pulses during the early Holocene before the 8.2 k event are more than 10 times smaller (Teller and Leverington, 2004)than that corresponds to the 8.2 k event, we consider the boreal emission change is not the major cause of CH_4 local minima.

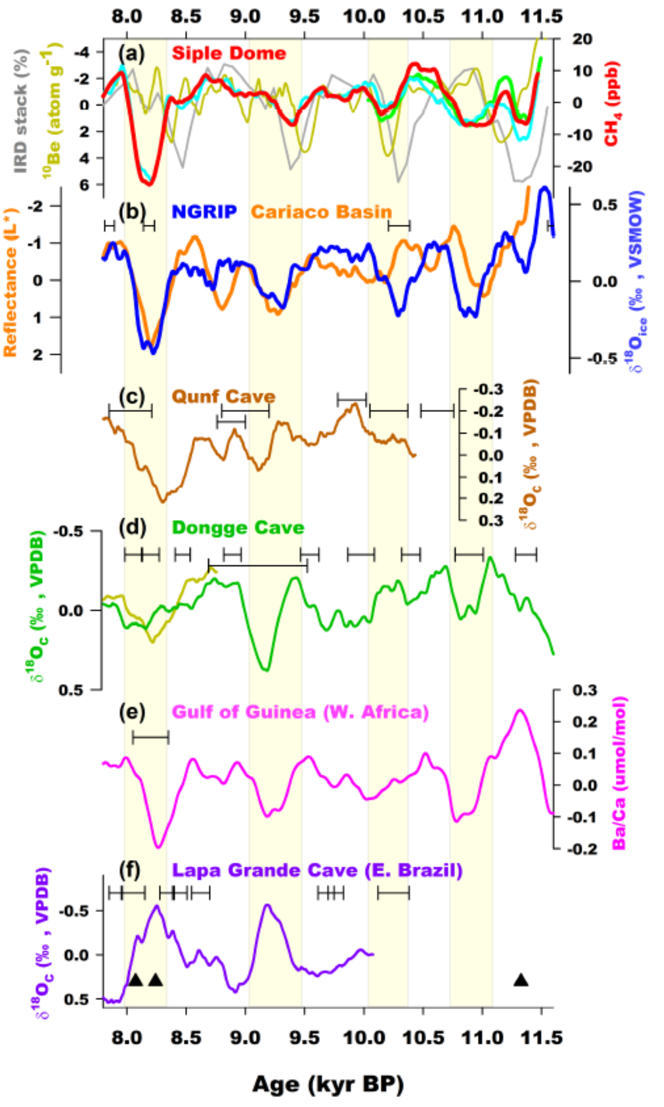

Figure R2. Millennial scale climate variability. All proxies present here were smoothed by 250-year running average and detrended by high-pass filter with 1/1800-year window. (a) Siple Dome CH_4 (red, this study), Greenland ^{10}Be (dark yellow, Finkel and Nishizumii, 1997), North

Atlantic IRD stack (grey, Bond et al., 2001). Also shown are WAIS Divide CH_4 data by discrete (cyan, WAIS Divide members, 2015) and continuous (yellow green, Rhodes et al., 2015) technique. (b) NGRIP stable water isotope ratio (blue, Rasmussen et al., 2006) and Cariaco Basin reflectance (orange, Deplazes et al., 2013). (c) Qunf Cave speleothem oxygen isotope (Fleitmann et al., 2007). (d) Dongge Cave speleothem oxygen isotope (green, Dykoski et al., 2005; dark yellow, Wang et al., 2005). (e) Gulf of Guinea planktonic Ba/Ca ratio (Weldeab et al., 2007). (f) Lapa Grande Cave speleothem oxygen isotope (purple, Strikis et al., 2011). Age tie-points are used to synchronize Siple Dome and WAIS Divide CH_4 data with the GICC05 time scale (black triangles).

c) Interpolar CH4 difference: The IPD is a tricky business and erroneous effects can be easily introduced by comparing CH4 data from different labs, different sites, or insufficient robustness of the results. Accordingly, this needs more supporting information and detail: In the method part it is mentioned that blank ice measurements show an offset with a very large scatter between 5 and 15 ppb. I read the text in such a way that a daily blank correction is applied based on 4 blank measurements per day. This needs more detailed discussion as a potentially erroneous correction, which varies by 10 ppb, has a huge influence on the IPD, which varies with a similar amplitude. Please add the following information/discussion:

- Can you be sure that the CH4 blank is coming from the extraction system and is not reflecting dissolved CH4 in the bubble-free ice? In the latter case you should not correct for this blank. Did you perform blank tests without bubble-free ice or did you repeat the extraction of bubble-free ice for a second or third time to see, whether the blank is constant or declining?

We've performed the gas extraction test using bubble-free ices to estimate how much air dissolves in ice melt. We found less than 5 mTorr of gas extracted after second melting-refreeze procedure. Considering the typical amount of standard air injected (30-40 Torr), the air extracted from the second melting-refreezing process should have CH_4 mixing ratio of >~100000 ppb to cause 20 ppb of blank offset. This is unlikely because (1) such high concentration cannot be explained by gas solubility effect, and (2) the bubble-free ices were trimmed outermost layer sufficiently before cutting the artificial ice samples to prevent causal contamination by ambient air dissolution. The microbial activity within bubble-free ice during storage is unrealistic, given that we use water distiller (Barnstead) and anti-bacterial membrane filter (Millipak Express) to produce deionized water, and the deionized water is boiled within a stainless steel chamber for degassing. Further, we found the blank offset is similar order from the blank test without bubble-free ices (9.4 ~ 21.6 ppb, n = 36) using various working standards (721.31, 895.03, and 1384.91 ppb CH_4 in NOAA04 scale). Therefore, we conclude that the CH_4 blank offset caused by dissolved CH_4 in bubble-free ice is not the case.

- Please add information on the time scale on which the blank changes. If my understanding is correct that a daily mean blank correction of 5-10 ppb is performed, it is important to know what the variability of the four blank ice measurements is within one day. If this intra-day variability is of the same size as the inter-day variability, then a daily blank correction varying between 5 and 15 ppb introduces offsets from one day to the other which only reflect the stochastic variability of the blank measurements themselves and not systematic day-to-day differences in the entire measurement system. In that case a long-term mean blank correction seems more appropriate. If the blank values are reproducible within one day, a daily correction seems justified.

The daily systematic offset was determined using a standard air injected into the sample flasks which have bubble-free blank ices. Even though the systematic offset varied daily, the 4 blank results were rather in a small range, yielding the intra-day standard error of the mean of the 4 blanks of 2.0±1.0 ppb on average. Our final CH_4 data were presented by averaging the results of duplicate sample analysis from the same depths. To test our correction method, we reanalyzed duplicate samples from the adjacent ices (~10 cm depth difference) at 8 depth intervals 8-80 days after the first analysis (see the table in the response to reviewer #1). The pooled standard deviation of the average of duplicates from first and second measurements was ±1.0 ppb. Given that we found a good reproducibility from the reanalysis, our blank correction method is robust.

- On the other hand if you have a long-term trend in this blank value, which may not reflect a trend in the extraction system but in the bubble-free ice quality, you introduce this error into the IPD. Did you use a randomized order to measure the samples to avoid such spurious trends.

Siple Dome ice samples were measured in a randomized order. As we described above, contamination by blank ice quality and/or by air occluded in blank ice is unlikely. Therefore, we decide not to apply this correction.

- There was an average offset of 3 ppb observed between Siple Dome data measured at SNU and OSU and the OSU data have been corrected by subtracting 3 ppb, but it is not discussed what the influence of this correction may be on the IDP. Note that the NEEM discrete data used to calculate the IPD are also measured at OSU. Accordingly, to avoid any systematic errors in the IPD it is mandatory to add the 3 ppb to the Siple Dome data measured at SNU and not to subtract 3 ppb from OSU data.

The offset comes from different correction methods between the two laboratories. As described in our response to the Reviewer #1, instead of simply adding 3 ppb to SNU data, we applied the similar solubility correction used in Mitchell et al. (2011) at OSU, and found that the average offset between SNU-OSU reduces to ~0.6 ppb, which lies within analytical uncertainty range of both institutes. Instead of adding 3 ppb, now we apply the OSU solubility correction methods to our data for IPD calculation.

- Comparing the continuous WAIS data with the discrete (or continuous) CH4 data from NEEM, it becomes apparent that the relative changes in CH4 concentrations in the northern and southern hemisphere are much more similar than when comparing Siple Dome and NEEM data after the Monte Carlo synchronization in Figure 3. For example the downward trend in NEEM between 11.3 and 10.9 kyr BP is also seen in the continuous WAIS data, while in Siple this time interval shows essentially constant CH4 after a first short peak. Consequently, the constant values in the Siple data lead to an erroneous downward trend in the IPD in this time interval. Vice versa, there is an upward trend from 10.9 to 10.4 kyr BP found in NEEM and continuous WAIS data. The same time interval in Siple looks more like a broad maximum, again with implications on the derived IPD. A similar observation holds for the maximum around 10 kyr BP. Note that on these centennial to millennial time scales, which are much longer than the atmospheric lifetime and the interhemispheric exchange time, you may have changes in the size of the IPD, however, it is extremely difficult to create a millennial trend in one hemisphere without a trend in the other. This is nicely illustrated in the high-resolution data by Mitchell et al., 2013. Obviously, the resolution and quality of the data in Figure 3 does not suffice to gain a robust IPD and/or the Monte Carlo synchronization fails to synchronize the records sufficiently. In fact, it seems crucial that the IPD analysis is performed not only on the Siple but also on the WAIS discrete and continuous data to study the robustness of the results gained from the Siple Dome core. Note that the WAIS very high-resolution data from continuous measurements can be used to much better synchronize WAIS to the continuous records available from NEEM. This would circumvent the synchronization problems apparent between Siple and NEEM. As a final remark on this topic, I do not agree with the authors' statement that the IPD values in Siple Dome over the time interval 9.5-11.5 kyr BP are in agreement with previous results. If you calculate the mean over this time interval in the Siple IPD data and calculate the standard error, this appears to clearly higher than the literature values. In summary, the IPD discussion needs more work before the manuscript should be published in CP.

We thank to the Reviewer #2 for pinpoint comment and useful suggestions. We will revise the IPD section of our manuscript based on the following discussions:

To test robustness of the early Holocene IPD change, we revised the previous IPD and calculated various IPD curves by using different data sets, including NEEM continuous, NEEM discrete, Siple Dome discrete, WAIS continuous, and WAIS discrete data (Figure R3). We calibrated NEEM and WAIS continuous data against to discrete measurements, as discrete measurements are more accurate than continuous ones in absolute values although they are worse in precision. However, we hesitate to equally consider all the IPDs because some CH_4 data sets are not sufficient for centennial to millennial IPD estimates (see below for detailed discussion on data reliability). Here we consider NEEM discrete, WAIS Divide continuous, and Siple Dome discrete records are reliable to draw IPDs. Also we address the inconsistency among the different ice core records for interval older than ~10.3 kyr BP, which make IPDs different each other.
Resulting IPD curve from NEEM discrete and WAIS continuous (green) shows long-term increase from the onset of Holocene to ~9.9 kyr BP, and it supports the NEEM-Siple IPD reconstruction (black). The good agreement implies that the millennial scale IPD increase trend during the early Holocene is robust. However, the IPD fluctuation during 10.8 – 11.2 kyr BP is not reproduced in the alternative IPD, hence we'll modify our original argument that IPD increase from ~10.7 to 9.8 kyr BP. Instead, we'll more focus on long term IPD increase. Except for during 10.8 – 11.2 kyr BP, both IPDs show concomitant increase with NH extratropical temperature and thermokarst lake CH_4 emission increase. These evidences show that boreal emission increased while tropical emission decreased (Table S1).

Finally, we'd like to note that mean value of the Siple Dome IPD is 41 ± 6 ppb over the 9.5-11.5 kyr BP period, which is consistent with previous results within uncertainty range.

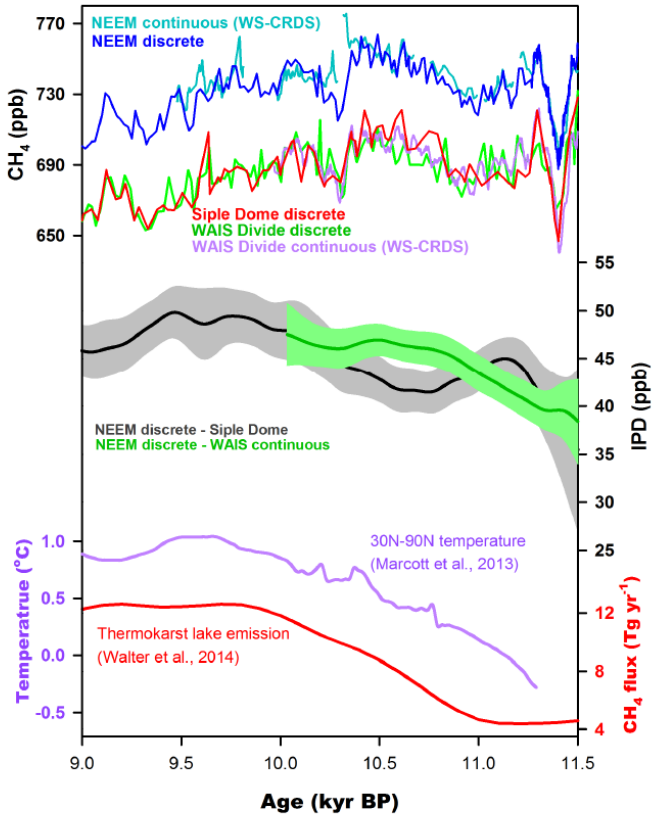

Figure R3. Inter-polar difference (IPD) reconstructions. Top: high resolution CH_4 records from Greenland and Antarctica, synchronized to NEEM gas age scale by Monte Carlo procedure. Middle: Millennial-scale IPD trends derived from Siple Dome (black) and WAIS Divide continuous (green) data. Shaded area indicates 95% significance interval. Bottom: Proxy-based temperature reconstruction for northern mid to high latitude and boreal CH_4 emission from northern thermokarst lakes. Note that this figure may be subject to change.

We excluded some dataset from our discussion for the following reasons. Rhodes et al. (2015) reported that WAIS continuous data are lower than OSU discrete measurements by 1.5-2.5% for 1804-2420m (9.8 – 17.1 kyr BP in WD2014 scale) interval. WAIS continuous data were calibrated against to Siple discrete data instead of WAIS data, because we consider that Siple data are more reliable during the early Holocene period. Even though the analytical method of Mitchell et al. (2011) has been regarded as a "benchmark" of discrete wet-extraction technique, but unfortunately, none of existing Antarctic CH_4 data for the early Holocene was measured by Mitchell et al. (2011) method. Most of WAIS discrete data covering the early Holocene were measured in a different institute (Penn State University, PSU) showing a noisy trend with pooled standard deviation of ~7.3 ppb (1σ), and there is an unexplained offset of 9.9 ppb between WAIS discrete data measured in OSU and PSU lab (Rhodes et al., 2015). As it lacks rigorous comparison between the two data sets during the early Holocene, there is no evidence to show that WAIS discrete data are more reliable. Meanwhile, it has been revealed that during the early Holocene interval, SNU Siple data (this study) agree well with OSU Siple data (Ahn et al., 2014) by comparison of the nearest data point from both labs. Furthermore, it should be noted that we used NEEM continuous data obtained by WS-CRDS (Wavelength Scanned Cavity Ring Down Spectroscopy, CFADS36, Picarro Inc.)) because the OF-CEAS (Optical Feedback Cavity Enhanced Absorption Spectroscopy, SARA, Laboratoire Interdisciplinaire de Physique, Universite Joseph Fourier, Grenoble, France) instrument was calibrated against different standard scale (CSIRO standard, Chappellaz et al., 2013), and WAIS continuous data were measured by the same instrument (WS-CRDS, 1804-2621m depth, 2012 campaign, Rhodes et al., 2015). In summary, we regard the NEEM discrete, Siple Dome discrete, and WAIS Divide continuous records as more reliable than the others during the early Holocene period.

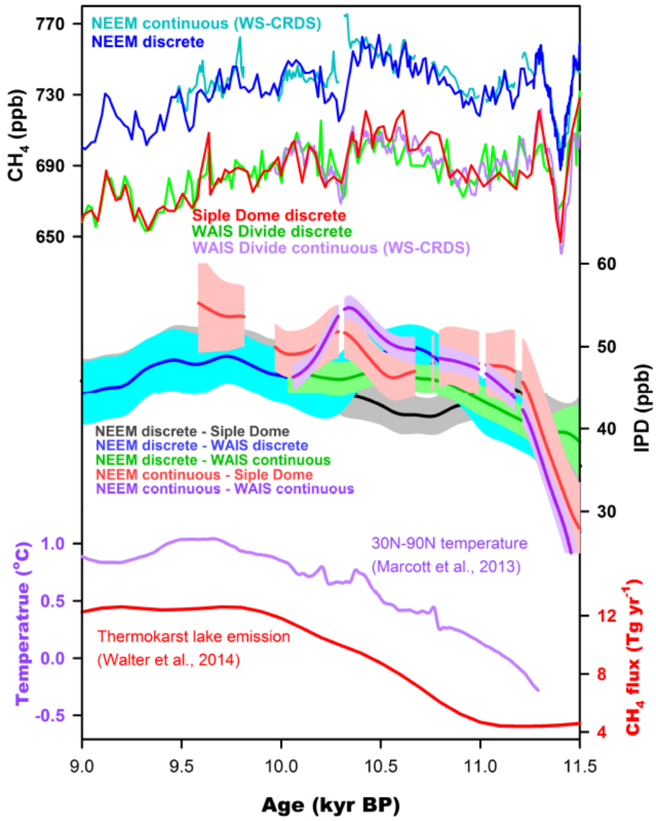

Figure R4. Same as Figure R3, but including various IPDs calculated from different dataset.

Table S1. 3-box source distribution model results of tropical (green, T) and boreal (red, N) boxes and boreal emission fraction (N/(T+N+S)) compared with previous results. Errors denote 95% confidence interval.

Ref.	N box	T box	N/(N+T+S) ratio
(ka)	($Tg\ yr^{-1}$)		(%)
Brook et al., 2000 (9.5-11.5 ka)	64 ± 5	123 ± 8	32 ± 3
Chappellaz et al., 1997 (9.5-11.5 ka)	66 ± 8	120 ± 9	33 ± 3
This study (9.5-11.5~~10.8~~ ka)	66 ± 4 ~~65 ± 2~~	120 ± 4 ~~122 ± 4~~	33 ± 2 ~~32 ± 1~~
This study (11.5 ka)	57 ± 6	119 ± 11	30 ± 4
This study (9.9 ka)	70 ± 4 ~~74 ± 2~~	115 ± 4 ~~110 ± 3~~	35 ± 2 ~~37 ± 1~~

Specific comments

I started to correct for English language issues, but had to stop at some point. Please ask your English speaking co-author for a thorough language check not only for typos but also to improve the clarity of the arguments. As major textual changes are still required for this manuscript, I will not comment on language issues here.

P(age) 2 l(ine) 2: Daniau et al., 2012 is not an appropriate reference in this respect (CH4 emissions)
We will replace the reference with citations below to deal with pyrogenic CH_4 emissions:

Ferretti, D. F., Miller, J. B., White, J. W. C., Etheridge, D. M., Lassey, K. R., Lowe, D. C., MacFarling Meure, C. M., Dreier, M. F., Trudinger, C. M., van Ommen, T. D., and Langenfelds, R. L.: Unexpected changes to the global methane budget over the past 2000 years, Science, 309, 1714-1717, 2005.
Andreae, M. O., and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cycles, 15, 955-966, 2001.
Hao, W. M., and Ward, D. E.: Methane production from global biomass burning, J. Geophys. Res., 98, 20657-20661, 1993.

P2 l20: Cite recent work by Baumgartner et al. CP 2014

We will add the citation.

P2: discuss in more detail previous work on the relationship between ITCZ changes and CH4 emissions

We will add a paragraph such as the following one in the introduction section:

"Relationship between the latitudinal shift of ITCZ and CH_4 emission varies with time scales. Landais et al. (2010) and Guo et al. (2012) suggested that ITCZ migration is not a dominant control of glacial-interglacial CH_4 cycle because long-term CH_4 trend does not follow well the precession change. Modelling studies found the southward shift of ITCZ coincides with reduced CH_4 in LGM and HS events, but changes in wetland area and surface hydrology were small (Weber et al., 2010; Hopcroft et al., 2011). They instead suggested that changes in temperature and/or plant productivity affected CH_4 production during those events.

Rather, ITCZ migration seems to be related with millennial- or submillennial scale CH_4 change. Brook et al. (2000) found that submillennial-scale CH_4 minima during the last deglacial period correspond with reduced precipitation recorded in Cariaco Basin sediment data, which indicates southward displacement of ITCZ (Hughen et al., 1996). It is supported by spectral analysis of CH_4 during the past 800 kyr record that found that ITCZ change becomes an important driver of millennial scale CH_4 change (Tzedakis et al., 2009; Guo et al., 2012)."

P3: discuss the difference in orbital parameters for the early and late Holocene and the potential implications for CH4 emissions

We will add a dedicated paragraph based on our comment to General comment a).

P4 methods: Is it correct that you use only a one standard calibration? Comment on the potential systematic error introduced by this approach

The GC linearity was tested by using working standards of 395.50, 721.31, 895.03, and 1384.91 ppb (in NOAA04 scale). We will add more details in the method section.

P4 l25-32: This paragraph should be moved to the methods section

We will move that paragraph to method section.

P5 1st paragraph. You say that you use a 250 year running average (and similar a high-pass filter with 1800 cut-off), however, your data is not equidistant. Please explain in more detail how you averaged the data

We interpolated the data annually and then averaged each 250-year interval. We will explain more about data filtering and averaging process.

P5 l14: Is the significance level of the correlation coefficient really taking the reduced degrees of freedom into account after averaging the data? Looking at the value, I am afraid it didn't and the significance is highly overestimated.

The p-value was estimated by a reduced degree of freedom. As described in Discussion Paper, Siple Dome CH_4 gas age was adjusted to GICC05 scale by matching to NGRIP $\delta^{18}O$ at 8.2 ka and PBO (e.g., Kobashi et al., 2007). We will note that it might overestimate the correlation coefficient.

P5: see comment on insufficient discussion of the effect of an ITCZ shift on CH4 emissions north and south of the equator. Please discuss also in more detail the dating uncertainties of the various archives and their potential impacts on the conclusions.

We will add age uncertainty (1σ) of each proxy used (Figure R2).

P5 l30: Reference Bjorck et al. is not in the reference list

The reference will be added.

Björck, S., Muscheler, R., Kromer, B., Andresen, C. S., Heinemeier, J., Johnsen, S. J., Conley, D., Koç, N., Spurk, M., and Veski, S.: High-resolution analyses of an early Holocene climate event may imply decreased solar forcing as an important climate trigger, Geology, 29, 1107-1110, 2001.

P6 l5-7: There is also variability in GRIP and GISP2. Please explain in more detail what you refer to.

We will delete the sentence.

P7 l1-17: This paragraph is highly speculative and lacks clarity and detail.

In this paragraph we intended to discuss possibility of solar forcing to observed CH_4 change. Several previous studies found evidences solar-induced climate change, but we observed that the timings of solar activity minima differ by 195 (8.2 ka), 278 (9.3 ka), 110 (10.3 ka), and 250 (11.0 ka) years to CH_4 minima and Greenland cooling. The maximum layer counting error of GRIP age scale (GICC05) is less than 100 years (Rasmussen et al., 2006), and the maximum gas age uncertainty of Siple Dome is ~150 years (This study). Therefore, age difference larger than ~180 years is not explained by chronological uncertainty.

P7 following l24: You disturbed the age of the data points by a Gaussian distribution with sigma=30 years. How did you make sure that the chronological order of all data points was ensured in your approach? How did you take the measurement uncertainty in each data point into account? Please explain in more detail.

We chose the sigma of 30 years given the mean temporal resolution of Siple data is ~27 years. We will describe the synchronizing process more detail.

P8 l1: there is a significant offset between your average and previous IPD estimates

Figure R3 (above) shows the IPD calculated from NEEM discrete and SDMA discrete data together with alternative IPDs from different data set. Previous IPD estimates lie within range of the new IPDs. The IPDs calculated from NEEM continuous data show higher values, which reflects the offset between NEEM continuous and discrete record.

P8 l10-12: not entirely clear to the outsider what you did, please clarify

We will modify the paragraph as below:

"~~To calculate the N-box CH_4, we subtracted the 7 % of IPD from Greenland CH_4 concentration, assuming the difference between Greenland and the mean latitude of N-box is ~7 % of IPD (Chappellaz et al., 1997).~~"

"The mean CH_4 concentration of N-box (30-90N) is not identical to that of Greenland ice core record, given the latitudinal CH_4 distribution (e.g., Fung et al., 1991). To derive the N-box CH_4, we followed the assumption of Chappellaz et al. (1997), where the authors assumed that difference between Greenland and the mean N-box CH_4 is 7% of IPD. Hence here the N-box CH_4 is calculated by subtracting 7% of IPD from the Greenland concentration."

P8 l19: the boreal sources increased

This phrase will be changed as below:

"This result supports our interpretation that the boreal sources ~~were less reduced than those in low latitudes~~ increased during the early Holocene."

P9 l10. You discuss the effect of the different age distributions in the Siple Dome and NEEM cores, but you do not follow up on this in your analysis. Either you use WAIS to compare with NEEM (as it has essentially the same enclosure characteristics) or you low-pass filter NEEM to the same enclosure characteristics as Siple. I would strongly recommend to do both to study the robustness of the results.

See our response to general comment above. We present alternative IPD reconstructions including WAIS and NEEM data.

P9 l20-21: The results by Fischer et al. (2008) on LGM biomass burning emissions result from the use of temporally constant isotopic source signatures in the box model approach. Moller et al., Nature Geoscience, 2013 showed that also the source signatures changed significantly over time and they revised the biomass burning estimates, showing that LGM emissions were lower than Holocene emissions.

We will insert below paragraphs (P9 l20-31):

"On the other hand, Fischer et al. (2008) argued nearly constant biomass burning emission of ~45 Tg yr^{-1} throughout the last glacial termination with a slight increase in PB, and also showed that the boreal sources were expanded during the YD-PB transition. However, Moller et al. (2013) pointed out the possibility of changing isotopic signature of each sources itself, and they found that less pyrogenic emission is required for LGM condition if they increased the $\delta^{13}C$-CH_4 signatures of tropical wetland and biomass burning. The triple isotopic … old carbon (e.g., Petrenko et al., 2009). ~~Therefore, the cause of the high IPD at the start of the Holocene still remains elusive.~~ The IPDs at the very start of the Holocene and during the PBO show large offset among each other that prevents us from drawing a reliable trend because IPD calculation could be sensitive to sample resolution and calibration."

P9 l26: why do you only refer to biomass burning in the tropics?

According to model estimation by Walter-Anthony et al. (2014), CH_4 emission from the thermokarst lakes started to increase more later than PBO. We will describe this in that sentence.

P20 l5 Chappellaz et al., 1997 not 2013
The citation will be corrected.

References that are not cited in discussion paper

Andreae, M. O., and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cycles, 15, 955-966, 2001.

Baumgartner, M., Kindler, P., Eicher, O., Floch, G., Schilt, A., Schwander, J., Spahni, R., Capron, E., Chappellaz, J., Leuenberger, M., Fischer, H., and Stocker, T. F., NGRIP CH_4 concentration from 120 to 10 kyr before present and its relation to an $\delta^{15}N$ temperature reconstruction from the same ice core, Clim. Past, 10, 2014.

Björck, S., Muscheler, R., Kromer, B., Andresen, C. S., Heinemeier, J., Johnsen, S. J., Conley, D., Koç, N., Spurk, M., and Veski, S.: High-resolution analyses of an early Holocene climate event may imply decreased solar forcing as an important climate trigger, Geology, 29, 1107-1110, 2001.

Dai, A., and Wigley, T. M. L.: Global patterns of ENSO-induced precipitation, Geophys. Res. Lett., 27, 1283-1286, 2000.

Ferretti, D. F., Miller, J. B., White, J. W. C., Etheridge, D. M., Lassey, K. R., Lowe, D. C., MacFarling Meure, C. M., Dreier, M. F., Trudinger, C. M., van Ommen, T. D., and Langenfelds, R. L.: Unexpected changes to the global methane budget over the past 2000 years, Science, 309, 1714-1717, 2005.

Flückiger, J., Blunier, T., Stauffer, B., Chappellaz, J., Spahni R., Kawamura, K., Schwander, J., Stocker, T. F., and Dahl-Jensen, D., N_2O and CH_4 variations during the last glacial epoch: Insight into global processes, Global Biogeochem. Cycles, 18, GB1020, 2004.

Guo, Z., Zhou, X., and Wu, H.: Glacial-interglacial water cycle, global monsoon and atmospheric methane changes, Glim. Dyn., 39, 1073-1092, 2012.

Hao, W. M., and Ward, D. E.: Methane production from global biomass burning, J. Geophys. Res., 98, 20657-20661, 1993.

Hopcroft, P. O., Valdes, P. J., and Beerling, D. J.: Simulationg idealized Dansgaard-Oeschger events and their potential impacts on the global methane cycle, Quat. Sci. Rev., 30, 3258-3268, 2011.

Hughen, K. A., Overpeck, J. T., Peterson, L. C., and Anderson, R. F.: The nature of varved sedimentation in the Cariaco Basin, Venezuela, and its palaeoclimatic significance, Geol. Soc. London Spec. Publ., 116, 171-183, 1996.

Landais, A., Dreyfus, G., Capron, E., Masson-Delmotte, V., Sanchez-Goñi, M. F., Desprat, S., Hoffman, G., Jouzel, J., Leuenberger, M., and Johnsen, S.: What drives the millennial and orbital variations of $\delta^{18}O_{atm}$?, Quat. Sci. Rev., 29, 235-246, 2010.

Lyon, B., and Barnston, A. G.: ENSO and the spatial extent of interannual precipitation extremes in tropical land areas, J. Clim., 18, 5095-5108, 2005.

Strikis, N. M., Cruz, F. W., Cheng, H., Karmann, I., Edwards, R. L., Vuille, M., Wang, X., Paula, M. S., Novello, V. F., and Auler, A. S., Abrupt variations in South American monsoon rainfall during the Holocene based on a speleothem record from central-eastern Brazil, Geology, 39, 2011.

Tzedakis, P. C., Palike, H., Roucoux, K. H., and de Abreu, L.: Atmospheric methane, southern European vegetation and low-mid latitude links on orbital and millennial timescales, Earth Planet. Sci. Lett., 277, 307-317, 2009.

Wang, X., Auler, A. S., Edwards, R. L., Cheng, H., Ito, E., and Solheid, M., Interhemispheric anti-phasing of rainfall during the last glacial period, Quat. Sci. Rev., 25, 3391-3403, 2006.

Wang, Y., Cheng, H., Lawrence Edwards, R., Kong, X., Shao, X., Chen, S., Wu, J., Jiang, X., Wang, X., and An, Z.: Millennial- and orbital-scale changes in the East Asian monsoon over the past 224,000 years, Science, 451, 1090-1093, 2008.

Weldeab, S., Lea, D. W., Schneider, R. R., and Andersen, N., Centennial scale climate instabilities in a wet early Holocene West African monsoon, Geophys. Res. Lett., 34, L24702, 2007.

Weber, S. L., Drury, A. J., Toonen, W. H. J., and van Weele, M.: Wetland methane emissions during the Last Glacial Maximum estimated from PMIP2 simulations: Climate, vegetation, and geographic controls, J. Geophys. Res., 115, D06111, 2010.

Zürcher, S., Spahni, R., Joos, F., Steinacher, M., and Fischer, H.: Impact of an abrupt cooling event on interglacial methane emissions in northern peatlands, Biogeosciences, 10, 1963-1981, 2013.