We thank the anonymous referee #1 and #2, and the Editor for her/his careful reviews of our paper. We appreciate the useful comments and believe the input improved greatly the manuscript. Below we attach our point-by-point response to all the comments. The original referee comments are copied in black, and the author's response to the comments are given in red italics. We add paragraphs from original discussion paper in green italics and our modifications in blue italics.

Editor Report----------

Comments to the Author:

Thank you for the very detailed responses to the comments from the two reviewers. Particularly through your more detailed treatment of the IPD it seems that you should be able to make a new version that will satisfy most of the concerns of the reviewers. I am therefore now happy to encourage you to prepare the new version, which I expect to send back for re-review

We sincerely appreciate the Editor for giving us chance to revise and update our manuscript.

On the analytical isde, I still don't feel that I understand how you can end up at a precision of 1 ppb when you have blanks varying by 10 ppb. Nor do I really understand how you arrive at a headline figure of 1 ppb when the mean absolute difference between replicate measurements was about 2 ppb. I think this might require more thought as my feeling is that the reviewers will share my concerns.

We revised the gas solubility correction scheme following the empirical method of Mitchell et al. (2011) for better fit to OSU-measured SDMA data (0.6 ppb difference instead of 3 ppb before). With new correction method, the pooled standard deviation between replicates increased slightly to 1.4 ppb, but it does not change the main plot of this manuscript. The good agreement between 1st and 2nd replicate measurements reveals that our correction method for daily blank offset is valid. The "intra-day" offset among the 4 blank is about 2 ppb, but given that all the samples were measured in duplicate, this figure should be reduced by a factor of $\sqrt{2}$, which yields similar uncertainty to the stated precision.

For the IPD, I do appreciate the multiple parallel calculations, but again have two concerns. The first is simply presentational. In Fig R2 the Siple based reconstruction, central to your paper is almost completely invisible. But now I look at it in Fig R3 and it is completely different from what you showed in the original paper (Fig 3). The absolute values of IPD (NEEM/SD) there ranged from ~42-52 ppb, now you show 30-48. The shape is different as well. This is not a question that WAISDivide differs from SD, but your SD/NEEM profile has changed considerably, and yet I can't find any discussion of this in the response. The IPD at 11.5 ka, using the same (NEEM/SD) data, has changed from 52 to around 35 ppb, a very considerable change. Have you resynchronised or changed some values? Please address this in the material you submit with the paper that addresses the review comments.

Since the initial manuscript submission, we extended IPD synchronization interval back to ~12 ka for better matching during the YD termination. This has not changed the essential findings of our manuscript, and the section 3.2.2. has been removed because it is out of main focus of our paper. Furthermore, the Supplementary Figure 3 (Figure S3) has been removed and replaced by a plot that shows the alternative IPDs not being discussed in the main text.

Some smaller points from me:
You say Fig R1 includes Fluckiger but i don't see it.

We included EPICA Dome C data set by Loulergue et al. (2008) instead of Flückiger et al. (2002), which is more recently published data from EDC core.

For the second rev, Fig R1, you should use the same y-scale for panels a and b so readers can compare the magnitude of variations directly.

We adjusted both y-axes to have the same scale and same size as suggested.

Referee Comment #1----------

Summary of manuscript

First of all, I would like to congratulate Ji-Woong Yang et al. for the excellent work they put into producing a high-resolution record of CH_4 mole fractions as well as their interpretation of the data. So far, the early Holocene is underrepresented in high-resolution CH_4 reconstructions and this paper will be a valuable addition to the literature. I hope the following comments will be helpful and I look forward to reading the revised version of the manuscript.

Ji-Woong Yang et al. reconstruct the CH_4 variability of the Early Holocene, from 11.6 to 8.5 ka before 1950, using a melt-refreeze extraction system coupled to a GC-FID analyser, which was newly developed at Seoul National University (SNU). The new method is very briefly described in this paper and not yet published. The authors show that the SNU data are in good agreement with two existing benchmark records from the WAIS divide ice core, where the latter two were measured using *i*) a similar technique (WAIS members 2015) and *ii*) a sample gas stream derived from a continuously melted ice core, analysed by an optical instrument (Rosen et al., 2015).

Ji-Woong Yang et al. observe millennial CH_4 minima besides the 8.2 ka event, which have not been identified in previous studies. The authors relate these CH_4 minima to events in other geological records that indicate climate variability in the low and high latitudes of the Northern Hemisphere. These records include: $\delta^{18}O$-H_2O (NGRIP), ice rafted debris, ^{10}Be, reflectance of Cariaco Basin sediments, $\delta^{18}O$-$CaCO_3$ (speleothems) and $\Delta\varepsilon$. The authors show convincingly that the millennial CH_4 variability correlates with millennial variations in $\delta^{18}O$-H_2O (NGRIP)/Greenland temperature, which is a new and interesting finding. The authors furthermore discuss the relation of CH_4 with the other records and suggest that Northern Hemispheric cooling and a concomitant southward shift of the ITCZ created a teleconnection pattern of reduced intensities of Asian and Indian monsoons. Thereby, the authors identified changes in CH_4 emissions from tropical wetlands as the most likely cause of the millennial CH_4 minima. The authors claim that this mechanism cannot explain the CH_4 minima around 10.2 ka alone.

In a next section, the authors review how the variability in external forcing during may cause an “El Nino-like” climate. They discuss some relation in the climate system but how this is hypothetically related to their CH_4 data remains unclear. The authors conclude this discussion cannot be developed further as there is no ENSO reconstruction for the Early Holocene. The purpose of this section is not entirely clear to me, also in the light of a range of existing publications on ENSO reconstructions (e.g.

Z. Liu et al., 2014, Nature, Vol. 515, p. 550-553)

In order to investigate the CH_4 record further, the authors calculate the inter-polar difference in CH_4 (IPD) using the presented SNU and previously published NEEM data. The calculated IPD record is close to the range of previously published estimates, but exhibits interesting features on millennial time scales. The authors suggest a high IPD at the onset of the Holocene, which then decreases between 11.1 and 10.7 ka and increases again between 10.7 and 9.9 ka to previous levels. They furthermore use a previously published box model to separate hemispheric and tropical CH_4 emissions. The authors discuss the variability of tropical and boreal source fractions over time in the light of other studies and conclude that the IPD increase between 10.7 and 9.9 ka is a result from Northern Hemispheric warming/thawing and expansion of boreal wetlands. This is in convincingly good agreement with previous reconstructions of increased CH_4 emissions from boreal wetlands.

The study of Ji-Woong Yang et al. is a valuable contribution and in the scope of Climate of the Past. I would recommend the publication of this manuscript but think that major revisions are required.

General comments:

1) Ice core: The authors could provide more complete information on the ice core and related logistics. For example, it is not explicitly mentioned that the samples were shipped from the USA to South Korea (just names of institutions). Neither did the authors mention the year the ice core was retrieved nor how long it was stored prior to analysis. Do the authors think that storage time affects the analysis? Do the authors think that extended storage time could help with the handling of samples from the brittle ice zone? The authors did not specifically address that their samples were from the brittle ice region. However, I would strongly recommend to raise this point and how it might or might not have affected the data.

Now we added more details on samples and ice logistics as below:

"In this study we used ice samples from Siple Dome A (SDMA) deep ice core drilled from 1997 to 1999 on the Siple Coast, West Antarctica (81.65°S, 148.81°W; 621m elevation) (Taylor et al., 2004). The SDMA samples were selected and cut at National Ice Core Lab (NICL), Denver from January to February 2013. Since brittle zone of SDMA ice starts below 400 m depth (Gow and Meese, 2007) that makes some part of ices fractured and/or cracked internally. Hence the samples were carefully collected from unbroken ices during the sample preparation at NICL. The samples were packed in isothermal foam boxes with numerous eutectic gels, and shipped to South Korea via expedited air freight. Temperature loggers were enclosed within the isothermal boxes to record the temperature change inside during the logistics, and it showed the temperatures were maintained below -25°C for each box. The boxes were picked up directly just after custom clearance at the airport and then the ice samples were stored in a walk-in freezer of Seoul National University (SNU) that maintained below -20°C."

2) The analytical system: Even though the authors are preparing another manuscript on the analytical method, a more detailed description of the analytical system would be helpful. For example, the authors describe their melt-refreeze method as "traditional", though, I am in doubt that most readers have a melt-refreeze technique in their lab, if they work in a lab at all. I find the term "traditional" misleading, as it might be taken as a support for the performance of the method.

We agree with the suggestion. We deleted the expression "traditional" and added more detailed description of the analytical method as follows:

"The air occluded in ice was extracted by melting and refreeze process under vacuum. Ice samples were prepared in a walk-in freezer in the morning of each experiment day, and trimmed the outermost >2 mm to eliminate possible contamination by ambient air during the storage. Then the samples were moved to the laboratory and placed in glass sample containers. The sample containers (sample flask hereafter) were custom-made glass flasks welded to stainless steel flange, and attached to the vacuum line with a copper gasket. ~~Each day we normally analysed 3 samples in duplicate and four blank samples (bubble free ice made in the laboratory, with standard air added to the sample flask prior to air extraction).~~ The sample flasks were partially submerged into a chilled ethanol bath during ice insert and attaching to the line for preventing temperature increase by laboratory air. After that all flasks were evacuated at least 40 minutes, the ice samples were melted by submerging the sample flasks in a warm water bath. Melting process was usually completed within 30 minutes. The sample flasks were then submerged in the cold ethanol bath chilled to around -82 ℃ more than 1 hour to refreeze. During the refreezing, we carried out GC pre-running (20 injections) and daily calibration that normally took 90 minutes. The ethanol temperature rises up to -55°C just after submerging the flasks, and it was chilled to below -65°C before expansion of the air in the flasks. The extracted air in the head space was expanded into a gas chromatograph (GC) equipped with a flame ionization detector (FID) to measure CH_4 mixing ratio. The GC linearity was tested by a series of inter-tank calibration using four working standard air cylinders (395.5, 721.3, 895.0, and 1384.9 ppb CH_4 in NOAA04 scale, Dlugokencky et al., 2005). Daily GC calibration curve was determined by measurements of a working standard having the closest CH_4 mixing ratio of expected value from the samples; in this study we used 721.3 ppb CH_4 standard for samples of the early Holocene. To account for system condition change throughout experiments (i.e., influence by water vapor), we calibrated with a standard air 6 times before and after sample measurements. The detailed configuration of the vacuum line and GC is described in another paper that is currently in preparation (Yang et al., in preparation). Gas extraction line was evacuated to under detection limit of 0.1 mTorr, and system leak check was done as a daily routine before sample preparation. If any pressure increase in 10^{-4} Torr scale for 30 seconds is detected, the experiment was stopped to figure out the leakage."

I think the presentation of the analytical performance needs to be developed further. The authors describe their system as similar to Mitchell et al., (2011). To my understanding, the system of Mitchell et al., (2011) is the benchmark GC-FID system in the community, with an estimate for measurement uncertainty of ±2.8 ppb, based on the pooled standard deviation of replicate samples that were measured with extended periods of time between the analysis of each sample pair. That is, the uncertainty estimate of Mitchell et al., (2011) can be understood as "worst case scenario". The method of Yang et al. is presented with an uncertainty of ±1.0 ppb, which is determined by the pooled standard deviation of 8 replicate measurements. While I am more than happy to be convinced that a newly developed method is superior in performance to an existing method, I feel strongly that this claim has to be proven. I think a more detailed description of the method to determine the uncertainty estimate is required, especially because the method of Yang et al. is supposed to be by far superior than that of Mitchell et al., (2011). In the light of a total number of 295 samples measured for this study, the authors need to describe why they chose these specific 8 replicate samples to determine the analytical uncertainty and why they did not chose other samples.

Our CH_4 data are presented by averaging the results of duplicate sample analysis from the same

depths. In order to estimate our data precision, we reanalyzed duplicate samples from the adjacent ices (~10 cm depth difference) at 8 depth intervals 8-80 days after the first analysis (see Table R1). The depth intervals were randomly chosen. The pooled standard deviation of the average of duplicates from first and second measurements was 1.5 ppb (mean difference of 1.9 ppb).
We note that Mitchell et al. (2011) used different solubility correction methods. Applying the solubility correction method described in Mitchell et al. (2011) yields pooled standard deviation of the average of duplicates at adjusted samples of 1.4 ppb. One of the main differences of our analytical system compared to the OSU one is that, while in OSU the blank test was done once in several days and the systematic offset was interpolated between the blank days (Mitchell et al., 2011), we measured at least 3-4 blank ices every day. By doing this, the systematic offset can be quantified more precisely that accounts for daily changing conditions of instrument.

Table R1. Summary of the first (original) and second (replicate) measurements from the depths used for system reproducibility test.

Depth (m)	*1st measurements*		*2nd measurements*		*Difference*	
	CH_4 (ppb)	Date (dd/mm/yy)	CH_4 (ppb)	Date (dd/mm/yy)	$1^{st} - 2^{nd}$ (ppb)	Time (days)
523.15	*631.8±0.1*	*27/01/14*	*632.7±1.6*	*24/02/14*	*-0.9*	*29*
530.95	*663.0±2.2*	*03/02/14*	*664.7±1.6*	*24/02/14*	*-1.7*	*22*
558.295	*676.7±2.1*	*14/03/14*	*679.3±4.5*	*02/04/14*	*-2.6*	*20*
559.85	*682.0±5.8*	*03/02/14*	*684.7±1.9*	*26/03/14*	*-2.7*	*52*
561.15	*685.0±0.8*	*14/03/14*	*683.7±3.1*	*02/04/14*	*1.3*	*20*
562.407	*682.8±0.8*	*26/03/14*	*684.3±1.5*	*02/04/14*	*-1.5*	*8*
575.913	*676.5±0.2*	*07/02/14*	*679.3±3.8*	*28/03/14*	*-2.8*	*50*
578.15	*676.2±4.3*	*04/02/14*	*674.9±2.3*	*24/04/14*	*1.3*	*80*

Furthermore, the method includes several corrections, including blank (determined with bubble-free ice, 5-15 ppb), gravitational fractionation (1.97±0.15 ppb) and correction for dissolved CH_4 (range should be specified). These corrections include uncertainties that should be represented in the uncertainty budget of the method. Furthermore, a blank of 5-15 ppb is enormous, both in absolute values as well as in the range, especially when the total performance is stated with ±1.0 ppb. For comparison, the method of Mitchell et al., (2011) has a blank of 1.1±0.5 ppb, while a blank of 5-14 ppb was interpreted as indicator of leakage. I think this issues need to be clearly demonstrated so that superior performance can be claimed.

The blank offset, which is calculated from the mean of the 3-4 blank results, reflects any errors by contaminants, leaks, or different GC conditions. The exact cause of the blank offset is not clear, but the important thing is that the 4 blank results agree well each other, yielding the intra-day standard error of the mean of 2.0±1.0 ppb. Since every single data point is obtained by analysis of at least in

duplicate, the intra-day blank offset for one depth is reduced by a factor of √2. Comparede to inter-day blank offset of 5-14 ppb, the small intra-day blank offset implies that the daily offsets of the 10 sample flasks are rather systematic. The robustness of our final results was proven by reanalysis of duplicates at adjacent depths (< 10 cm) 8-80 days after the first analysis. The difference of mean of duplicates between the time intervals was 1.9 ppb on average (see the table above).

System leakage is unlikely. We carried out system leak check as a daily routine before starting gas extraction. If any pressure increase in 10^{-4} Torr scale for 30 seconds is detected, the experiment was stopped to figure out the leakage.

Further issues that could be clarified include the uncertainty of the standard air (available for each NOAA04 cylinder), how the linearity of the system is controlled (additional air standards covering the analytical CH_4 range?) and what reason the authors base their decision on to subtract 3 ppb from the OSU data instead of adding 3 ppb to the SNU data or to take the 3 ppb as temporal signal? If the measurement uncertainty claimed by both institutes is realistic, both data should be on the NOAA04 scale and therefore be in close agreement. I think this is in the order of expected disagreement, but I feel the manuscript is stronger if these issues are clearly addressed.

It seems that the 3 ppb offset is due to different correction for gas solubility effect. Unlike to Mitchell et al. (2011), we calculated the expected gas enrichment by assuming that equilibrium stateis achieved between air-water during the melting process. In OSU, the gas solubility effect is empirically estimated by measuring CH_4 mixing ratio from the air that is re-dissolved after melt-refreeze process (Mitchell et al., 2011). The directly-measured OSU correction method seems more realistic, but the uncertainty is large (~10%) due to small amount of air. After applying the OSU solubility correction scheme, the average difference between SNU and OSU reduces to less than 1 ppb. Therefore, for the better fitted composite record, we applied the OSU correction method to SNU data set.

3) CH_4 data: The authors mention 295 measurements. However, Figure 1 seems to show a much smaller number. If the displayed measurements are averages of duplicates or if other measurements are not displayed in Figure 1, the number would have to be revised.

The number was corrected as below. Basically we made duplicates for all depths, and the average of the replicates was used for the data.

"We measured 295 samples from 156 depth intervals from 518.87 to 718.83 m, covering from 8.36 to 20.25 kyr BP after synchronizing to the Greenland Ice Core Chronology 2005 (GICC05). All samples were duplicated, so that our final CH_4 data were presented by averaging the results of duplicate sample analysis from the same depths and analytical uncertainty is deduced from standard error of the mean of duplicate pairs. ~~We measured a total of 295 samples on 143 depth intervals from 518.87 to 623.38 m, of Siple Dome ice core, West Antarctica.~~"

Otherwise, please clarify. How long is the overlapping period between OSU and SNU data? Maybe Figure 1 could show this in a detailed Figure?

We added an enlarged figure in Figure 1 (See Figure R1). The overlapping period is from ~8.4 to ~9.1 ka. To make the SDMA CH_4 composite, we used the OSU data from 7.7 to 8.5 ka, due to higher mean temporal resolution of the OSU data.

Figure 1 has a reference in the captions (Brook and Sowers, 2016) that is new to me and that I cannot find in the reference list.

Reference was modified.

The authors may or may not consider to show data from previous publications (e.g. Brook et al., 2000, Flueckiger et al., 2002) to highlight the superiority of their temporal resolution.

Figure 1 was modified as Figure R1 that includes the data from Tayler Dome, EPICA Dome C, and Talos Dome (see below).

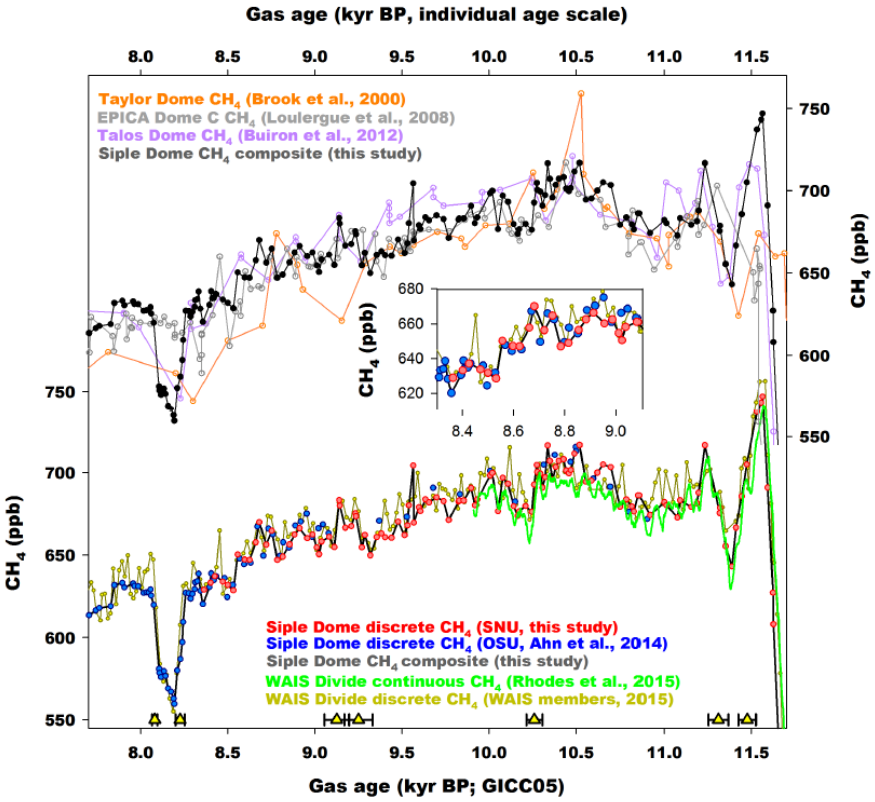

Figure R1. CH_4 reconstructions during the early Holocene. Top: new high-resolution Siple Dome CH_4 data (black, this study) compared with previous records from Taylor Dome (orange, Brook et al., 2000), EPICA Dome C (grey, Loulergue et al., 2008), and Talos Dome (purple, Buiron et al., 2012). Bottom: Siple Dome CH_4 records measured in OSU (blue, Ahn et al., 2014) and SNU (red, this study). Siple Dome composite (black line) is plotted with WAIS Divide discrete (dark yellow, WAIS Divide project members, 2015) and continuous technique (green, Rhodes et al., 2015). Inset: Enlarged plot showing overlapped interval between OSU and SNU Siple Dome data.

4) IPD: IPD is a powerful concept, but very one has to be very careful in its reconstruction and interpretation. The authors mention the potential for ill-calculated IPDs based on errors in the gas age scale of the CH_4 records. Therefore, the authors developed a tool to synchronize the CH_4 records, which I think is a very good approach! However, it is not clear to me why the authors chose to calculate the IPD based on the Siple Dome data? My concerns have several reasons:

i) The authors state themselves that the histories of gas enclosure is more similar between NEEM and WAIS. The authors state that, based on just this fact, the amplitude of CH_4 variations is 10-20 ppb larger in the WAIS than in the Siple Dome record. Therefore, I understand that both the NEEM and the Siple Dome CH_4 records are altered by physical processes during gas enclosure that are different for each core. I understand that a dampened amplitude in the Siple Dome CH_4 record

would create a IPD variation. Ideally, gas enclosure effects should be identical in both records so that they would cancel. Since the WAIS record is in that sense more similar to the NEEM record than the Siple Dome record is, I would suggest to use the WAIS record for the IPD reconstruction.

ii) The WAIS record is of even higher temporal resolution than the Siple Dome record. I would expect that the IPD reconstruction based on NEEM and WAIS would be more robust.

iii) The comparison of the CH_4 records from Siple Dome and WAIS in Figure 1 shows two periods (~10.5 and ~10.7 ka) where Siple Dome CH_4 exceeds WAIS CH_4 by up to ~20 ppb. During this period, the smoothed CH_4 variations (Figure 2) also show an ice core specific pattern of disagreement. Around 11 ka, the pattern of agreement is different. Here, the continuous CH_4 record from WAIS seems to agree better with CH_4 from Siple Dome, while the GC-FID record from WAIS contains a number of samples that exceed the former records by ~20 ppb. The difference between the records during these times exceeds the stated measurement uncertainty by far. It is also important that the difference seems to be in the order of the presented IPD variability.

All of the above mentioned reasons directly impact on the IPD reconstructions. The choice of the authors to use their Siple Dome data for the IPD reconstruction is justified and understandable. However, I fear that the interpretation is sensitive to the choice of CH_4 record so that this choice could impact on the IPD result for the above mentioned reasons. Therefore, I suggest to calculate the IPD also using both WAIS records as three independent sets of IPD reconstructions as a sensitivity test. This will make the interpretation of reconstructed IPDs more robust and will furthermore give valuable insights into the IPD technique on high temporal resolution records for future studies. Because this concerns one of the main outcomes of this manuscript, I would consider this essential.

We appreciate the Referee #1 for her/his useful and reasonable comment on IPD reconstruction. Calculating IPD from different ice core data sets would draw more objective conclusion. We provided alternative IPD reconstructions from various records (NEEM discrete, NEEM continuous, WAIS discrete, WAIS continuous, and Siple Dome discrete data) to test robustness of early Holocene IPD trend (We denote the original IPD between NEEM discrete and SDMA discrete data as "IPD-1" hereafter).

As mentioned in comment iii) above, the Siple- and WAIS ice core records do not agree at some period, and this offset could lead erroneous IPD change. However, here we consider Siple data show more reliable result by following reasons. First, it was shown that SNU Siple Dome discrete data (this study) have a good agreement with OSU Siple Dome discrete data (Ahn et al., 2014) during the early Holocene interval, while PSU (Penn State University) WAIS discrete data that covers most of the early Holocene period show offset up to ~9.9 ppb to OSU data (Rhodes et al., 2015). Second, PSU WAIS discrete data show a pooled standard deviation (for depth-adjacent samples) of 7.3 ppb (1sigma), which is larger than SNU Siple data. In addition, NEEM continuous- and discrete data do not agree well in some intervals (Figure R2), even though NEEM continuous data were calibrated against to discrete measurements carried out at OSU (Chappellaz et al., 2013). Further, the NEEM continuous record is not exactly "continuous", that may introduce uncertainty into synchronization. Hence, here we regard the NEEM discrete, Siple Dome discrete, and WAIS Divide continuous records as more reliable ones than the others during the early Holocene period (black and green curves in Figure R2). Resulting IPD curve from NEEM discrete and WAIS continuous (green, IPD-2 hereafter) shows long-term increase from the onset of Holocene to ~10.0 kyr BP. This indicates that contribution from

boreal sources increased during ~11.5 to 9.9 kyr BP, which is consistent with increase of northern extratropical temperatures and thermokarst lake CH_4 emissions. The revised 3-box model results obtained from IPD-1 (Table R1) show elevated emission from N-box and slight decline in T-box. However, since a small IPD increase during ~10.8-11.2 kyr BP observed in Siple Dome IPD is not supported by alternative IPDs, it remains unclear the short-term IPD change during this time period. Therefore, we modified descriptions and findings on early Holocene IPD trend.

Table R1. 3-box source distribution model results from IPD-1. Tropical (green, T) and boreal (red, N) boxes and boreal emission fraction (N/(T+N+S)) are 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~~

Table R2. 3-box source distribution model results from IPD-2.

Ref.	N box	T box	Boreal source fraction
(ka)	(Tg yr^{-1})		(%)
This study (11.5 ka)	60 ± 7	134 ± 16	29 ± 4
This study (10.0 ka)	71 ± 7	115 ± 11	35 ± 4

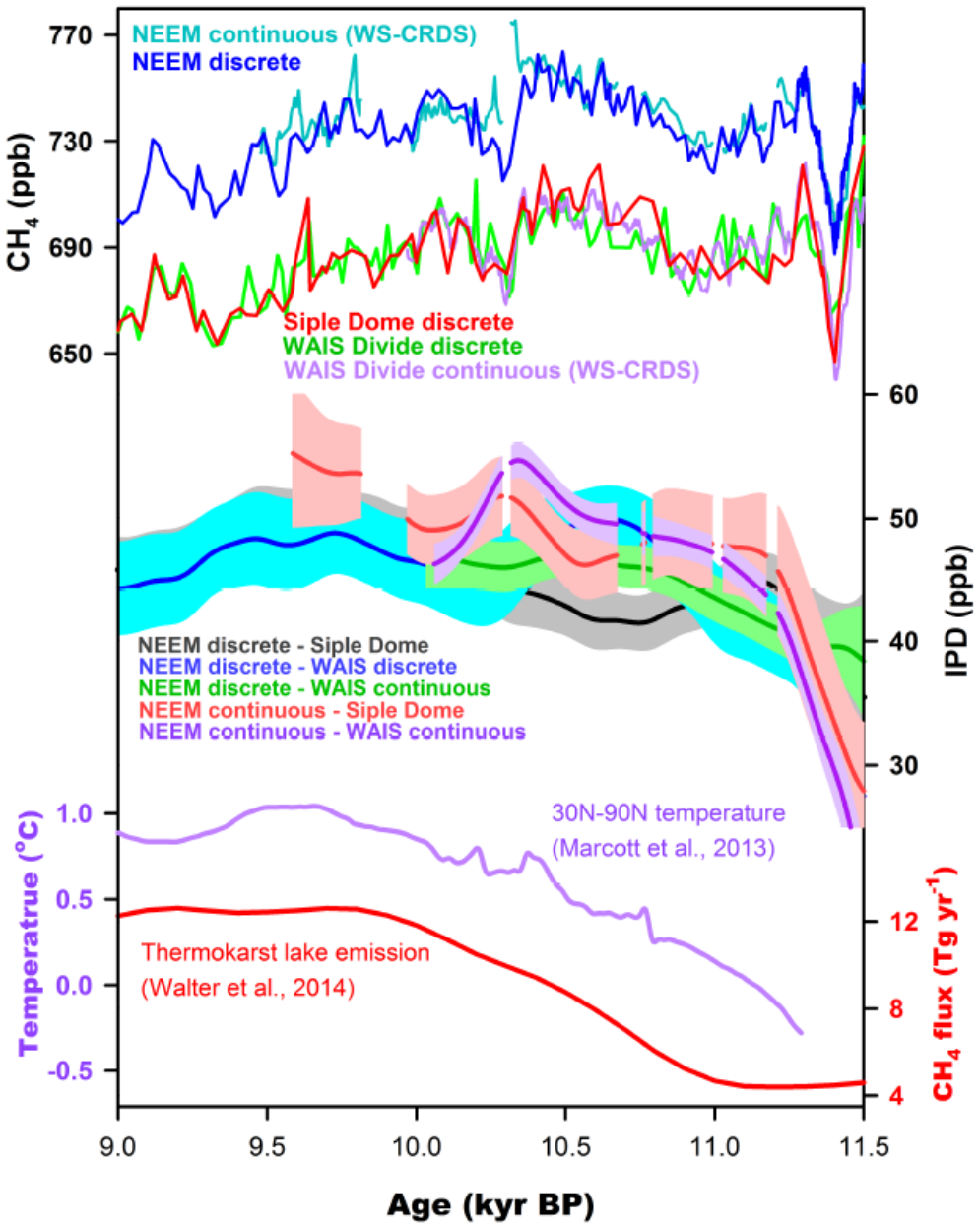

Figure R2. Inter-polar difference (IPD) reconstructions. Top: high resolution CH_4 records from Greenland and Antarctica, synchronized to NEEM gas age scale by Monte Carlo procedure. Middle: Millennial-scale IPD trends derived from various pairs of data set. Shaded area indicates 95% significance interval. Bottom: Proxy-based temperature reconstruction for northern mid to high latitude and boreal CH_4 emission from northern thermokarst lakes.

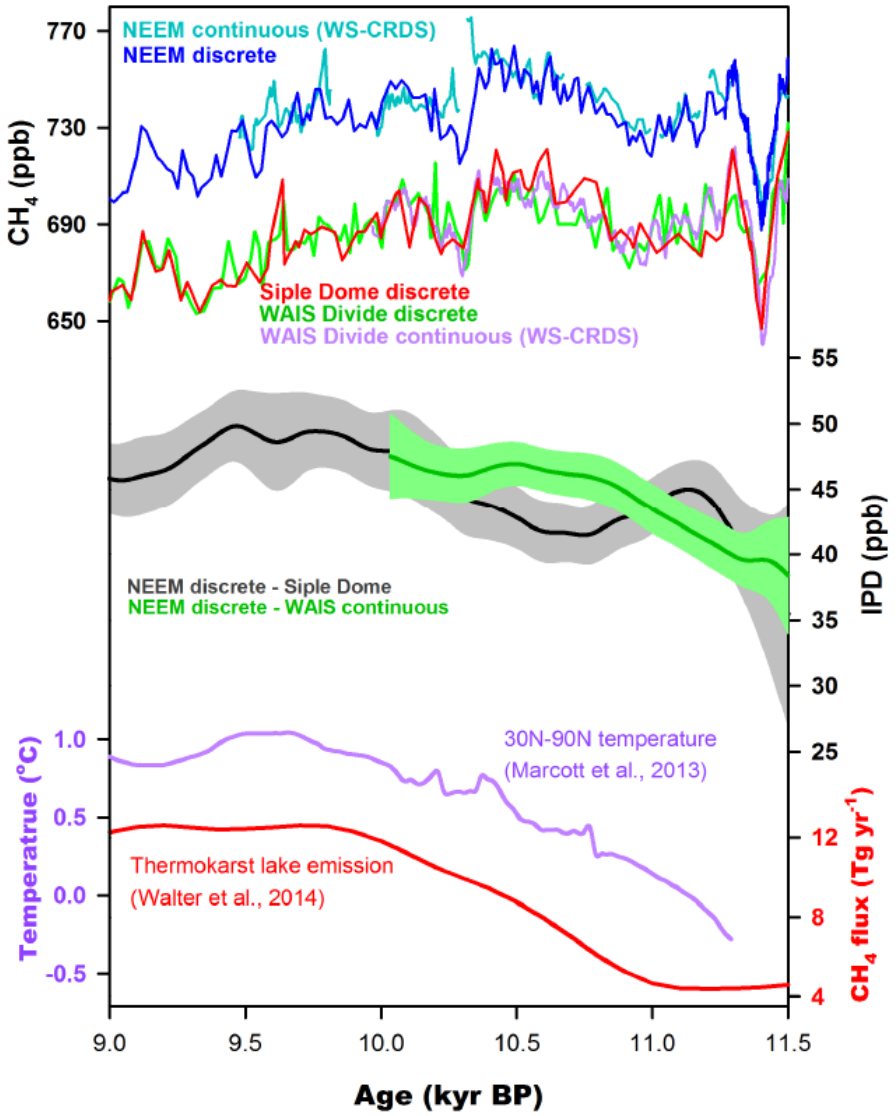

Figure R3. Same as Figure R2, but shown are IPD-1 (black, NEEM discrete – SDMA discrete) and IPD-2 (green, NEEM discrete – WAIS continuous) only.

5) Consideration of significant publications: *i*) The authors claim that no correlation between CH_4 and tropical monsoon signals has been reported on shorter time scales, however, I feel this is not accurate. Both Cruz et al., (2005, Nature, Vol, 434, p. 63-65) and Sperlich et al., (2015, Global Biogeochemical Cycles, 29) have related CH_4 and $\delta^{13}C$-CH_4 records to South American speleothem records, respectively. Both publications would principally support the interpretation of this study. *ii*) The authors discuss that their IPD reconstruction suggests increasing boreal source fractions during the early Holocene and support their finding with studies on boreal wetland dynamics. However, their finding of increased boreal source fractions is in line with the interpretation of $\delta^{13}C$-CH_4 data by Fischer et al., (2008) and Sowers, (2010). Again, both publications would principally support the interpretation of this study while Sowers (2010) had the same finding for the early Holocene previously. *iii*) The authors stated that there is currently no ENSO reconstruction for the early Holocene, even though a range of them exist (e.g. Z. Liu et al., 2014, Nature, Vol. 515, p. 550-553, Clement et al., 1999, Paleoceanography, Vol. 14, p. 441-456).

We checked the suggested publications and take them into consideration for further developing discussions.

i) The suggested citations were added:

P37 L28-31: "It has been found that tropical monsoon activities were closely related to orbital-scale CH_4 change (e.g., Brook et al., 1996; Chappellaz et al., 1990), especially reported are Asian monsoon (e.g., Loulergue et al., 2008) and South American monsoon (e.g., Cruz et al., 2005)."

P44 L14-15: "Sperlich et al. (2015) also found that a sharp CH_4 peak at Greenland Interstadial 21.2 (~85 ka) was occurred by emission from Asian and South American wetlands."

ii) We removed the discussion on YD-Holocene transition and isotopic mass balance because it seems out of main focus of this paper.

iii) We appreciated the suggestion, but unfortunately, the mentioned publications (Liu et al., 2014; Clement et al., 1999) are dealing with climate modelling result, not proxy-based reconstructions. Instead, we cited Kirby et al. (2010) for PDO, and Moy et al. (2002) and Rodbell et al. (1999) for Holocene ENSO reconstructions. According to Holocene ENSO activity reconstructions by Moy et al. (2002), no ENSO event was recorded during the early Holocene until around 7 ka, except weak ENSO events during 10.4-10.1 ka, where abrupt CH_4 decrease is observed without significant changes in ITCZ and NH monsoon intensities. Mitchell et al. (2011) observed a significant positive correlation between CH_4 and PDO variability during the late Holocene. It has been reported that Pacific Decadal Oscillation (PDO) modulates the wet/dry impact of ENSO depending on phase relationship between ENSO and PDO (e.g., Wang et al., 2014 and references therein). The Holocene PDO reconstruction from sediment grain size analysis by Kirby et al. (2010) shows PDO-related drying intervals in North America during 9.5-9.1, 8.9-8.6, and 8.3-7.8 ka, which overlap the CH_4 minima at 8.2 and 9.3 ka present in this study.

6) Chosen data filtering technique: I suggest to provide more information why a 250-year window width was used. What is the effect of other window widths on the data you use and on your resulting interpretation? The authors state on p5L2-5 that the 250 year window was also used in other studies. However, that is not necessarily a satisfying justification. The window width should be carefully chosen in dependence on the time scales you are investigating.

Power spectrum (REDFIT, Schulze and Mudelsee, 2002) of Siple Dome CH_4 data indicates moderate (over 90% significance level) powers at ~1340, 401, 309 and 96-year period. Considering ~42 years of gas age distribution of Siple Dome (Ahn et al., 2014), it is not reliable to study centennial scale variability. Thus we applied 250-year window to smooth out any high frequency component having shorter period than 309 years. We added below paragraph:

"To extract millennial-scale variability, we carried out spectral analysis using REDFIT program (Schulze and Mudelsee, 2002) and moderate (over 90% significance level) powers were found at ~1340, 401, 309, and 96-year period. Given the ~42 years of gas age distribution of SDMA (Ahn et al., 2014), it would not reliable to study centennial scale variability. Thus we produced annual data by interpolation and then calculated 250-year running means to smooth high frequency components having shorter period than 309-year. Then the smoothed time series was filtered with a high-pass

window (cut off period of 1800 years) to study millennial scale variability throughout the early Holocene."

7) Sometimes, I have trouble to understand the point the authors intend to make, e.g. p1, 12–18; p5, 18–29; p6, 25–p7, 5; p10, 6–12, p22, 1-2. Please consider re-wording.

We thoroughly revised the mentioned sentences.

8) I have difficulties to follow the discussion and the display of CH_4 and other records. For example, the authors discuss why there is agreement/disagreement between some records within the uncertainty of the age model. However, I find it tough to see this in Figure 2. For example, the CH_4 minima are highlighted with yellow bars. During 9.4 and 10.2 ka, the yellow bars include both local minima and local maxima of the other records, e.g. of the Cariaco Basin record, of $\Delta\varepsilon$, of Dongge Cave. Other local extrema, e.g. in the Cariaco Basin record have no correspondence in CH_4 during 8.5-8.7 ka or 10.5 ka, which is not mentioned at all. Therefore, I feel this discussion needs to be further developed to provide more guidance to the reader. Is ^{10}Be really important here? Sometimes it seems to correlate, other times it is anti-correlated. Could figure clarity increase without it?

Not every single variation of the Cariaco Basin reflectance record corresponds to abrupt CH_4 change, because the Cariaco Basin record could also have local signal. What we wanted to show in this Figure is that abrupt cooling occurred around Greenland changes tropical rain belts and hence CH_4 emission. ^{10}Be and IRD proxies were included to discuss trigger of abrupt Greenland cooling, but data resolution and dating uncertainty (±100 to 150 years, Bond et al., 2001) prevent us from drawing rigorous conclusion. By taking into consideration with the comment #12 (Figure R4), we modifiedthe Figure 2 and relevant discussions.

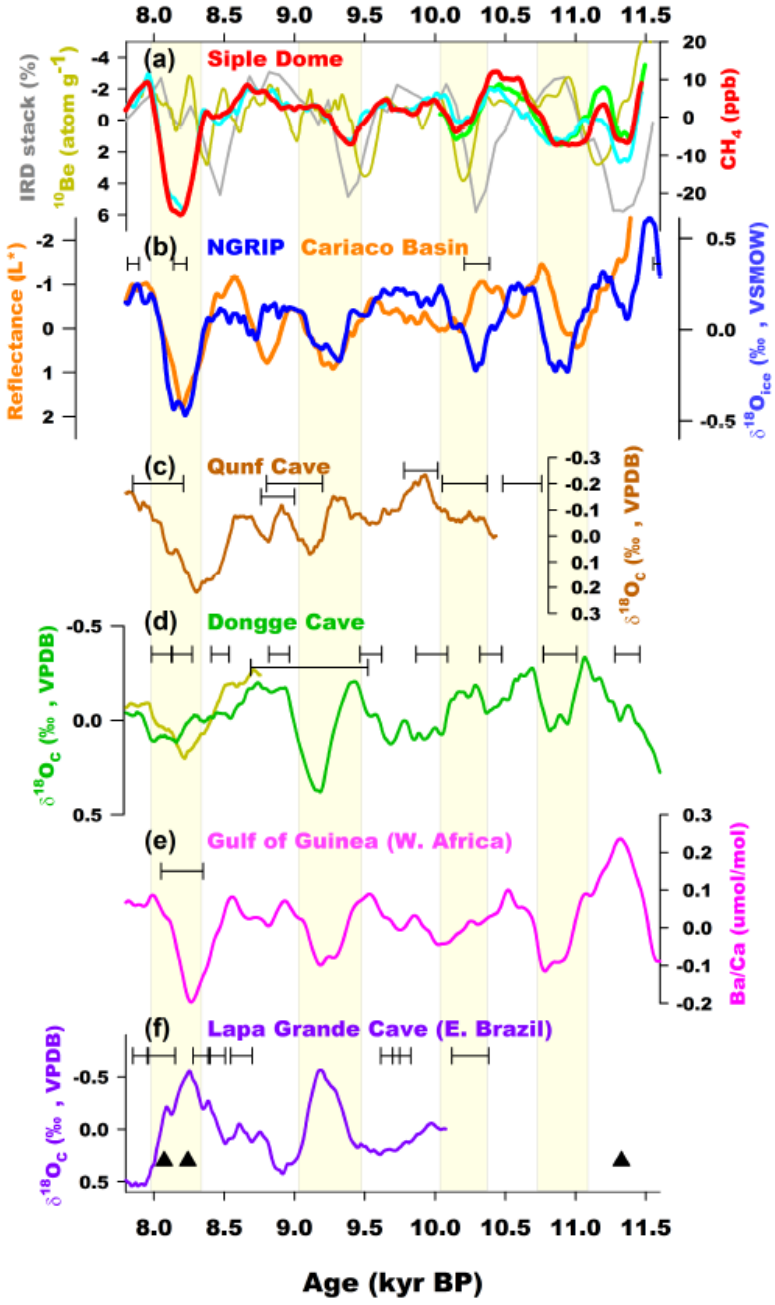

Figure R4. Millennial scale climate variability. All proxies present here were smoothed by 250-year running average and detrended by high-pass filter with 1/1800-year window. (a) Siple Dome CH_4 (red, this study), Greenland ^{10}Be (dark yellow, Finkel and Nishizumii, 1997), North Atlantic IRD stack (grey, Bond et al., 2001). Also shown are WAIS Divide CH_4 data by discrete (cyan, Buizert et al., 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 used to adjust Siple Dome and WAIS Divide CH_4 data to GICC05 scale are marked in black triangles.

9) Figure 2: Presented is CH_4 anomaly. I don't see an advantage of anomaly over CH_4 mole fractions. Also, how is anomaly of 0 defined for each record?

We refer "anomaly" in Figure 2 as detrended time series after filtered with high-pass window. We used "detrended data" instead of "anomaly" to clarify it.

10) Structure: I would suggest to avoid three levels, e.g. 3.1.1 and 3.1.2 but make 3. Millennial scale variability, 4. Latitudinal distribution…. to keep structure with max. two levels.

We simplified the structure in two levels.

11) In many places of the manuscript, the author review literature, e.g. on pattern of climate teleconnections, for which they allow extensive text sections. While I think it is important to review in such detail, I feel the authors could improve their discussion of how they think this is linked/relevant to their CH_4 interpretation. A good example for this is the entire section 3.1.3. I would like to encourage the authors to consider this throughout the entire manuscript, even though this probably means either adding to, or re-writing many sections of the manuscript.

We re-structured the 3.1.2. section.

12) Understanding the variability in tropical wetlands is crucial for the understanding of CH_4 source regions and tropical CH_4 fractions. (The same rule applies for boreal wetlands.) The authors fully acknowledge this throughout the manuscript. However, I note that the authors exclusively focus their interpretation on Asian/Indian monsoon systems. It has been described previously that the African monsoon system and wetland extension changed tremendously throughout the Holocene (e.g. Sahara region etc). There are also several publications on South American monsoon systems besides the Cariaco Basin reflectance, which I understand the authors only use as proxy for ITCZ migration, but not for their interpretation of hydrological changes in South American wetlands. Including further records (e.g. Cruz 2005) might allow for a more comprehensive evaluation of hydrological changes. Based on $\delta^{13}C$-CH_4 data, the South American monsoon system has recently been suggested to be a controlling factor in rapid CH_4 changes leading up to DO21 (Sperlich 2015). I would strongly recommend to either include a complete representation of tropical wetlands or to discuss why you think monsoon systems other than the Asian/Indian are not relevant.

It is possible that the monsoon system of other regions could play a role in CH_4 change. However, considering that mean position of ITCZ was moved to northward than glacial condition (e.g., Deplazes et al., 2013), the rainfall and CH_4 emission of Asian/Indian monsoon regions should be stronger than southern hemisphere monsoon regions, for example, South America and Africa. This makes boundary condition different from those arguing that Southern Hemisphere emission leads abrupt CH_4 increase during Heinrich Stadial 1, 2, 4, and 5 (Rhodes et al., 2015) or DO21 (Sperlich et al., 2015). Both studies are dealing with abrupt CH_4 change under glacial condition. In the meanwhile, one of main conclusion of our paper is that abrupt cooling in Greenland lead ITCZ mean position change and tropical CH_4 emission. Therefore, it seems sufficient to support the idea with the Asian/Indian monsoon records and Cariaco Basin reflectance data.

Lapa Grande Cave (Eastern Brazil, 14°25'22"S) records demonstrate clear $\delta^{18}O$ depletion at 8.2 and 9.2 kyr BP (Strikis et al., 2011), indicating that ITCZ rain belts temporarily migrated southward and induced wet condition over eastern Amazonia region. This evidence agrees well with Cariaco Basin rainfall reconstructions and does not go back beyond ~10.2 kyr BP, therefore we thought that adding the South American monsoon proxy would not improve our conclusions. The monsoon record of

northern Peru (El Condor and Cueva del diamante, Cheng et al., 2013) and southern Brazil (Botuvera, 27°13'24"S, Cruz et al., 2005) are not highly resolved enough to see abrupt changes. Otherwise, the reconstructions of West African monsoon (Gulf of Guinea, Ba/Ca ratio of planktonic foraminifera, Weldeab et al., 2007) and Indian monsoon entire early Holocene (Qunf Cave, Fleitmann et al., 2007) track well the millennial scale CH_4 minima. Furthermore, Australian-Indonesian monsoon rainfall records (not shown) from Borneo (Partin et al., 2007) and Liang Luar (Griffiths et al., 2009) do not show clear evidence of abrupt change that coincides with Greenland cooling and CH_4 decrease. This may reflect that the rainfall in tropical western Pacific region was affected by both northern- and southern hemispheric climate change (Griffiths et al., 2009; Partin et al., 2007).

13) Data availability: I understand that Copernicus has developed a new policy for authors to provide either descriptions on data access, or to provide the data through international data-bases or supplementary information. Copernicus requires a dedicated section that describes this in detail, which the authors might want to consider during their revisions.

Thanks for this information. If our manuscript is accepted to publish, we will make our new data available online in NOAA Paleoclimate Data Center and PANGAEA Data repository.

Specific comments

The manuscript may be subject to considerable re-writing. Therefore, the specific comments will not include comments on grammar, wording or writing that might be subject to change. Since I am not a native English language speaker myself, I am not sure to what extend my comments would help to make it better or worse. Please understand suggested re-formulations as suggestions, only.

p1L1: Understanding processes controlling atmospheric methane

The sentence was changed as below:

"Understanding ~~the atmospheric methane (CH_4) change~~ processes controlling atmospheric methane (CH_4) mixing ratio is crucial to predict and mitigate the future climate change.

p2L2: reference Daniau et al., is it possible to provide a reference on palaeo-fire or a reference that is more specific on pyrogenic gas emissions?

Citation was changed.

p2L5: sink strength and light availability? e.g. polar winter

We added below sentence in Introduction:

P36 L35-36: "Further, since the OH is produced by photo-dissociation reaction, the sink strength is affected by light availability and tropospheric ozone (e.g., Levy, 1971). Polar winters may affect the CH_4 sink strength by reducing OH production rate, but this seasonal-scale cycles are not resolvable in ice core records due to gas dispersion in firn layers".

p2L16: Lisiecki and Raymo 2005, though this reference is not on CH_4, general I think a reference on CH_4 and Northern Hemisphere temperature would be useful here

We changed the citation as below and added the reference:

(e.g., Brook et al., 2000; Chappellaz et al., 1993; Huber et al., 2006; Loulergue et al., 2008).

Huber, C., Leuenberger, M., Spahni, R., Fluckiger, J., Schwander, J., Stocker, T. F., Johnsen, S., Landais, A., and Jouzel, J.: Isotope calibrated Greenland temperature record over Marine Isotope Stage 3 and its relationship to CH_4, Earth Planet. Sci. Lett., 243, 504-519, 2006.

p2L18: See comment 5) in general comments

See Author Comment to general comment #5.

p2L18-19: too weak. The correlation between CH_4 and NH temperature (δ^{18}O-ice) is well established

It is true for longer time scales. The CH_4 – $\delta^{18}O_{ice}$ of Greenland has yet been revealed in shorter time scales during the early Holocene period.

p3L15: here and everywhere else: It is recommended to restrict the use of "concentrations" to public debate but to use "mole fraction" or "mixing ratio" in scientific literature (WMO, recommendations of GGMT experts)

This was updated as suggested.

p3L17: here and everywhere else, δ^{n}X, the δ is supposed to be *italicised*, same for Δ *Δ* (Coplen 2011, DOI: 10.1002/rcm.5129)

This was updated as suggested.

p3L27: there is still some ice left in Greenland

We modified the sentence as below:

"~~We note~~ It should be noted that environmental boundary conditions of the early Holocene were not identical to those of the late Holocene.~~, given that the global sea level rise continued throughout the early Holocene as the last sections of the northern hemisphere glacial ice sheets melted.~~ The global sea level was rising throughout the early Holocene and there is still some ice left in Greenland."

p3L30: ensure stated, used and displayed sample numbers agree, give age interval with depth

Please refer to our response to General Comment #3.

p3L32: (NICL, city, state, country), (SNU, city, state, country)

This was modified as suggested.

p4L3: "are described in…", referring to a paper that is currently in preparation as XYZ et al., (in prep.) seems strange as it is not useful to look it up. "A manuscript that describes the method in detail is currently in preparation."

We removed this sentence and added more details on our analytical system. Instead, we added below phrases regarding the detailed technical settings of GC and calculation of correction factors:

P41 L18-19: "The detailed configuration of the vacuum line and GC is described in another paper that is currently in preparation (Yang et al., in preparation)."

P42 L16-17: "Further details on correction method will be discussed in our manuscript in preparation (Yang et al., in preparation)."

p4L7: was the standard air added before or after the bubble-free ice was melted?

The standard air was injected before melting the bubble-free ice. We revised the wording for clarity.

p4L7: "traditional" melt-refreeze seems misleading to me, traditional can be left out

We removed the word "traditional".

p4L7-17: see point 2) in general comments

See Author Comment to general comment #2.

p4L17: provide reference how you calculated gravitational fractionation

Relevant citation and reference was added: Craig et al., 1988

Craig, H., Horibe, Y., and Sowers, T.: Gravitational separation of gases and isotopes in polar ice caps, Science, 242, 1675-1678, 1988.

p4L19: provide reference for GICC05 time scale

Reference was added: Rasmussen et al. (2006)

p4L25: "one of the high resolution data sets" sounds vague and strange to me, where do you draw the line between high and low resolution? The temporal resolution of your data is higher than some but lower than both WAIS records. "It has the currently second/third highest temporal resolution of Antarctic CH_4 records covering the early Holocene."

The sentence was modified accordingly as below:

"Our new Siple Dome CH_4 ~~record is the one of the high resolution data set~~ data has the currently third highest temporal resolution of Antarctic CH_4 records covering ~~from 11.6 to 8.5 ka, apart from the WAIS Divide records (Rhodes et al., 2015; WAIS Divide members, 2015)~~ the early Holocene after the WAIS Divide continuous (~2 years, Rhodes et al., 2015) and discrete (~20 years, WAIS Divide members, 2015) records."

p4L26: develop a more complete representation of analytical uncertainty

Please refer to our response to general comment #2, as well as our response to the Referee #2.

p4L30: describe the overlapping interval and describe why you think the OSU record should be adjusted to match the SNU data. Why not the other way around, why not accepted as real difference? Both should be on NOAA04?

We attribute the SNU-OSU offset to different correction method for solubility effect. After applying revised solubility correction used in OSU (Mitchell et al., 2011), the mean offset reduces 0.6 ppb which lies well within analytical uncertainty of both institutes.

p5L3: this comparison example only makes sense if you look at variations on similar time scales. Otherwise the argument that you use the same filter as has been applied for the WAIS record is not valuable, but could be misleading.

Here we hesitate to remove the comparison. As we describe in this sentence and Figure 2, to ensure robustness of millennial scale variability from other Antarctic ice core record is important. We used filtered data because both data set have different time resolution, and they show different fluctuations in short time scales as shown in Figure 1.

p5L14: add references that show anthropogenic signal in LPIH CH_4, e.g. Ferretti 2005, Mischler 2009, Sapart 2012)

We add the relevant citations:

"Mitchell et al. (2011) found no significant correlation with Greenland climate in multi-decadal time scale during the late pre-industrial Holocene (LPIH), possibly due to growing anthropogenic emissions (e.g., Ferretti et al., 2005; Mischler et al., 2009; Mitchell et al., 2013; Sapart et al., 2012)."

p5L16: "even though this conclusion is less robust as there are no age tie-points…"

The sentence was modified as suggested:

"In contrast, we observe a moderate positive correlation between the Siple Dome CH_4 and NGRIP $\delta^{18}O_{ice}$ during the early Holocene, which implies that natural CH_4 budget is closely connected with Greenland climate in millennial timescales, even though this conclusion is less robust as there is no age tie-point between the 8.2 ka episode and the Preboreal oscillation (Fig. S1)."

p5L21-22: shift this sentence to after the following sentence to keep logical flow from NH to tropics

Done.

p5L25-29: describe the meaning for CH_4, develop the discussion towards CH_4, what does a ITCZ shift mean for South American CH_4 source regions?

South American monsoon proxy shows concurrent intensification at similar timings of ~8.2 and 9.3 ka CH_4 drop (Strikis et al., 2011), where precipitations in Cariaco Basin and other NH monsoon regions decreased. For older period it is difficult to draw robust conclusion due to lack of high resolution data at the timing of preparing this paper. The southward ITCZ migration may lead reduction in NH wetland emission and enhanced in SH. However, given the orbital parameters that show maximum summer insolation in NH while minimum in SH during the early Holocene, it can be inferred that contribution of SH wetland emission was relatively low and cancelled by reduction of NH emission. We revised the paragraph as below:

"~~Moreover~~ The ^{18}O enrichments of Asian (Dongge) and Indian (Hoti and Qunf) cave stalagmites occurred at similar timing with abrupt cooling in Greenland, which indicate the reduction of monsoonal rainfall at northern tropical wetlands. The speleothem records from Chinese and Oman caves seem to lag by ~100-200 years after the CH_4 change at ~9.3 ka, but this lies within chronological uncertainties of ~200-400 years at around 9.0 ka (Dykoski et al., 2005; Fleitmann et al., 2007). Moreover, sediment Ba/Ca ratio from Gulf of Guinea demonstrates concurrent decrease of west African monsoon (Weldeab et al., 2007). These evidences indicate that precipitation over the major wetland area was reduced and in turn it would lower the wetland CH_4 emissions in NH. In the meanwhile, an inverse relationship is observed from the Eastern Brazilian speleothem data (Lapa Grande Cave, Strikis et al., 2011) that demonstrate the increasing of precipitation at the time of abrupt CH_4 drop occurred as a result of southward migration of ITCZ. Considering the orbital parameter that shows maximum summer insolation in NH while minimum in SH during the early Holocene, it can be inferred that contribution of SH wetland emission was relatively low and cancelled by reduction of NH emission."

p6L4: there are other monsoon systems that Asian/Indian that should be considered

See our response to general comment #12.

p6L10: the monsoon intensity change. (delete Asian, include other monsoon systems)

We modified the sentence as below:

"Given the weak reduction of precipitation over the Asian, Indian, and African monsoon regions (Figure R4), ~~This~~ it may imply CH_4 ~~reduction~~ drop was controlled by other processes than the ~~Asian~~ monsoon intensity change."

p6L20: even though the Cariaco Basin record is shown, it is presented only as indicator for ITCZ migration, without direct connection to CH_4. I feel the assumed passiveness of South American CH_4 source regions during the study period might not be a natural assumption and should be explained.

Considering the orbital configurations that show maximum summer insolation in NH while minimum in SH, contribution of SH wetland emission had to be reduced during the early Holocene.

p6L23-p7L5: describe relevance for CH_4, what is the CH_4 controlling process chain? A sentence that says "the proxies show this and that which could explain the increase/decrease in CH_4 during time period XY".

We added more details on proxies and its relevance to CH_4 change. Adding following sentence at the end of the paragraph will be more explained: "Above evidences indicate that the early Holocene CH_4 minima were triggered by anomalous low solar activity, but future study is warranted to draw more conclusive result."

The entire paragraph was revised as below:

"Below it is discussed the possible impact by external forcing. Bond et al. (1997) reported four large ice-rafted debris (IRD) drifts occurred at ~8.1, 9.4, 10.3 and 11.1 ka caused by surface cooling of North Atlantic Ocean. They found that the ocean surface cooling and the IRD events are closely related to cooling over the Greenland. Figure 2 shows that each IRD event (maxima in hematite stained grain) occurred concurrently with minima of NGRIP $\delta^{18}O_{ice}$ record within age uncertainty. Then the Greenland cooling leads southward shift of ITCZ and in turn it changes wetland CH_4 emission in low latitudes. Bond et al. (2001) found that IRD maxima during the Holocene coincide with solar activity minima. The authors suggested that solar forcing could affect the climate change around the North Atlantic Ocean (and Greenland), through amplification by changes in sea ice and/or deep water formation. A close interplay between solar activity and monsoon intensity has been observed in previous studies using the Chinese and Oman speleothem records during the Holocene (Neff et al., 2001; Wang et al., 2005; Gupta et al., 2005), even on multi-decadal time scales (Agnihotri et al., 2002). However, the forcing mechanism of solar activity on the North Atlantic and global climate is not well understood. Jiang et al. (2015) found positive correlations between North Atlantic SST and solar forcings inferred from plaeoproxies (^{14}C and ^{10}Be) for he last 4000 years, while the correlation disappears during the mid- and early Holocene. They hypothesized that climate sensitivity to solar forcing is high for cooler climate. As evidenced above, the early Holocene CH_4 minima were likely triggered by anomalous low solar activity, but future study is needed to make it more conclusive."

p7L8-9: you could add Cruz et al., 2005 to the reference list, as they discussed the interplay of solar radiation, monsoon intensity and CH_4 mole fractions

Here we hesitate to cite Cruz et al. (2005) here because the time scale they are dealing with is quite different.

p7L16-17: see above comment regarding references on ENSO variability during Holocene period

Please refer to our response to general comment #5.

p7L23: provide temporal resolution of NEEM record, 1 sample in how many years?

Done.

p7L24: consider IPD reconstructions with CH_4 records from WAIS

We added alternative IPD reconstructions from more reliable records; NEEM discrete, WAIS continuous, and Siple Dome discrete data. Please refer to our response to Referee #2.

p7L25-30: NICE approach!

p8L2: …show an increase by XYZ ppb from…

We removed that sentence.

p8L4: …in both hemispheres during…

We removed this sentence, but we changed the word "poles" to "hemispheres" in other phrases.

p8L6: …from both hemispheres and…

This sentence was modified as below:

"~~Given the new high resolution CH_4 records from both poles and IPD,~~ By using our new IPD and the reliable highly resolved CH_4 records (NEEM discrete – SDMA discrete / WAIS continuous), we ran a simple 3-box CH_4 source distribution model to quantify how much the boreal and tropical source strengths were changed."

p8L8-9: extra-tropical latitudes (30N or 30S is not high latitude, rather extra-tropical)

This was modified.

"Briefly, the model contains 3 boxes; northern ~~high~~ extra-tropical latitude (30-90°N, N-box), tropical (30°S-30°N, T-box), and southern ~~high~~ extra-tropical latitude boxes (30-90°S, S-box). CH_4 concentrations in 3 boxes (in Tg box^{-1}) were determined from CH_4 mixing ratio of Antarctica and Greenland."

p8L13-14: develop description of model assumptions and impacts, e.g. what life times did you assume and why? did you tune life times to match previous flux estimates?

We used the identical parameters described in Chappellaz et al. (1997) and Brook et al. (2000). Previous estimates are averaged values throughout the early Holocene, so that it is difficult to compare and match our higher resolution IPD to the previous results.

p8L15-16: quantify and discuss flux estimates, otherwise meaningless

Done.

p8L18: in tropical emissions by XYZ Tg. (quantify)

Done.

p8L25-30: increased CH_4 emissions from boreal wetlands were previously suggested by Sowers 2010, that agreement should be acknowledged

Please refer to our response to General Comment #5.

p8L29: explain "conventional" northern CH_4 emissions

We deleted this sentence.

p9L1: the isotope records are already published and need to be acknowledged (Sowers, 2010). these isotope data are available and should be added to the figures of this manuscript.

We thoroughly revised the paragraph where we cited the Sowers 2010 publication.

p9L10-15: therefore, IPD should be calculated with WAIS records as well

See our response above and Figure R2 and R3.

p9L30: there is also no explanation for the drop in IPD if I am not mistaken?

The slight decrease in IPD between ~10.3 and 10.8 ka is not observed in the alternative IPDs. Although it is argued above that Siple Dome CH_4 is more reliable data than others, we cannot rule out the possibility that the Siple data deteriorate during this period. At present it is difficult to say the drop in our IPD represents the real change or not.

p10L5-6: why can the 10.2 ka event not be explained by low latitude hydrology, but the other events can? I feel this is a section where great care has to be taken to prevent from over interpretation. There is no quantitative estimate for low latitude emissions during other events, i am not convinced that the presented records allow for a partial explanation of the CH_4 minima and that there is only a missing bit. I would recommend to re-formulate. Even if the revised IPD reconstruction supports the current discussion, this might seem as two results are made to fit together. I feel this can be toned down and still be strong a conclusion.

The 10.2 ka event does not bring corresponding abrupt change in Cariaco Basin record, Dongge Cave speleothem data, and Siple Dome $\Delta\varepsilon_{LAND}$. Thus we speculated that CH_4 drop at 10.2 ka was caused by source reduction in boreal regions and/or Southern Hemisphere.

Assessing the latitudinal source change from IPD is not reliable, given each ice core gas record has had own characteristic smoothing process at firn, because it could lead erroneous result for abrupt changing intervals such as 10.2 ka event.

p10L6-12: I am not sure I understand this section

We reworded the sentences to better clarity.

p10L11: the quantification 20-40 ppb is mentioned for the first time here. The conclusions cannot include information that have not been presented earlier in the manuscript. I am not sure how you quantify ppb changes? Is that from the box model?

The amplitude of each millennial scale CH_4 drop is quantified by CH_4 anomaly curve shown in Figure 2. We added this into earlier part of manuscript.

p16L19: I didn't check all references, but Sowers 2010 is not correct. This is "Atmospheric methane isotope records covering the Holocene period, Quaternary Science Reviews 29, 213-221, 2010". The title/journal name in your references refers to his 2006 paper

Done.

p18L4: add reference to the list or check reference

Done.

p18F1: show overlapping period, show minor ticks on both axes

Please see our response to the General Comment #1.

p19F2: define 0 ppb in anomaly or show CH_4 mole fractions, check width of yellow bars, can be confusingly wide

The Figure was updated.

p20F3: add IPD with WAIS data

Please see our response above, and Figure R2 and R3.

p22T1: caption is confusing to me, also what is this table supposed to add? how can this agreement be explained, life time?

The table and caption was updated according to the revised IPD reconstructions.

Referee Comment #2----------

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

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 added a dedicated chapter based on discussion below:

"We compared amplitude of CH_4 variability between the early- and the late Holocene in multi centennial to millennial time scales. Figure 5 shows amplitude spectrum and root mean square (RMS) amplitude for the early Holocene and the late Holocene, respectively. The amplitude of the early Holocene CH_4 change is ~10 ppb and does not change greater except for PBO and the 8.2 ka event, while the late Holocene spectrum shows smaller amplitude than early Holocene for shorter-term change and larger for longer-term fluctuation. Late Pre-Industrial Holocene (LPIH) CH_4 amplitude is elevated to early Holocene level from ~0 C.E. (~2.0 ka), and increases up to higher from ~1450 C.E. (~0.5 ka).

The reason of low amplitude variability during 3.5 to 1.2 ka, or 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). Combined with elevated summer insolation in Northern Hemisphere (NH) and with climate warming in NH extratropics, the amplified variability of the early Holocene may suggest that CH_4 control by NH wetlands was likely stronger than the late Holocene period. Meanwhile, lower summer insolation during the late Holocene might induce diminished CH_4 amplitude. 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 during 3.5-1.2 ka (604.9 ppb) is similar to that during 9.0-7.6 ka (628.6 ppb), and that anthropogenic emission is greater in later Holocene than the early Holocene. Abrupt increase of CH_4 amplitude since ~800 C.E. (1.2 ka) 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 greater."

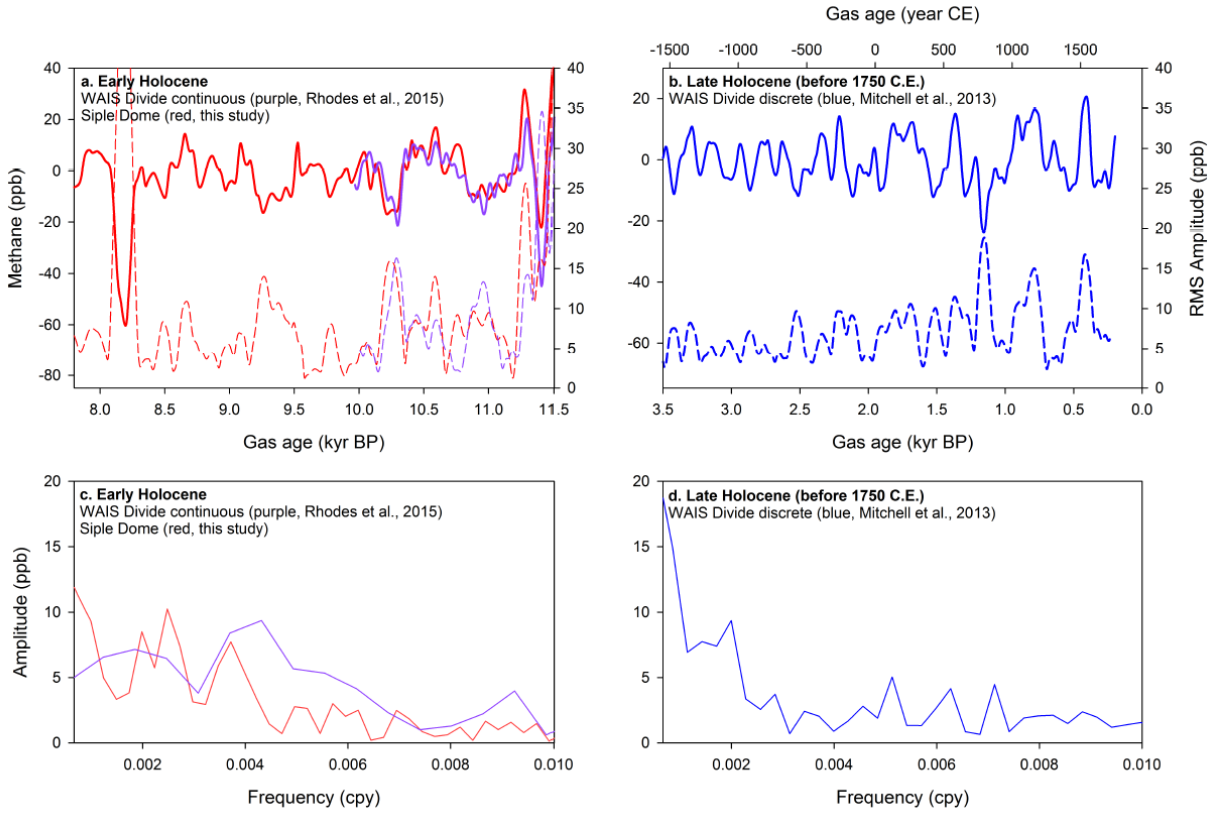

Figure R5. Upper: Detrended (75 to 1800-year band-pass filtered) CH_4 for the early (a) and late (b) Holocene from Siple Dome (red, this study), WAIS divide continuous (purple, Rhodes et al., 2015), and WAIS divide discrete (blue, Mitchell et al., 2013) data. Dashed lines are root mean square (RMS) amplitude 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. 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.

The Figure R4 (see above our response to Reviewer #1) 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.

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 of 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 1.4 ppb. This implies a good reproducibility and good precision of our analytical method.

Despite it is true that daily blank offsets are larger than the stated precision, the good agreement between the original- and replicate measurements does reveal that our blank correction method is reliable.

- 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 revised 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 above). 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 a

ll 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 modified our original argument that IPD increase from ~10.7 to 9.8 kyr BP. Instead, we focused 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 R1).

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.

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.

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)

Citation has been changed.

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

Citation added.

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

Following paragraphs have been added in the introduction section:

P37 L9-20: "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 precessional insolation change in the northern hemisphere. Modelling studies found the southward shift of ITCZ coincides with reduced CH_4 in Last Glacial Maximum (LGM) and Heinrich Stadial (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 appears to be related with millennial- or sub-millennial 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 ka 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

Please refer to our response 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 395.5, 721.3, 895.0, and 1384.9 ppb (in NOAA04 scale). This has been added in method section.

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

The paragraph was moved 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. This has been added.

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.

We add more details on it. Before the synchronization to GICC05, the correlation coefficient between the filtered SDMA CH_4 and NGRIP $\delta^{18}O_{ice}$ was calculated as $r = 0.57$ ($p = 0.06$). After adjusting to GICC05 scale by matching with NGRIP $\delta^{18}O_{ice}$ at 8.2 ka event and PBO interval (three tie-points), this correlation increases to $r = 0.74$ ($p < 0.01$).

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 added age uncertainty (1σ) of each proxy used.

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

The reference was 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.

Unlikely to NGRIP data, GRIP and GISP2 records show even smaller variability at 10.9 ka than that of 10.3 ka.

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.

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

Figure R2 (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 modified the paragraph as below:

P47 L33-38: "~~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 mole fraction 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 CH4 is calculated by subtracting 7% of IPD from the Greenland mixing ratio."

P8 l19: the boreal sources increased

We removed the paragraph.

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 removed the entire paragraph on LGM-Holocene transition, because it is out of main focus of this paper.

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

We removed the entire paragraph on LGM-Holocene transition, because it is out of main focus of this paper.

P20 l5 Chappellaz et al., 1997 not 2013

We removed the comparison.

References that are not cited in discussion paper----------

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Atmospheric methane control mechanisms during the early Holocene

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Abstract. Understanding processes controlling atmospheric methane (CH_4) mixing ratio ~~the atmospheric methane (CH_4) change~~ is crucial to predict and mitigate the future climate change. In spite of recent studies using various approaches for the last ~1000 to 2000 years, control mechanisms of CH_4 still remain unclear, partly because the late Holocene CH_4 budget is comprised of natural and anthropogenic emissions. In contrast, the early Holocene was a period when human influence should have been substantially smaller, so that it allows us to elucidate the natural controls under interglacial conditions. Here we present new high resolution CH_4 records from Siple Dome, Antarctica, covering from 11.6 to 7.7 thousands of years before 1950 AD (ka). We observe several local CH_4 minima on a roughly 1000-year spacing, which~~. Each CH_4 minimum~~ correspond~~s~~ to cool periods in Greenland. We hypothesize that the cooling in Greenland forced the Intertropical Convergence Zone (ITCZ) to migrate southward, reducing rainfall in northern tropical wetlands although there is no obvious change was observed in low latitude hydrology corresponding to abrupt CH_4 reduction at ~10.3 ka. Inter-polar difference (IPD) of CH_4 shows a gradual increase from the onset of the Holocene to ~9.9 ka, which implies growth of boreal source strength following the climate waring in the northern extratropics during that period. Finally, we find that amplitude of centennial- to millennial scale CH_4 variability of the early Holocene is larger on average than that of the earlier part of the late Holocene (3.5 – 1.2 ka).

~~A high resolution inter-polar difference (IPD) during the early Holocene increased from ~10.7 to 9.9 ka, and remained high until ~9.3 ka. With a simple three-box model results, our new IPD records suggest that the ratio of northern high latitude to tropical sources increased due to a boreal source expansion following the deglaciation.~~

1 Introduction

Methane (CH_4) is a potent greenhouse gas whose atmospheric mixing ratio has ~~been~~ increased more than 2.5 times since the Industrial Revolution (Dlugokencky et al., 2009). Although lower in abundance compared to carbon dioxide (CO_2), CH_4 has ~28 times higher global warming potential (GWP) on centennial timescales, and even higher GWP on shorter time scales due to the shorter lifetime in the atmosphere (Stocker et al., 2013). Hence the knowledge of control mechanisms of CH_4 is important to predict ~~foresee~~ and mitigate the future climatic and environmental changes.

Naturally, CH_4 is mainly produced by microbial decomposition by methanogens in anaerobic environments, such as waterlogged soil, wetlands, or sediments of lakes and rivers. Even though CH_4 can be oxidized and emitted as CO_2, a considerable amount of CH_4 is still released into the atmosphere through vascular plants, diffusion and ebullition processes (e.g., Joabsson and Christensen, 2001). Geological CH_4 released from mud volcanoes and

gas seepages through faults is the second most important natural source (e.g., Etiope et al., 2008 and references therein). Additionally, a portion of CH_4 is produced by termites and wild animals via microbial digestive process (e.g., Sanderson, 1996), and by pyrogenic sources such as wildfire and biomass burning (e.g., ~~Daniau et al., 2012;~~ Andreae and Merlet, 2001; Ferretti et al., 2005; Hao and Ward, 1993). The oceanic CH_4 flux is considered as too small to create a significant change in global budget compared to the other sources (Rhee et al., 2009). The major sink of atmospheric CH_4 is photochemical reactions (oxidation) with the hydroxyl radical (OH), which is mainly controlled by atmospheric temperature, humidity, and ~~concentration~~ mixing ratio of non-methane volatile organic compound (NMVOC) (e.g., Levine et al., 2011 and references therein). The air temperature affects air humidity, limiting the production of OH. Both NMVOCs and CH_4 are competing for OH to be oxidized, that is, increase in NMVOC emission reduces the available OH, so it increases the lifetime of CH_4 in the atmosphere (Valdes et al., 2005). Further, since the OH is produced by photo-dissociation reaction, the CH_4 sink strength is affected by light availability and tropospheric ozone (e.g., Levy, 1971). Polar winters may affect the CH_4 sink strength by reduced OH production rate, but the seasonal-scale cycles are not resolvable in ice core records due to gas dispersion in firn layers. However, recent model studies suggested the dominant role of source changes rather than sink in controlling atmospheric CH_4 during the past climate changes (Weber et al., 2010; Levine et al., 2011). Therefore, the sink changes are not considered here.

Since the direct CH_4 monitoring of modern air samples only covers the late 20th and early 21st centuries (Dlugokencky et al., 1994, 2011), investigation further back in time requires the unique archive of polar ice that preserves the ancient atmospheric air. Paleoatmospheric CH_4 levels have been reconstructed for the last 800 ~~thousand of years (kyr)~~ ka from Antarctic- and Greenland ice cores ~~(e.g., Spahni et al., 2005; Loulergue et al., 2008).~~. Given the relatively long lifetime in troposphere (11.2 ± 1.3 years at present, e.g., Prather et al., 2012) compared to atmospheric mixing time, ice core CH_4 records represent well-mixed global signatures. From the 800 ka records, it was revealed that ~~A time series of the past CH_4 changes found that~~ the past CH_4 change generally followed the glacial-interglacial cycles, being ~~was~~ low during glacial periods and high in interglacials, as well as the shorter orbital cycles of obliquity and precession ~~, generally following the glacial-interglacial cycles and related global ice volume changes (Lisiecki and Laymo, 2005;~~ (e.g., Spahni et al., 2005; Loulergue et al., 2008). Those earlier studies have suggested that the changes in climate and hydrology on tropical wetlands induced by the orbital changes controlled the CH_4 emissions. The resemblance between water stable isotopes from Greenland ice cores, a proxy for Greenland climate change, and global CH_4 mixing ratio has been largely reported. This implies that local temperature change around Greenland could affect the major CH_4 sources in low latitudes (e.g., Brook et al., 1996; Chappellaz et al., 1993; Huber et al., 2006). The similarity is also held in short time scale climate events. Previous works reported that rapid CH_4 increases were coincident with abrupt climate changes in Dansgaard-Oeschger (DO) events during the glacial period (e.g., Brook et al., 1996; EPICA Community Members, 2006; Grachev et al., 2007, 2009). The coincidence between abrupt CH_4 and Greenland climate change was also found in 8.2k cooling event, Preboreal Oscillation (PBO), Younger Drays (YD), and Bølling-Allerød (BA) periods (Brook et al., 2000; Kobashi et al., 2007, 2008).

Intensive precipitation changes on the low latitude summer monsoon area by insolation changes (e.g., Asian monsoon) have been suggested as an important control during the glacial period (e.g., Chappellaz et al., 1990). From time series analysis of past CH_4 records, Guo et al. (2012) found that the tropical monsoon circulations are primary control of relatively shorter (millennial) time scale variability, while long-term (multi-millennial to orbital

scale) variations are dominated by solar insolation changes. ~~For shorter time scale variability, the tropical monsoon activity and solar insolation changes were proposed as primary controls (Guo et al., 2012).~~ It has been found that tropical monsoon activities were closely related to orbital-scale CH_4 change (e.g., Brook et al., 1996; Chappellaz et al., 1990), especially reported are Asian monsoon (e.g., Loulergue et al., 2008) and South American monsoon (e.g., Cruz et al., 2005). ~~in shorter time scales,~~ However, no direct correlation between CH_4 and tropical monsoon signals has been reported for the early Holocene, although demonstrated were the positive relationships between Greenland climate and tropical monsoons (e.g., Chiang et al., 2008), and between Greenland climate and CH_4 (e.g., Spahni et al., 2005; Wang et al., 2005; Mitchell et al., 2011) have been discussed. Given that waterlogged wetlands are the largest natural CH_4 source, this complex relationship may imply that tropical monsoons are not the sole, primary controls; wetlands in northern high latitude and southern hemisphere might act as a secondary role.

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 precessional insolation change in the northern hemisphere. Modelling studies found the southward shift of ITCZ coincides with reduced CH_4 in Last Glacial Maximum (LGM) and Heinrich Stadial (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 appears to be related with millennial- or sub-millennial 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 ka record that found that ITCZ change becomes an important driver of millennial scale CH_4 change (Tzedakis et al., 2009; Guo et al., 2012).

The ice core scientists started to apply high-resolution CH_4 mixing ratio and stable isotope data to discern governing mechanisms of Holocene CH_4 variation, but currently the high-resolution records cover a part of the Holocene. High-resolution CH_4 records from Law Dome and WAIS Divide ice cores (Antarctica) show characteristic variability in multi-decadal to centennial time scale, apart from long-term gradual increasing trend (MacFarling-Meure et al., 2006; Mitchell et al., 2011). The high-resolution records were compared with various temperature- and precipitation proxies, but the previous works found no strong correlations that explain the observed decadal- to centennial scale variabilities. Limitation was that the late Holocene CH_4 budget may have been comprised of both natural and anthropogenic terms, making it difficult to distinguish between them. Mitchell et al. (2011) pointed out that some of abrupt CH_4 decreases could have resulted by historical events, such as Mongolian invasion, Plagues, or Spanish invasion. Later, Mitchell et al. (2013) made simultaneous measurement of Antarctic (WAIS Divide) and Greenland (GISP2) ices to derive IPD, and extended their high-resolution records back to ~4 ka. They used eight-box atmospheric methane model (EBAMM) and anthropogenic- and natural emission scenarios to investigate CH_4 control factors. Their results showed that the late Holocene CH_4 evolution can be explained by a combination of natural- and anthropogenic emissions. In the other hand, stable isotope ratios of CH_4 help us to distinguish the types of sources – biogenic, pyrogenic, and geologic. Sowers (2010) reconstructed CH_4 mixing ratio and stable isotope ratio of CH_4 ($\delta^{13}C$-CH_4 and δD-CH_4) throughout the entire Holocene and suggested several possible control factors, such as boreal wetlands and thermokarst lakes, changing

C_3/C_4 plant ratio of CH_4-emitting ecosystems, and changing composition of methanogenic communities. However, temporal resolution of the data (~138 years in average during 7.0 – 11.3 ka) was not sufficient to capture sub-millennial scale variability. Former studies have shown the reduction of pyrogenic emission and increased agricultural emission during the last millennia (Ferretti et al., 2005; Mischler et al., 2009). In later work using δ^{13}C-CH_4 records from NEEM ice core, Sapart et al. (2012) found that the centennial-scale variations during the last two millennia were caused by changes in pyrogenic- and biogenic emissions. Ruddiman et al. (2011) and Sapart et al. (2012) estimated CH_4 emission change due to anthropogenic land use changes, which shows a good agreement with long-term CH_4 increasing trend. However, there is no high-resolution reconstruction of past population and land use area, and consequently large uncertainties of CH_4 emission from land use change impede identification of any shorter scale changes.

The early Holocene is suitable period to study natural CH_4 controls under Holocene interglacial climate condition. Since there was only negligible human population and relevant CH_4-emitting anthropogenic activities (e.g., Goldewijk et al., 2010; Kaplan et al., 2011) during this time, the early Holocene CH_4 changes must have occurred mostly due to the natural causes. Understanding natural controls could contribute to better constrain the human-induced CH_4 changes. However, high-resolution study for the early Holocene has not been carried out extensively so far, except for the prominent cooling event at 8200 years BP (Spahni et al., 2003; Kobashi et al., 2007; Ahn et al., 2014). Earlier studies mainly focused on long-term change, attributing the major control to low latitude hydrology based on regional climate records that show wetter climate in tropics during the early Holocene (Blunier et al., 1995; Brook et al., 2000; Chappellaz et al., 1993, 1997). Therefore, in this study we present a new high-resolution CH_4 record during the early Holocene to investigate natural control mechanisms under interglacial condition. It should be noted that environmental boundary conditions of the early Holocene were not identical to those of the late Holocene. The global sea level was rising throughout the early Holocene and there was still remnant ice sheets in the north America.

~~Detailed studies of CH_4 variability during the early Holocene period are limited, except for the prominent 8.2 ka event (Blunier et al., 1995; Brook et al., 1996; Spahni et al., 2003; Kobashi et al., 2007; Ahn et al., 2012), which is thought to be caused by abrupt fresh water input from Lakes Agassiz and Ojibway into the North Atlantic, and consequently changes in North Atlantic Deep Water (NADW) formation and meridional heat transfer (e.g., Alley and Agustsdottir, 2005). Otherwise, earlier studies mainly focused on the multi-millennial variability, attributing the major control for the Holocene CH_4 to low latitude hydrology based on regional climate records that showed wet condition in tropical regions during the early Holocene (e.g., Blunier et al., 1995; Chappellaz et al., 1993, 1997; Brook et al., 2000). Climate simulation studies using the atmospheric chemistry and vegetation coupled models also confirm the previously suggested 'low latitude control' hypothesis (Harder et al., 2007; Singarayer et al., 2011).~~

~~Humans may also be an another important factor in the Holocene CH_4 budget. Ruddiman et al. (2008) proposed that an increase of agricultural activity (i.e. rice cultivation) was a major driver of the anomalous CH_4 rise after ~5 ka, based on radiocarbon dating of evidence for rice agriculture. This hypothesis was argued against later by Singarayer et al. (2011) who suggested that the insolation induced monsoon intensification in southern hemisphere could explain the late Holocene CH_4 increase. On the other hand, Sowers (2010) attempted to disentangle the Holocene CH_4 change by taking advantage of stable isotope ratios of CH_4 ($^{13}C/^{12}C$ and D/H) recovered from polar ice cores and suggested some possible control factors: northern emission from thermokarst~~

~~lakes and wetlands, changing in C_3/C_4 plant ratio of the CH_4-emitting ecosystem, and composition of methanogenic communities. The temporal resolution of the data was not sufficient to understand the underlying mechanisms of sub-millennial scale CH_4 variability.~~

~~Recently, emergence of high-resolution measurements permits us to study the CH_4 variability on multi-decadal to sub-millennial time scales, especially for the last 2000 years (Ferretti et al., 2005; MacFarling-Meure et al., 2006; Mitchell et al., 2011, 2013; Sapart et al., 2012; Rhodes et al., 2013). Mitchell et al. (2011) reported a new, decadally resolved CH_4 records for the last millennia from WAIS Divide, Antarctica. They tested the previous hypothesis by comparing proxies of temperature and precipitation of various regions, and found no strong correlation with CH_4. Their approach was optimal for validating the hypothesis, but the limitation was that the late Holocene CH_4 budget may have been comprised of both natural and anthropogenic terms, making it difficult to distinguish between them. Considering together with Antarctic- and Greenlandic CH_4 concentrations, deriving the inter-polar difference (IPD) of CH_4, Mitchell et al. (2013) demonstrated that the late Holocene CH_4 evolution can be explained by a combined emission of natural and anthropogenic sources. Sapart et al. (2012) analysed CH_4 concentration simultaneously with $^{13}C/^{12}C$ isotope ratio of CH_4 ($\delta^{13}C$-CH_4) using the ice core samples of North Greenland Eemian Ice Drilling (NEEM) and calculated isotopic mass balance to separate biogenic, pyrogenic and geologic emissions. Furthermore, Ruddiman et al. (2011) and Sapart et al. (2012) estimated CH_4 emission change due to anthropogenic land use changes, which shows a good agreement with long term CH_4 increasing trend. However, there is no high-resolution reconstruction of past population and land use area, and consequently large uncertainties of CH_4 emission from land use change impede identification of any shorter scale changes.~~

~~In this paper, we present a new high-resolution CH_4 record during the early Holocene to study the natural control mechanisms under interglacial conditions. Since there was only negligible human population and relevant CH_4 emitting anthropogenic activities (e.g., Goldewijk et al., 2010; Kaplan et al., 2011) during this time, the early Holocene CH_4 changes must have occurred mostly due to the natural causes. We note that environmental conditions were not identical to those of the late Holocene, given that the global sea level rise continued throughout the early Holocene as the last sections of the northern hemisphere glacial ice sheets melted.~~

2 Materials and Methods

In this study we used ice samples from a Siple Dome deep ice core (SDMA) drilled from 1997 to 1999 on the Siple Coast, West Antarctica (81.65°S, 148.81°W; 621m elevation) (Taylor et al., 2004). The SDMA samples were collected and cut at National Ice Core Laboratory (NICL, Denver, Colorado, USA) from January to February of 2013. Since brittle zone of SDMA ice starts below 400 m depth (Gow and Meese, 2007) that makes some part of ices fractured and/or cracked internally. Hence the samples were carefully collected from unbroken ices during the sample preparation at NICL. The samples were packed in isothermal foam boxes with numerous eutectic gels, and shipped to South Korea via expedited air freight. Temperature loggers were enclosed within the isothermal boxes to record the temperature change inside during the shipping, and it showed the temperatures were maintained below -25℃. The boxes were picked up directly just after custom clearance at the airport and then the ice samples were stored in a walk-in freezer at Seoul National University (SNU, Seoul, South Korea) that was maintained below -20℃. We measured 295 individual ice samples from 156 depth intervals from 518.87 to 718.83 m, covering from 8.36 to 20.25 ka after synchronizing to the Greenland Ice Core Chronology 2005 (GICC05,

Rasmussen et al., 2006). All samples were duplicated, so that our final CH_4 data were presented by averaging the results of duplicate analysis from the same depth and the analytical uncertainty of each data point is estimated by standard error of the mean of duplicate pairs. ~~We measured a total of 295 samples on 143 depth intervals from 518.87 to 623.38 m_i of Siple Dome ice core, West Antarctica. Siple Dome ice samples were cut and packed in insulated boxes and with eutectic gel packs at National Ice Core Laboratory (NICL), and shipped to Seoul National University (SNU) via expedited air freight. Automated temperature loggers were enclosed in the boxes to check the temperature during the shipping. The temperatures within the boxes were kept below -20 ℃ and the ice samples were preserved in SNU cold storage maintained below -20 ℃.The basic principles of gas extraction and CH_4 analysis are described in Yang et al. (in preparation). Briefly, the ice samples were cut by a clean band saw and trimmed outermost 2 mm to remove possible dissolution of modern ambient atmosphere. In case of cracks, the sample was trimmed along the fractures. The typical sample size is ~2.5 × 2.5 × 10 cm and the weight varies from 35 to 55 g depending on sample availability.~~The air occluded in ice was extracted by a melting and refreeze process under vacuum. Ice samples were prepared in a walk-in freezer in the morning of each experiment day, and trimmed the outermost >2 mm to eliminate potential contamination by ambient air during the storage. Then the samples were moved to the laboratory and placed in glass sample containers. The sample containers (sample flask hereafter) were custom-made glass flasks welded to stainless steel flange, and attached to the vacuum line with a copper gasket. ~~Each day we normally analysed 3 samples in duplicate and four blank samples (bubble-free ice made in the laboratory, with standard air added to the sample flask prior to air extraction).~~ The sample flasks were partially submerged into a chilled ethanol bath during ice insert and attaching to the line for preventing temperature increase by laboratory air. After that all flasks were evacuated at least 40 minutes, the ice samples were melted by submerging the sample flasks in a warm water bath. Melting process was usually completed within 30 minutes. The sample flasks were then submerged in the cold ethanol bath chilled to around -82℃ for more than an hour to refreeze. During the refreezing, we carried out GC pre-running (20 injections) and daily calibration that normally took 90 minutes. The ethanol temperature rose up to -55℃ just after submerging the flasks, and it was chilled to below -65℃ before expansion of the air in the flasks. The extracted air in the head space was expanded into a gas chromatograph (GC) equipped with a flame ionization detector (FID) to measure CH_4 mixing ratio. The GC linearity was tested by a series of inter-tank calibration using four working standard air cylinders (395.5, 721.3, 895.0, and 1384.9 ppb CH_4 in NOAA04 scale, Dlugokencky et al., 2005). Daily GC calibration curve was determined by measurements of a working standard having the closest CH_4 mixing ratio of expected value from the samples; in this study we used 721.3 ppb CH_4 standard for samples of the early Holocene. To account for system condition change throughout experiments (i.e., influence by water vapor), we calibrated with a standard air six times before and after sample measurements. The detailed configuration of the vacuum line and GC is described in another paper (Yang et al., in preparation). ~~Air was extracted by a traditional melt-refreeze technique, and the extracted air was expanded to gas chromatograph (GC), where CH_4 was separated and measured by a flame ionization detector (FID). The GC was calibrated daily with a standard air of 721.31 ppb CH_4 on the NOAA04 scale (Dlugokencky et al., 2005). The blank measurements of four bubble-free blank ice samples show a daily offset of 5–15 ppb, which is subtracted from all measurements. Preferential dissolution of CH_4 into meltwater (gas solubility effect) was corrected by Henry's law, assuming that the equilibrium state was accomplished within the sample container being a closed system.~~

To estimate our daily blank offset, we used four bubble-free blank ices every day, instead of interpolating between days of blank analysis (Mitchell et al., 2011). The blank ice was made by chilling the degassed ultrapure water (resistivity > 18.2 MΩ·cm at 25°C) slowly from bottom in a closed stainless steel chamber. The daily blank offset is calculated from the mean of the four blank results ranging from 5 to 15 ppb, and it reflects any daily offsets by contaminants, leaks, and any different GC conditions. The exact cause of this blank offset is currently not clear, but the four blank results agree well each other, yielding the intra-day standard error of the mean of 2.0 ± 1.0 ppb. This 'intra-day' blank offset is much smaller than the 'inter-day' offset, which implies that conditions of each sample flask are rather constant within a day, and vary systematically day by day. Since every single data point is obtained by analysis of at least in duplicate, the intra-day blank offset for one depth is reduced by a factor of √2. ~~This implies that offset among the flasks is not stochastic, and caused by daily systematic condition drift.~~ The robustness of our final results was proven by re-analysis of eight duplicates at adjacent depths (< 10 cm) 8 to 80 days after the first analysis. The difference of the mean of duplicates between the time intervals was 1.9 ppb on average (pooled standard deviation of 1.4 ppb). The good reproducibility of our results demonstrates that our blank correction method is reliable. ~~The exact mechanism that draws good reproducibility is currently unknown, but it seems that the replicate measurements do not follow exactly the normal (Gaussian) distribution. Similar results were obtained from the measurements of different ice cores (see Supplement).~~

~~M~~ass dependent (gravitational) fractionation within firn layer (Craig et al., 1988; Schwander, 1989) was corrected by ~~Our data were corrected for mass dependent (gravitational) fractionation by diffusion within the firn layer (Craig et al., 1988; Schwander, 1989). The gravitational fractionation effect was corrected~~ using the nitrogen isotope ratio ($\delta^{15}N$) of atmospheric ~~nitrogen (~~N_2~~)~~ occluded in bubbles. Siple Dome $\delta^{15}N$ records show a mean enrichment of 0.23 ± 0.01 ‰ during the early Holocene (Severinghaus et al., 2009) and result in a slight decrease of CH_4 by 1.97 ± 0.15 ppb, which we added to all our measurements to correct the gravitational fractionation.

Different solubility of each gas species cause preferential dissolution of a gas having higher solubility than others, and consequently it makes the mixing ratio of extracted air different from that trapped originally within the ice (solubility effect hear after). As solubility of CH_4 is higher than the other major component of air – nitrogen (N_2), oxygen (O_2), Argon (Ar), the solubility effect lowers the CH_4 mole fraction of the extracted air and needs to be corrected properly. At the early stage of method development, we derive theoretically the solubility effect by using Henry's Law in a closed- and chemically equilibrated condition. After applying this theoretical solubility correction, we observed that SDMA CH_4 data measured at SNU are lower than SDMA CH_4 records from OSU by ~3 ppb in average. Hence we compared the theoretical solubility correction with that obtained empirically from the second gas extraction (following the method described in Mitchell et al., 2011) and found that a correction factor of 1.0058 from the theoretically- to empirically-driven solubility effect. Further details on correction method will be discussed in our manuscript in preparation (Yang et al., in preparation).

Our new Siple Dome CH_4 data has the currently third highest temporal resolution of Antarctic CH_4 records ~~record is the one of the high resolution data set~~ covering the early Holocene after the WAIS Divide continuous (~2 years, Rhodes et al., 2015) and discrete (~20 years, WAIS Divide members, 2015) records. ~~from 11.6 to 8.5 ka, apart from the WAIS Divide records (Rhodes et al., 2015; WAIS Divide members, 2015). Duplicate measurements for 8 depths show ±1.0 ppb precision (1 sigma; pooled standard deviation).~~ To check reliability of the record we compared our data set with previous SDMA measurement ~~carried out~~ at Oregon State University

(OSU) for 8.4 to 9.1 ka period when the two records are overlapped. ~~using the same ice core.~~ The OSU CH_4 record was measured with a temporal resolution of 8 years with precision of ±2.8 ppb (Mitchell et al., 2011; Ahn et al., 2014). Resulting mean offset between two data set is ~0.6 ppb, which lies within analytical uncertainty range of both institutes. Therefore, we merge the two records to make the SDMA CH_4 composite data. Fig. 1 presents the SDMA data points during the early Holocene period (119 depths, 518.87 – 623.38 m).

3. Result and Discussion

3.1 Millennial scale variability

To extract millennial-scale variability, we carried out spectral analysis using REDFIT program (Schulze and Mudelsee, 2002) and moderate (over 90% significance level) powers were found at ~1340, 401, 309, and 96-year period. Given the ~42 years of gas age distribution of SDMA (Ahn et al., 2014), it would not reliable to study centennial scale variability. Thus we produced annual data by interpolation and then calculated 250-year running means to smooth high frequency components having shorter period than 309-year. Then the smoothed time series was filtered with a high-pass window (cut off period of 1800 years) ~~We applied a 250-year running average and high-pass filter (cut-off period of 1800 years) to Siple Dome CH_4 composite~~to study millennial scale variability throughout the early Holocene. For comparison, the same ~~signal~~ processing scheme was applied to WAIS Divide time series and we observed that Siple Dome and WAIS Divide CH_4 anomalies share similar millennial scale variability, confirming reliability of both our data and observed millennial scale changes (Fig. 2).

~~3.1.1 Low latitude hydrology~~

The CH_4 anomalies demonstrate millennial scale minima at ~8.2, 9.3, 10.2 and 11.0 ka, which occurred in nearly 1000-year spacing. Each minimum ~~was~~is accompanied by depletion of water stable isotope ratio ($\delta^{18}O_{ice}$) from NGRIP ice core, which implies climate cooling in Greenland. A close relationship between CH_4 and Greenland $\delta^{18}O_{ice}$ has been previously reported in glacial-interglacial cycles and Dansgaard-Oeschger (DO) events during the last glacial period (e.g., Brook et al., 1996, 2000; Blunier and Brook, 2001; Chappellaz et al., 1993, 2013; EPICA Community Members, 2006). However, it has not been confirmed at interglacial climate conditions during the Holocene. Mitchell et al. (2011) found no significant correlation with Greenland climate in multi-decadal scale during the late pre-industrial Holocene (LPIH), possibly because LPIH CH_4 budget is also affected substantially by anthropogenic emissions (e.g., Ferretti et al., 2005; Mischler et al., 2009; Mitchell et al., 2013; Sapart et al., 2012). In contrast, we observe a ~~moderate~~significant positive correlation ($r = 0.57$~~ 0.66~~, $p = 0.06$~~ 0.0013~~) between the millennial-scale change of Siple Dome CH_4 and NGRIP $\delta^{18}O_{ice}$ during the early Holocene~~, which~~. The correlation coefficient between the smoothed- and filtered time series of SDMA CH_4 (before synchronization to GICC05) and NGRIP $\delta^{18}O_{ice}$ was calculated for the 7.8-11.5 ka by interpolating to the original ages of SDMA CH_4 composite, with a reduced degree of freedom.

T~~he gas age scale, which was previously constrained (Brook et al., 2005),~~ The previous SDMA gas chronology (Brook et al., 2005; Severinghaus et al., 2009) was synchronized to ~~the Greenland Ice Core Chronology 2005 (GICC05)~~ GICC05 age scale by setting age tie-points with stable water isotope ($\delta^{18}O$) record from the North Greenland Ice Core Project (NGRIP) ice cores during the abrupt climate change events of the Preboreal

Oscillation (PBO) and the 8.2 ka event, given that both events have been proved to be synchronous with CH_4 change (Kobashi et al., 2007, 2008). The synchronization between the tie points was done by linear interpolation of age differences between the synchronized- and the previous ones, which range from -114 to 28 years. After synchronizing to GICC05 scale, the correlation coefficient between SDMA CH_4 composite and NGRIP $\delta^{18}O_{ice}$ increases to $r = 0.74$ ($p < 0.01$). ~~It implies that natural CH_4 budget is closely connected with Greenland climate on millennial timescales, even though this conclusion is less robust as there is no age tie points between the 8.2 ka episode and PBO~~ (Fig. S1).~~the Preboreal oscillation. (Fig. S1).~~

According to atmospheric modelling studies, abrupt cooling in the North Atlantic regions can alter atmospheric circulation and to cause southward migration of the mean latitudinal position of the Intertropical Convergence Zone (ITCZ) (e.g., Chiang and Bitz, 2005; Broccoli et al., 2006; Cvijanovic and Chiang, 2012). The climatic teleconnection between northern North Atlantic and low latitude regions is shown by climate proxies. ~~The southward displacement of ITCZ leads further weakening of Asian and Indian summer monsoons and probably reduces CH_4 emission from northern tropical wetlands.~~ Sediment reflectance record from Cariaco Basin shows increased rainfall and humidity – which is due to southward displacement of ITCZ – corresponding to the 8.2, 9.3, and 10.9 ka abrupt cooling event, ~~each abrupt cooling event~~,as revealed in previous studies for the different time periods (Peterson et al., 2000; Haug et al., 2001; Fleitmann et al., 2007; Deplazes et al., 2013). The southward displacement of ITCZ leads further weakening of Asian and Indian summer monsoons and probably reduces CH_4 emission from northern tropical wetlands. ~~Moreover,~~ The ^{18}O enrichments of speleothem in Dongge Cave (China), Qunf Cave (Oman), and Hoti Cave (Oman, not shown, Neff et al., 2001) ~~Asian (Dongge) and Indian (Hoti and Qunf) cave stalagmites~~ occurred at similar timing with abrupt cooling in Greenland at 8.2, 9.3, and 10.9 ka, which indicate the reduction of monsoonal rainfall at northern tropical wetlands. The speleothem records from Chinese and Oman caves seem to lag by ~100-200 years after the CH_4 change at ~9.3 ka, but this lies within chronological uncertainties of ~200-400 years at around ~9.0 ka (Dykoski et al., 2005; Fleitmann et al., 2007). Moreover, sediment Ba/Ca ratio from Gulf of Guinea demonstrates concurrent decrease of west African monsoon (Weldeab et al., 2007). The record indicates that precipitation over the major wetland area was reduced and in turn it would lower the wetland CH_4 emissions in NH. In the meanwhile, an inverse relationship is observed from the Eastern Brazilian speleothem data (Lapa Grande Cave, Strikis et al., 2011) that demonstrate the increase of precipitation at the time of abrupt CH_4 decrease occurred as a result of southward migration of ITCZ. Rhodes et al. (2015) pointed out that strong southward migration of ITCZ could induce an abrupt CH_4 increase from southern hemisphere during the HS 1, 2, 4, and 5 events. Sperlich et al. (2015) also found that a sharp CH_4 peak at Greenland Interstadial 21.2 (~85 ka) was occurred by emission from Asian and South American wetlands. However, considering the orbital parameters that indicate maximum summer insolation in NH while minimum in SH during the early Holocene, it can be inferred that contribution of SH wetland emission was relatively weak and overcompensated by reduction of NH emission.

The possibility that the observed CH_4 minima were caused by reduction of northern extra-tropical sources is not supported by previous modelling study. Zürcher 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 decrease (~80 ppb) during the 8.2 ka event. If we assume linear scaling of the 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 ka

event are more than 10 times weaker (Teller and Leverington, 2004) than that corresponding to the 8.2 ka event, we consider the boreal emission change is not the major cause of CH_4 local minima.

Previously, Björck et al. (2001) found the climate cooling in the northern Atlantic and Santa Barbara Basin occurred with solar-forcing change at ~10.3 ka. However, ~~in the proxy data, there is~~ the proxy data in Figure 2 show no clear indication of southward migration of ITCZ position and ~~reduction of~~ changes in Asian, Indian, African, and South American summer monsoon intensity ~~corresponding~~ associated to ~10.2 ka cooling (Fig. 2b-f). ~~(Fig. 2).~~ Moreover, there was no distinct change in $\Delta\varepsilon_{LAND}$ at that time, a proxy of global terrestrial respiratory fractionation of atmospheric O_2, which is affected by low latitude surface hydrology (Severinghaus et al., 2009). This paleoproxy record suggests that changes in precipitation and surface hydrology in the northern tropics may have not changed significantly during around the 10.2 ka. Instead, there are two small decreases at ~9.9 and ~10.6 ka as shown in Dongge cave deposit record (Fig. 2d). These episodes are not likely associated with the CH_4 minimum at 10.2 ka because the timing differences between the CH_4 minimum and reductions of Asian summer monsoon intensity are beyond the chronological uncertainty. The age uncertainty of Dongge Cave deposits is ±77 years (2 sigma error; Dykoski et al., 2005), and ~~that~~ the estimated error of SDMA CH_4 gas age in this study is less than ~150 years (see Supplement and Fig. S3). ~~the Siple Dome age uncertainty is likely less than ~100 years (see above and Fig. S1).~~ The climate teleconnection between North Atlantic and tropical hydrology at 10.2 ka might not have been ~~was not~~ sufficiently strong enough to change the low latitude climate. Weak cooling around the North Atlantic region can be a candidate, given that NGRIP $\delta^{18}O_{ice}$ records demonstrate smaller amplitude negative anomaly during ~10.2 ka event than those of 8.2 and 9.3 ka.~~, but this~~ The amplitude of $\delta^{18}O_{ice}$ changes at 10.2 ka of the high-pass filtered GRIP and GISP2 records does show smaller variability than those at 8.2 and 9.3 ka cooling events, but larger than the variability at 10.9 ka (Fig. S2). Hence, the "weak cooling" speculation is not fully supported by the other Greenland ice core records. ~~However, this is not supported by other Greenland ice core records such as Greenland Ice Core Project (GRIP) and Greenland Ice Sheet Project 2 (GISP2)., because the high-pass filtered GRIP and GISP2 $\delta^{18}O_{ice}$ records show even smaller variability at ~10.9 ka (Fig. S2[JA1]).~~

Although there appears to have been no strong change in low latitude hydrology at 10.2 ka, the amplitude of CH_4 decrease at 10.2 ka is similar order to the other millennial events. Given that no clear reduction ~~weak reduction~~ of the Asian, Indian, and African monsoon intensity is observed, ~~This~~ it may imply the CH_4 ~~reduction~~ decrease at 10.2 ka was controlled by other processes than the monsoon circulation change. ~~the Asian monsoon intensity change.~~ If the climate proxy from Dongge cave reflects rather regional climate changes, monsoonal rainfalls and surface hydrology of other regions could be responsible for CH_4 decrease. ~~The speleothem $\delta^{18}O$ records from Mawmluh Cave show no weakening of the Indian monsoon (Berkelhammer et al., 2012), moreover, there was no distinct change in $\Delta\varepsilon_{LAND}$, a proxy of global terrestrial respiratory fractionation of atmospheric oxygen, which is affected by low latitude surface hydrology (Severinghaus et al., 2009). This paleo proxy record suggests that changes in precipitation and surface hydrology in the northern tropics may have not changed significantly during around the 10.2 ka.~~

~~3.1.2 External forcing~~

There should be an ultimate cause of the CH_4 and climate change in the early Holocene. Previous works have suggested an important role of solar forcing during the Holocene (e.g., Björck et al., 2001; Bond et al., 1997,

2001). Bond et al. (1997) reported four large ice-rafted debris (IRD) drifts occurred at ~8.1, 9.4, 10.3 and 11.1 ka caused by surface cooling of North Atlantic Ocean. They found that the ocean surface cooling and the IRD events are closely related to cooling over the Greenland. Figure 2 shows that each IRD event (maxima in hematite stained grain) occurred concurrently with minima of NGRIP $\delta^{18}O_{ice}$ record within age uncertainty. Then the Greenland cooling leads southward shift of ITCZ and in turn it changes wetland CH_4 emission in low latitudes. Bond et al. (2001) found that IRD maxima during the Holocene coincide with solar activity minima. The authors suggested that solar forcing could affect the climate change around the North Atlantic Ocean (and Greenland), through amplification by changes in sea ice and/or deep water formation. A close interplay between solar activity and monsoon intensity ~~is confirmed by~~ has been observed in previous studies using the Chinese and Oman speleothem records during the Holocene (Neff et al., 2001; Wang et al., 2005; Gupta et al., 2005), even on multi-decadal time scales (Agnihotri et al., 2002). However, the forcing mechanism of solar activity on the North Atlantic and global climate is not well understood. Jiang et al. (2015) found positive correlations between North Atlantic SST and solar forcings inferred from plaeoproxies (^{14}C and ^{10}Be) for he last 4000 years, while the correlation disappears during the mid- and early Holocene. They hypothesized that climate sensitivity to solar forcing is high for cooler climate. As evidenced above, the early Holocene CH_4 minima were likely triggered by anomalous low solar activity, but future study is needed to make it more conclusive.

~~Figure 2(a) shows a possible cause of the observed millennial scale climatic changes and abrupt cooling recorded in Greenland ice cores. Four large ice rafted debris (IRD) drift deposits occurred during the early Holocene at ~8.5, 9.3, 10.3 and 11.3 ka (Bond et al., 2001). This record lacks a large IRD deposit that corresponds to 8.2 ka cooling (Bond et al., 2001). Later study found that increase of hematite stained glass (HSG) at the timing of 8.5 ka should be revised to 8.2 ka based on quartz to plagioclase ratio analysis (Moros et al., 2004). Additionally, Bond et al. (2001) found that 1500 year cycle of IRD in the North Atlantic are concurrent with the global climate cooling and the negative solar activity inferred by ice core ^{10}Be and $\Delta^{14}C$ records. From this evidence the authors speculated that the solar influence should be amplified by changes of sea ice and/or in deep water formation in the North Atlantic. However, the forcing mechanism of solar activity on the North Atlantic and global climate is not well understood during the early Holocene. Renssen et al. (2006) suggested that low solar activity (in terms of total solar irradiance) can induce sea ice expansion around the Nordic Seas and weakening of deep water formation and cooling in North Atlantic region. Nevertheless, the anti correlation between solar forcing and sea ice expansion (and hence deep water formation weakening) is not strong during the early Holocene due to relatively warm climate conditions. Jiang et al. (2015) also found a negative correlation between North Atlantic SST and solar forcing proxies (^{14}C and ^{10}Be), which is statistically significant for the last 4000 years, while the correlation disappeared during the mid- and early Holocene. They hypothesized that climate sensitivity to solar forcing is high for cooler climate.~~ Meanwhile, shifting to El Nino-like SST condition was suggested as another mechanism that changes tropical rainfall pattern (Marchitto et al., 2010). According to modern atmospheric observation, El Niño condition leads drying conditions in low latitude wetlands in Africa, Asia, and America (e.g., Dai and Wigley, 2000; Lyon and Barnston, 2005; Hodson et al., 2011), which reduces tropical CH_4 emissions. Thus, we could speculate that both the ITCZ migration and El Niño-like SST change affected the tropical surface hydrology and CH_4 emission. According to Holocene ENSO activity reconstructions by Moy et al. (2002), no ENSO event was recorded during the early Holocene until around 7 ka, except weak ENSO events during 10.4-10.1 ka, where abrupt CH_4 decrease is observed without significant changes in ITCZ

and NH monsoon intensities. Mitchell et al. (2011) observed a significant positive correlation between CH_4 and Pacific Decadal Oscillation (PDO) variability during the late Holocene. It has been reported that PDO modulates the wet/dry impact of ENSO depending on phase relationship between ENSO and PDO (e.g., Wang et al., 2014 and references therein). The Holocene PDO reconstruction from sediment grain size analysis by Kirby et al. (2010) shows PDO-related drying intervals in North America during 9.5-9.1, 8.9-8.6, and 8.3-7.8 ka, which overlap the CH_4 minima at 8.2 and 9.3 ka present in this study.

~~Marchitto et al. (2010) suggested that negative solar forcing induces so called "El Niño like" conditions; warming in East Tropical Pacific and weakened Asian and Indian summer monsoons. A close interplay between solar activity and monsoon intensity is confirmed by previous studies in Chinese and Oman speleothem records (Neff et al., 2001; Wang et al., 2005; Gupta et al., 2005), even on multi decadal time scales (Agnihotri et al., 2002). Marchitto et al. (2010) also suggested a connection between "El Niño-like" climate and IRD events (except for 8.2 ka event), through reorganization of ocean currents around the North Atlantic due to intensified El Niño Southern Oscillation (ENSO) driven by the solar forcing so that it may have driven more IRD episodes (Emile-Geay et al., 2007). According to modern climate conditions it has been found that the El Niño state generally induces wetter conditions in tropical land area and vice versa (e.g., Dai and Wigley, 2000; Lyon and Barnston, 2005; Hodson et al., 2011). However, since there is no ENSO- and PDO index reconstructions back to the early Holocene at present, with different climate boundary conditions, we cannot test this hypothesis.~~

3.2 ~~Latitudinal source distribution~~ Inter-polar difference of CH_4 during the early Holocene

~~3.2.1 Inter-polar difference of CH_4 and source distribution model~~

We calculated inter-polar difference (IPD) of CH_4 to trace the latitudinal source distribution change during the early Holocene. The currently available high-resolution CH_4 records covering the early Holocene are SDMA discrete (this study), WAIS Divide discrete (WAIS Divide project members, 2015), WAIS Divide continuous (Rhodes et al., 2015), NEEM discrete (Chappellaz et al., 2013) and NEEM continuous data (Chappellaz et al., 2013). Among the Antarctic records, we consider WAIS continuous records are more reliable than WAIS discrete ones because WAIS discrete data covering the early Holocene were measured in a different institute (Penn State University, PSU), showing a pooled standard deviation of ~7.3 ppb (1σ). In addition, there is an unexplained offset between WAIS CH_4 measured in OSU and PSU lab by ~9.9 ppb (Rhodes et al., 2015), which is larger than that observed between SNU and OSU SDMA data sets. Regarding the Greenland side, we use NEEM discrete records because there are discrepancies between continuous- and discrete data in some intervals, but also because the NEEM continuous record is not exactly "continuous". Hence, here we regard the NEEM discrete, Siple Dome discrete, and WAIS Divide continuous data as more reliable ones than the others to reconstruct IPD during the early Holocene. ~~In this study, the IPD was calculated by using our Siple Dome CH_4 record and a NEEM high resolution discrete CH_4 record (Chappellaz et al., 2013).~~

Precise synchronization is crucial for direct comparison between data sets which have high frequency variations. For synchronizing between Antarctic (Siple Dome and WAIS Divide continuous) and NEEM records, ~~T~~the NEEM CH_4 record (~11 years resolution on average) is chosen as reference. ~~because the mean time resolution is higher than our data set.~~ Synchronization was done by two steps: First, we made initial synchronization between the Antarctic and NEEM data by setting ~~7~~match points at the midpoint of abrupt CH_4 change, and then we linearly

interpolated the age offset of each match point for the rest of data points. Then we applied a Monte Carlo simulation to find a maximum correlation. Both data sets were resampled every 30 years, and each point was randomly disturbed (assuming a normal distribution with 1 sigma of 30 years). By doing so 1000 different time series were created, and one set having a maximum correlation with NEEM data was chosen. Criteria for "best fit" is correlation coefficient of 0.8 with NEEM original age scale, so that a maximum correlation less than 0.8 was discarded. This procedure was repeated to make 20 sets of maximum correlation time series, and the mean ages of 20 replicate simulations were set to synchronized age scale. Temporal uncertainty (synchronizing error) was determined for each point as 1 standard deviation of 20 replicates and CH_4 uncertainty includes analytical error of the both records (4.3 ppb for NEEM discrete ~~and~~ /1.4 ppb ~~1.0 ppb~~ for SDMA / 1.5 ppb for WAIS continuous, 1 sigma). Since IPD calculation is very sensitive to high frequency variability of CH_4 records from both poles, and it is difficult to reconstruct reliable IPD in short time scales, all IPD records in this study were filtered by a 1000-year low-pass window to discuss multi-centennial to millennial scale change. As discrete measurements are regarded as more accurate than continuous ones in absolute sense, WAIS continuous data were calibrated against to Siple Dome data instead of WAIS discrete record, because Siple Dome records were more rigorously tested for its reproducibility, and also compared with OSU measurements that shows small offset during the early Holocene interval.

The IPDs from those three data sets are plotted in grey (NEEM discrete – Siple Dome, IPD-1 hereafter) and green (NEEM discrete – WAIS continuous, IPD-2 hereafter) in Figure 3. Resulting IPD-1 and IPD-2 show long-term increase from 11.5 to 9.9 ka, which indicates that boreal source contribution enhanced. However, IPD-1 shows a sharp increase during PBO followed by decrease until ~10.7 ka, and in the latter case both IPDs differ beyond 95% envelope (from 10.4 to 10.8 ka). This is because Siple Dome discrete data are higher than WAIS continuous data during this interval. Similarly, the small peak of IPD-1 at ~11.1 ka that is not seen in IPD-2 may be caused by offset between Siple- and WAIS data during 11.0 to 11.3 ka. ~~Fig. 3 shows our IPD results with 95 % significance envelope. Our IPD agrees with the previous low resolution estimates for the earlier part of the Holocene (9.5~11.5 ka) (Chappellaz et al., 1997; Brook et al., 2000). Our results show an increase from ~10.7 ka to ~9.9 ka, which was not previously reported. Considering the long-term decreasing trend of CH_4 mixing ratio in both poles during the early Holocene, the increasing IPD implies that the amount of boreal emission reduction should have been less than that of low latitude emissions.~~

Given the new high resolution CH_4 ~~records from both poles and IPD,~~ By using our new IPDs and the reliable highly resolved CH_4 records (NEEM discrete – SDMA discrete / WAIS continuous), we ran a simple 3-box CH_4 source distribution model to quantify how much the boreal and tropical source strengths were changed. Here we used the same box model employed in Chappellaz et al. (1997) and Brook et al. (2000). Briefly, the model contains 3 boxes; northern ~~high~~ extra-tropical latitude (30-90°N, N-box), tropical (30°S-30°N, T-box), and southern ~~high~~ extra-tropical latitude boxes (30-90°S, S-box). CH_4 ~~concentrations~~ mixing ratios in 3 boxes (in Tg box^{-1}) were determined from CH_4 mixing ratio of Antarctica and Greenland. ~~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 mole fraction 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 mixing ratio. T-box ~~concentration~~ mixing ratio is inferred by assuming that the S-box emission is constant of 15 Tg yr^{-1} (Fung et al., 1991). Emission from each box (Tg yr^{-1}) is then estimated by using the ~~concentration~~mixing ratios of the boxes, lifetime of CH_4 in each box, and transport times among the boxes. The modelled emission changes in NH extratropical- and tropical boxes from IPD-1 are plotted in Figure 4 (see Fig. S3 for results from IPD-2). The model reveals decreasing tropical sources (accounting for the largest portion in CH_4 budget), while enhancing NH extratropical emissions. The tropical emission was elevated by ~8 Tg yr^{-1} from the onset of the Holocene to its maximum at 10.6 ka, followed by ~15 g yr^{-1} reduction to ~111 Tg yr^{-1} at 9.5 ka. Tropical emission decrease is also observed in IPD-2 from 134 to 115 Tg yr^{-1} during the 11.5-10.0 ka, but this change is not significant in 95% confidence range (Fig. S3 and Table S2). The long-term decreasing trend follows the NH summer insolation change. This covariation may reflect the insolation-driven changes in multi-millennial timescale (e.g., Loulergue et al., 2008; Guo et al., 2012). CH_4 flux from NH extratropical box increased from ~57 Tg yr^{-1} (11.5 ka) to ~70 Tg yr^{-1} (9.5 ka), showing a local minimum of ~63 Tg yr^{-1} at 10.7 ka. Also plotted in Figure 4 is boreal source fraction, defined as ratio of N-box emission to total source emissions. It shows a significant increase from ~30% at 11.5 ka to ~35% at 9.5 ka. The box model results from IPD-2 demonstrate increase of NH extratropical emission from 60 to 71 Tg yr^{-1}, and hence increase of the boreal source fraction from 29 to 35% during the 11.5 to 10.0 ka interval.

~~The results of our model are consistent with previous estimates by Chappellaz et al. (1997) and Brook et al. (2000). Although the early studies reported average value for the 11.5 9.5 ka interval, our IPD records show similar value before the IPD starts to rise at ~ 10.7 ka (Fig. 4 and Table 1). After that, our results show an increase of boreal emission by 9 Tg yr^{-1} and a decrease in tropical emission. Boreal source fraction, a ratio of boreal emission to total emission, reveals an increase by 5 %. This result supports our interpretation that the boreal sources were less reduced than those in low latitudes.~~ This conclusion is supported by proxy-based temperature reconstructions that indicate a gradual warming in ~~northern high latitude~~ northern extratropical region (30N-90N) until ~9.6 ka, while tropical temperature remains stable (Marcott et al., 2013). The climate warming in northern high latitudes caused ice sheet retreat (e.g., Dyke, 2004) and enhanced CH_4 emission from boreal permafrost by forming new wetlands in mid- to high latitudes (e.g., Gorham et al., 2007; Yu et al., 2013) and accelerating microbial decomposition of organic materials (e.g., Christensen et al., 2004; Schuur et al., 2015). Thermokarst lakes created by thawing ice wedges and ground ices in Alaskan- and Siberian permafrost ~~are~~ has been suggested as a source of CH_4 (e.g., Walter et al., 2006, 2007; Brosius et al., 2012). The modelled enhancement of NH extratropical emission of ~13 Tg yr^{-1} is greater than the CH_4 release of 8.2 Tg yr^{-1} from thermokarst lake thawing, which is estimated based on present-day observations (Walter et al., 2014). Since most thermokarst lakes are located in NH high latitude regions (e.g., Walter et al., 2006; 2014), it may indicate that other sources, such as northern peatlands or mid-latitude wetlands, should contribute to increasing NH extratropical emission. Our results are consistent with previous findings of CH_4 stable isotope analysis. Fischer et al. (2008) found that increase of boreal source contribution is required to explain the more depleted $\delta^{13}C$-CH_4 during Preboreal Holocene than the Younger Dryas interval. Sowers (2010) extended the CH_4 isotopic ratio into the entire Holocene that displayed a gradual decrease of $\delta^{13}C$-CH_4 by ~2 ‰ from 10.5 to 4 kyr BP, which was attributed to progressive expansion of NH high latitude sources.

~~Indeed, the increased boreal CH_4 emission of 9 Tg yr^{-1} is in similar order of the CH_4 release of 8.2 Tg yr^{-1} from thermokarst lake reported by Walter Anthony et al. (2014). However, it should be noted that the CH_4 release~~

estimates from the thermokarst lakes are based on present-day CH_4 flux measurements in Siberian- and Alaskan lakes and that 9 Tg yr^{-1} is a small change in the budget that could be driven by conventional northern CH_4 emission. A recent study also argued a possibility of underestimation of such CH_4 emission measurements (Wik et al., 2016). We could not estimate the IPD for the later part of the record (7.7 ~ 8.8 ka) due to a lack of high resolution CH_4 from Greenland ice cores. However, since the first-generation lakes produce CH_4 more actively than later-generation lakes formed after drainage (Brosius et al., 2012), it is unlikely that thermokarst lake CH_4 emission would remain higher after 9.0 ka. Future study should include extending high resolution CH_4 record from Greenland, as well as CH_4 isotope ratio data for the younger time period.

3.2.2 IPD during the Pre-Boreal Oscillation (PBO)

We also observed a high IPD at the earliest part of the Holocene, where CH_4 records from both hemispheres poles show a large variability. This could be due to mismatching of synchronized time scales and different surface conditions of drilling site and hence signal attenuation process within firn. A sensitivity test on synchronizing error has been carried out by shifting the reference age scale (in this study, NEEM chronology) 15 years back and forth given that the initial age match points were resampled every 30 years. The IPDs calculated with shifted age scales (plus 15 years, control, and minus 15 years) are plotted in Fig. S3, showing a consistent high IPD values during ~11.0 to 11.2 ka interval, while for the earlier part IPD seems to be highly sensitive to synchronization. Nevertheless, this might be a result of different gas enclosing processes within firn layers in both ice cores. As the width at half height of the gas age distribution at NEEM site was reported as ~32 years (Buizert et al., 2012), which is ~23 % narrower than that of Siple Dome (Ahn et al. 2014). It means that the NEEM signal has been less attenuated than Siple Dome one, which could result in higher (lower) IPD at the period where rapid CH_4 increase (decrease) is observed. Indeed, the discrete and continuous CH_4 record from WAIS Divide, which has a mean accumulation rate similar to NEEM (Buizert et al., 2013), shows ~10 to 20 ppb higher amplitude variability.

Our new CH_4 data confirms the abrupt doubling at the Younger Dryas termination. Previous studies using stable isotopes of CH_4 have shown contradictory results. Previous studies that, using the stable isotopic composition of C and H in CH_4 that aimed to disentangle the cause of abrupt CH_4 increase during the earliest period of the Holocene have shown contradictory results. Schaefer et al. (2006) calculated isotopic ($\delta^{13}C$ CH_4) mass balance model to discern major source term that caused a slight enrichment in ^{13}C during the Younger Dryas termination, suggesting tropical wetland emission as a dominant source. The authors also proposed biomass burning, geologic CH_4 and enhanced sink process at marine boundary layer as alternatives, but less probable scenarios. 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, Möller et al. (2013) pointed out the possibility of changing isotopic signature of each source 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 of biomass burning. The triple isotopic mass balance model using $\delta^{13}C$ CH_4, δD CH_4 and $\Delta^{14}C$ CH_4, Melton et al. (2012) suggested the biomass burning and thermokarst lakes are the most important additional sources. The enhanced biomass burning agrees with global charcoal influx, an independent proxy for wildfire, which shows intensified wildfire in northern tropical regions (Daniau et al., 2012). However, it is unlikely that the increased pyrogenic emission in tropics leads to higher IPD. Brosius et al. (2012), using an isotopic mass balance model including thermokarst lake sources, suggested another scenario that the enhanced boreal wetland emission contributed largely for the CH_4 overshoot. In the meanwhile,

~~the boreal emission hypothesis was refuted by a recent study of ^{14}C-CH_4 change during the YD termination that revealed the major carbon source for abrupt CH_4 doubling was not the permafrost-origin old carbon (e.g., Petrenko et al., 2009, 2015). Therefore, the cause of the high IPD at the start of the Holocene still remains elusive.).~~

3.3. Comparison with late Holocene variability

We compared amplitude of CH_4 variability between the early- and the late Holocene in multi centennial to millennial time scales. Figure 5 shows amplitude spectrum and root mean square (RMS) amplitude for the early Holocene and the late Holocene, respectively. The amplitude of the early Holocene CH_4 change is ~10 ppb and does not change greater except for PBO and the 8.2 ka event, while the late Holocene spectrum shows smaller amplitude than early Holocene for shorter-term change and larger for longer-term fluctuation. Late Pre-Industrial Holocene (LPIH) CH_4 amplitude is elevated to early Holocene level from ~0 C.E. (~2.0 ka), and increases up to higher from ~1450 C.E. (~0.5 ka).

The reason of low amplitude variability during 3.5 to 1.2 ka, or 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). Combined with elevated summer insolation in Northern Hemisphere (NH) and with climate warming in NH extratropics, the amplified variability of the early Holocene may suggest that CH_4 control by NH wetlands was likely stronger than the late Holocene period. Meanwhile, lower summer insolation during the late Holocene might induce diminished CH_4 amplitude. 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 during 3.5-1.2 ka (604.9 ppb) is similar to that during 9.0-7.6 ka (628.6 ppb), and that anthropogenic emission is greater in later Holocene than the early Holocene. Abrupt increase of CH_4 amplitude since ~800 C.E. (1.2 ka) 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 greater.

4. Conclusion and summary

~~In this study w~~We reconstructed a new high resolution CH_4 record during the early Holocene from Siple Dome ice core, Antarctica, to study millennial-scale CH_4 variability and its natural controls under Holocene interglacial condition. ~~the early Holocene CH_4 time series in high resolution to discuss natural processes that control the millennial scale CH_4 variations in the past atmosphere.~~ Since the new SDMA data agree well with previous measurements at OSU, we made SDMA CH_4 composite data covering ~7.7 to 11.6 ka. We found ~~Our results show a series of the~~ four millennial scale CH_4 minima accompanied with Greenland cooling, changes in ITCZ position and reduced Asian and Indian monsoon intensities.~~,~~ The observed evidences suggest that low latitude hydro climate changes were closely related to millennial scale CH_4 minima. ~~and the evidence suggests that the low latitude source changes were the major causes of the early Holocene CH_4 minima.~~ Further, this study presented the millennial scale change of IPD, which was calculated from high resolution discrete dataset of NEEM and SDMA. Here we reported for the first time the IPD increase from the onset of the Holocene to ~9.9 ka

following the temperature rise in NH extra-tropical regions. The three-box model demonstrates that elevated emission from NH extratropics and reduction of tropical sources, resulting the increased contribution of the NH extra-tropical sources. Finally, we observed that RMS amplitude of earlier part of the late Holocene is smaller than that of the early Holocene, which may be attributed to different orbital parameters.

~~However, the North Atlantic-induced changes in low latitude hydrology cannot fully explain the CH_4 minimum at ~10.2 ka. High resolution IPD and 3-box source distribution model results indicate that fraction of boreal sources increased by 5 % during the early Holocene, which indicates that fraction of boreal sources increased from ~10.7 ka and remained high until ~9.3 ka. To summarize, the millennial scale variability of CH_4 during the early Holocene was primarily controlled by low latitude climatic and surface hydrological conditions, while relative boreal source contribution increased during 10.7-9.3 ka by newly developed high latitude sources following terrestrial deglaciation. Further, our observations imply that ~20-40 ppb of CH_4 change could be induced naturally by low latitude hydroclimate changes.~~

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Data availability

The early Holocene Siple Dome CH_4 data will be available on NOAA Paleoclimatology database and PANGAEA data repository.

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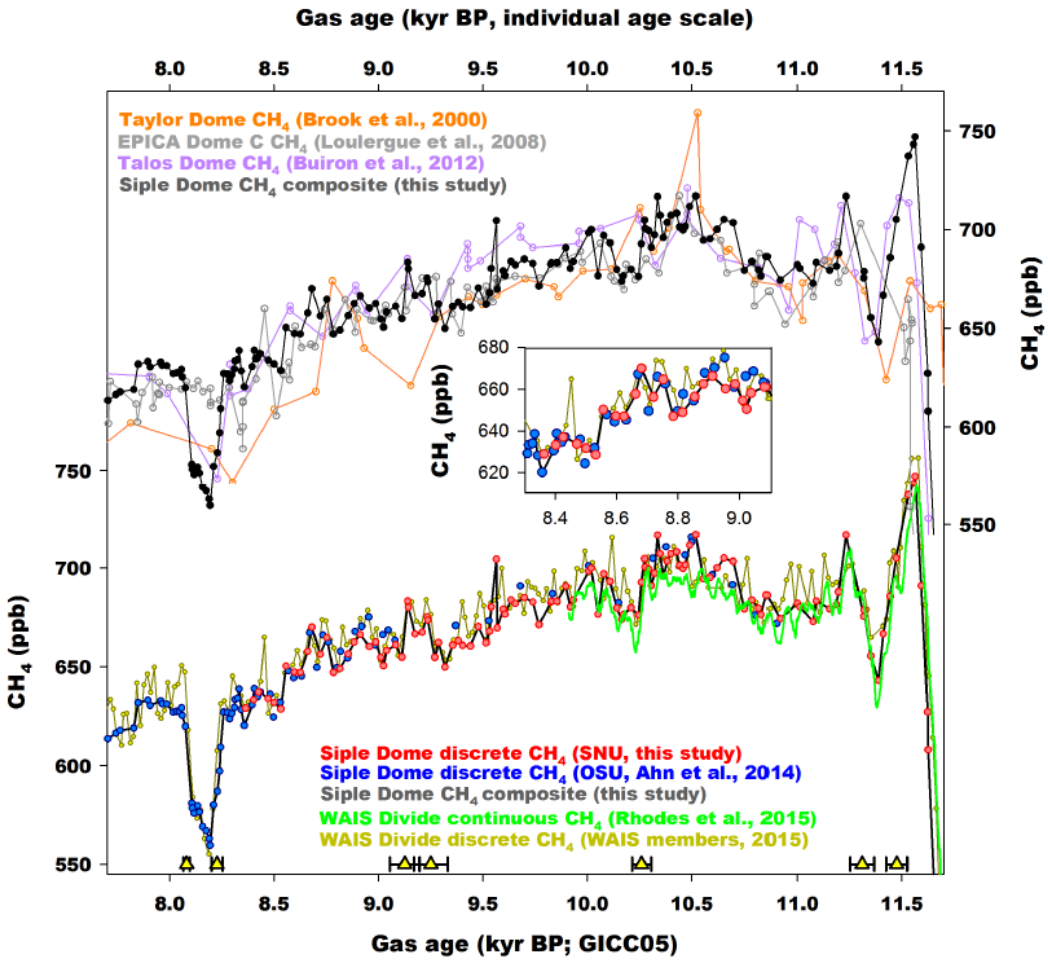


Figure 1. Atmospheric CH_4 concentration reconstructions during the early Holocene. Top: new high-resolution Siple Dome composite (black, this study and Ahn et al., 2014) compared with previous records from Taylor Dome (orange, Brook et al., 2000), EPICA Dome C (grey, Loulergue et al., 2008), and Talos Dome (purple, Buiron et al., 2012). Bottom: Siple Dome CH_4 records measured in OSU (blue, Ahn et al., 2014) and SNU (red, this study). Siple Dome composite (black line) is plotted with WAIS Divide discrete (dark yellow, WAIS Divide project members, 2015) and continuous measurement records (green, Rhodes et al., 2015). Inset: Enlarged plot showing overlapped interval between OSU and SNU Siple Dome data. ~~Siple Dome CH_4 time series measured by Seoul National University (red, this study) and Oregon State University (blue, Ahn et al., 2014). The Siple Dome composite data is shown in black line. For comparison, WAIS Divide CH_4 records reconstructed by discrete (yellow, Brook and Sowers, 2016) and continuous measurements (green, Rhodes et al., 2015) are plotted together. The WAIS Divide data set are synchronized to GICC05 age scale by CH_4 correlation. The yellow triangles and horizontal error bars indicate the age tie points and age displacements, respectively.~~

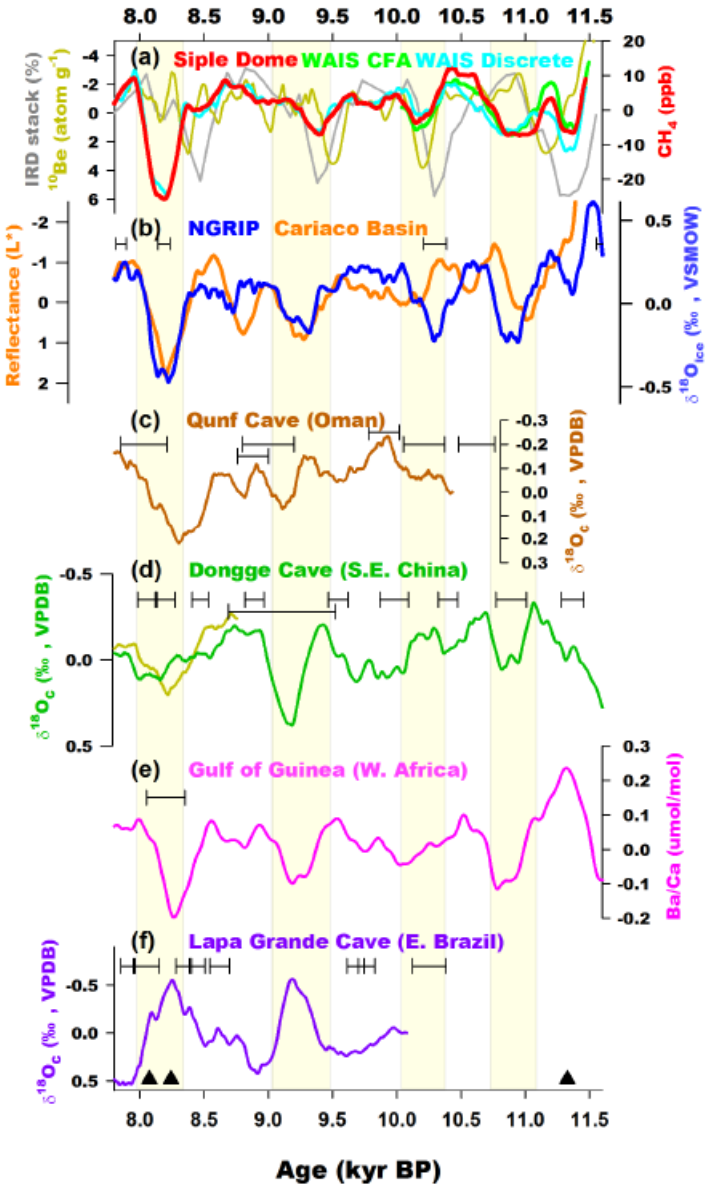


Figure 2. Millennial scale climate variability. All proxies present here were smoothed by 250-year running average and detrended by high-pass filter with 1/1800-year window. (a) Siple Dome CH_4 (red, this study), Greenland ^{10}Be (dark yellow, Finkel and Nishizumii, 1997), North Atlantic IRD stack (grey, Bond et al., 2001). Also shown are WAIS Divide CH_4 data by discrete (cyan, denoted "WAIS Discrete", WAIS Divide project members, 2015) and continuous (yellow green, denoted "WAIS CFA", 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). Black solid triangles are age tie-points used to adjust Siple Dome and WAIS Divide CH_4 data to GICC05 scale. ~~Millennial scale variability of CH_4 and other climate proxies. All climate proxies are smoothed with 250-year window after filtered in 1/1800 year high-pass window. (a) Millennial scale Siple Dome CH_4 anomaly (red) is plotted with North Atlantic IRD stack (grey, Bond et al., 2001) and Greenland composite ^{10}Be concentration from GRIP and GISP2 (olive yellow, Finkel and Nishizumii, 1997; Yiou et al., 1997). GRIP and GISP2 ice core chronologies are synchronized to GICC05 scale by visual matching between $\delta^{18}O_{ice}$ time~~

~~series. WAIS Divide CH_4 anomalies are shown in green (Rhodes et al., 2015) and cyan (WAIS Divide Project Members, 2015). (b) NGRIP $\delta^{18}O_{ice}$ on GICC05 time scale (dark blue, Rasmussen et al., 2006) and Cariaco basin sediment reflectance (orange, Deplazes et al., 2013). Black error bars indicate maximum age uncertainty of GICC05 scale as stated in Rasmussen et al. (2006). (c) Oman speleothem records from Qunf (purple, Fleitmann et al., 2007) and Hoti cave (grey, Neff et al., 2001) on their own chronology. (d) Siple Dome Δe_{LAND} (red), Severinghaus et al., 2009) and Dongge Cave speleothem $\delta^{18}O$ composite (green) from Dykoski et al. (2005) and Wang et al. (2005).~~

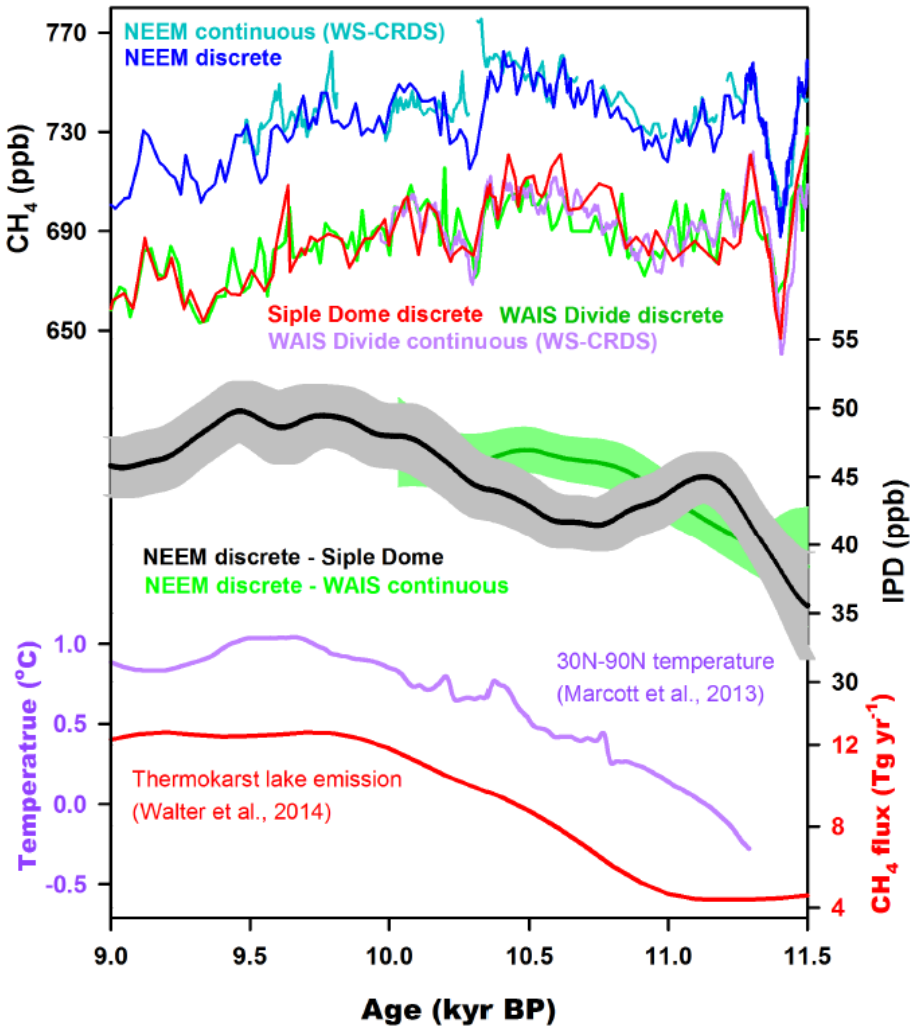


Figure 3. **CH_4 inter-polar difference (IPD) and high latitude CH_4 sources.** ~~(from top to bottom)~~ **Top:** High resolution CH_4 discrete measurements from NEEM discrete (blue, Chappellaz et al., 2013), NEEM continuous (turquoise, Chappellaz et al., 2013), WAIS Divide discrete (yellow green, WAIS Divide project members, 2015), WAIS Divide continuous (purple, Rhodes et al., 2015), and Siple Dome (red, this study) ice core records. Middle: ~~IPD (light grey) and 500~~1000-year low-pass filtered IPD-1 (black) and IPD-2 (green) with 95 % significance interval (shaded). Bottom: ~~Previous estimates are marked in green and orange (Brook et al., 2000; Chappellaz et al., 2013).~~ Proxy-based temperature reconstruction for 30°N-90°N and 30°S-30°N latitude (blue, Marcott et al., 2013). CH_4 flux estimate from Siberian- and Alaskan thermokarst lakes (red, Walter-Anthony et al., 2014).

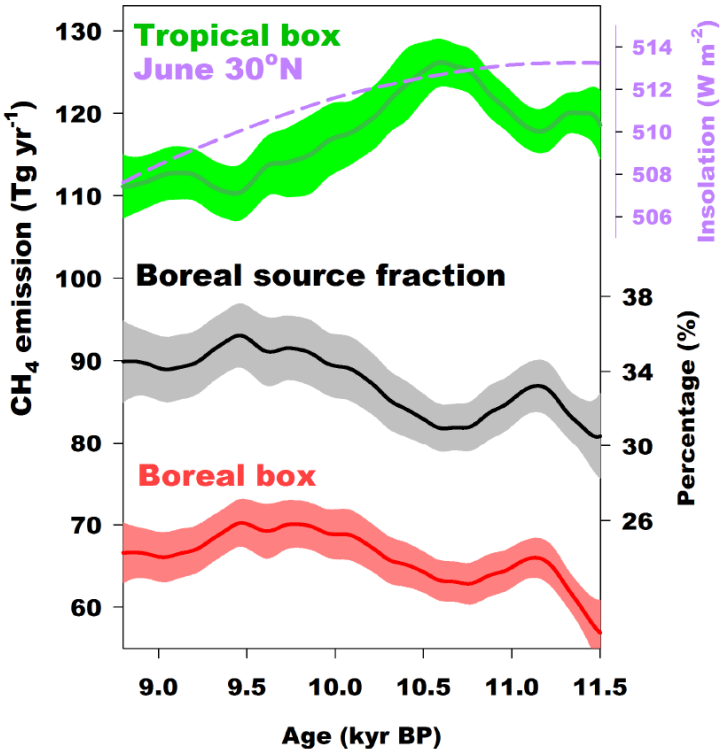


Figure 4. 3-box source distribution model results of tropical (green) and boreal (red) boxes. Black line shows the boreal to total source fraction (see text). Purple dashed line plotted with tropical emission is summer insolation in 30°N (Berger and Loutre, 1991).

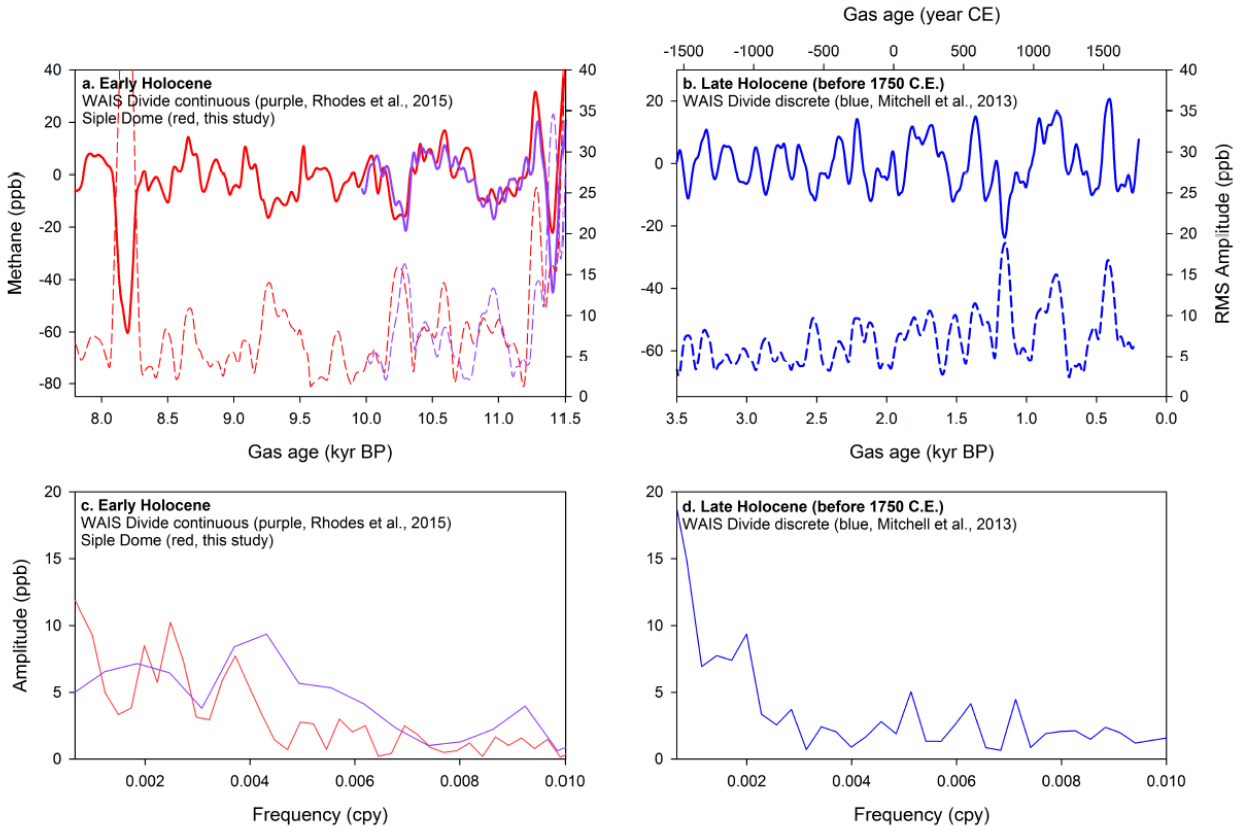


Figure 5. Upper: Detrended (75 to 1800-year band-pass filtered) CH_4 for the early (a) and late (b) Holocene from Siple Dome (red, this study), WAIS divide continuous (purple, Rhodes et al., 2015), and WAIS divide discrete (blue, Mitchell et al., 2013) data. Dashed lines are root mean square (RMS) amplitude running averaged by 75-year window. Lower: Amplitude spectrum of Early (c) and Late (d) Holocene CH4 records. Note that CH_4 data before 1750 C.E. are used for the preindustrial late Holocene.

Table 1. 3-box source distribution model results of tropical (green, T) and boreal (red, N) boxes and boreal source fraction (N/(T+N)) compared with previous results. Errors denote 95% confidence interval.

Ref.	N box	T box	Boreal source fraction N/(N+T)
(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 ka) ~~(10.8 ka)~~	66 ± 4 ~~65 ± 2~~	120 ± 4 ~~122 ± 4~~	33 ± 2 ~~32 ± 1~~
This study (11.5 ka)	47 ± 9	149 ± 10	22 ± 5
This study (9.9 ka) ~~(9.8 ka)~~	65 ± 8 ~~74 ± 2~~	119 ± 9 ~~110 ± 3~~	33 ± 5 ~~37 ± 1~~

Supplementary text

Reproducibility test from Styx glacier ice core

Here we present results of reproducibility test by using different ice core samples in the same manner as used for Siple Dome ices, to demonstrate the reliability of our analytical system. The ice core was drilled at Styx glacier (73° 51.10'S, 163° 41.22'E, 1623 m a.s.l) in 2014-2015 austral summer, and mean snow accumulation rate was estimated as 0.13 Mg m^{-2} yr^{-1} (Han et al., 2015). The replicate measurements were carried out at randomly-chosen 7 depths with time interval of 51 to 226 days. Depth difference between the replicate pairs is less than 10 cm. Results show the mean absolute difference between the original- and replicate measurements of 1.9, which is as good as the Siple Dome results. The daily blank offset (see the main text) ranges from ~6 to 19 ppb, and the intra-day blank offset is 2.4 ppb (standard error of the mean). Again, these results reveal the robustness of our blank correction methods.

Maximum SDMA gas age unceratinty

In this paper we used a modified gas age scale from the previous one based on $\delta^{15}N$ measurements (Severinghaus et al., 2009) by setting 3 age tie-points and interpolating the age offset between the tie-points (Fig. S1). To estimate gas age uncertainty, we compared the SDMA modified gas age (this study) to the new gas age determined by CH_4 correlation with NEEM discrete CH_4 data measured at OSU (Chappellaz et al., 2013). Figure S3 shows the offset between the two age scales, which it should be moved to adjust the NEEM CH_4 in GICC05modelext-NEEM-1 age. In addition, we take into account the maximum layer counting uncertainty of 99 years (Rasmussen et al., 2006) and delta-age uncertainty of 30 years (Rasmussen et al., 2013) during the early Holocene. Error progagation gives us the maximum uncertainty of the early Holocene SDMA gas age of ~147.4 years.

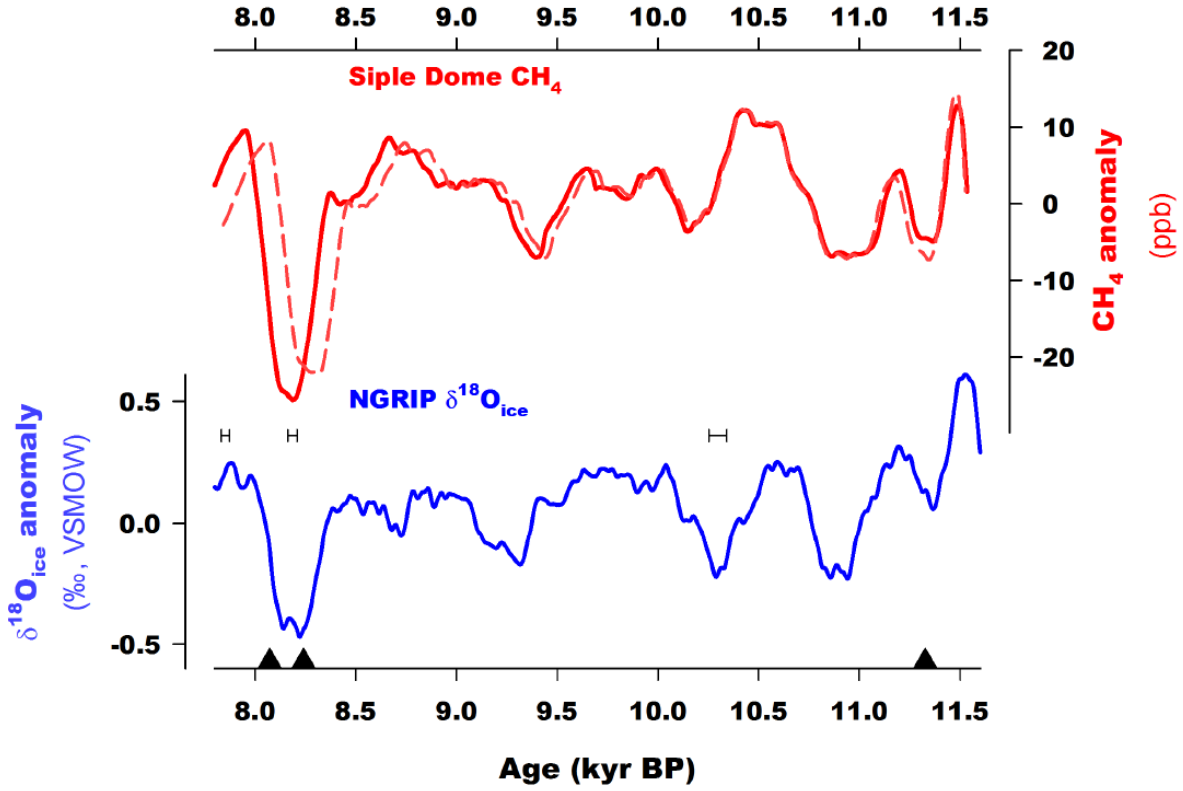


Figure S1. Upper: Comparison between Siple Dome CH_4 anomalies plotted with gas age adjusted to GICC05 (red, solid) and previous gas age (red, dashed; Brook et al., 2005). Lower: NGRIP $\delta^{18}O$ anomaly in GICC05 scale. The horizontal error bars denote the age uncertainty of GICC05 chronology (Rasmussen et al., 2006), and the black triangles are age tie points used to adjusting the Siple Dome age scale to GICC05 scale.

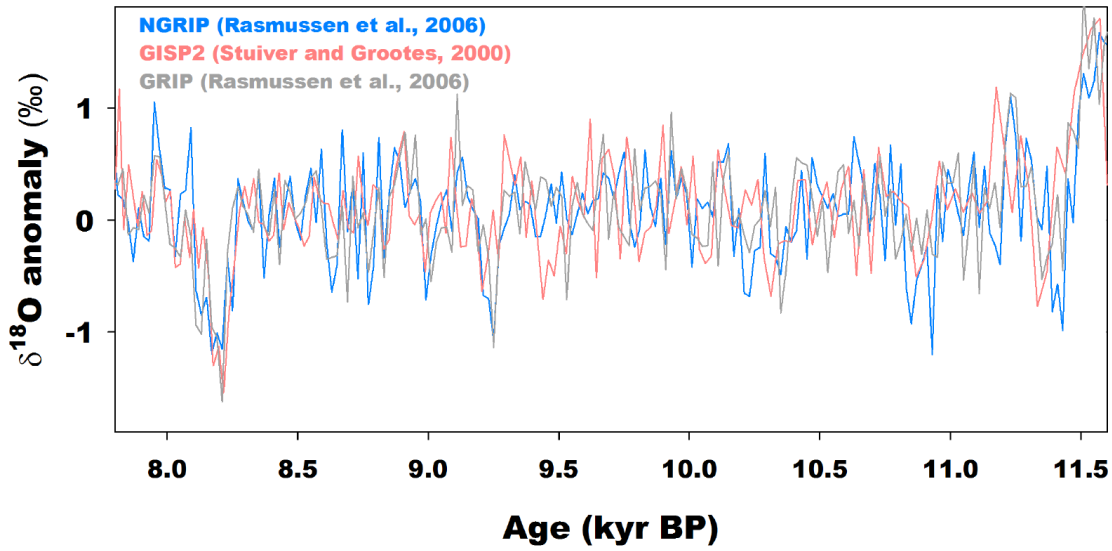
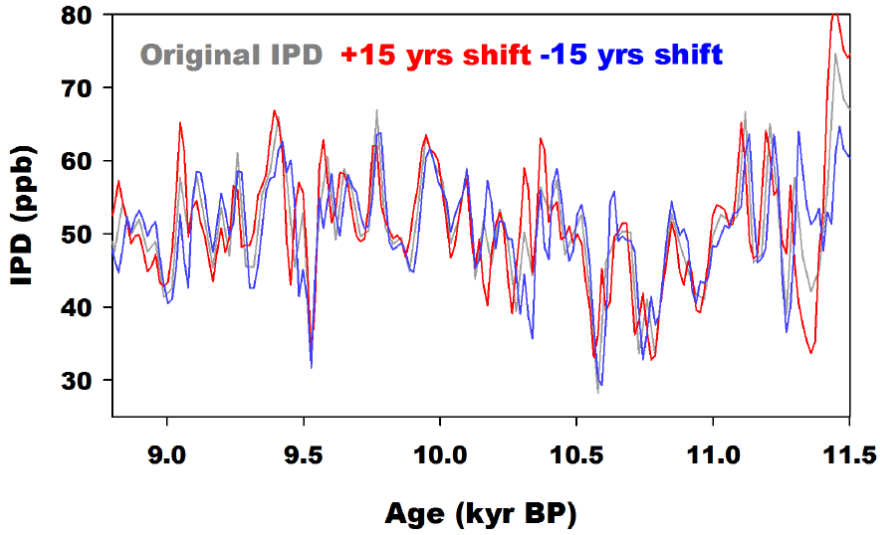


Figure S2. Comparison of Greenland oxygen isotope ratios from NGRIP (blue, Rasmussen et al., 2006), GRIP (grey, Rasmussen et al., 2006) and GISP2 (red, Stuiver and Grootes, 2000). All time series were high-pass filtered with 1/1800-year window. Note that the cooling amplitude at 10.3 ka is smaller than 8.2 and 9.3 ka events in NGRIP records, but this is not clear in GRIP and GISP2 ice cores.



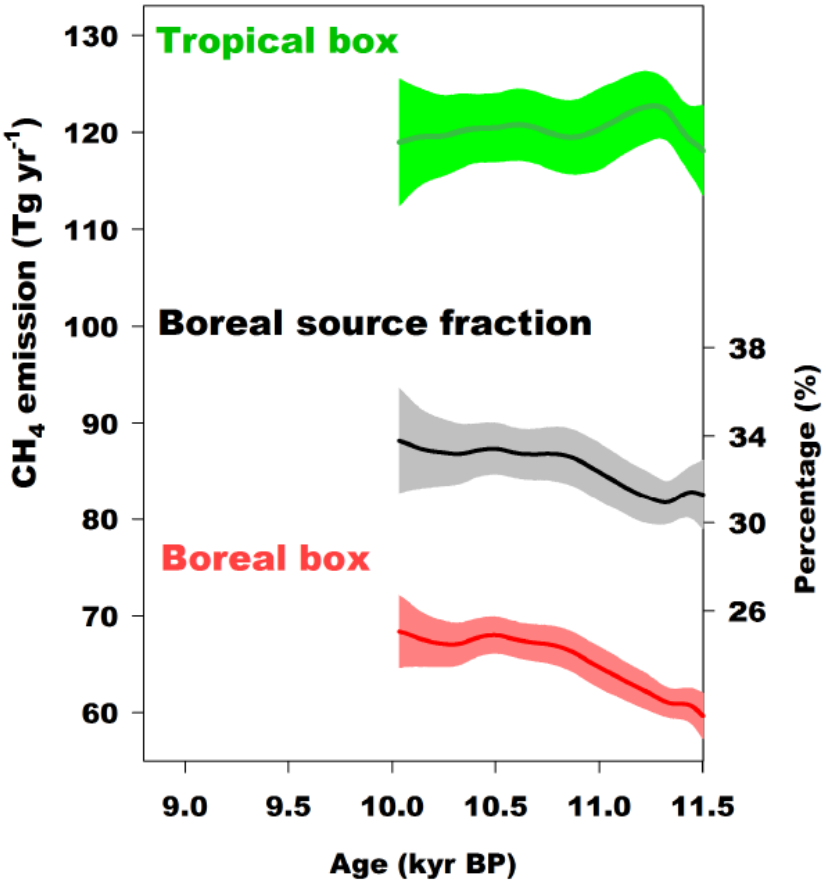


Figure S3. 3-box source distribution model results from IPD-2. ~~IPD sensitivity test. The IPD calculated by the best match chronology (grey line) is plotted with ±15 years shifted chronology (red and blue, respectively).~~

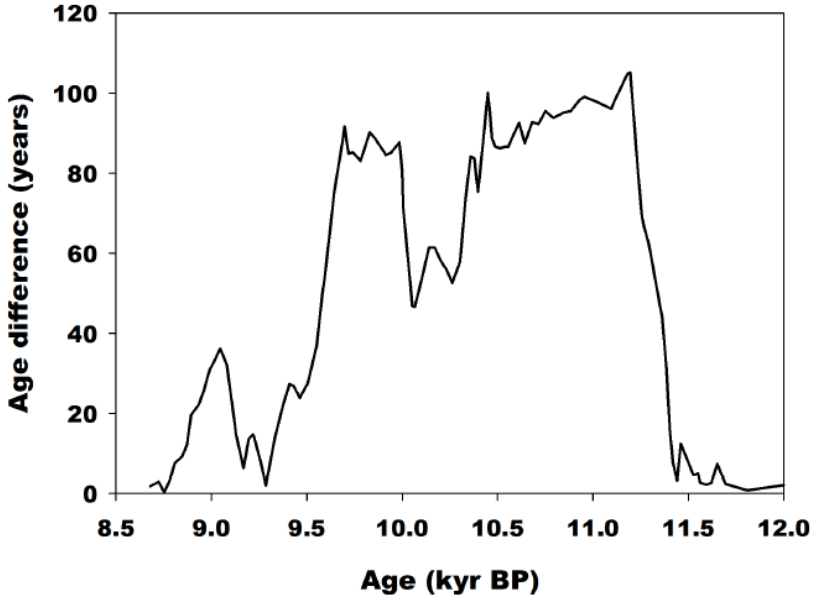


Figure S4. Age difference between the new gas age scale adjusted to GICC05 by Monte Carlo matching with NEEM discrete CH_4 (Chappellaz et al., 2013) and the original gas age based on $\delta^{15}N$ records (Severinghaus et al., 2009).

Table S1. 3-box source distribution model results of tropical (green, T) and boreal (red, N) boxes and boreal source fraction obtained from IPD-2. Errors denote 95% confidence interval.

Ref.	N box	T box	Boreal source fraction
(ka)	(Tg yr^{-1})		(%)
This study (11.5 ka)	60 ± 7	134 ± 16	29 ± 4
This study (10.0 ka)	71 ± 7	115 ± 11	35 ± 4

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