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Atmospheric methane control mechanisms during the early Holocene

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Abstract. Understanding 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 of millennial scale CH_4 variability 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. Each CH_4 minimum corresponds 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. 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 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 sources (e.g., Etiope et al., 2008 and references therein). Additionally, a portion of CH_4 is

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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). 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 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). However, recent model studies suggested the dominant role of source changes rather than sink in controlling atmospheric CH_4 for 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) from Antarctic- and Greenland ice cores (e.g., Spahni et al., 2005; Loulergue et al., 2008). A time series of the past CH_4 changes found that CH_4 mixing ratio was low during glacial periods and high in interglacials, generally following the glacial-interglacial cycles and related global ice volume changes (Lisiecki and Laymo, 2005; Loulergue et al., 2008). For shorter time scale variability, the tropical monsoon activity and solar insolation changes were proposed as primary controls (e.g., Guo et al., 2012). However, no direct correlation between CH_4 and tropical monsoon signals has been reported in shorter time scales, 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.

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

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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 C3/C4 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

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. 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

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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. 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). 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.

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.

The gas age scale, which was previously constrained (Brook et al., 2005), was converted to the Greenland Ice Core Chronology 2005 (GICC05) age scale by setting the 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 scale, which range from -114 to 28 years.

3. Result and Discussion

Our new Siple Dome CH_4 record is the one of the high-resolution data set covering 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 measurement carried out at Oregon State University (OSU) 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). Both records agree well with each other for overlapping interval of the mean difference between two data sets (OSU – SNU) of 3 ppb. We subtracted 3 ppb from the OSU data to combine with our new data. Fig. 1 shows the resulting CH_4 composite extending from 11.6 to 7.7 ka with a mean temporal resolution of 26 years.

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3.1 Millennial scale variability

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 the millennial scale minima at ~8.2, 9.3, 10.2 and 11.0 ka, which occurred in nearly 1000-year spacing. Each minimum was 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 for the Holocene interglacial condition. 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. In contrast, we observe a moderate positive correlation ($r = 0.66$, $p = 0.0013$) 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 there is no age tie-points between the 8.2 ka episode and the Preborial 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 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 each abrupt cooling event, as revealed in previous studies for different time periods (Peterson et al., 2000; Haug et al., 2001; Fleitmann et al., 2007; Deplazes et al., 2013). Moreover, ^{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).

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 no clear indication of southward migration of ITCZ position and reduction of Asian summer monsoon intensity corresponding to ~10.2 ka cooling (Fig. 2). Instead, there are two

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small decrease at ~9.9 and ~10.6 ka, but these episodes are likely beyond the chronological uncertainty, considering that the age uncertainty of Dongge Cave deposits is ±77 years (2 sigma error; Dykoski et al., 2005), and that the Siple Dome age uncertainty is likely less than ~100 years (see above and Fig. S1). This could be because the climate teleconnection between North Atlantic and tropical hydrology 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 is not supported by other Greenland ice core records such as Greenland Ice Core Project (GRIP) and Greenland Ice Sheet Project 2 (GISP2) (Fig. S2).

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. This may imply CH_4 reduction was controlled by other processes than 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 evidence suggests that changes in precipitation and surface hydrology in the northern tropics may have not changed significantly during around the 10.2 ka.

Increased boreal source contribution is a probable compensating mechanism for smaller reduction in tropical CH_4 emission. Terrestrial proxies in boreal regions showed the formation of peatlands and thermokarst lakes, which occurred first in Alaska and northern Canada from ~11.0 ka, followed by Siberia and Asia (MacDonald et al., 2006; Brosius et al., 2012; Yu et al., 2013). Exposing a new land by deglaciation (Dyke, 2004) and temperature increase over northern high latitude (Marcott et al., 2013) established a favourable climate condition for thawing permafrost, thus releasing CH_4 from there. We will discuss it further below.

3.1.2 External forcing

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

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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.

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). Mitchell et al. (2011) reported a moderate positive correlation between LPIH CH_4 and reconstructed Pacific Decadal Oscillation (PDO), which has been known as a long-period ENSO pattern in Pacific Ocean (Mantua and Hare, 2002). 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

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. 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. The NEEM CH_4 record is chosen as a 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 Siple Dome and NEEM data by setting 7 match points, 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 and 1.0 ppb for SDMA).

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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, 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 latitude (30-90°N, N-box), tropical (30°S-30°N, T-box), and southern high 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. 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). T-box concentration 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 of the boxes, lifetime of each box, and transport times among the boxes.

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 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 suggested as a source of CH_4 (e.g., Walter et al., 2006, 2007; Brosius et al., 2012). 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

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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 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.

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. 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.

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4. Conclusion and summary

In this study we reconstructed 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. Our results show a series of the millennial scale CH_4 minima accompanied with Greenland cooling, changes in ITCZ position and reduced Asian and Indian monsoon intensities, and the evidence suggests that the low latitude source changes were the major causes of the early Holocene CH_4 minima. 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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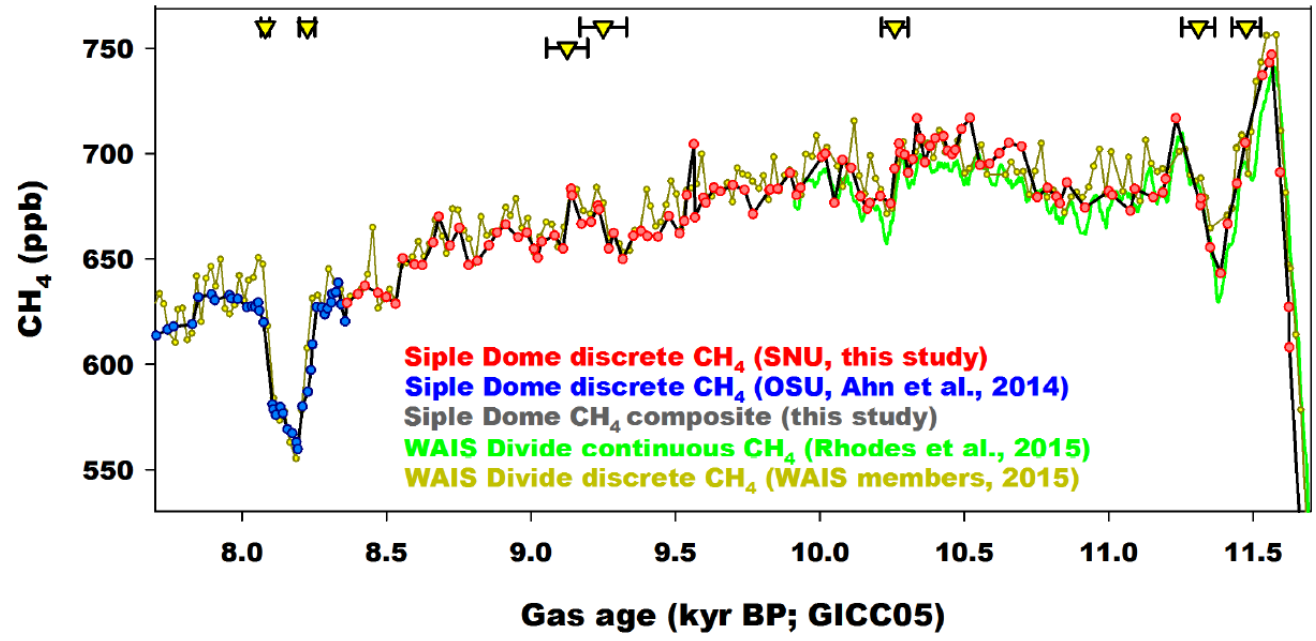

Figure 1. Siple Dome CH_4 time series measured by Seoul National University (red, this study) and Oregon State University (blue, Ahn et al., 2014). The Siple Dome composite data is shown in black line. For comparison, WAIS Divide CH_4 records reconstructed by discrete (yellow, Brook and Sowers, 2016) 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.

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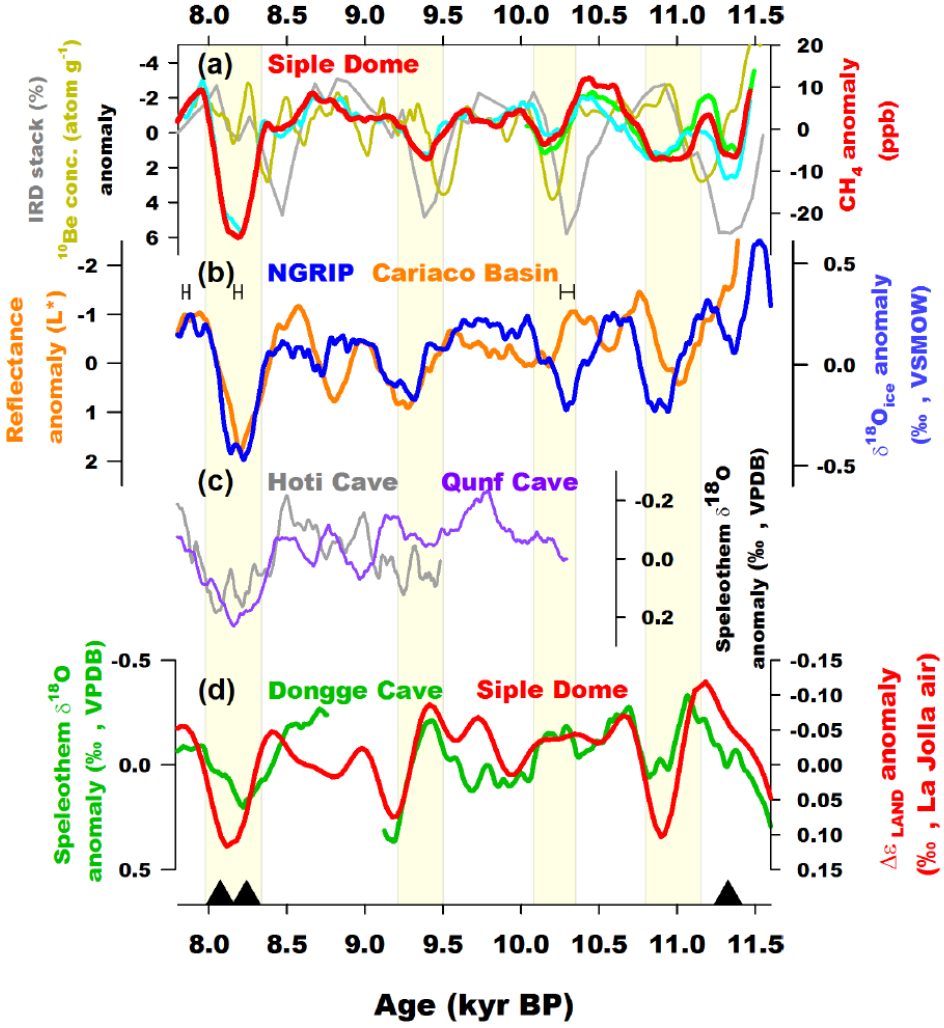

Figure 2. Millennial scale variability of CH_4 and other climate proxies. All climate proxies are smoothed with 250-year window after filtered in 1/1800 year high-pass window. (a) Millennial scale Siple Dome CH_4 anomaly (red) is plotted with North Atlantic IRD stack (grey, Bond et al., 2001) and Greenland 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 $\Delta\varepsilon_{LAND}$ (red), Severinghaus et al., 2009) and Dongge Cave speleothem $\delta^{18}O$ composite (green) from Dykoski et al. (2005) and Wang et al. (2005).

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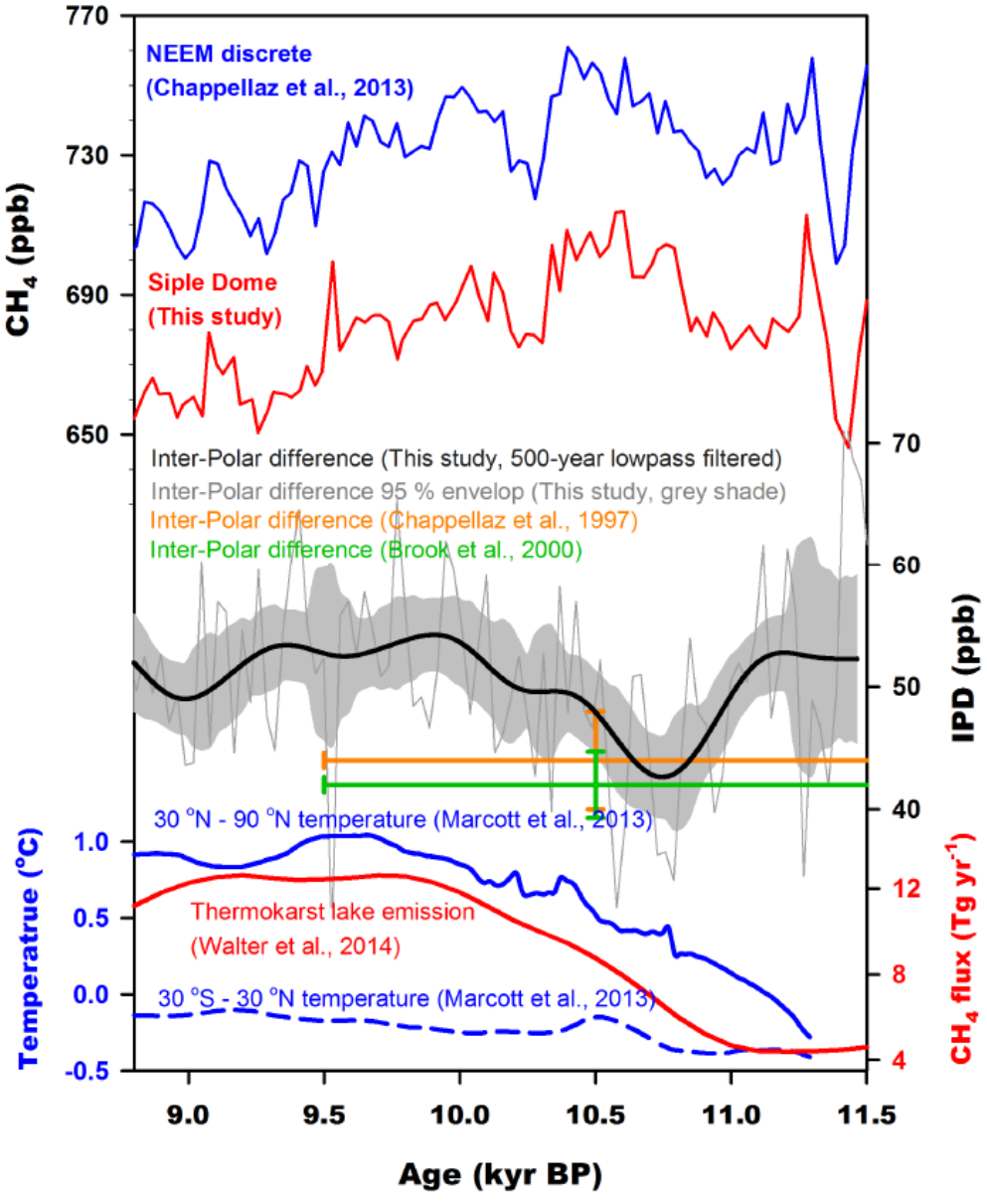

Figure 3. (from top to bottom) High resolution CH_4 discrete measurements from NEEM (blue, Chappellaz et al., 2013) and Siple Dome (red, this study). IPD (light grey) and 500-year low-pass filtered IPD (black) with 95 % significance interval (grey shaded). Previous estimates are marked in green and orange (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).

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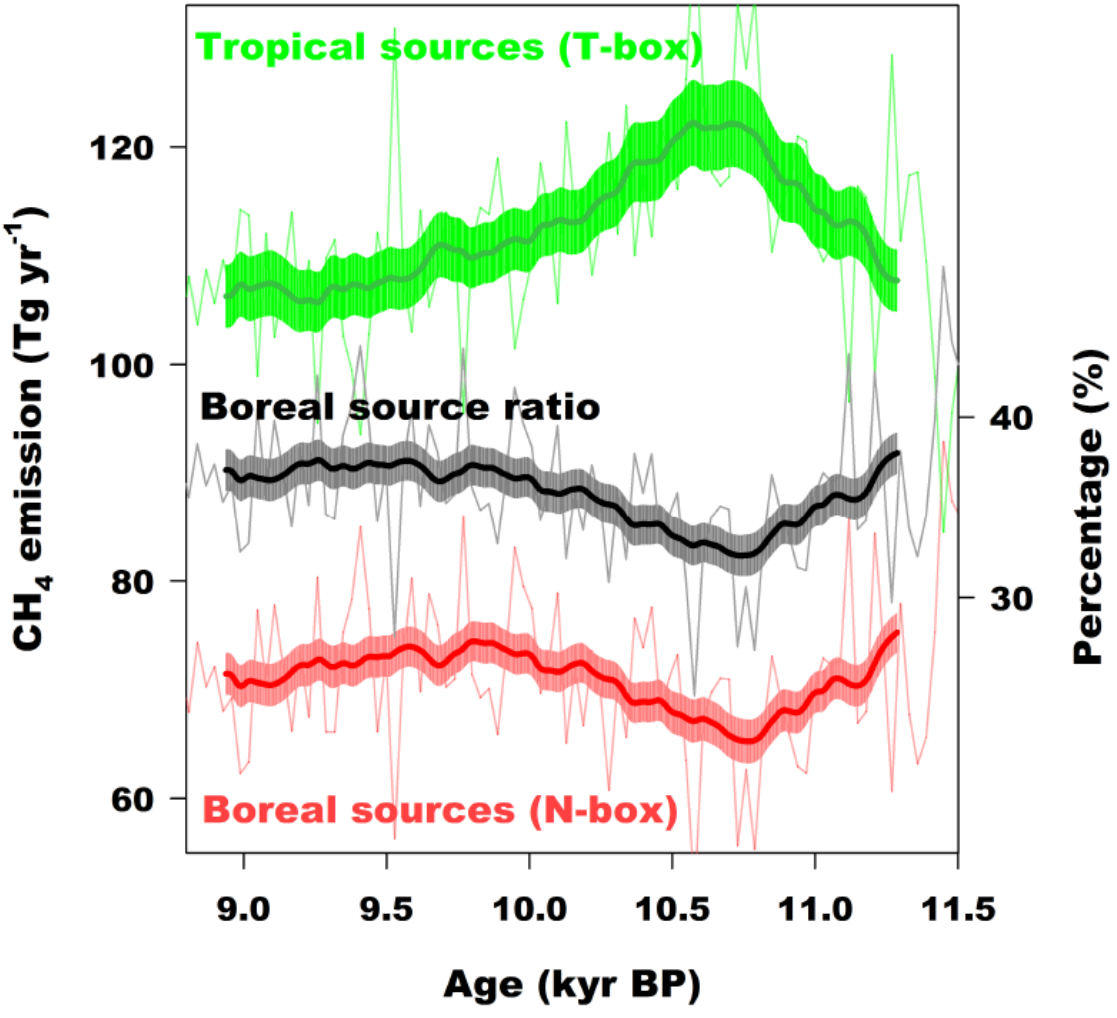

Figure 4. 3-box source distribution model results of tropical (green) and boreal (red) boxes. Black line shows the boreal source fraction (see text).

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Ref.	N box	T box	N/(N+T) 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 (10.8 ka)	65 ± 2	122 ± 4	32 ± 1
This study (9.8 ka)	74 ± 2	110 ± 3	37 ± 1

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