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Climate of the Past Discussions

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

Anonymous Referee #2

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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.

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.

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?

- 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.

- 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.

- 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.

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.

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)

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

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

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

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

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

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.

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.

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

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

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

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.

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

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

P8 l19: the boreal sources increased

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.

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.

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

P20 l5 Chappellaz et al., 1997 not 2013

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