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.

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.

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. 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. 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.

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

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.

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).

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.

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.

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?

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?

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.

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.

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.

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.

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

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?

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

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

p2L18: See comment 5) in general comments p2L18-19: too weak. The correlation between CH_4 and NH temperature (δ^{18}O-ice) is well established

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)

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

p3L27: there is still some ice left in Greenland

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

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

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.”

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

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

p4L7-17: see point 2) in general comments

p4L17: provide reference how you calculated gravitational fractionation

p4L19: provide reference for GICC05 time scale

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.”

p4L26: develop a more complete representation of analytical uncertainty

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?

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.

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

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

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

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?

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

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

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".

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

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

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

p7L24: consider IPD reconstructions with CH_4 records from WAIS

p7L25-30: NICE approach!

p8L2: …show an increase by XYZ ppb from…

p8L4: …in both hemispheres during…

p8L6: …from both hemispheres and…

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

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?

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

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

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

p8L29: explain "conventional" northern CH_4 emissions

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.

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

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

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.

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

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

p18L4: add reference to the list or check reference

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

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

p20F3: add IPD with WAIS data

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