Interactive comment on "Substrate potential of Eemian to Holocene permafrost organic matter for future microbial greenhouse gas production" *by* Janina G. Stapel et al.

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Responses to the reviewer's comments: "Substrate potential of Eemian to Holocene permafrost organic matter for future microbial greenhouse gas production"

By Janina G. Stapel et al.

We thank the reviewer for his/her thoughtful and very constructive comments and suggestions on our manuscript which will improve the clarity and the quality of the paper. Below, we will address all listed issues, which are relevant for discussion.

Reviewer #1 To Introduction:

1) The link between the general introduction on permafrost thaw, its consequences, and the scope of this study can use some improvement. For example, Bol'shoy Lyakhovsky Island is suddenly mentioned on P2, L15. Later it appears to be the study site, but it needs some more context in the introduction. Similar for the 'Eemian deposits in this study...' (P2, L30). Also include acetate, and the difference between free and bound acetate.

We agree with the reviewer and will improve the first part of the general introduction by rephrasing, condensing and/or combining sentences, and better outlined the relation between global warming, permafrost thawing, OM availability, greenhouse gas production and it feedback on global warming and permafrost thaw. We agree to describe why Bol′shoy Lyakhovsky Island was selected as study site earlier in the introduction and why Eemian deposits are of specific interest. We will add some sentences on acetate as a quality indicator for greenhouse gas production and the difference between free and bound acetate into the introduction. Finally, we will improve the context of our scientific question: "The feedback between climate warming and microbial greenhouse gas generation from thawing permafrost is a topic of intensive modern scientific debate (Zimov et al., 2006; Koven et al., 2011; Schuur et al., 2015). Especially the contribution of OM from thawing permafrost deposits of different ages to the climate carbon feedback cycle is still an open question. Therefore, the aims of this study are (1) to compare the stored potential for microbial greenhouse gas production in permafrost deposits from different glacial/ interglacial and stadial/ interstadial periods, and (2) to assign the substrate potential of different permafrost units to characteristic OM parameters and palaeoenvironmental deposition conditions. Furthermore, the Eemian deposits in this study are used as model for an interglacial period containing information on how an ongoing warming climate in the Arctic may affect permafrost OM degradation."

To Stratigraphy:

2) The composite core consists of different lithologies, i.e. lacustrine (MIS1and MIS5), floodplain deposits (MIS4), and also contains cryostructures. I miss a discussion on

how different sources may influence OM parameters, lipid abundance and distribution, or acetate availability? For example, GDGTs in lacustrine (MIS1 and MIS5) and flood-plain deposits (MIS4) may have a mixed soil and aquatic origin. This may influence your results, especially when considering that an earlier study has shown that Siberian thermokarst lakes (probably comparable to the MIS5 deposits in this study?) can contain >200 times the concentration of branched GDGTs compared to Yedoma (Peterse et al., 2014, JGR-B). Similarly, isoGDGTs, archaeol, and PLFA concentrations may be influenced too.

Our pyrolysis data indicate that the OM is mainly of terrestrial origin with some variations in the aliphatic character. Mainly samples deposited during the Late Pleistocene (LP) glacial period show this higher aliphatic character. We already discussed these differences in the aliphatic character as a result of different environmental conditions (higher soil moisture favorable for algae growth vs dry conditions, anaerobic soil conditions/reduced OM decomposition, cold vs warmer conditions etc.) rather than of different depositional settings (see chapter 5.1), which seems sometimes confusing. For instance, Holocene as well as Eemian samples were both deposited in a kind of thermokarst lakes, but they show significant differences in the amount and quality of the OM. Thus, the influence of the depositional settings on the OM parameters applied here is not clear yet and environmental impact on OM production and degradation might play a more important role. We will add a few sentences on this into the manuscript. In our study we cannot really see that the Holocene GDGTs or archaeol concentrations (deposited in thermokarst lakes) are significantly more abundant than in the LP glacial deposits. The data are in the same range in all time intervals. However, we will add a sentence on this into the manuscript. PLFAs are generally high in active layers and significantly lower in permafrost sequences. Thus, the PLFA signal is mainly determined by the thaw-front in summer and not so much from the depositional environment.

3) Another way to check sources could be to calculate BIT index values (Hopmans et

al., 2004, EPSL). Peterse et al., 2014 (JGR-B) found that Yedoma has a significantly lower BIT index than in soils. Similar changes should be visible throughout the composite core studied here.

We calculated the BIT index (according to Hopmans et al., 2004) and will integrated it into the text as well as provide the data in the table in the supplement (S1). The BIT index is usually 1 and only decrease in four MIS 3 samples (and in one AL of MIS 4) to 0.91 and, therefore, it supports the overwhelming terrestrial soil character. Our study found that the BIT in the Yedoma deposits (MIS 3 and 4) did not much differ from the BIT values indicated in the active layers or in MIS 1 and 5e. The results presented by Peterse et al. (2014), indicate a much lower BIT value of 0.82 for Yedoma deposits from the Duvannyi Yar cliff. According to Strauss et al. (2012), the Yedoma at Duvanny Yar is of polygenetic origin formed on a floodplain of the Kolyma River, where alluvial and fluvial processes were the controlling processes. On the other hand, the Yedoma deposits on Bol′shoy Lyakhovsky Island were not influenced by dynamic fluvial processes during deposition (except for the lowermost 40 cm of core L14-03, where gravel in the sediments might indicate stronger fluvial influence during deposition). Furthermore, the investigated permafrost deposits are not floodplain deposits and therefore are less influenced by alluvial processes (Andreev et al., 2009). That explains why - other than in Peterse et al., 2014 - we observe a higher BIT index in the Yedoma deposits at Bol′shoy Lyakhovsky Island and do not see relevant differences in the BIT index by comparing the individual deposits.

To Methods:

4) P4, L30: were samples decalcified prior to determining TOC? Otherwise total carbon is reported instead of TOC. P5, L5: was there any pre-treatment of the sample material prior to pyrolysis?

Samples for TOC analyses were decalcified before the measurement. We will add this information into the text. For both Rock-Eval pyrolysis and open-system pyrolysis

sample material was freeze-dried and ground (as indicated in the text). There was no additional pre-treatment of the samples before the measurement. Free-biomolecules were thermally removed before pyrolysis of the macromolecular organic matrix. We will add this information into the text, too.

To Discussion:

5) P9, L3: How/in what figure/parameter is the contribution of aquatic OM reflected? Similar for the input of aquatic OM during MIS3 (P9, L32).

Generally, based on our results the deposited OM in all investigated cores is of terrestrial origin. However, the results from Rock-Eval analysis (Hydrogen Index) indicate that the samples have varying proportion of a hydrogen-rich component (Fig. 3a). Open-system pyrolysis shows that the samples with a higher HI reveal a higher aliphatic character (Fig. 3b). In contrast to aromatic compounds aliphatic compounds are assumed to be better degradable, thus, samples with higher HI (more aliphatic rich) seem to have a better OM quality in terms of biodegradability. This material also contains higher concentrations of potential substrates (e.g. acetate). A source of more aliphatic-rich OM could be algae, living in surface ponds or water saturated soils. Periods of increased soil-moisture for the Yedoma deposits were already indicated by Sher et al. (2005). Thus, higher HI point to a higher aliphatic character in permafrost OM which may reflect a higher proportion of aquatic OM. (We will add this to the text.) Figure 3a (after Eglinton et al., 1990) classifies the OM by different types and provides hints on the origin of the OM; whereby OM type I and II are characteristic for aquatic or marine OM. Especially, Yedoma samples (Fig. 3a; e.g. AL, LP1 and LP2) show a stronger proportion of hydrogen-rich OM (type II OM). The higher aliphatic character seems to be related to increased soil moisture favorable for algae growth during time of deposition or as it is observed today in the active layer during the thawing period. Figure 3b assigns the higher aliphatic character to the LP glacial samples. We will add some further information on the parameters and figures indicating "aquatic OM" to the discussion.

6) Please clarify. Again P12, L15-16: what indicates the link to moist depositional conditions?

We will delete "moist depositional conditions" from the text here, since it was not important at this position. As described under point 5) above the interpretation on moist depositional environment derived from the increased aliphatic character and from literature information for the LP glacial period.

7) P.9, L27: I can't follow this sentence. Check grammar/order of words. Also: if glacial conditions would slow down degradation, how does it influence its production?

We will rephrase this sentence as suggested.

8) P.10, L15 and following: How can you be certain that GDGTs reflect past microbial biomass? In this study, only the core lipids are analyzed, whereas part of the GDGT pool may present as IPL, and thus derive from living biomass. Other studies have reported an IPL contribution of >30% to the GDGT pool in OC-rich soils (e.g. Peterse et al., 2011 Org Geochem).

IPL with an ether bond moiety are less suitable to act as life markers due to their significantly increased stability (Logemann et al., 2011). Due to the similar structural moieties something similar can be suggested for the br-GDGTs and iso-GDGTs. In contrast IPLs with ester moieties are known to rapidly decrease after cell death and thus, since microbial communities usually consist of both bacteria and archaea, we use the PLFAs as life markers for the microbial community in general here. Since life marker and past marker do not match we interpret that past marker signal is a paleo-signal. We will add this information to the discussion in chapter 5.2.

9) Furthermore, the manuscript refers to both microbial biomass and methanogens, of which the latter is of course more specific. Please go through the manuscript and check which level of specificity is relevant.

Methanogens are more relevant when discussing acetate as a substrate for green-

house gas production. We will go through the manuscript to be more precise.

10) Fig. 2: Is there a reason why archaeol is plotted together with isoGDGTs? They do not necessarily share the same source. Instead, it would be more logical to plot archaeol next to e.g. GDGT-0/cren, which are both indicators of methanogenesis. These data can then also be used to compare with the acetate data.

The reason was to have one parameter for the past archaeal biomass and to safe some vertical space for the figure. Iso-GDGTs are mainly dominated by iso-GDGT-0 (supplement table S1) and both iso-GDGT-0 and archaeol are used for the presence of methanogens in permafrost regions (Bischoff et al., 2014; Pancost et al., 2011), although their ratio differs throughout different methanogenic genera (Koga and Mori, 2006). We will add some sentences on this to chapter 5.2. Thus, we would like to keep this combined archaeal parameter in Fig. 2, but provide all GDGT data in the supplement table S1.

11) L 20: Can you explain how exactly GDGT concentration data provides information on the activity of microbial biomass in the past?

Sure the GDGTs are not a direct measure of microbial activity. However, we can assume that if microorganisms can be found that they have been active. Thus, the presence of past markers concomitantly suggests the activity of these past microbial communities (when they formed the active layer community). To avoid further confusions, we will delete this part of the sentence from the text.

12) L23: Given that GDGT concentrations and TOC seem to covary (i.e. both higher in Yedoma?), it makes sense to normalize GDGT concentrations on TOC to distinguish between high GDGT concentrations due to high TOC content and actual elevated microbial biomass. Do the trends and conclusions still hold?

Yes they do. By normalizing the GDGT and archaeol concentrations to TOC, the same trends within the depth profiles of the cores are visible. Depths of increased TOC and

OM quality (e.g. core L14-02 at about 1.5 m with a HI of 246 mg HC/ g TOC and TOC of 4.7 wt%) correspond with increased concentrations of br-GDGTs. On the opposite, depths of decreased TOC but increased HI (e.g. core L14-03 at about 1.4 m with a HI of 254 mg HC/ g TOC and TOC of 1.9 wt%) reveal increased br-GDGT and archaeol concentrations indicating that the concentration of living microorganisms in the past not only depends on the amount of TOC but also on the quality of the OM (HI values).

13) The choice of 'excellent' as description for substrate (in abstract and P10, L31) seems odd. In my opinion something can turn out to be, or has proven to be an excellent substrate, but you cannot select something as an excellent substrate if it isn't compared to anything else.

Acetate is well-known to be intensively used as a substrate for microbial metabolism. Thus, based on this background knowledge acetate was classified as being "an excellent substrate" especially for methanogenesis (thus, compared to what we know from literature). However, we will replace "excellent" by "appropriate".

14) The discussion on microbial activity in the active layer leads to the conclusion that MIS1, 3, 4 provide most substrate upon thaw. However, it is important to mention that active layer sediments included in this study, and that there are no active layer samples from MIS2 and MIS5 permafrost are included. There should be a few words on how representative this sample selection is.

Reading this comment, we agree that this sentence can be interpreted in this way. However, what we wanted to say is that there are already "natural" examples that old material can stimulate microbial life again. We are aware that we cannot say anything about the potential of MIS 5e material, since there was no active layer for the MIS 5e available in the field. We will add the following sentences:"…representing the overlaying soil as part of the active layer above the permafrost deposits from MIS 1, 3 and 4. Due to the stratigraphic settings at the study site on Bol'shoy Lyakhovsky Island, active layers containing OM from MIS 2 and MIS 5e could not be obtained in the field".

15) I furthermore miss the link between substrate availability and OM quality or composition and microbial biomarker abundance. I also miss a comparison with data from the literature. I have already mentioned Peterse et al 2014, JGR-B (Branched glycerol dialkyl glycerol tetraether in Arctic lake sediments: sources and implications for paleothermometry at high latitudes), but there are more biomarker papers on Siberian (or Arctic in general) permafrost soils, and there must be on microbial community composition and OM properties, too. To name a few: Bischoff et al 2013, GBC. Response of methanogenic archaea to Late Pleistocene and Holocene climate changes in the Siberian Arctic. Knoblauch et al., 2013, GCB, Predicting long-term carbon mineralization and trace gas production from thawing permafrost of Northeast Siberia. Blaud et al., 2015, Res in Microb. Arctic soil microbial diversity in a changing world. And papers citing those.

We will include a paragraph on the relation of OM quality or composition and microbial abundance, and a sentence on OM quality and substrate availability. Furthermore we will add a paragraph were we compare our results with literature data.

16) P11, L31: Can you support the drawn similarities between TOC and free acetate with statistics? Is their relation stronger during MIS3 and 4 compared to during MIS1 and 5?

The statistical similarity of the bound acetate fraction with TOC (average R^2 for all cores= 0.7-0.8) is much better than for the free acetate (for all cores $R^2 < 0.5$), which was already observed in Stapel et al., 2016. We think that the free acetate might be much more influenced by external factors (adsorption, release, consumption (e.g. in active layer), diffusion and maybe transport). The factors are not clear yet. We will integrate the statistics between free acetate and TOC to the text of chapters 4.1 and 5.3.

17) P12, L2-5: check the grammar of this sentence, I can't follow the reasoning. I think 'favoured' needs to be replaced with 'a result of'....caused by the onset of the Holocene,

when a warming climate caused unstable environmental conditions ....

We will correct the sentence according to the reviewer's suggestions.

18) P12, L5-9: This sentence also seems to lack punctuation marks, verbs, logical order of words. Please check.

We will rewrite and split this sentence.

Specific comments:

We thank the reviewer for his/her comments and will follow all his/her suggestions.

Terminology:

P.11, L20: what is old freeze-look permafrost; - it has to be freeze-locked permafrost

P. 11, L 27: what is a thermos terrace?; - It has to be "thermo terrace" which is also known as "thermo-erosional valley" (Schirrmeister et al., 2011).