We sincerely thank the reviewer for the comments, which helped us to improve our manuscript. Please find the comments (blue) and our reply (black) below.

## General comments

The article "Greenhouse gas production in degrading ice-rich permafrost deposits in northeast Siberia" by Josefine Walz et al. discusses the important issue of permafrost aggradation history and organic matter quality on greenhouse gas (CO2 and CH4) production from degrading yedoma deposits. The findings are based on short-term (134 days) and longer-term (785 days) incubation of samples collected at three locations, and the measured CO2 and CH4 production is linked to a wide array of measurements on geochemical characteristics and the stratigraphy of soil/sediment cores.

The potential future C release from thawing permafrost soils, especially yedoma, is connected to large uncertainties. Only a limited number of studies address this topic and I particularly value the authors' efforts to asses the longer-term production potential. This topic clearly is of interest to the broader scientific community and I thus consider this manuscript highly relevant for the journal. The manuscript is carefully written, however, it would benefit from some streamlining, especially of the results and discussion section as outlined in my comments below, in order to further improve readability and scientific value of the manuscript.

## Specific comments

1) First of all, the results section is rather detailed, partly repeating values presented as figures and reporting many numbers, making it difficult to follow. I would recommend providing part of the information as tables, e.g. an overview table with site names, site codes, ages, mean CO2 and CH4 production rates etc., helping the reader to get an overview of the differences between the three locations.

We added Tabel 2 as a summary table as suggested and removed some of the detailed numbers from the results section to improve readability.

Chapters 4.2, 4.3, 4.4 should be presented under a sub-heading, e.g. "greenhouse gas production".

Chapters 4.2, 4.3, 4.4 are now presented under the sub-heading, "Greenhouse gas production potentials" as suggested.

To improve readability, I would further recommend dropping the numbers in the site codes, e.g. just MUO, BK, and L instead of MUO12, BK8, L14.

We decided to keep the numbers in the site codes, because the same numbers and codes were used for the same sample material in other studies that are referenced in this study. Hence, keeping codes and numbers facilitates the comparison of data from different studies.

2) the conclusions drawn in the discussions are partly based on results obtained in this study, but also quite heavily rely on detailed analyses reported in previously published literature (radiocarbon age, plant macrofossils and soil microbial analyses), e.g. L285-303. I suggest to emphasize results measured within this study throughout the discussion. Additionally, section 5.1 of the discussion is rather lengthy and would benefit from some streamlining to more clearly emphasize the main results from this study.

We rewrote parts of Section 5.1 and 5.2 to shorten and streamline the discussion and to put more emphasis on the incubation results of this study, see. lines 285–299, lines 319–320, or 359–363.

3) The CO2 and CH4 production potential was assessed using separately incubated soil samples, excluding effects of vegetation (e.g. input of fresh OM to the soil system, atmospheric CO2 uptake), and processes among different layers in the soil profile (e.g. diffusion and leaching, as well as priming of old OM). How would the authors relate the gas production measured in these soil incubations to gas release to the atmosphere under in situ conditions? I would appreciate some more discussion on this part.

Post-thaw processes and the introduction of fresh organic are important points. We included some discussion of the priming effects on organic matter decomposability in section 5.1, L307-312.

## Technical corrections / line edits

Abstract L31: if more than 80% were produced during the first 134 of the long-term incubation, shouldn't that rather highlight the importance of the labile C pool, rather than the slowly decomposing C pool?

Yes, the labile pool is important for the production in the initial incubation. But this pool is very small. So over longer time scales, the slowly decomposing C pool will become relevant. In the abstract we now refer to the non-linearity of the decomposition processes and discuss the importance of fast vs slowly decomposing C in the discussion.

#### Introduction L40: give depth range for C stocks, is it 0-3m?

This is the combined C stock of soils, refrozen thermokarst and Holocene cover deposits in the top 3 m as well as sediments and deltaic deposits below 3 m. We added this information in the text for clarity.

L44: "The changes" is slightly vague, please specify.

We replaced "The changes" with "The effects of elevated atmospheric greenhouse gas concentrations and temperatures on processes in soils and sediments".

L54: what about MIS 4 and 6, are they not preserved in this region?

MIS4 deposits are preserved in the region, but not all MIS 4 deposits are ice complex deposits, e.g. in the Kuchchugui Suite on Bol'shoy Lyakhovsky. In the text, we added the information, that "at some locations the accumulation of Yedoma material may have already started between 80 and 60 ka BP, i.e. during MIS 4" (L 55–56). Dating constraints of older, non-yedoma ice complexes, make it difficult to differentiate if the deposition occurred during the MIS7 or early MIS 6. In the text, we added the possibility of ice complex formation during both late MIS 7/early MIS 6 and MIS 5 (L57-59).

L60: Either separate sentence by colon ":" or add reference.

We added a reference.

L62-64: Are those C stocks representative of the whole yedoma deposits, or a certain depth range?

Those C stocks are for the whole Yedoma domain. We clarified this in the text.

L66: Consider replacing "thawed out", e.g. "exposed by degradation of ice-rich permafrost" We changed "is thawed out from degrading ice-rich permafrost deposits" to "will be exposed by degradation of ice-rich permafrost" as suggested.

L67: decomposed to the greenhouse gases carbon dioxide (CO2) and methane (CH4) We changed the sentence as suggested.

L77: Do they authors mean "permafrost aggradation"?

Yes. We corrected this typo.

Methods L97: Consider replacing "modern" with "current"

We replaced "modern" with "current" as suggested.

L155: Might the low temperatures during storage (-18oC) have had an effect on soil microbial community functioning during the incubations? -11oC seems to be the minimum naturally occurring permafrost temperature in this region.

We do not expect that the storage temperature will considerably affect the soil microbial community functioning because -11 °C is the ground temperature at the level of zero amplitude, which is at about 20 meters depth. Above that point, temperatures will be lower in winter, with colder temperatures in the upper permafrost and the coldest temperatures in the active layer reaching values of below -30°C.

L164/165:

Some specifications about the gas sampling would be useful, e.g. how many mL of gas were sampled from the headspace for GC analysis? Did gas sampling cause underpressure in the headspace?

We always worked with slight overpressure. In individual cases of underpressure, which occasionally occurred in the longer incubations, we added 5-10 mL of N2 gas to reestablish overpressure in the bottle. We removed 1 mL of headspace gas for each individual GC measurement and corrected for the cumulative removed gas during sampling. We added this information in the Section 3.3.

L171-174: Was the temperature dependency of gas solubility taken into account? I would suggest to provide some more details on the solubility/temperature coefficients used for CO2 and CH4.

Carroll et al. (1991) and Yamamoto et al. (1976) provide temperature-dependent solubility for CO2 and CH4, respectively. In the text, we added the information "Solubility for CO2 and CH4 in water at 4 °C".

Results L236: "anaerobic CO2 production"?

Yes. We added "production"

L243: increased 30-fold over what time frame?

We added the "between 134 and 785 incubation days".

Discussion L282-284: This seems like an overall conclusion of the study and does not belong in the opening paragraph of the discussion

We moved this sentence to the conclusion section and replaced it here with a more appropriate introductory sentence.

L369-372: Using the term "longterm" for a period of ca. 2 years is slightly questionable, I advise some caution with the use of this term throughout the manuscript.

Were appropriate, we replaced "long-term" throughout the text.

Figures L675 (Fig. 3): adding both y-axes (height and depth) to each figure panel, as well as using the same x-axis scaling (e.g. 0-80?) would improve readability of the figure. Since CH4 production is included as a third panel for the other cores, please mention in figure caption why it is not included here. L684 (Fig. 4 and Fig. 5): Please mention AL thickness in figure caption.

We tried adding both v-axes to each figure panel in Fig. 3, 4, and 5, but this made the figures overly busy, so we decided to keep the y-axes as is. We added the AL thickness in figure captions 4 and 5.

Cited references:

Carroll, J. J., Slupsky, J. D. and Mather, A. E.: The solubility of carbon dioxide in water at low pressure, J. Phys. Chem. Ref. Data, 20(6), 1201, doi:10.1063/1.555900, 1991. Yamamoto, S., Alcauskas, J. B. and Crozier, T. E.: Solubility of methane in distilled water and seawater, J. Chem. Eng. Data, 21(1), 78-80, doi:10.1021/je60068a029, 1976.

Thank you for the helpful suggestions on this manuscript. Please find the comments (blue) and our reply (black) below.

Authors using the word "glacial" very often do not specify that they mean age but not the origin of the deposits they studied. It might confuse the readers who are not familiar with the paleoenvironmental conditions of the area of investigation. I suggest to use marine isotopic stages or regional stratigraphic units.

Where necessary, we changed "glacial" to refer only to age and not origin to avoid ambiguity, e.g. line 23 and line 274.

Lines 64 and 65. I would recommend to authors include in the review of the assessments of the carbon pools in different stratigraphic horizons research published by Shmelev et al (Shmelev, D., Veremeeva, A., Kraev, G., Kholodov, A., Spencer, R. G., Walker, W. S., & Rivkina, E. (2017). Estimation and Sensitivity of Carbon Storage in Permafrost of North-Eastern Yakutia. Permafrost and Periglacial Processes, 28(2), 379-390.).

We included this information as suggested in lines 69–72.

Line 95. It is not so important for this study, but permafrost temperature in this region varies with the topographic forms and consist of -9 within the thermokarst depressions and -10.5 at the yedoma hills (Kholodov, A., Gilichinsky, D., Ostroumov, V., Sorokovikov, V., Abramov, A., Davydov, S., & Romanovsky, V. (2012, June). Regional and local variability of modern natural changes in permafrost temperature in the Yakutian coastal lowlands, Northeastern Siberia. In Proceedings of the Tenth International Conference on Permafrost, Salekhard, Yamal-Nenets Autonomous District, Russia (pp. 25-29).)

Thank you for this reference. However, we agree that the temperature differences between topographic forms are not of utmost importance for the current study.

For the Results section, I also recommend authors to insert the graphs of dynamics of the greenhouse gases production during the experiment to give readers a better idea about dynamics of the process of organic matter decay.

Thank you for this important comment. We will deposit all the data of the current manuscript on PANGAEA (<u>https://doi.pangaea.de/10.1594/PANGAEA.892950</u>) so we decided not to insert the graphs of dynamics of the greenhouse gas production during the incubation of the samples in the manuscript. Since we incubated 117 individual samples, this cannot be done in a reasonable way.

Please find below a list of all relevant changes made in the manuscript:

- We added Table 2 as a summary table and removed some of the detailed numbers from the results section to improve readability.
- Chapters 4.2, 4.3, 4.4 are now presented under the sub-heading, "Greenhouse gas production potentials"
- We rewrote parts of Section 5.1 and 5.2 to shorten and streamline the discussion and to put more emphasis on the incubation results of this study, see. lines 285–299, lines 319– 320, or 359–363.
- We included some discussion of the priming effects on organic matter decomposability in section 5.1, lines 307-312.
- We added details about ice complex formation in lines 55-59.
- We added details about the incubation procedure in Section 3.2, lines 161-164.
- Where necessary, we changed "glacial" to refer only to age and not origin to avoid ambiguity, e.g. line 23 and line 274.
- We added information about carbon pools in lines 69–72.
- We deposited all data of the current manuscript on PANGAEA
   <u>https://doi.pangaea.de/10.1594/PANGAEA.892950</u>) and added this reference

# 1 Greenhouse gas production in degrading ice-rich permafrost deposits in northeast Siberia

2 Josefine Walz<sup>1,2</sup>, Christian Knoblauch<sup>1,2</sup>, Ronja Tigges<sup>1</sup>, Thomas Opel<sup>3,4</sup>, Lutz Schirrmeister<sup>4</sup>, Eva-Maria

3 Pfeiffer<sup>1,2</sup>

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<sup>5</sup> <sup>1</sup>Institute of Soil Science, Universität Hamburg, Hamburg, 20146, Germany

- <sup>6</sup> <sup>2</sup>Center for Earth System Research and Sustainability, Universität Hamburg, Hamburg, 20146, Germany
- <sup>7</sup> <sup>3</sup>Permafrost Laboratory, Department of Geography, University of Sussex, Brighton, BN1 9RH, UK
- 8 <sup>4</sup>Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research, Periglacial Permafrost
- 9 Research Section, 14473 Potsdam, Germany
- 10 Correspondence to: Josefine Walz (josefine.walz@uni-hamburg.de)
- 11

## 12 Abstract

- 13 Permafrost deposits have been a sink for atmospheric carbon for millennia. Thaw-erosional processes,
- 14 however, can lead to rapid degradation of ice-rich permafrost and the release of substantial amounts of
- organic carbon (OC). The amount of the OC stored in these deposits and their potential to be microbially decomposed to the greenhouse gases carbon dioxide ( $CO_2$ ) and methane ( $CH_4$ ) depends on climatic
- and environmental conditions during deposition and the decomposition history before incorporation into
- 18 the permafrost. Here, we examine potential greenhouse gas production in degrading ice-rich permafrost
- 19 deposits from three locations in the northeast Siberian Laptev Sea region. The deposits span a period
- 20 of about 55 kyr and include deposits from the last glacial period and Holocene interglacial periods.
- 21 Samples from all three locations were incubated under aerobically and anaerobically incubated
- 22 conditions for 134 days at 4 °C. Greenhouse gas production was generally higher in deposits from glacial
- 23 periods, where 0.2-6.1% of the initially available OC was decomposed to CO2. glacial than Holocene
- 24 deposits. In contrast, only 0.1-4.0% of initial OC In permafrost deposits from the were decomposed in
- 25 permafrost deposits from the Holocene and the late glacial transition., only 0.1-4.0% of the initially
- 26 available OC could be decomposed to CO<sub>2</sub>, while 0.2–6.1% could be decomposed in glacial deposits.
- Within the glacial deposits from the Kargin interstadial period (Marine Isotope Stage 3), local depositional environments, especially soil moisture, also affected the preservation of OC. Sediments deposited under
- environments, especially soil moisture, also affected the preservation of OC. Sediments deposited under
   wet conditions contained more labile OC and thus produced more greenhouse gases than sediments
- 29 wet conditions contained more labile OC and thus produced more greenhouse gases than sediments
- 30 deposited under drier conditions. To assess the long-termgreenhouse gas production potentials over
- 31 longer periods, deposits from two locations were incubated for a total of 785 days. However, more than

50% of total CO<sub>2</sub> production over 785 days occurred within the first 134 days under aerobic conditions 32 while even 80% were produced over the same period under anaerobic conditions, which emphasizes 33 the non-linearity of the OC decomposition processes of the aerobically produced and more than 80% of 34 anaerobically produced CO2 after 785 days of incubation were already produced within the first 134 days, 35 emphasizing the non-linearity of the decomposition processes. highlighting the quantitative importance 36 of the slowly decomposing OC pool in permafrost. CH<sub>4</sub> production Methanogenesis was generally 37 observed in active layer samples but only sporadically in permafrost samples and was several orders of 38 39 magnitude smaller than  $CO_2$  production. 40 Key words: Permafrost carbonthaw, greenhouse gasesCO2 and CH4, incubation, Yedoma, Siberian 41

- 42 Arctic
- 43

#### 44 **1** Introduction

Permafrost, i.e. ground that is at or below ≤0 °C for at least two consecutive years (van Everdingen, 45 2005), may preserve organic matter (OM) for millennia (Ping et al., 2015). The current organic carbon 46 (OC) pool of soils and sediments in permafrost-affected landscapes, soils, refrozen thermokarst, and 47 Holocene cover deposits in the top 3 m as well as sediments and deltaic deposits below 3 m in permafrost 48 landscapes is estimated to be about ~1300 Pg, of which about ~800 Pg are perennially frozen (Hugelius 49 et al., 2014). However, warming-induced environmental changes and permafrost degradation could lead 50 to rapid thaw of substantial amounts of currently frozen OM, microbial decomposition of the thawed 51 52 materialOM, and rising greenhouse gas fluxes to the atmosphere (Natali et al., 2015; Schuur et al., 2015). The changes effects of elevated atmospheric greenhouse gas concentrations and temperatures on 53 processes in soils and sediments are expected to be most pronounced in near-surface layers (Schneider 54 von Deimling et al., 2012). However,, but-thermo-erosion of ice-rich permafrost, i.e. permafrost with 55 >more than 20 vol% ice (Brown et al., 1998), also enables deep thaw of several tens of meters 56 (Schneider von Deimling et al., 2015). 57

Ice-rich permafrost deposits, also called ice complex deposits, accumulated in unglaciated Arctic lowlands. During cold stages, fine grained organic-rich material of polygenetic origin was deposited on predominantly flat plains (Schirrmeister et al., 2013). The deposits are dissected by large ice wedges, which can amount for up to 60\_vol% (Ulrich et al., 2014). The most prominent ice complex deposits, referred to as Yedoma, accumulated during the late Pleistocene between ><u>approximately</u>55 and 13 ka before present (BP), i.e. during the Marine Isotope Stages (MIS) 3 and 2 (Schirrmeister et al., 2011)<sub>T</sub>.
Age-depth correlations, however, indicate that at some locations the accumulation of Yedoma material
may have already started between 80– and 60 ka BP, i.e. during MIS 4 (Schirrmeister et al., 2002b).
Locally, however, remnants of older ice complex deposits of both late MIS 7/early MIS 6 or and MIS 5
age are also preserved (Opel et al., 2017; Schirrmeister et al., 2002a; Wetterich et al., 2016), but not
studied yet in terms of greenhouse gas production.

The thickness of Yedoma deposits in Siberia (Grosse et al., 2013) and Alaska (Kanevskiy et al., 2011) 69 70 can reach more than >50 m. At the time of deposition; rapid sedimentation and freezing incorporated relatively undecomposed OM into the permafrost (Strauss et al., 2017). However, owing to the high ice 71 content, Yedoma deposits are highly susceptible to warming-induced environmental changes, erosion, 72 73 and ground subsidence following permafrost thaw (e.g. Morgenstern et al., 2013). Only 30% of the Yedoma region (about ~416,000 km<sup>2</sup>) is considered intact, (Strauss et al., 2013) while the other 70% 74 have already undergone some level of permafrost degradation (Strauss et al., 2013)-(e.g.-Morgenstern 75 et al., 2013). Today, the whole Yedoma region domain stores 213-456 Pg of OC, of which 83-269 Pg 76 are stored in intact Yedoma and 169-240 Pg in thermokarst and refrozen taberal deposits (Hugelius et 77 al., 2014; Strauss et al., 2013, 2017; Walter Anthony et al., 2014; Zimov et al., 2006). For an about 78 88,000 km<sup>2</sup> large area along the Bolshaya Chukochya and Alazeya RrRiver basins and the eastern parts 79 of the Yana-Indigirka and Kolyma lowlands in northeast Siberia, Shmelev et al. (2017) estimate the size 80 of the total carbon pool in the upper 25 m to be 31.2 Pg, of which 3.7 Pg are stored in Yedoma deposits. 81 But<u>However</u>, high spatial and temporal variability result in large uncertainties about of how much OC will 82 be exposed by degradation of ice-rich permafrost is thawed out from degrading ice-rich permafrost 83 deposits and how much of this OC can be microbially decomposed to the greenhouse gases carbon 84 dioxide  $(CO_2)$  or methane  $(CH_4)$  after thaw. 85 In addition to the quantity of OCM, its decomposability will influence how fast the OC in Yedoma 86

permafrost deposits can be transformed into  $CO_2$  or  $CH_4$  after thaw (Knoblauch et al., 2018; MacDougall and Knutti, 2016). Since plants are the main source of OM in soils, vegetation composition plays an important role for OM decomposability (Iversen et al., 2015). Furthermore, OM has undergone different degradation processes before being incorporated into permafrost depending on permafrost formation pathways (Harden et al., 2012; Waldrop et al., 2010). In epigenetic permafrost, that is permafrost aggradation through intermittent freezing after the material was deposited, OM has already undergone some level of transformation and easily decomposable, labile OC compounds are decomposed and lost

to the atmosphere prior to incorporation into the permafrost (Hugelius et al., 2012). In contrast, OM in 94 95 syngenetically frozen Yedoma, i.e. concurrent material deposition and permafrost aggradationaggregation, had little time to be transformed prior to freezing and may thus contain high 96 amounts of labile OCM, which may be quickly decomposed to greenhouse gases after thaw (Dutta et 97 al., 2006). In this case, the amount and decomposability of the fossil OM is controlled by the OM source, 98 i.e. predominantly vegetation, which in turn depends on paleo-climatic conditions (Andreev et al., 2011). 99 The decomposability of permafrost OM is often assessed based on OM degradation proxies, total OC 100 101 (TOC) content, total organic carbon to- total nitrogen ratios (C/N), or stable carbon isotopes ( $\delta^{13}C_{org}$ ) with contradictory results (Strauss et al., 2015; Weiss et al., 2016). Only few studies have measured CO<sub>2</sub> and 102 CH<sub>4</sub> production potentials from Siberian Yedoma deposits under laboratory conditions (Dutta et al., 2006; 103 Knoblauch et al., 2013, 2018; Lee et al., 2012; Zimov et al., 2006). In this study, we present incubation 104 data from late Pleistocene Yedoma and Holocene interglacial deposits from three locations in northeast 105 Siberia. We hypothesize that OM deposited during glacial periods experienced little pre-freezing 106 transformation and thus provides a more suitable substrate for future microbial decomposition and 107 greenhouse gas production post-thawing than Holocene deposits. 108

109

### 110 2 Study region and sample material

Three locations in the Laptev Sea region in northeast Siberia were studied (Fig. 1). The whole region is 111 underlain by continuous permafrost reaching depths of 450-700 m onshore and 200-600 m offshore 112 (Romanovskii et al., 2004) with ground temperatures of -11 °C for terrestrial permafrost (Drozdov et al., 113 2005) and -1 °C for submarine permafrost (Overduin et al., 2015). Long, cold winters and short, cool 114 115 summers characterize the modern-current climate. Mean annual (1971-2000) temperatures and precipitation sums are -13.3 °C and 266 mm at the central Laptev Sea coast (Tiksi, WMO station 21824) 116 and -14.9 °C and 145 mm in the eastern Laptev Sea region (Mys Shalaurova, WMO station 21647, 117 Bulygina and Razuvaev, 2012). Modern vegetation cover is dominated by erect dwarf-shrub and in 118 places by sedge, moss, low-shrub wetland vegetation or tussock-sedge, dwarf-shrub, moss tundra 119 vegetation (CAVM Team, 2003). A compilation of the regional stratigraphic scheme used in this work 120 with paleoclimate and vegetation history is summarized in Table 1. 121

The first study location is on Muostakh Island (71.61° N, 129.96° E), an island in the Buor Khaya Bay 40 km east of Tiksi. Between 1951–2013, the area and volume of Muostakh Island, which is subject to major coastal erosion (up to -17 m a<sup>-1</sup>) and thaw subsidence, decreased by 24% and 40%, respectively (Günther et al., 2015). The entire sedimentary sequence of Muostakh Island (sample code MUO12) was
sampled in three vertical sub-profiles on the northeastern shore (Meyer et al., 2015). In the current study,
we used 14 sediment samples from the entire MUO12 sequence between 0.5–15.6 meters below surface
(mbs), which corresponds to19.5–4.4 meters above sea level (masl).

The second study location is on the Buor Khaya Peninsula (71.42° N, 132.11° E). Thermokarst 129 processes affect 85% of the region, which resulted in more than >20 m of permafrost subsidence in some 130 areas (Günther et al., 2013). Long-term (1969-2010) coastal erosion rates along the western coast of 131 the Buor Khaya Peninsula are ---about -1 m a<sup>-1</sup> (Günther et al., 2013). On top of the Yedoma hill, 132 approximately 100 m from the cliff edge, a 19.8 m long permafrost core (sample code BK8) was drilled 133 (Grigoriev et al., 2013). Detailed cryolithological, geochemical, and geochronological data (Schirrmeister 134 135 et al., 2017), palynological analysis (Zimmermann et al., 2017b), and lipid biomarker studies (Stapel et al., 2016) were previously published for the BK8 site. In the current study, 20 sediment samples spread 136 evenly between the surface and 19.8 mbs (or 34-to-14.2 masl) were analyzed, excluding an ice wedge 137 between 3.2-8.5 mbs. 138

The third sampling location is on Bol'shoy Lyakhovsky Island (73.34° N; 141.33° E), the southernmost 139 island of the New Siberian Archipelago. Four cores (sample code L14) were drilled on the southern coast 140 (Schwamborn and Schirrmeister, 2015). Core descriptions, geochronological data -as well as, and pollen 141 and plant DNA analyses can be found in Zimmermann et al. (2017a), while biomarkers and pore water 142 analysis can be foundare reported in Stapel et al. (2018). Based on previous stratigraphic studies from 143 144 this location (e.g. Andreev et al., 2009; Wetterich et al., 2009, 2014) we focused on two cores, which represent the here investigated MIS 1-MIS 3 period. The first core, L14-05, was recovered from inside 145 a thermokarst basin, 4 km west of the Zimov'e River mouth, with Holocene thermokarst deposits 146 overlying thawed and refrozen taberal Yedoma deposits (Wetterich et al., 2009). Five sediment samples 147 between 0–7.9 mbs (11.5–3.6 masl) were analyzed for the current study. The second core, L14-02, was 148 taken on a Yedoma hill about 1 km west of the Zimov'e River mouth. The entire core was 20.0 m long, 149 including wedge ice below 10.9 mbs. Five sediment samples from the top to a depth of 10.9 mbs (32.2-150 21.3 masl) were incubated for the current study. 151

152

#### 153 3 Methods

#### 154 **3.1. Dating**

Radiocarbon dating was performed on plant macro fossils for MUO12 (Meyer et al., unpublished data),
BK8 (Schirrmeister et al., 2017), and L14 samples (Zimmermann et al., 2017a) using the AMS facilities
of University of Poznan and Cologne University. Additionally, feldspars grains from the BK8 core at 12.6–
12.75 mbs, 16.0–16.35 mbs 18.5–18.7 mbs were dated by infrared-stimulated luminescence (IRSL)
(Schirrmeister et al., 2017).

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### 161 **3.2.3.1.** Geochemical characteristics

Gravimetric water contents were calculated as the weight difference between wet and dried (105 °C) 162 163 samples. pH values were measured in a suspension of 5 g thawed sediment in 12.5 ml distilled water (CG820, Schott AG, Mainz, Germany). For sediment chemical analyses, bulk samples were dried at 164 70°C and milled. Total carbon (TC) and total nitrogen (TN) contents were measured with an element 165 analyzer (VarioMAX cube, Elementar Analysensysteme GmbH, Hanau, Germany), while TOC contents 166 were measured with a liquiTOC II coupled to a solids module (Elementar Analysensysteme GmbH, 167 Hanau, Germany). The  $\delta^{13}C_{org}$ -values were measured with an isotope-ratio mass spectrometer (Delta V, 168 Thermo Scientific, Dreieich, Germany) coupled to an elemental analyzer (Flash 2000, Thermo Scientific, 169 Dreieich, Germany) after samples were treated with phosphoric acid to release inorganic carbon. 170

171

#### 172 **3.3.3.2.** Incubation

Frozen samples were slowly thawed from -18 °C to 4 °C over 48 h in a refrigerator, -and-homogenized 173 174 and divided into triplicates. Anaerobic incubations were prepared under a nitrogen atmosphere in a glove box. Approximately 15-30 g thawed sediment was weighed into glass bottles and sealed with rubber 175 stoppers. Anaerobic samples were saturated with 5-20 ml of nitrogen-flushed, CO2-free distilled water 176 and the headspace was exchanged with molecular nitrogen. The headspace of aerobic incubation bottles 177 was exchanged with synthetic air (20% oxygen, 80% nitrogen). We added enough molecular nitrogen 178 179 and synthetic air to establish a slight overpressure inside each bottle. In occasional cases of-negative pressure differences between headspace pressure and underpressureambient pressure, inside a bottle 180 over the course of the incubation, we added 5–10 mL of molecular nitrogen to reestablish overpressure. 181 Samples from all three study locations were incubated for 134-incubations days at 4 °C. During this 182 time, the headspace CO<sub>2</sub> and CH<sub>4</sub> concentrations were measured weekly to biweekly. The incubation of 183

samples from the Buor Khaya Peninsula and Bol'shoy Lyakhovsky Island continued until 785 days and 184 the gas concentrations were measured . The measuring intervals gradually decreased to every 8-12 185 weeks for the remaining incubation period. To determine the gGas concentrations inside each bottle, 1 186 mIL of headspace gas was removed by a syringe and injected into a were determined by a gas 187 chromatograph (GC 7890 Agilent Technologies, Santa Clara, USA) equipped with a 500 µL sample loop, 188 a nickel catalyst to reduce CO<sub>2</sub> to CH<sub>4</sub>, and a flame ionizing detector (FID). Gases were separated on a 189 PorapakQ column with helium as carrier gas. If the headspace concentration of CO<sub>2</sub> in aerobic incubation 190 191 bottles approached 3%, the headspace was again exchanged with synthetic air.

The amount of gas in the headspace was calculated from the concentration in the headspace, headspace volume, incubation temperature, and pressure inside the bottle using the ideal gas law. The amount of gas dissolved in water was calculated from the gas concentration in the headspace, pressure inside the bottle, water content, pH, and gas solubility. Solubility for  $CO_2$  and  $CH_4$  in water at  $4 \, ^\circ C$  was calculated after Carroll et al. (1991) and Yamamoto et al. (1976), respectively. To account for the dissociation of carbonic acid in water at different pH values, we used dissociation constants from Millero et al. (2007).

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## 200 **3.4.3.3** Statistics

Differences in mean values were analyzed with the Kruskal-Wallis test followed by multiple post-hoc Mann-Whitney tests with Bonferroni adjustment for multiple group comparisons. We tested for differences between deposits from different periods as well as for differences between deposits from the same period but from different locations. In both cases, the number of post-hoc comparisons was three, giving an adjusted significance level of 0.017. All statistical analyses were performed using MATLAB® (MATLAB and Statistics Toolbox Release 2015b, The MathWorks Inc., Natick, MA, USA).

207

#### 208 4 Results

## 209 4.1. Chronostratigraphy and geochemical characteristics

The sedimentary sequence on Muostakh Island was divided into three sections, which were separated by two erosional contacts and sharply intersecting ice wedges (Meyer et al., 2015). Based on radiocarbon ages (Meyer et al., unpublished data), these sections could be separated into three periods (Fig. 3). Deposits from the uppermost section between 0.5–2.4 mbs were classified as Holocene deposits from the MIS 1 and deposits from the late glacial to early Holocene transition, confirmed by radiocarbon ages of 7.5 and 13.2 ka BP for samples at 1.3 and 2.4 mbs, respectively. The middle section between 4–10 mbs yielded radiocarbon ages of 16.1–18.9 ka BP and were therefore classified as Sartan stadial deposits from the MIS 2. The lowermost section between 11.3–15.6 mbs yielded radiocarbon ages of 41.6–45.9 ka BP and represents the MIS 3 Kargin interstadial.

The BK8 core from the Buor Khaya Peninsula was subdivided into four sections (Fig. 4). The first 219 section between 0-0.5 mbs represents the seasonally thawed active layer. The subdivision of the 220 permafrost deposits below the active layer was based on previously published radiocarbon and infrared-221 222 stimulated luminescence (IRSL) ages (Schirrmeister et al., 2017). Deposits from the second section between 0.5–3.2 mbs yielded radiocarbon ages between 9.7–11.4 ka BP, which corresponds to the late 223 224 glacial transition to the early Holocene. The third section between 3.2-8.5 mbs consisted of an ice 225 wedge, which was not sampled for the current study. The fourth section between 8.5–18.9 mbs yielded infinite radiocarbon ages of ->older than 50 ka BP. The additional IRSL ages of feldspar grains yielded 226 deposition ages of about ~45 ka BP. Thus, sediments from this section were classified as deposits from 227 the Kargin interstadial. 228

The upper 0.5 m from both cores from Bol'shoy Lyakhovsky Island represent the active layer. 229 Radiocarbon ages of the L14-05 core from the thermokarst basin ranged between 2.2–10.1 ka BP for 230 the upper core section between 0-1.7 mbs and 51.2-54.6 ka BP for deposits below 5.8 mbs 231 (Zimmermann et al., 2017a). Based on these ages, stratigraphic interpretations from a nearby outcrop 232 (Wetterich et al., 2009), and the available palynological data (Zimmermann et al., 2017a), the L14-05 233 core was divided into two parts (Fig. 5). The upper part between 0-5.5 mbs were deposited during the 234 235 Holocene and late glacial transition, while deposits below 5.5 mbs was deposited duringoriginate from the Kargin interstadial. Deposits from the L14-02 core from the Yedoma hill yielded radiocarbon ages 236 between 33.1–62.8 ka BP, which corresponds to deposition during the MIS3 Kargin interstadial. 237

Overall, the permafrost deposits showed a wide range in TOC contents (0.8 – 6.3 wt%), C/N (4.6 – 239 29.4), and  $\delta^{13}C_{org}$  (-29.0 – -22.8 ‰VPDB, (Fig. 2). Generally higher TOC contents and C/N were found 240 in deposits from the Holocene and Kargin interstadial than in deposits from the Sartan stadial (Mann-241 Whitney test, p < 0.017), while the  $\delta^{13}C_{org}$ -values were significantly higher in Sartan stadial deposits 242 (Mann-Whitney test, p < 0.001).

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## 244 4.2. Greenhouse gas production potentials

## 245 **4.2.1.Muostakh Island**

Based on the TOC content, CO<sub>2</sub> production after 134 incubation days from sediment samples from the 246 MUO12 sequence ranged between 4.8–60.7 mg CO<sub>2</sub>-C g<sup>-1</sup> OC under aerobic conditions and 0.5–20.9 247 mg CO<sub>2</sub>-C g<sup>-1</sup> OC under anaerobic conditions (Fig. 3). Higher aerobic CO<sub>2</sub> production was generally 248 observed in the lowermost Kargin deposits between 11.3-15.6 mbs (Table 2) but elevated CO2 249 production rates werewas also observed at 1.6 mbs, 6 mbs, and 10 mbs. Under anaerobic conditions, 250 251 the highest production was observed at 6 mbs  $(19.3 \pm 1.4 \text{ mg CO}_2 - \text{C g}^4 - \text{OC})$ , which was nearly twice as high as in most other samples. No methanogenesis was observed in any Muostakh Island samples over 252 253 the 134-day incubation period.

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#### 255 4.2.2. Buor Khaya Peninsula

After 134 incubation days, CO<sub>2</sub> production in BK8 core samples ranged between 2.2–64.1 mg CO<sub>2</sub>-C g<sup>-1</sup> OC aerobically and 2.2–17.1 mg CO<sub>2</sub>-C g<sup>-1</sup> OC anaerobically (Fig. 4), which is within the same range as production in samples from Muostakh Island over the same incubation period (<u>Table</u>\_2). The highest production was observed in the active layer. Production then decreased sharply between 0.5–3.2 mbs but increased again in Kargin interstadial deposits below the ice-wedge. Methanogenesis was only observed in the active layer, but in much smaller quantity than anaerobic CO<sub>2</sub> production (0.37 ± 0.22 mg CH<sub>4</sub>-C g<sup>-1</sup> OC compared to 13.3 ± 3.6 mg CO<sub>2</sub>-C g<sup>-1</sup> OC).

To assess the long-term decomposability of OC over longer periods, all BK8 core samples were 263 incubated for a total of 785 days. After 785 incubation days, CO<sub>2</sub> production ranged between 4.6–131.1 264 mg CO<sub>2</sub>-C g<sup>-1</sup> OC under aerobic conditions and 2.2–43.0 mg CO<sub>2</sub>-C g<sup>-1</sup> OC under anaerobic conditions. 265 CO<sub>2</sub> production rates, however, decreased sharply within the first weeks of incubation. On average, 58 266  $\pm$  12% of the aerobically and 86  $\pm$  24% of the anaerobically produced CO<sub>2</sub> after 785 incubation days was 267 268 already produced within the first 134 days. In contrast, CH<sub>4</sub> production in the active layer increased 30fold between 134 and 785 incubation days to 11.4 ± 3.0 mg CH<sub>4</sub>-C g<sup>-1</sup>-OC. Additionally, two out of three 269 replicates at 10 mbs also showed active methanogenesis between 134 and after 785 days. The total 270 271  $CH_4$  production after 785 days accounted for 17 and 50% of the total carbon production in those samples, 272 respectively-(4.0 mg CH<sub>4</sub>-C g<sup>-4</sup> OC and 12.7 mg CH<sub>4</sub>-C g<sup>-4</sup> OC, respectively).

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#### 274 4.2.3. Bol'shoy Lyakhovsky Island

Aerobic CO<sub>2</sub> production after 134 incubation days in samples from the L14 cores ranged between 3.7– 275 18.9 mg CO<sub>2</sub>-C g<sup>-1</sup> OC (Fig. 5). The mean aerobic CO<sub>2</sub> production in all MIS 3 Kargin interstadial deposits 276 from Bol'shoy Lyakhovsky Island (9.2 ± 4.7 mg CO<sub>2</sub>-C g<sup>-1</sup> OC) was significantly lower (Mann-Whitney 277 test, p < 0.001) than CO<sub>2</sub> production in MIS 3 deposits from Muostakh Island  $(32.2 \pm 15.6 \text{ mg CO}_2 - \text{C g}^4)$ 278  $\frac{OC}{C}$  and the Buor Khaya Peninsula (Table 226.0 ± 12.6 mg CO<sub>2</sub>-C g<sup>-1</sup> OC). Anaerobic CO<sub>2</sub> production 279 in Kargin deposits from Bol'shoy Lyakhosvky Island ranged between 3.2–11.6 mg CO<sub>2</sub>-C g<sup>-1</sup> OC, which 280 281 was within the same range as production observed from the other two locations. No CH<sub>4</sub> production was observed in any L14 samples after 134 days. 282

After 785 incubation days, aerobic and anaerobic  $CO_2$  production ranged between 11.0–55.2 mg CO<sub>2</sub>-C g<sup>-1</sup> OC and 3.0–27.0 mg CO<sub>2</sub>-C g<sup>-1</sup> OC, respectively. Active methanogenesis was only observed in two2 out of three3 replicates from the active layer from the L14-05 core. <u>However, (0.41 mg CH<sub>4</sub>-C g<sup>-1</sup>)</u> <sup>4</sup>-OC and 0.63 mg CH<sub>4</sub>-C g<sup>-1</sup> OC). CH<sub>4</sub> production was therefore an order of magnitude lower than anaerobic CO<sub>2</sub> production in the same sample (5.7 mg CO<sub>2</sub>-C g<sup>-1</sup> OC and 4.6 mg CO<sub>2</sub>-C g<sup>-4</sup> OC) and also an order of magnitude smaller than CH<sub>4</sub> production in the active layer from the Buor Khaya Peninsula.

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## 291 4.3. Decomposability of permafrost OM deposited under different climatic regimes

Overall, permafrost OM deposited during the MIS 3 Kargin interstadial supported the highest greenhouse 292 gas production (Fig. 6). After 134 days of aerobic incubation, 0.2–6.1% of the initially available OC (mean 293  $2.3 \pm 1.4\%$ ) was decomposed to CO<sub>2</sub>. This was significantly more (Mann-Whitney test, p < 0.001) than 294 in deposits from the Holocene and late glacial transition, where production ranged between 0.4-4.0% 295 296 (mean 1.2  $\pm$  0.8%). The aerobic CO<sub>2</sub> production in MIS 2 Sartan stadial deposits ranged between 0.5– 4.2% (mean 1.7  $\pm$  1.2 %). Anaerobically, 3.3 times less CO<sub>2</sub> was produced (Pearson correlation 297 298 coefficient r = 0.63, p < 0.001). The lowest production was observed in Holocene and late glacial transition deposits, where 0.1–1.1 % of the OC was anaerobically decomposed to  $CO_2$  (mean 0.5 ± 0.3). 299 This was significantly less (Mann-Whitney test, p < 0.01) than in <u>Yedoma-glacial</u> deposits, where 0.4– 300 301 2.1% (mean 0.9  $\pm$  0.5%) and 0.2–1.6 % of initial OC (mean 0.7  $\pm$  0.3%) were decomposed in Sartan stadial and Kargin interstadial deposits, respectively. 302

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### 304 **5 Discussion**

## 305 5.1. Organic matter decomposability

The ice-rich permafrost deposits of Muostakh Island, the Buor Khaya Peninsula, and Bol'shoy 306 Lyakhovsky Island are typical for northeast Siberia and the geochemical OM characteristics (TOC, C/N, 307  $\delta^{13}C_{ord}$ ) were all within the range of other permafrost deposits in the region (Schirrmeister et al., 2011). 308 However, a better understanding of the differences in OM decomposability is needed to estimate the 309 310 future-contribution of thawing permafrost landscapes to future greenhouse gas fluxes. We hypothesized, 311 that the climatic conditions during deposition affected the amount and decomposability of preserved OM and thus greenhouse gas production potentials after thaw. OM decomposability in degrading ice-rich 312 permafrost therefore needs to be interpreted against the paleo-environmental background. 313

- The highest CO<sub>2</sub> production potentials from permafrost samples in the BK8 core were observed below 314 315 the ice wedge between 8.35-16 mbs (Fig. 3). For this core section, which was deposited during the MIS 3 Kargin interstadial (Schirrmeister et al., 2017), (Zimmermann et al., (2017b) report a high taxonomic 316 richness of vascular plants with high proportions of swamp and aquatic taxa, pointing towards a water-317 saturated environment at the time of deposition, likely a low-centered ice-wedge polygon. Furthermore, 318 (Stapel et al., (2016) report high concentrations of branched glycerol dialkyl glycerol tetraether (br-319 (GDGT), a microbial membrane compound, at 10 mbs, 11.2 mbs, and 15 mbs, indicative of a soil microbial 320 community, which developed when the climate was relatively warm and wet. Together with higher TOC 321 contents at these depths, this suggests accumulation of relatively undecomposed OM under anaerobic 322 conditions, which can be quickly decomposed after thaw (de Klerk et al., 2011), resulting in higher CO2 323 324 production. One way to analyze the OM source in more detail is sedimentary ancient DNA (sedaDNA), 325 which can be used to reconstruct local plant communities and infer predominant climatic conditions (Willerslev et al., 2004). In the BK8 core, a total of 134 vascular plants and 20 bryophytes were identified 326 327 (Zimmermann et al., 2017b). Salix, Poaceae and Cyperaceae, whose roots are a main OM source in tundra soils (lversen et al., 2015), are present throughout the core. The taxonomic richness was highest 328 between 8.35–16 mbs, where also high CO<sub>2</sub> production was observed. This core section, which belongs 329 to the MIS-3 Kargin interstadial (Schirrmeister et al., 2017), was dominated by swamp and aquatic taxa, 330 pointing towards a water-saturated environment, likely a low-centered ice-wedge polygon (Zimmermann 331 et al., 2017b). Together with higher TOC contents at these depths, this suggests accumulation of 332 relatively undecomposed OM under anaerobic conditions, which can be quickly decomposed after thaw 333 (de Klerk et al., 2011). Furthermore, high concentrations of branched glycerol dialkyl glycerol tetraether 334
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(br-GDGT), a microbial membrane compound, are indicative of a soil microbial community, which 335 developed when the climate was relatively warm and wet (Stapel et al., 2016). Overall, br-GDGT 336 concentrations were highest at 10 mbs, 11.2 mbs, and 15 mbs (Stapel et al., 2016), which corresponds 337 to the same levels where the highest CO<sub>2</sub> production was observed. In contrast, lower abundance of 338 swamp taxa and higher abundance of terrestrial taxa at 8.8 mbs and >below 15 mbs (Zimmermann et 339 al., 2017b), suggest that intermittently drier conditions existed. This resulted in accelerated OM 340 decomposition under aerobic conditions prior to OM incorporation into the permafrost and therefore lower 341 342 TOC contents as well as lower  $CO_2$  production potentials at these depths as overserved in this study.

Sediments above the ice-wedge in the BK8 core showed similar TOC contents, C/N, and  $\delta^{13}C_{org}$ -343 values compared to the rest of the core, but CO<sub>2</sub> production was consistently low in this section. This ~3 344 m long core section yielded radiocarbon ages of 11.4-10.1 ka BP (Schirrmeister et al., 2017), which 345 corresponds to the late glacial-early Holocene transition. After the Last Glacial Maximum (LGM), 346 temperatures were favorable for increased microbial decomposition of active layer OM, which led to the 347 preservation of comparatively stable OM fractions after the material was incorporated into the 348 permafrost. Similar conclusions can be drawn for Holocene deposits in thermokarst landforms or on top 349 350 of Yedoma deposits. On the one hand, they received fresh OM inputs, which explains their relatively high TOC contents (Strauss et al., 2015). On the other hand, intensive thermokarst development during the 351 352 late glacial transition and the early Holocene likely resulted in higher decomposition rates in thawed soils and the loss of labile OM compounds before the sediments were refrozen when climate conditions 353 354 deteriorated after the Holocene climate optimum. If these sediments were to thaw again in the future, 355 results from the current study suggest that the decomposability of the remaining OM will be comparatively 356 low. However, deeper rooting, cryoturbation, and post-thaw leaching of labile OM from vegetation could stimulate the decomposition and greenhouse gas production from the more stable OM through positive 357 priming (Fontaine et al., 2007). Both the chemical structure (Di Lonardo et al., 2017) and the frequency 358 of labile organic matterOM inputs (Fan et al., 2013) influence the size of the priming effect. For permafrost 359 soils, it has also been shown, that the priming effect is larger at lower temperatures (Walz et al., 2017). 360 Although-Thus, climatic conditions influence the vegetation composition and OM source on a regional 361 level, but the local depositional environment as well as post-depositional processes likely also control 362 the amount and decomposability of the OM that is presently incorporated into permafrost. 363 364 First results of *in situ* CO<sub>2</sub> fluxes from Muostakh Island were published by Vonk et al. (2012). Based

365 on the downslope decrease in OC contents, ‡they estimate that 66% of the thawed-out Yedoma OC

from degrading ice-rich permafrost deposits can be decomposed to greenhouse gasesCO<sub>2</sub> and released 366 back to the atmosphere before the material is reburied in the Laptev Sea. This is an order of magnitude 367 more than what the results from current incubation study suggest, where after 134 days only 0.4-6.0% 368 of the Yedoma OC from Muostakh Island were aerobically decomposed to CO<sub>2</sub>. However, nNo further 369 detailed palynological or microbial biomarker studies are yet available for the MUO12 sequence. The 370 closest reference locations is the comprehensive permafrost record at the Mamontovy Khayata section 371 on the Bykovsky Peninsula (Andreev et al., 2002; Sher et al., 2005). Between 58- and 12 ka BP 372 373 (Schirrmeister et al., 2002b), fine-grained material accumulated on the large flat foreland plain of the 374 today Bykovsky Peninsula area that was exposed at a time of lower sea level (Grosse et al., 2007). Sea 375 level rise after the last glacial period, coastal erosion, and marine ingression of thermokarst basins 376 formed the Buor Khaya Bay and eventually separated Muostakh Island from the Bykovsky Peninsula (Grosse et al., 2007; Romanovskii et al., 2004). Today, the distance between the northern tip of Muostakh 377 Island and the southern tip of the Bykovsky Peninsula is about 16 km. Between 58-12 ka BP 378 (Schirrmeister et al., 2002), fine-grained material accumulated on the large flat foreland plain of the today 379 380 Bykovsky Peninsula area that was exposed at a time of lower sea level (Grosse et al., 2007). The 381 distance between Muostakh Island and the Buor Khaya Peninsula is about 80 km. It is therefore likely that the deposition regimes on Muostakh Island and the Buor Khaya Peninsula were similar to the regime 382 at the Bykovsky Peninsula. This conclusion is also supported by similar OM decomposability. After 134 383 incubation days, the amount of aerobic and anaerobic CO2 production did not differ significantly (Mann-384 Whitney test, p = 0.339) between MIS 3 Kargin deposits from Muostakh Island  $(3.2 \pm 1.6\% \text{ of initial OC})$ 385 aerobically and 0.7 ± 0.6% anaerobically) and the BK8 coreBuor Khaya Peninsula (Table 22.7 ± 1.2% 386 aerobically and  $0.8 \pm 0.3$  % anaerobically), which suggests that the deposits formed under similar 387 388 conditions. In contrast, Under aerobic conditions, CO<sub>2</sub> production of in MIS 3 deposits from Bol'shoy Lyakhovsky Island in the eastern Laptev Sea was nearly three times lower (0.9 ± 0.5% of the initial OC 389 390 after 134 days) than observed for Muostakh Island and the Buor Khaya Peninsula in the central Laptev Sea. Considerably lower temperatures and precipitation characterize the modern-current climate on 391 Bol'shoy Lyakhovsky Island. It is also likely that regional differences between the eastern and central 392 Laptev Sea region would have affected the paleo-climate (Anderson and Lozhkin, 2001; Lozhkin and 393 Anderson, 2011; Wetterich et al., 2011, 2014). Different summer temperatures, precipitation, thaw depth, 394 and vegetation composition could explain regional differences in OM quantity and decomposability. 395 Interestingly, the differences in the amount of OC that was aerobically decomposed were mostly due to 396

397 differences in the initial CO<sub>2</sub> production rates. Maximum CO<sub>2</sub> production rates during the first weeks of incubation of Muostakh Island and Buor Khaya deposits were up to four times higher than in deposits 398 399 from Bol'shoy Lyakhovsky Island. However, long-term production rates after >130 incubation days did no longer differ considerably between the different locations (median 23.3 µg CO<sub>2</sub>-C g<sup>-1</sup>-OC d<sup>-1</sup>). These 400 rates are within the range of other long-term production rates from Yedoma deposits in northeast Siberia 401 (Dutta et al., 2006; Knoblauch et al., 2013) and Alaska (Lee et al., 2012). Considering the large slowly 402 decomposing permafrost OC pool (Schädel et al., 2014), long-term decomposition rates are likely to 403 404 provide more reliable projections of future greenhouse gas emissions from degrading permafrost 405 landscapes.

406 A distinctive feature of the Muostakh Island sequence is the preservation of MIS 2 Sartan deposits, 407 which are only sparsely preserved in northeast Siberia (Wetterich et al., 2011). Interestingly, mean aerobic CO<sub>2</sub> production in Sartan deposits from Muostakh Island was lower than in Kargin deposits, but 408 slightly higher under anaerobic conditions, but the difference was not statistically significant (Mann-409 Whitney test, p = 0.205). The rapid deposition of 8 m thick comparatively coarse-grained material in just 410 a few thousand years between 20 and -16 ka BP were unfavorable for the development of a stable land 411 surface and the establishment of a vegetation cover comparable to the Kargin interstadial or Holocene 412 periods (Meyer et al., unpublished data). Pollen analysis from the corresponding sections on the 413 Bykovsky Peninsula (Andreev et al., 2002) and Kurungnakh Island in the Lena River Delta (Schirrmeister 414 et al., 2008; Wetterich et al., 2008) suggest relatively cold and dry summer conditions during this stadial 415 with sparse vegetation. Relatively undecomposed OM was quickly buried, before it could be transformed 416 to greenhouse gases. 417

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## 419 5.2. Long-term production potentialsMulti-annual incubation

The 785-day incubation of permafrost samples from the Buor Khaya Peninsula and Bol'shoy 420 421 Lyakhovsky Island revealed Long-term greenhouse gas production measurements after 785 days showed that 51% of the aerobically and 83% of the anaerobically produced CO<sub>2</sub> were already produced 422 within the first 134 incubation days, highlighting the non-linearity of OM decomposition dynamics 423 (Knoblauch et al., 2013; Schädel et al., 2014) and the importance of the labile OC pool in short term 424 incubations. Maximum CO<sub>2</sub> production rates were generally reached within the first 100 incubation days. 425 After the initial peak, CO<sub>2</sub> prouction rates remained consitently low (median 23.3 µg CO<sub>2</sub>-C g<sup>-1</sup> OC d<sup>-1</sup> 426 aerobically and 3.2 µg CO2-C g<sup>-1</sup> OC d<sup>-1</sup> anaerobically). These rates are within the range of other multi-427

428 <u>annual production rates from Yedoma deposits in northeast Siberia (</u>Dutta et al., 2006; Knoblauch et al.,
 429 2013) <u>and Alaska (Lee et al., 2012).</u>

Assuming no new input of labile OM (e.g. from modern-the current vegetation), decomposition rates 430 are likely to remain low after the labile pool is depleted. Short-term greenhouse gas production and 431 release from thawing ice-rich permafrost will therefore mainly depend on the size of the labile pool. A 432 synthesis study of several incubations studies from high-latitude soils, including Yedoma deposits, 433 estimated the size of the labile OC pool to be generally less than <5% of the TOC (Schädel et al., 2014). 434 435 For Yedoma deposits on nearby Kurungnakh Island in the Lena River delta, Knoblauch et al. (2013) 436 estimated the size of the labile pool to be even smaller (less than <2%). -Considering the large slowly decomposing permafrost OC pool (Schädel et al., 2014), long-term decomposition rates are therefore 437 438 likely to provide more reliable projections of future greenhouse gas emissions from production in degrading permafrost landscapes. 439

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## 442 **5.3. Methanogenesis**

CH<sub>4</sub> production from Yedoma deposits, or the lack thereof, is a highly controversial topic in permafrost 443 research (Knoblauch et al., 2018; Rivkina et al., 1998; Treat et al., 2015). In the current work, active 444 methanogenesis was only observed in the active layer and 2two out of 38 Yedoma samples from the 445 BK8 core. , but only after a long lag-phase. Within 134 incubation days, no samples from Muostakh 446 Island produced any CH4... In those samples showing active methanogenesis, CH4 production continued 447 to rise over the 785 incubation days, which is in contrast to anaerobic CO<sub>2</sub> production, which decreased 448 with increasing incubation time. Rising CH<sub>4</sub> production rates indicate that methanogenic communities 449 450 still grow in these samples and were not limited by substrate supply. Chemical pore water and bulk sediment analyses from the BK8 core showed that there are high concentrations of both free and OM-451 452 bound acetate present in Yedoma deposits, indicating a high substrate potential for methanogenesis (Stapel et al., 2016). Knoblauch et al. (2018) showed that the small contribution of methanogenesis to 453 overall anaerobic permafrost OM decomposition found in short-term incubation studies (Treat et al., 454 2015) is due to the absence of an active methanogenic community. On a multi-annual timescale, 455 methanogenic communities become active and equal amounts of CO<sub>2</sub> and CH<sub>4</sub> are produced from 456 permafrost OM under anaerobic conditions. Under future climate warming and renewed thermokarst 457 activity, high levels of CH<sub>4</sub> production can locally be expected, but depend on favorable conditions such 458

as above-zero temperatures and anaerobic conditions. It can be expected that the development of an
active methanogenic community, e.g. by growth or downward migration of modern methanogenic
organisms, will lead to elevated long-term CH<sub>4</sub> production in these deposits (Knoblauch et al., 2018).

## 463 **6** Conclusion

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In this study, we investigated greenhouse gas production potentials in degrading ice-rich permafrost 464 deposits from three locations in northeast Siberia. We hypothesized, that the climatic conditions during 465 deposition affected the amount and decomposability of preserved OM and thus greenhouse gas 466 production potentials after thaw. OM decomposability in degrading ice-rich permafrost therefore needs 467 468 to be interpreted against the paleo-environmental background. It could be shown that Yedoma deposits 469 generally contained more labile OM than Holocene deposits. However, in addition to the regional climate conditions at the time of OM deposition, local depositional environments also influenced the amount and 470 decomposability of the preserved fossil OM. Within the deposits of the MIS 3 Kargin interstadial, 471 sediments deposited under wet and possibly anaerobic conditions produced more CO<sub>2</sub> than sediments 472 deposited under drier aerobic conditions. Further, deposits from the central Laptev Sea region produced 473 2-3 times more CO<sub>2</sub> than deposits from the eastern Laptev Sea region. It is therefore likely, that OM 474 decomposability of the vast Yedoma landscape cannot be generalized solely based on the stratigraphic 475 position. Furthermore, it is expected that CH<sub>4</sub> production will play a more prominent role after active 476 methanogenic communities have established since abundant substrates for methanogenesis were 477 present. 478

479

## 480 Data availability

All shown data sets as well as the temporal evolution of CO<sub>2</sub> and CH<sub>4</sub> production over the whole incubation period is available at https://doi.pangaea.de/10.1594/PANGAEA.892950 (Walz et al.,

483 2018).https://www.pangaea.de/ (follows after acceptance and includes all shown datasets)

484

#### 485 Author contributions

JW and CK designed the study. TO collected sediment samples on Muostakh Island and LS collected cores from the Buor Khaya Peninsula and Bol'shoy Lyakhovsky Island. JW and RT performed the laboratory analyses, with guidance from CK and EMP. JW performed data analyses and wrote the manuscript with contributions from all authors.

# 491 Acknowledgements

492 This research was supported by the German Ministry of Education and Research as part of the projects CarboPerm (grant no. 03G0836A, 03G0836B) and KoPf (grant no. 03F0764A). We further acknowledge 493 the financial support through the German Research Foundation (DFG) to EMP and CK through the 494 Cluster of Excellence "CliSAP" (EXC177), University Hamburg and to TO (grant OP 217/3-1). We also 495 thank the Russian and German participants of the drilling and sampling expeditions, especially Mikhail 496 497 N. Grigoriev (Melnikov Permafrost Institute, Yakutsk), Hanno Meyer and Pier Paul Overduin (both Alfred-Wegener-Institute, Potsdam). Additional thanks go to Georg Schwamborn (Alfred-Wegener-Institute, 498 Potsdam) for his assistance with core subsampling and Birgit Schwinge (Institute of Soil Science, 499 500 Hamburg) for her help in the laboratory. We are also immensely grateful for the two anonymous reviews on a previous version of this manuscript. 501

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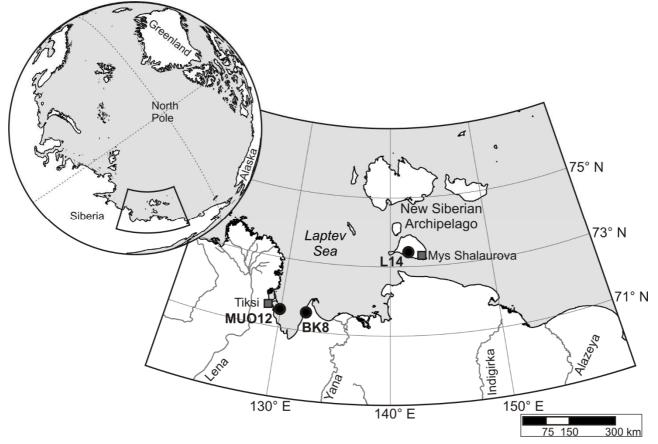
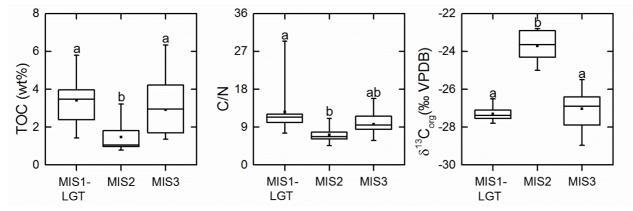


Figure 1. Overview map of the Laptev Sea region with the study locations at Muostakh Island (sample code MUO12), the Buor Khaya Peninsula (BK8) and Bol'shoy Lyakhovsky Island (L14).



755 Figure 2. Boxplot of total organic carbon (TOC), total organic carbon to total nitrogen ratio (C/N) and  $\delta^{13}C_{ord}$ -values of permafrost deposits from the MUO12 sequence, the BK8 core, and the two L14 cores 756 from the Holocene interglacial (MIS 1), including the late glacial transition (LGT) (n = 12), the Sartan 757 stadial (MIS 2) (n = 6), and the Kargin interstadial (MIS 3) (n = 27). The whiskers show the data range 758 and the box indicates the interquartile range. The vertical line and square inside the boxes show the 759 median and mean, respectively. The letters above the whiskers indicate statistically significant 760 differences in geochemical characteristics between the deposits of different ages (Mann-Whitney test, p 761 < 0.016 for TOC and C/N, p < 0.001 for  $\delta^{13}C_{org}$ ) 762

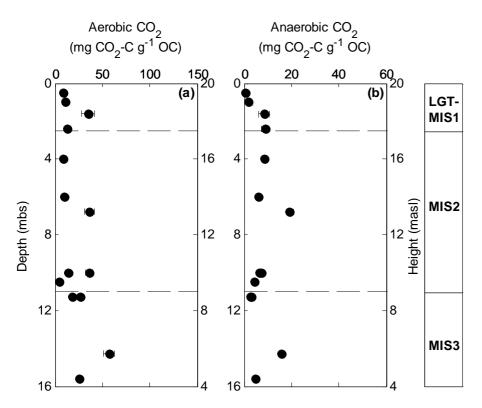




Figure 3. Depth profiles of total aerobic (a) and anaerobic (b)  $CO_2$  production per gram organic carbon (g<sup>-1</sup> OC) in sediment samples from the MUO12 sequence after 134 incubation days at 4 °C for deposits from the Holocene interglacial (MIS 1), including the late glacial transition (LGT), the Sartan stadial (MIS 2), and the Kargin interstadial (MIS 3). Data are mean values (n = 3) and error bars represent one standard deviation. Note the different scales. No CH<sub>4</sub> production was observed during the 134-days incubation period.



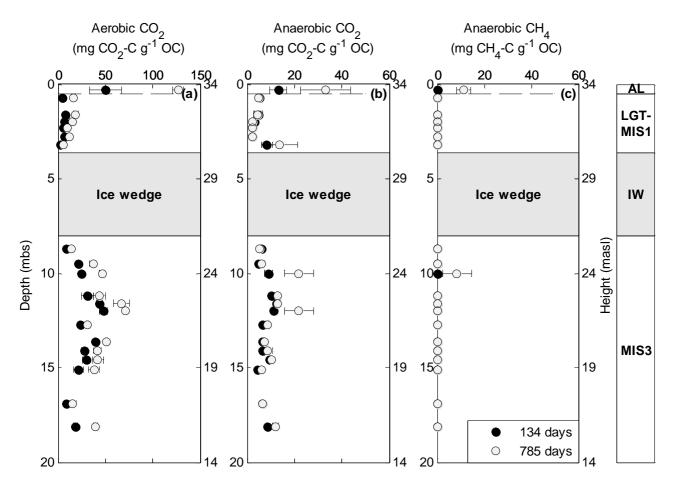


Figure 4. Depth profiles of total aerobic  $CO_2$  (a), anaerobic  $CO_2$  (b) and anaerobic  $CH_4$  (c) production per gram organic carbon (g<sup>-1</sup> OC) in sediment samples from the BK8 core after 134 (closed symbols) and 785 incubation days (open symbols) at 4 °C for the active layer (AL), which is considered to be 0.5 <u>m thick</u>, and permafrost deposits from the Holocene interglacial (MIS 1), including the late glacial transition (LGT) and the Kargin interstadial (MIS 3). Data are mean values (n = 3) and error bars represent one standard deviation. Note the different scales.

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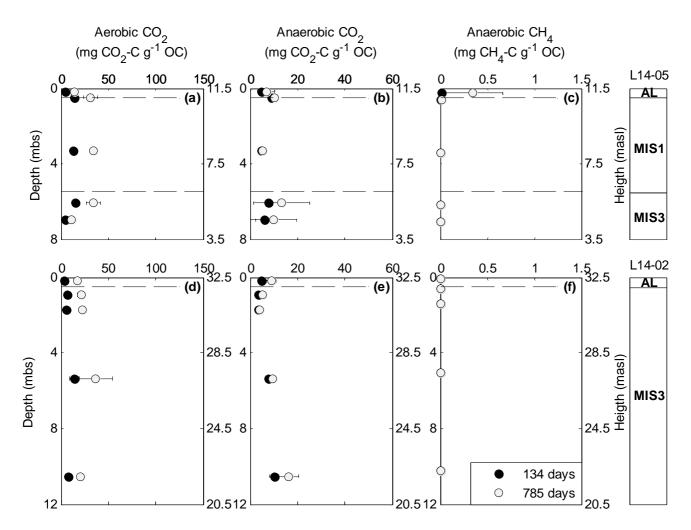


Figure 5. Depth profiles of total aerobic  $CO_2$  (a), anaerobic  $CO_2$  (b) and anaerobic  $CH_4$  (c) production per gram organic carbon (g<sup>-1</sup> OC) in sediment samples from the L14-05 (a,b,c), and L14-02 cores (d,e,f) after 134 (closed symbols) and 785 incubation days (open symbols) at 4 °C for the active layer (AL), which is considered to be 0.5 m thick, and permafrost deposits from the Holocene interglacial (MIS 1) and the Kargin interstadial (MIS 3). Data are mean values (n = 3) and error bars represent one standard deviation. Note the different scales.



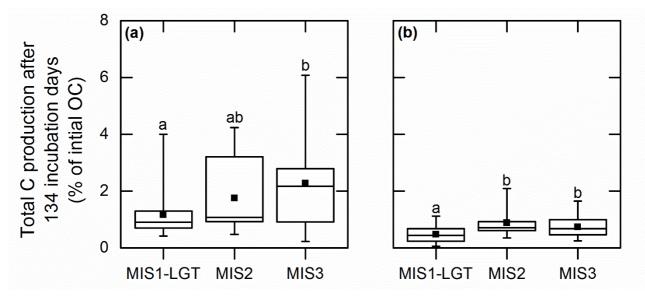


Figure 6. Total aerobic (a) and anaerobic (b)  $CO_2$ -C production after 134 incubation days from permafrost deposits from the MUO12 sequence, the BK8 core, and the two L14 cores from the Holocene interglacial (MIS 1), including the late glacial transition (LGT) (n = 22), the Sartan stadial (MIS 2) (n = 15), and the Kargin interstadial (MIS 3) (n = 50). The whiskers show the data range and the box indicates the interquartile range. The vertical line and square inside the boxes show the median and mean, respectively. The different letters indicate significant differences (Mann-Whitney test, p < 0.016) between deposits from different periods.

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# 797 Tables

Table 1. Compilation of the regional chronostratigraphy of the Laptev Sea region used in this work with paleoclimate (summer) and vegetation history based on an overview by Andreev et al. (2011) and references therein.

800										
	Age	Period		Regional chrono- stratigraph	• •				n	
	ka BP									
	<10.3	Holocene		Holocene	MIS 1	Holoce	Climate amelioration during the early Holocene; shrub-tundra vegetation gradually disappeared ca 7.6 ka BP			
	ca 10.3– 13	0,				Maxim	Climate amelioration after the Last Glacial Maximum; transition to shrubby tundra vegetation			
	ca 13–30	ca 13–30 Late Weichselian glacial (stadial)		Sartan	MIS 2		Cold and dry summer conditions, winter colder than today; open tundra steppe			
	ca 30–55	Middle Weichselian (interstadial)	•	Kargin I	MIS 3		Relatively warm and wet summers; open herb and shrub dominated vegetation			
801		(								
802 803 804	Production a	after 785 day	<u>ample</u> <u>n</u>	not determin material from <u>Aerobic (</u> <u>product</u> (mg CO <sub>2</sub> -C	ned (n.d.) for <u>n the Buor Kh</u> <u>CO<sub>2</sub> <u>ion</u> g<sup>-1</sup> OC)</u>	the Muosta naya Penins Anaerob produ	oduction after 134 and 785 incubation daysMuostakh Island sequence. MIS 2 deposita Peninsula and Bol'shoy Lyakhovsky IslandAnaerobic CO2Anaerobic CH4productionproductiong CO2-C g <sup>-1</sup> OC)(mg CH4-C g <sup>-1</sup> OC)days785 days134 days785 days			
	Muostakh	Active layer	Not s	ampled		-				
	<u>Island</u>	MIS 1	<u>12</u>	17.0 ± 11.9	<u>n.d.</u>	<u>4.9 ± 4.2</u>	<u>n.d.</u>	<u>0</u>	<u>n.d.</u>	
	<u>(MUO12)</u>	<u>MIS 2</u>	<u>17</u>	17.4 ± 12.9	<u>n.d.</u>	<u>8.8 ± 5.2</u>	<u>n.d.</u>	<u>0</u>	<u>n.d.</u>	
		<u>MIS 3</u>	<u>12</u>	<u>32.2 ± 15.6</u>	<u>n.d.</u>	<u>6.6 ± 5.6</u>	<u>n.d.</u>	<u>0</u>	<u>n.d.</u>	
		Active layer	<u>3</u>	50.2 ± 16.9	<u>126.8 ± 6.0</u>	<u>13.3 ± 3.5</u>	<u>33.1 ± 10.6</u>	<u>0.4 ± 0.2</u>	<u>11.4 ± 3.0</u>	
	Peninsula	<u>MIS 1-LGT</u>	<u>15</u>	<u>6.1 ± 1.8</u>	<u>13.4 ± 4.5</u>	<u>4.1 ± 2.2</u>	<u>4.5 ± 4.1</u>	<u>0</u>	<u>0</u>	
	<u>(BK8)</u>	<u>MIS 2</u>	Not p	resent in the	core materia	al				
		<u>MIS 3</u>	<u>38</u> 2	<u>27.2 ± 11.7</u>	<u>42.2 ± 15.9</u>	<u>7.9 ± 2.5</u>	<u>11.0 ± 5.8</u>	<u>0</u>	<u>0.4 ± 2.1 *</u>	
	Bol'shoy	Active layer	<u>6</u>	<u>4.3 ± 0.6</u>	<u>15.9 ± 2.8</u>	<u>5.0 ± 1.3</u>	<u>8.0 ± 2.7</u>	<u>0</u>	<u>0.2 ± 0.3 *</u>	
	Lyakhovsky	MIS 1-LGT		<u>13.8 ± 2.4</u>	<u>32.6 ± 4.8</u>		<u>7.8 ± 2.9</u>	<u>0</u>	<u>0</u>	
	<u>Island</u> (L14)	I) <u>MIS 2</u> <u>Not present in the core material</u>								
		<u>MIS 3</u>	<u>18</u>	<u>9.2 ± 4.7</u>	<u>24.7 ± 11.1</u>		<u>9.7 ± 7.0</u>	<u>0</u>	<u>0</u>	
805	* Methanog	<u>enesis was o</u>	only ob	served in two	o out of three	e replicates				