

Authors reply by Loeka Jongejans, on behalf of all authors (loeka.jongejans@awi.de)

We thank this referee for the positive feedback on our manuscript and their constructive comments. We will revise our manuscript according to your suggestions. Below, we addressed and replied to all the suggestions and questions that were raised.

Referee Comment (RC): This manuscript is a characterization of different deposit types in a permafrost landscape in Western Alaska. The study is very sound and provides the necessary level of detail to be useful to the research community. The results from this study are a good addition to existing datasets and the authors do a nice job putting the results from Baldwin Peninsula in a larger context within the permafrost zone.

Authors Reply (AR): We thank the referee for their positive feedback and for acknowledging the importance of our study.

RC: A simple but crucial fix would be to use meaningful abbreviations for the different deposits throughout the manuscript (in all figures, tables, and text). I am sure the current naming system means something to the authors but it is very disruptive and confusing to have to read BAL16-B2 and BAL16-UPL1-L1 for two different types of deposits. It should be possible to understand tables and figures without having to read the part about what the different labels mean. The flow of the manuscript would be much better with a simpler naming system.

AR: We thank the referee for their suggestion. We decided to keep the names of the study sites, as these are the names given to the samples during the field work and laboratory analyses and it would complicate data management and overview when we would rename the samples. Following the suggestion of the reviewers, we explain the naming system by adding Table 1 (see below) with the study sites with corresponding names in chapter 2.3. We think that a consistent and defined naming system improves the reading flow as then a stratigraphical orientation is clear by reading the sample/site name.

Table 1: Overview of study sites of yedoma exposure, drained thermokarst lake basin (DTLB) exposure and thermokarst lake sediments including coordinates and number of samples. BAL: Baldwin Peninsula, 16: year of expedition, 2016, B: bluff sampling, UPL: upland sampling, L: lake sampling.

Study site ID	Landscape unit	Coordinates	Samples, n
BAL16-B2	Yedoma exposure	66.73262°N, 62.49450°W	18
BAL16-B4	DTLB exposure	66.73644°N, 62.50208°W	31
BAL16-UPL1-L1	Thermokarst lake sediments	66.74220°N, 62.41310°W	9

RC: Section 3.1.4 Statistical significance: The statistics are too simplistic. I have to assume (you are not mentioning it or showing it) that your data are not normally distributed and that you therefore choose a non-parametric test, correct? The description of data distribution and statistical procedure is insufficient. Also, it is not appropriate to do pairwise comparisons when there are more than two groups without at least correcting for multiple testing. The minimum would be to perform a Kruskal-Wallis test and if significant to add a pairwise Wilcoxon-test, which would calculate pairwise comparisons between groups (you would have to include a correction for multiple testing). But, I think even that approach is too simplistic. You have multiple depths at different sites and so it does not make sense to compare one site with the other when you are not comparing the same thing. You could consider binning your data to different depths or different ages per site and then perform statistical analyses, preferably an ANOVA or something. The statistical results are not the core of your manuscript and that is fine, it still needs to be accurate.

AR: Thank you for the suggestion. We improved our explanation on the applied statistics in the text and the supplement. We now also performed a normality test. However, non-parametric tests also work for normally distributed data. We see some conflicts with the aim of our study concerning the binning of our data. Our goal is to compare the different parameters between the different landscape

units in order to see the range and variation of the data per landscape unit. Therefore, we do not bin the data to different depths but take the landscape unit as a whole. We explained this better in the revised manuscript. We added the results of the Kruskal-Wallis tests to the supplementary information.

RC: C/N already says it is a ratio and you do not need to add ratio afterwards

AR: We changed this throughout the manuscript. Thank you for the suggestion.

RC: The photographs in the Supplementary material are useful

AR: Thank you

RC: Table 1: I don't understand what Mean cal ages and rounded 14C ages are and why only a few samples had a +/- . You need to explain what +/- is

AR: The Mean cal ages (changed to Calibrated ages in the revised manuscript) are the mean values of the age range that were derived from the 14C calibration software (using CALIB 7.1 and IntCal13 calibration curve). The calibrated ages are shown including one standard deviation (σ) uncertainty (\pm). The calibration is not possible for samples with infinite ages ($>50,000$ years) and therefore, no uncertainty is given for these samples. We clarified this in the revised version of our manuscript.

RC: Why are you showing 14C Ages again in Fig. 3? Isn't it the same as in Figure 2? It seems redundant but maybe there is a reason for it and an explanation is needed

AR: The reviewer is right; the 14C Ages in Fig. 3 are indeed the same as in Fig. 2, which we clarified in the figure caption. We chose to show the ages again in Fig. 3 as the timeline inferred from them support the discussion on the biomarkers present in the sediments.

RC: Fig. 2 and 3, it would be better to show dots for all the other variables as well and not just for the age column. You are only measuring a few data points along the profile and it gives a slightly wrong impression if you show lines as you are not continuously measuring

AR: We thank the reviewer for their comment and revised the graphs accordingly.

RC: Fig. 3, why did you not measure biomarkers in the lake sediment? In the method section you say that you only measured it in those two but you don't say why

AR: The initial focus of the project was on the terrestrial deposits and therefore the biomarker analysis is too. We later decided to also include lacustrine sediments to cover all three main landscape units of the Baldwin Peninsula. However, only general biogeochemical properties were analyzed for this landscape type.

RC: When describing results along a depth gradient I think it is much better to go from the surface downwards and not the other way around. All soils have a surface but they go to a different depths and that just makes it confusing

AR: We agree with the reviewer that soils are generally formed from the surface downwards and that age increases with depth. However, the permafrost deposits in this study, and the yedoma in particular, have built up with time. Therefore, we decided to follow the geological timeframe, also considering that the time of deposition has a big influence on the studied sediments.

RC: Section 3.1.3, add that the data to this section are shown in Figure 2 (bottom panels)

AR: We added this reference as suggested.

RC: Table S2, here you introduce new acronyms for Yedoma when previously you have used this awkward BAL16-B2 naming, I very strongly suggest that you use informative labels in all figures, tables, and throughout the text

AR: We changed the names in Table S2 and S3 to match the names used throughout the manuscript.

RC: Figure 4: can probably be moved to the Supplementary Material. Is the number n-C29 or n-C31 that is indicated in the x-axis the dominant chain? I find this figure confusing.

AR: The figure was moved to the Supplements as suggested. The number below the x-axis is indeed the dominant chain per sample, which we clarified in the revised manuscript.

RC: The grain size distribution figures in the supplementary material require more explanation in the revised manuscript. Should S7 be for BAL16-B4 and not B2? You show BAL16-B2 in S6. What is B2.1 through B2.42 in S6 and B4.1 through B4.31 in S7? You need to write out what f., m., c., v.c. means.

AR: The first graph in the section Grain size distribution (previously S7, S8 in the revised version) shows the grain size distribution of the yedoma exposure, the second graph (previously S8, now S9) the drained thermokarst lake basin. Numbers B2.1 through B2.42 stand for the subsamples of the exposure. In the revised manuscript, we indicated the depths instead. The same changes were made for the drained thermokarst lake basin graph. The letters f., m., etc. refer to different grain size classes, which we explained in the revised manuscript as suggested.

RC: Table S2, S3, and S4, what is “outcome of Mann-Whitney-Wilcoxon test”? P-values? Which software did you use? Please also use a consistent number of digits after the comma. I would refrain from adding stars to non-significant outcomes as that is usually used to indicate significance. The table would be so much easier to read if you had less numbers per cell, why not just indicate p-values as <0.05, <0.01, and <0.001 or something like that.

AR: We calculated the Kruskal-Wallis-Test and Mann-Whitney-Wilcoxon using R (version 3.4.3) (`kruskal.test` and `wilcox.test`, respectively). To improve clarity, we changed the notation of the p-values in Table 2, 3 and 4 as suggested (<0.05, <0.01 and <0.001).

RC: Figure 6 could be moved to Supplementary Materials

AR: As suggested, we moved Figure 6 to the Supplements and we added the references to the text.

RC: Discussion: the discussion is very hard to read because it often is a listing of results followed by another listing of results from other publications. Some re-organization and focus on the important results would help the story line.

AR: Thank you for the comment. We restructured the discussion as suggested and shifted the focus to comparing our data to those of other studies.

RC: I think it is useful that you compare the results from Baldwin Peninsula with previous studies, I am hesitant to believe the statistical results at this point because of my previous comments in regard to statistics

AR: In order to identify the differences or similarities between the landscape units between this and other studies in Alaska. We performed pairwise comparisons (Mann-Whitney-Wilcoxon test) and used these results in the discussion.

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We thank this referee for the valuable feedback on our manuscript. We went through the discussion section and incorporated their suggestions. We hope to clarify the central theme of our manuscript in our replies to their open questions.

Referee Comment (RC): Permafrost affected soils and sediments of the Northern hemisphere are a major terrestrial C reservoir, highly vulnerable to climate change. A better knowledge on the amount and composition of organic matter is thus crucial (e.g. to improve earth system models). Thus the authors report on a very important topic in biogeochemical research. However, the authors miss to get a clear central theme. It seems the group of authors tried to include a bit of everything in a very descriptive manner rather than providing a synthesis of the extensive data set. Another major drawback is the rather one-sided citation of studies either from the co-author list or affiliated colleagues. Especially with respect to organic matter quantity and quality a growing number of biogeochemical basic research is going on in the Arctic. For instance Gentsch et al. worked on the bioavailability of specific OM in Siberia, or Mueller et al. worked rather “close by” on OM quantity and quality on thaw lake basins in the Alaskan North slope region.

Authors Reply (AR): In our study, we aim to characterize the OC properties in permafrost deposits in order to assess the vulnerability of the permafrost to climate change and contribute to a better estimate of the terrestrial C reservoir in this part of the Arctic. We highlighted this theme better in the revised version of the manuscript. Regarding the comment on one-sided citation, in particular the OC quantity, we compared our OC budget estimates to other studies that studied similar deposits (yedoma or DTLB) and were expressed in the same units (kg/m³). In the revised manuscript we added the suggested study of Mueller et al. (2015). Regarding the OC quality, we elaborated on bioavailability of OC in high-latitude soils and included more studies such as the suggested study of Gentsch et al. (2015), and Vonk et al. (2010).

RC: line 21 Volumetric OC content in your case is OC stock. With giving soil OC stocks you are closer to what gets reported for soils.

AR: With volumetric OC content we mean the OC density, as the values are expressed per unit weight over unit volume. The carbon stock, however, is expressed per unit weight, which we report in megaton (Mt).

RC: page 3 line 6, To which OC pool do you refer here? Are you aiming to model specific OC pools with respect to decomposability, or are you just aiming to differentiate OC stocks with respect to different research sites?

AR: We here refer to the OC pool in the yedoma, DTLB and thermokarst lake sediments. We aim to estimate the stocks and decomposability, i.e. size and quality of OC pools. We clarified this in the revised version of the manuscript.

RC: page 3, line 7 The used biomarkers only represent a minor portion of the organic matter. Although useful for reconstruction of OM origin, these proxies are lower in explanatory power for the bioavailability of the sequestered OM. So I would not speak of "molecular composition of each OC pool and its quality" as it only represents a minor part of the bulk OC.

AR: We agree with the reviewer and rephrased this throughout the manuscript.

RC: page 3, line 10-23 - What was the reason to go to this site? How representative is it for Arctic permafrost soil landscapes with respect to the studied OC distribution and composition?

AR: This is the first time yedoma deposits on Baldwin Peninsula were described. Therefore, this study contributes to a better and more precise approximation of the OC of yedoma deposits for this part of the Arctic. We made this clearer in the revised version of the manuscript. The yedoma deposits were

discovered on the coast of the peninsula during a reconnaissance campaign, after which this coastal bluff and those of the drained thermokarst lake basin deposits were sampled. We show that the total organic carbon content of the yedoma deposits is in the range of other yedoma studies in Siberia and Alaska. Also, the higher quality of yedoma OC compared to that in DTLB deposits was shown before. We address the representativity of our study site in the outlook, the past paragraph of the discussion.

RC: page 3 line 28 - What do you mean by representative? How did you test representativity?
How are the five locations connected to each other with respect to the choice of sampling spots?

AR: With representative, we mean that we tried to cover the whole yedoma exposure. Due to the difficult terrain and the fact that the samples were taken in summer – the fast thawing of the deposits limits the accessibility – it was not possible to sample the whole exposure in one straight profile. Therefore, we sampled different portions of the exposure wherever accessible and compiled a composite profile. We changed this explanation in the manuscript.

RC: page 5 line 2 - How were the samples pre-treated? Did the authors test for Carbonates in the samples, or is the TC representing OC and IC?

AR: We did not pre-treat the samples. We measure the TC and TOC in different devices. To measure the TOC, the samples are combusted at a much lower temperature compared to the TC measurements, so that the inorganic part of the sample is not combusted, and hence not measured in the device. The total carbon (TC) represents the sum of the organic (TOC) and inorganic carbon (TIC).

RC: page 5 line 8 - Was it not possible to increase the sample amount to get into the measurement range?

AR: The sensitivity of the Elementar Vario Max C is 0.1 wt%. This means that with a large sample amount, the weight percentage would be similar and therefore also below the detection limit of the device. We measured two aliquots per sample where we allowed a standard deviation of <5%; we measured multiple times when this criterion was not met.

RC: page 5 line 15-28 - You are extracting free lipids, and thus you can make assumptions about the composition of the extractable lipid fraction of your samples. You cannot draw conclusions about the "molecular composition of the OC" in general as proposed. Please be more precise in the writing.

AR: You are right, we rephrased as suggested.

RC: page 6/7 line 30 and following - You are taking some samples at one small edge of the Island and estimate based on this the OC stocks for the whole Island? Do you have any data on the representativity of the sampled locations for the rest of the Island? And what is the aim of such a very vague approximation? I miss a consequent central theme in the manuscript. Is it the quantification of OC stocks in a permafrost affected landscape? If yes, you clearly miss representivity (e.g. just one lake core!). Or is it the study of the composition of the extractable lipids in concert with C and N contents? If yes, you could possibly dig deeper into that by looking for correlations between all the measured data.

AR: Arctic fieldwork is expensive and it is difficult to get to the remote places for sampling. Therefore, we sampled the three main landscape units of the peninsula to get an initial overview of the thermokarst processes influencing the topography and the organic carbon characteristics. The sample sites are exposed at the coast, allowing us to study OC characteristics of deep permafrost deposits. Sampling sites on top of the deposits, however, would require drilling. This was not possible as no drilling rig was available during the fieldwork. Using the stratigraphical land cover classification map that we made and remote sensing, we indeed generated a first estimate of the characteristics and size of the OC pool in this part of the Arctic.

RC: Results - How are all the single proxies/data correlated? You are just reporting every single measured proxy, but how are things related to each other?

AR: Our aim is to characterize the OC pool in the different landscape units on Baldwin Peninsula by assessing the OC pool size and quality. In order to assess the organic carbon quantity, we analyzed the total organic carbon content and – using the stratigraphical landcover classification map we made and bootstrapping techniques – we estimated the OC stock of the different landscape units (based on the wedge-ice volume, bulk density, total organic carbon content and the coverage and thickness of the deposits). In order to assess the OC decomposability, i.e. the quality, we analyzed the carbon-nitrogen ratio and stable carbon isotopes. Using the differences between the landscape units, we show that the C/N ratio and $\delta^{13}\text{C}$ show both OC source as well as quality. We used the n-alkane derived ACL index to distinguish between terrestrial land plants, algae and bacteria, and the CPI index as an indicator for OC decomposability, where a higher CPI suggests well preserved material. We clarified the link between the parameters in the revised manuscript. However, we would like to keep the results section as it is to keep it factual.

RC: page 9 line 7 - What is the uncertainty based on the spatial heterogeneity of sediment and soil properties including BD, C content, horizon depths etc.? How did you account for the spatial heterogeneity on the Island with respect to only 5 sampling spots at the edge of the research area?

AR: This study represents the first characterization of yedoma deposits on Baldwin Peninsula. Even though spatial heterogeneity exists both between and within landscape units (e.g. Zona et al., 2011), we were able to collect a total of 91 samples at 5 different locations that we used for all analyses. We believe that this is sufficient for an initial characterization of the OC pool in this part of the Arctic, and the objective of our study. The uncertainties of the estimations are included by repeated artificial subsampling for the OC stock calculation using bootstrapping. A detailed assessment of potential spatial heterogeneity is beyond the scope of this paper. However, following the suggestions of the reviewers, we sharpened the existing focus of the paper in the revised manuscript.

RC: page 9 line 20 and following - What does this paragraph in its extensive form have to do with "organic carbon characteristics" as proposed in the title? I recommend to at least shorten the "origin of the material" section, or put very reduced parts of it into the site description in the M&M section. The parts with ^{14}C and ^{13}C etc. should go into a condensed discussion of the OM composition in the subsequent section.

AR: This section describes the depositional environment of the island to provide a framework for the interpretation of the OC data. In order to assess the vulnerability of carbon in permafrost deposits on Baldwin Peninsula, it is crucial to know the source and properties of the deposits in which this OC is stored. Especially for OC in old, deep permafrost deposits like yedoma, it is highly important to report on sedimentary origin as well as the ages of the deposits. These both describe the regional geological context of the deposits, as well as it gives an insight in the origin of the material. Hence, we prefer to keep the content of this paragraph as it is.

RC: page 11 line 2 - So if it is comparable, why should one keep on reading? Put your data first and get the central theme out of it, not just repeat other peoples work at a new fancy sampling location.

AR: Thank you for the suggestion. We restructured the paragraph.

RC: page 11 line 8 - "a significant OC pool is expected" - do you have data to prove it? Otherwise stay away from vague approximations.

AR: We wanted to stress the importance of the volume of the yedoma deposits compared to the relatively shallow thermokarst deposits, after which we report on the absolute numbers. To avoid further confusion, we rephrased this sentence as suggested.

RC: page 13 line 13-27 - This whole paragraph is purely hypothetical. You have no data on OC vulnerability to climate warming nor for OC bioavailability. What is the central theme of your work? It reads like the authors wanted to have a bit of everything in it, paleo reconstruction, large scale OC estimates and OC composition. It would be great to get a synthesis of these parts rather than a descriptive manuscript.

AR: Giving a synthesis of the OC pool size and composition in permafrost on Baldwin Peninsula is exactly what we tried to do in the first part of the discussion, where we report on the quantity and quality of OC based on organic geochemical, sedimentological and also palaeoecological methods. The paragraph following this is meant as an outlook and to put the study and also the study area in a larger perspective. We put more stress on the actual data rather than on the more hypothetical part in the revised version of our manuscript.

RC: Conclusions – This is just a summary of your findings, but what are the take home messages and especially the implications of your work?

AR: We agree with the reviewer: we indeed summarized our findings in the conclusion. With these main findings, we show the answer to our research question and our main message: the first estimate of the total OC pool on Baldwin Peninsula, the relative contribution of the different landscape units (answer to our first research question), as well as the finding that OC in yedoma is most vulnerable to decomposition (answer to our second research question).

Organic carbon characteristics in yedoma and thermokarst deposits on Baldwin Peninsula, West-Alaska

Loeka L. Jongejans¹, Jens Strauss¹, Josefine Lenz^{1,2}, Francien Peterse³, Kai Mangelsdorff⁴, Matthias Fuchs^{1,5} and Guido Grosse^{1,5}

5 ¹Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research, Permafrost Research Section, Potsdam, Germany

²University of Alaska Fairbanks, Institute of Northern Engineering, Fairbanks, USA

³Utrecht University, Department of Earth Sciences, Utrecht, Netherlands

⁴Helmholtz Centre Potsdam - German Research Centre for Geosciences, Germany

10 ⁵University of Potsdam, Institute of Earth and Environmental Sciences, Potsdam, Germany

Correspondence to: Loeka L. Jongejans (loeka.jongejans@awi.de)

Abstract. As Arctic warming continues and permafrost thaws, more soil and sedimentary organic carbon (OC) will be decomposed in northern high latitudes. Still, uncertainties remain in the quantity and quality of OC stored in different deposit types of permafrost landscapes. This study presents OC data from deep permafrost and lake deposits on the Baldwin Peninsula which is located in the southern portion of the continuous permafrost zone in West-Alaska. Sediment samples from yedoma and drained thermokarst lake basin (DTLB) deposits as well as thermokarst lake sediments were analyzed for cryostratigraphical and biogeochemical parameters and their lipid biomarker composition to identify the size and quality of belowground OC pools in ice-rich permafrost on Baldwin Peninsula. We provide the first detailed characterization of yedoma deposits on Baldwin Peninsula. We show that three quarters of soil organic carbon in the frozen deposits of the study region (total of 68 Mt) is stored in DTLB deposits (52 Mt) and one quarter in the frozen yedoma deposits (16 Mt). The lake sediments contain a relatively small OC pool (4 Mt), but have the highest volumetric OC content (93 kg m⁻³) compared to the DTLB (35 kg m⁻³) and yedoma deposits (8 kg m⁻³), largely due to differences in the ground ice content. The biomarker analysis indicates that the OC in both yedoma and DTLB deposits is mainly of terrestrial origin. Nevertheless, the relatively high carbon preference index of plant leaf waxes in combination with a lack of degradation trend with depth in the yedoma deposits indicates that OC stored in yedoma is less degraded than that stored in DTLB deposits. This suggests that OC in yedoma has a higher potential for decomposition upon thaw, despite the relatively small size of this pool. These findings highlight the importance of [molecular OC analysis-lipid biomarker analysis](#) for determining the potential future greenhouse gas emissions from thawing permafrost, especially because this area close to the discontinuous permafrost boundary is projected to thaw substantially within the 21st century.

1 Introduction

The Arctic region is warming twice as fast as the global mean (Overland et al., 2017). Ice-rich permafrost soils are particularly vulnerable to climate warming and susceptible to large-scale thermokarst processes. Thermokarst is the subsidence of ground resulting from the thawing of ice-rich permafrost (Grosse et al., 2013; Günther et al., 2013). Thermokarst lake development often starts with the coalescence of polygonal ponds after the degradation of ice wedges (Grosse et al., 2013). This is followed by the formation of a body of unfrozen ground underneath the lake (i.e. talik) and subsequently, both the lake and talik grow and deepen. Thermokarst lake development can cease by drainage through lateral outflows formed by thermal erosion of ice wedge networks, by tapping of lakes due to coastal erosion, by vertical outflow through open taliks in regions with thin permafrost, or by infilling with sediment or encroaching vegetation (Burn_ and -Smith, -1990; Jones and Arp, 2015; Lenz et al., 2016c). After lake loss, the remaining basins largely become vegetated and eventually permafrost can reform (Jones et al., 2012). Due to the different stages of lake development, thermokarst landscapes are highly dynamic and form complex patterns of landscape units (Jones et al., 2012; Lenz et al., 2016a).

Late Pleistocene, ice-rich syngenetic permafrost, known as yedoma, is especially prone to rapid and deep thaw processes (Schirmermeister et al., 2013). These deposits cover large regions of Siberia and Alaska (Kanevskiy et al., 2011; Schirmermeister et al., 2013; Strauss et al., 2017) and can reach a thickness of up to 50 m (Sher, 1997; Shur et al., 2012). Yedoma contains large syngenetic ice-wedges and can have a ground ice content of up to 80 vol%, thus yedoma deposits are highly vulnerable to thermokarst processes (Kanevskiy et al., 2016; Ulrich et al., 2014).

Permafrost landscapes store large quantities of OC (Hugelius et al., 2014; Strauss et al., 2013) as the generally low decomposition rates, due to low soil temperatures and poor drainage, inhibit decomposition of organic matter (Davidson and Janssens, 2006). Permafrost thaw and talik formation, however, allow microbial decomposition of the previously freeze-locked OC, resulting in increased greenhouse gas emissions (in particular carbon dioxide and methane) into the atmosphere and enhancing the initial warming (Koven et al., 2015; Schuur et al., 2015; Strauss et al., 2017). Rapid and large-scale permafrost degradation of ice-rich permafrost, such as yedoma deposits, may constitute a positive feedback to atmospheric warming.

Due to the freezing conditions during and after accumulation, the OC stored in yedoma deposits is highly decomposable (Knoblauch et al., 2013; Schädel et al., 2014; Strauss et al., 2017). Yedoma deposits contain on average less OC than DTLB deposits (e.g. Strauss et al., 2013), however higher respiration rates from yedoma deposits have been observed compared to DTLB deposits (Dutta et al., 2006; Lee et al., 2012; Zimov et al., 2006). Moreover, previous studies found that the state of OC decomposition is more dependent on OC properties, rather than on OC age (Knoblauch et al., 2013; Stapel et al., 2016). It is therefore crucial to characterize the quantity and quality of OC pools in both undisturbed as well as disturbed ice-rich permafrost landscapes, so that the potential contribution to future greenhouse gas release following permafrost degradation can be better constrained for each of the different landscapes.

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This study aims to characterize the belowground OC stored in thermokarst affected areas by analyzing sediment samples from yedoma deposits, drained thermokarst lake basin deposits and thermokarst lake sediments. Two main questions are addressed: 1) how much OC is stored and 2) what is the quality of this OC? In this study, we particularly refer to belowground OC and the quality of OC refers to the decomposability of the material, respectively the vulnerability to future decomposition. In our analysis, we focus on cryostratigraphical (bulk density, absolute ice content) and biogeochemical parameters (total organic carbon, total nitrogen, ~~carbon-nitrogen C/N~~ ratio and stable carbon isotopes) to estimate the OC pool size ~~of the yedoma, DTLB and thermokarst lake sediments~~. In addition, lipid plant and soil specific biomarkers (*n*-alkanes and brGDGTs, respectively) were used to determine the ~~source and quality of the OC pools, molecular composition of each OC pool and its quality~~.

10 2 Material and methods

2.1 Study area

The study sites are located on the western coast of the Baldwin Peninsula, which is surrounded by the Kotzebue Sound in Northwest Alaska (66°40' N, 162°15' W) (Figure 1). The Baldwin Peninsula is part of the former landmass Beringia that remained unglaciated during the Late Pleistocene (Hopkins, 1982). The peninsula is located in southern portion of the continuous permafrost zone and therefore close to the discontinuous permafrost boundary (Jorgenson et al., 2008). According to geological maps, the peninsula is largely composed of a sequence of marine, fluvial and glaciogenic sediments which are well exposed along coastal bluffs and in some regions covered by loess-like deposits (Hopkins et al., 1961; Huston et al., 1990; Pushkar et al., 1999). This study represents the first characterization of yedoma deposits on Baldwin Peninsula. A field campaign in summer 2016 revealed several yedoma exposures in active retrogressive thaw slumps on the western coast of the peninsula that consisted of fine-grained, ice-rich permafrost deposits penetrated by large syngenetic ice wedges. These yedoma exposures were located in upland remnants, a setting which is typical for yedoma hills found in northeast Siberia (Schirmermeister et al., 2013). A large portion of the peninsula has been affected by severe permafrost degradation and multiple thermokarst lake basin generations visible in satellite imagery (Figure 1). The present-day climate is subarctic with an average annual precipitation of 280 mm and a mean annual air temperature of -5.2°C (US Climate Data, 2017).

25 2.3 Field work

Sampling of yedoma and drained thermokarst lake basin (DTLB) exposures and thermokarst lake sediments was carried out on the western coast of the Baldwin Peninsula in August 2016 (Table 1+). Photographs of the exposures and the lake core are shown in Supplement 2.1. The samples were named as follows: the geographical region (BAL for Baldwin Peninsula), the year of sampling (16 for 2016), the setting (B for bluff, UPL for upland), the site numbers (here: 2, 4) and the sampling of an aquatic environment was indicated (L for lake) (Table 1+).

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A yedoma coastal bluff (BAL16-B2) of 16 m thickness was sampled. This bluff is characterized by sediments containing large ice wedges, ice bands and ice lenses, which are overlying a separate unit of ice-rich silty sediments. Due to the difficult terrain and fast thawing of the deposits in summer, it was not possible to sample the whole exposure in one straight profile. Therefore, 25 representative-samples of the yedoma deposits were collected using a handheld power drill (ø 57 mm) at five different sampling locations along the exposure, forming a composite profile. Three additional samples were taken from the separate depositional unit underlying the yedoma which was not as ice-rich and did not contain in-situ formed ice wedges. Exposures of DTLB deposits ranging in thickness from 3 to 8 m were sampled from three exposed profiles (BAL16-B3, BAL16-B4 and BAL16-B5). 31 samples were collected from the 8-m-high DTLB exposure BAL16-B4 consisting of mostly laminated sediments that contained various cryotextural and organic features (e.g. ice inclusions, lenses, organic matter inclusions). The DTLB exposures BAL16-B3 (9 samples) and BAL16-B5 (4 samples) were sampled to generate a broader sample base for the estimates of the DTLB OC pool (the results of the two DTLB additional profiles BAL16-B3 and BAL16-B5 are shown by depth in Supplement 2.3). All samples from the yedoma and DTLB exposures were kept frozen until laboratory analysis at Alfred Wegener Institute (AWI), Helmholtz Centre for Polar and Marine Research, in Potsdam.

In addition, a 34-cm-long sediment core (BAL16-UPL1-L1) was retrieved from a shallow thermokarst lake (1.35 m deep at coring site) with a N-W extent, a length of about 1,400 m and a width of 800 m. Unfrozen, slightly layered, clayey silty lake sediments were retrieved in a PVC tube (ø 60 mm) using a piston corer and were kept cool until sub-sampling (9 samples) and laboratory analysis.

2.4 Bulk physical properties

In the laboratory, the sediment samples were freeze-dried. The absolute ice content was derived from the weight difference of the frozen and dry samples after Eq. (1) and is expressed in weight percentage (wt%).

$$\text{Absolute ice content [wt\%]} = \frac{\text{wet weight} - \text{dry weight}}{\text{wet weight}} * 100 \quad (1)$$

For the frozen samples, the bulk density BD_1 was calculated following Eq. (2).

$$BD_1 [10^3 \text{ kg m}^{-3}] = (\varphi - 1) * -\rho_s \quad (2)$$

where φ is porosity (fraction) and ρ_s is the mineral density in $[10^3 \text{ kg m}^{-3}]$. The porosity is derived from the volumetric ratio of ice and dry sample whereby a constant ice density was assumed of $0.91 * 10^3 \text{ kg m}^{-3}$ (at 0°C) (Lide, 1999). For the mineral density, the density of quartz of $2.65 * 10^3 \text{ kg m}^{-3}$ was taken (Rowell, 1994).

For the unfrozen thermokarst lake sediments, BD_2 was calculated following Eq. (3). The volume of the sediment was corrected to account for the compression of the sediment during sampling. The volume of the excess water in the core tube after sampling was measured and included in the volume calculation of the core sediment.

$$BD_2 [10^3 \text{ kg m}^{-3}] = \frac{\text{dry weight}}{\text{volume}} \quad (3)$$

where dry weight is given in [g] and volume in $[\text{cm}^3]$, respectively in $[10^3 \text{ kg}]$ and $[\text{m}^3]$.

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2.5 Radiocarbon dating

Macrofossils of 15 samples were dated for accelerator mass spectrometry (AMS) radiocarbon dating in the Radiocarbon Laboratory in Poznan, Poland (Goslar et al., 2004). The radiocarbon ages were calibrated using the CALIB 7.1 software and the IntCal13 calibration curve (Stuiver et al., 2017). All calibrated ages are expressed as calibrated kilo years before present (cal ka BP), including the standard deviation (\pm). Infinite ages (>50,000 a) cannot be calibrated and hence, no uncertainty could be given.

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2.6 Nitrogen and carbon content and composition

Homogenized samples were analyzed for total nitrogen (TN), total carbon (TC) and total organic carbon (TOC) using an elemental analyzer (Elementar Vario EL III and Elementar Vario Max C) and are expressed in [wt%]. The TOC/TN (weight) ratio was calculated and will be referred to as C/N ~~ratio~~. The stable carbon isotopic composition was determined by measuring $\delta^{13}\text{C}$ (ThermoFisher Scientific Delta-V-Advantage gas mass spectrometer equipped with a FLASH elemental analyzer EA 2000 and a CONFLO IV gas mixing system). The ratio is compared to the standardized Vienna Pee Dee Belemnite (VPDB) and expressed in per mille (‰ vs. VPDB). For samples with a TOC below the analytical accuracy (0.1 wt%), no C/N ~~ratio~~ nor $\delta^{13}\text{C}$ was measured.

The combination of the C/N and $\delta^{13}\text{C}$ has been widely used as indicator of OC source. A higher C/N suggests an enhanced input of terrestrial land plants whereas algal produced matter is generally characterized by a lower C/N. The C/N and $\delta^{13}\text{C}$ of OC furthermore allow to distinguish between marine and lacustrine algae, where marine algae generally have a higher $\delta^{13}\text{C}$ (e.g. Lenz et al., 2016a; Meyers, 1994, 1997). Moreover, the C/N and $\delta^{13}\text{C}$ have been used as an indicator of OC decomposition, where a lower C/N and higher $\delta^{13}\text{C}$ indicate further degraded material (i.e. lower quality) (Gentsch et al., 2015; Gundelwein et al., 2007; Schädel et al., 2014; Weiss et al., 2016).

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In order to compare the cryostratigraphy and biogeochemistry (BD, TOC, C/N and $\delta^{13}\text{C}$) of the three stratigraphical landscape units (yedoma, DTLB and thermokarst lake deposits), the non-parametric Kruskal-Wallis test and Mann-Whitney-Wilcoxon test ~~was/were~~ performed using the R statistical environment to test for significant differences ~~of the biogeochemical parameters. The non-parametric between the stratigraphic landscape units.~~ When the p-value exceeds 0.05, the null hypothesis (H₀: There is ~~a no~~ statistically significant difference between the sample means of the different stratigraphic landscape units) cannot be rejected. In these statistical tests, the landscape units are treated separately and therefore, the internal differences of the study sites with depth and age are not included.

More extensive method descriptions of the elemental analysis can be found in Supplement 1.2.

2.7 Lipid biomarker analysis

2.7.1 Extraction and separation

Biomarker analysis was carried out to identify the ~~molecular composition of the OC in order to analyze the~~ OC composition and quality (Andersson and Meyers, 2012; Strauss ~~et al.~~, 2015). In total, 13 samples (6 from BAL16-B2 and 7 from BAL16-B4) were analyzed at the German Research Centre for Geosciences (GFZ) for *n*-alkanes (long-chained, single bonded hydrocarbons) and branched glycerol diacyl glycerol tetraethers (brGDGTs; bacterial membrane lipids). The methods were adapted from Schulte et al. (2000) and Strauss et al. (2015). About 8 g of each sample was extracted (Dionex 200 ASE extractor) using dichloromethane/methanol (DCM/MeOH) (99:1 v/v) (heating phase 5 min, static phase 20 min at 75°C and 106 Pa). Excess solvent was evaporated under N₂. A known amount of four internal standards was added: 5 α -androstane, ethylpyrene, 5 α -androstane-17-one and erucic acid. The samples were passed over a sodium sulfate column with *n*-hexane prior to separation by medium-pressure liquid chromatography (MPLC) (Radke et al., 1980) into three fractions: aliphatic hydrocarbons, aromatic hydrocarbons and nitrogen-, sulfur-, and oxygen (NSO) containing compounds. The dissolved extracts were injected into the MPLC system where they were led through a column and pre-columns (thermally deactivated silica 100 63-200 μ m and 200-500 μ m on top with ratio ~7:1) with *n*-hexane. The NSO-fraction was further separated manually into a polar and acid fraction using a KOH-impregnated silica gel column with DCM.

2.7.2 Measurements

The *n*-alkanes were measured as part of the aliphatic fraction using gas chromatography (GC) - mass spectrometry (MS) (Trace GC Ultra and MS DSQ, Thermo Electron Corporation) using helium as a carrier gas. The samples were vaporized (50°C to 300°C with 10°C/s, 10 min isothermal holding) and led through a capillary column (BPX5; 22 mm x 50 m, film thickness 0.25 μ m) (Peters et al., 2005). The oven was programmed from 50°C to 310°C (3°C/min and 30 min isothermal holding). The GC was linked to the MS to enable compound identification (ionization mode at 70 eV, 230°C). Full scan mass spectra were obtained from *m/z* 50 to 600 Da (2.5 scans/s). Using the software XCalibur, the peaks in the GC-MS total ion current chromatogram were integrated manually. The *n*-alkanes were quantified by comparing the peak area of the target compounds to the applied internal standards.

The brGDGTs were measured as part of the acid fraction using high-performance liquid chromatography (Shimadzu LC-10AD HPCL device coupled to a Finnigan TS 7000 mass spectrometer with APCI interface). The compounds were separated at 30°C over a Prevail Cyano column (2.1x150 mm, 3 μ m; Alltech) preceded by a precolumn filter of the same material, which does not separate 5- and 6-methyl isomers. Each fraction was eluted isocratically with *n*-hexane A and isopropanol B (5 min. 99% A and 1% B, linear gradient to 1.8% B within 40 min, in 1 min to 10% B holding for 5 min and back to initial conditions in 1 min, held for 16 min) with a flow rate of 0.2 mL/min. The APCI device has a corona current of 5 μ A, voltage of 5 kV, the vaporizer temperature is 350°C and the capillary temperature 200°C. The source operates with nitrogen sheath gas at 60 psi

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without auxiliary gas. Full mass spectra were obtained at a scan rate of 0.33 s. The integration was performed in XCalibur and the quantification was performed by comparing the compound peaks to an Archaeol run.

2.7.3 Biomarker derived indices

The carbon preference index (CPI) of the *n*-alkanes was calculated after Marzi et al. (1993) following Eq. (4), and the average chain length (ACL) after Poynter and Eglinton (1990) following Eq. (5).

$$CPI_{23-33} = \frac{\sum_{i=n}^m C_{2i+1} + \sum_{i=n+1}^{m+1} C_{2i+1}}{2 * (\sum_{i=n+1}^{m+1} C_{2i})} \quad (4)$$

where *n* is the starting dominating chain length divided by 2, *m* the ending dominating chain length divided by 2 and *i* the carbon number index.

$$ACL_{23-33} = \frac{\sum i C_i}{\sum C_i} \quad (5)$$

where *i* is the carbon number and *C* the concentration. For the calculation of both CPI and ACL, the interval of C₂₃ to C₃₃ was used. The ACL indicates OC source, where higher land plants (i.e. vascular plants) are dominated by long-chain *n*-alkanes of 25 to 30 carbon atoms, whereas bacteria and algae contain mainly shorter chains of 15 to 20 carbon atoms (Killops and Killops, 2013; Strauss et al., 2015). The CPI is the odd-to-even predominance of the hydrocarbons and indicates the degree of degradation of the OC, where a lower value indicates further degraded material (Andersson and Meyers, 2012; Glombitza et al., 2009).

2.8 Landscape organic carbon pool estimation

A map of key landscape units (yedoma hills, DTLBs and thermokarst lakes) of the northern part of Baldwin Peninsula was developed in order to calculate the coverage for each landscape unit and to allow a first-order estimate of the belowground OC storage. Using a Landsat 8 satellite image (false color image with short-wave infrared, near-infrared and deep blue bands (bands 7-5-1), pixel resolution 30 m) as well as a digital terrain model (DTM; grid cell resolution 5 m), the three main landscape units were manually mapped and digitized for the northern part of the Baldwin Peninsula (~450 km²). A similar approach of operator-driven thermokarst mapping by manually digitizing landforms from remote sensing imagery in combination with a DTM was successfully applied by Morgenstern et al. (2011) and Farquharson et al. (2016).

The total OC pool on Baldwin Peninsula was then estimated based on the deposit thickness, the spatial coverage, wedge ice volume (WIV), BD and TOC from the sampled exposures and cores (BAL16-B2 to B5 and BAL16-UPL1-L1) following Eq. (6).

$$Total\ OC\ pool\ [Mt] = \frac{thickness * coverage * \frac{100-WIV}{WIV} * BD * \frac{TOC}{100}}{10^6} \quad (6)$$

where the deposit thickness is given in [m], spatial coverage in [m²], WIV in [vol%], BD in [10³ kg m⁻³] and TOC in [wt%]. The deposit thickness is based on few field observations and for this first-order assessment assumed constant over the whole peninsula. Deeper deposits below frozen yedoma and DTLB are excluded due to unknown spatial coverage and thickness.

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Also, the deposits below lake sediments, potentially unfrozen in a talik, are not included in the calculation. The spatial coverage per landscape unit was calculated from the land coverage classification map. Strauss et al. (2013) calculated ~~the average wedge-ice volume~~ *WIV* for yedoma and DTLB deposits based on polygon size and ice-wedge width and depth. *WIV* was assumed to be zero for the thermokarst lake sediments. In order to compensate for possibly non-continuous field sampling, weighted *BD*TOC* values over depth were used by value replication depending on depth interval. The data were extrapolated to the northern part of the Baldwin Peninsula (i.e. the mapped part). The volumetric *OC pool* was estimated according to Eq. (7).

$$\text{Volumetric OC pool [kg m}^{-3}] = \left(\frac{100 - \text{WIV}}{\text{WIV}} * \text{BD} * \frac{\text{TOC}}{100} \right) * 10^3 \quad (7)$$

The calculation for each landscape unit was performed separately using bootstrapping techniques, which included 10^4 iterations of random sampling with replacement, after which the mean and standard deviation were calculated. Because *BD* and *TOC* are correlated, paired values were used during the random sampling.

3 Results

3.1 Biogeochemical and biomarker proxies

3.1.1 Yedoma exposure

Radiocarbon dates for yedoma exposure BAL16-B2 range from >50,000 to 10,000 cal a BP (Table 2). There is no consistent age-depth relationship: while a few samples, including the near-surface sample, have an infinite age, the second youngest sample is found at a depth of 1600 cm. The cryostratigraphical and biogeochemical parameters are presented by depth (Figure 2). Depths are measured from the cliff top downwards for the yedoma and DTLB exposures, and from the sediment surface downwards for the thermokarst lake sediments. The *BD* of the yedoma deposits (mean: $0.80 * 10^3 \text{ kg m}^{-3}$, sd: 0.18) shows most variation in the bottom part up to 1117 cm. The *TOC* ranges between 0.1 and 4.7 wt% (mean: 1.9 wt%, sd: 1.1). The *C/N ratio* varies between 4.4 and 14.0 (mean: 10.1, sd: 2.9) and does not show a trend with depth. The $\delta^{13}\text{C}$ is in the range of -24.8 to -28.3 ‰ (mean: -25.9 ‰, sd: 0.9) and shows most variation up to 1080 cm. Exposure BAL16-B2 is very ice-rich with an average absolute ice content of 45 wt% (70 vol%).

The biomarkers concentrations and index values are presented by depth in Figure 3. The *n*-alkane concentration in the yedoma deposits ranges from 0.5 to 1.8 $\mu\text{g mg}^{-1}$ *TOC* (mean: 1.0 $\mu\text{g mg}^{-1}$ *TOC*, sd: 0.5). The *ACL*₂₃₋₃₃ varies between 28.8 in the bottom sample and 28.2 in the top with a peak of 30.0 at 1117 cm (mean: 28.8, sd: 0.6). The samples are dominated by *n*-alkane chains with a high number of carbon atoms: the dominating chains are *n*-C₂₉ and *n*-C₃₁ (Figure 4). The *CPI*₂₃₋₃₃ is in the range of 9.0 to 13.6 (mean: 11.6, sd: 2.0) and does not show a trend over depth. The sample at 1399 cm has the maximum *brGDGT* concentration of 1.3 $\mu\text{g mg}^{-1}$ *TOC* whereas the other samples have relatively low concentrations (mean: 0.5 $\mu\text{g mg}^{-1}$ *TOC*, sd: 0.5).

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3.1.2 DTLB exposure

Radiocarbon ages for DTLB exposure BAL16-B4 (Table 2) are in the range from >-50,000 cal a BP (788 to 434 cm) to 240 cal a BP (22 cm) and show no age inversions: the lower samples are of infinite age and the near-surface sediments are the youngest and show Holocene ages. The BD decreases upwards in the profile and ranges between 1.27 and $0.29 \cdot 10^3 \text{ kg m}^{-3}$ (mean: $0.91 \cdot 10^3 \text{ kg m}^{-3}$, sd: 0.3), a particularly strong decrease occurs from 280 cm to 215 cm (Figure 2). The TOC (mean: 6.6 wt%, sd: 9.4) is higher in the interval from 250 to 22 cm than in the lower part, and strongly increases in the top 8-cm. The C/N ratio increases towards the top, ranging from 11.3 at the bottom to 29.0 at the top (mean: 14.9, sd: 4.2). The $\delta^{13}\text{C}$ displays an opposite trend with higher values in the lower part (-25.8‰) and a decrease near the surface (-28.7‰) (mean: -27.2‰, sd: 0.7), and a sudden decrease at 280 to 250 cm. The exposure has an average absolute ice content of 41 wt% (67 vol%).

The *n*-alkane concentration reaches its maximum of $16.0 \mu\text{g mg}^{-1}$ TOC in the bottom sample whereas the other samples have much lower concentrations (mean: $4.3 \mu\text{g mg}^{-1}$ TOC, sd: 5.4) (Figure 3). The ACL₂₃₋₃₃ is higher than 28 for all samples except at 583 cm and 430 cm (mean: 28.3, sd: 0.4). The dominating *n*-alkane chains are *n*-C₂₇ and *n*-C₃₁ (Figure S7). The CPI₂₃₋₃₃ increases towards the top from 5.7 to 12.6 (mean: 8.8, sd: 2.1). The brGDGT concentration varies between 0 and $3.8 \mu\text{g mg}^{-1}$ TOC with the peak at 160 cm below the surface (mean: $1.5 \mu\text{g mg}^{-1}$ TOC, sd: 1.4).

3.1.3 Thermokarst lake sediments

The radiocarbon age of the thermokarst lake core BAL16-UPL1-L1 (Table 2) is 2,010 cal a BP at a depth of 26.5-27.5 cm and 480 cal a BP at 19-20 cm. The sedimentological and biogeochemical parameters (Figure 2) have a low variability along the core profile with a mean for BD of $0.78 \cdot 10^3 \text{ kg m}^{-3}$ (sd: 0.01), TOC of 14.4 wt% (sd: 0.5), C/N ratio of 22.5 (sd: 0.6) and $\delta^{13}\text{C}$ of -28.5 ‰ (sd: 0.2). The $\delta^{13}\text{C}$ shows a slight decrease towards the lake sediment surface.

3.1.4 Statistical significance

We found significant differences ($p < 0.05$) for all (pairwise) comparisons of the biogeochemical parameters between the yedoma, DTLB and thermokarst lake deposits on Baldwin Peninsula (Supplement 2.6). The DTLB deposits have the highest BD and the thermokarst lake sediments the lowest. The TOC values and C/N ratios are highest in the thermokarst lake sediments followed by the DTLB and then the yedoma deposits, whereas the $\delta^{13}\text{C}$ values shows an opposite trend.

3.2 Organic carbon pool estimation

We produced a land cover classification map distinguishing between yedoma, DTLBs, thermokarst lakes and lagoons (Figure 4). For the lakes on yedoma uplands and lagoons, no field information is available, so that those areas are excluded from further OC pool calculations. The total area mapped is about 450 km^2 of which ~65% is covered by DTLB, ~30% by yedoma and ~5% by thermokarst lakes (Table 3). The input parameters of the bootstrapping for the OC pool estimates are shown in Table 3. The volumetric and total OC pool per stratigraphic landscape unit for the frozen deposits and unfrozen thermokarst

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lake sediments are presented in Table 3, where the WIV is included. The OC pool estimates without WIV are also reported to allow comparison with other studies (Table 3). The yedoma deposits contain $8.0 \pm 0.40.8$ kg OC m^{-3} ($15.3 \pm 0.81.6$ excl. WIV) and the DTLB deposits $34.7 \pm 1.42.9$ kg OC m^{-3} ($37.4 \pm 1.63.1$ excl. WIV), which corresponds to a total OC pool of $16.3 \pm 0.91.7$ Mt ($31.4 \pm 1.73.3$ excl. WIV) in yedoma and $51.5 \pm 2.34.3$ Mt ($55.4 \pm 2.34.6$ excl. WIV) in DTLB deposits. The total estimate of the total OC pool of the frozen sediments on northern Baldwin Peninsula is ~ 68 Mt OC. The thermokarst lake sediments contain $92.9 \pm 0.40.8$ kg OC m^{-3} which adds up to a total pool of 3.9 ± 0.0 Mt for all thermokarst lakes in the study area.

4 Discussion

4.1 Landscape development and carbon dynamics

4.1.1 Sediment facies

10 The yedoma deposits of BAL16-B2 have been accumulated in a stable predominantly aeolian depositional environment, as shown by the grain size distribution (detailed method description and results of the grain size analysis are given in Supplement 1.1 and 2.5.1). The grain size distributions indicate that these deposits are characterized by a stronger aeolian influence than northeastern Siberian yedoma sites (Schirrmeister et al., 2008b; Strauss et al., 2012). Field observations suggest that the lower part of exposure BAL16-B2 (1,600 to 1,870 cm) is a separate unit. The significant change in sediment type from very coarse silt to medium silt (p -value < 0.05), reflected in the grain size distributions (Supplement 2.5.1), confirms this distinction.

15 The formation of yedoma deposits on the west coast of the Baldwin Peninsula at study site BAL16-B2 possibly started before 50 cal ka BP. The large range of radiocarbon ages in these yedoma deposits suggests that they are mixed with ancient or younger organic material. According to Vasil'chuk and Vasil'chuk (2017), contamination with ancient organic material is common in yedoma deposits, due to the syngenetic character of the deposits. Therefore, they proposed to take the youngest age of the sediments, justified by the fact that age rejuvenation is not likely, due to the undisturbed character of the deposits. 20 However, it is unlikely that relatively young sediments (~ 17 cal ka BP at 1600 cm) are overlain by a few meters of older sediments (~ 45 cal ka BP at 1117 cm and infinite ages at 1399 and 1500 cm), suggesting that the young ages could be the result of re-deposition. Considering that BAL16-B2 is a coastal bluff, re-deposition of young material cannot be neglected. Furthermore, the ages fall in the range of previous studies from Siberian yedoma deposits (> 57 to 13 ka BP) (Schirrmeister et al., 2002a, 2002b, 2003; Strauss et al., 2013) and Alaskan yedoma deposits (> 48 to 14 ka BP) (Kanevskiy et al., 2011), except for the sample at 945 cm (~ 10 cal ka BP). Generally, care should be taken with the interpretation of dating of yedoma deposits.

25 The lower part of the DTLB exposure BAL16-B4 has a radiocarbon age of 46 cal ka BP or older, which suggests that the lowest deposits have been accumulated during the same time as yedoma formation. However, the TOC content and C/N ratio are significantly lower in BAL16-B2 than in BAL16-B4 whereas the $\delta^{13}C$ is significantly higher (Supplement 2.6). This suggests that the DTLB deposits are a mixture of former yedoma deposits thawed and partially decomposed in a lake talik, 30 lake sediments and post-drainage terrestrial peat. The grain size signal for both exposures is very similar ($p > 0.05$), suggesting

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a connection between the BAL16-B2 and BAL16-B4 sediments, which can be explained by the Holocene reworking of the yedoma sediments in the lake and in the talik affected by subsidence. The branched and isoprenoid tetraethers (BIT) index (detailed method description and results of the biomarker climatic indicator are given in Supplement 1.3 and 2.5.2) is lower for BAL16-B2 than for BAL16-B4, which may indicate that the sediments in BAL16-B2 have been deposited in a drier climate (Dirghangi et al., 2013), in line with earlier paleoenvironmental reconstructions (e.g. Andreev et al., 2011; Lenz et al., 2016c).

In exposure BAL16-B4 a change was observed in the proxy records (age, BD, TOC, $\delta^{13}\text{C}$) between 280 and 250 cm: the sediments above 250 cm have lower BD and $\delta^{13}\text{C}$ and a higher TOC and C/N ratio compared to the sediments below 280 cm. Furthermore, there is a change in sediment type, as shown by the different grain size distributions below 280 and above 250 cm (Supplement 2.5.1), and in the depositional environment, as shown by a change in the relative brGDGT distribution, reflected in the methylation of branched tetraethers (MBT) index (Supplement 1.3 and 2.5.2). These changes suggest the initiation of a thermokarst lake: the sediments below 280 cm likely are former yedoma sediments that were thawed in situ in a talik (taberite), whereas the upper sediments are lake sediments. The lake initiation likely happened during the Holocene. The drainage of the lake is not recorded in the data; the drainage event took place after 2.08 cal ka BP.

The thermokarst lake sediments from BAL16-UPL1-L1 have been accumulated during the Holocene (2,010 and 480 a BP). Assuming a recent age for the surface sediments, the calculated sedimentation rate between 2,010 and 480 years BP was $\sim 6 \text{ cm ka}^{-1}$ and the sedimentation rate for the last 480 years $\sim 50 \text{ cm ka}^{-1}$. For comparison, Lenz et al. (2016b) found a mean sedimentation rate for Peatball Lake, Alaska, of $\sim 70 \text{ cm ka}^{-1}$ for the last 1,400 years. The BD, TOC, C/N ratio and $\delta^{13}\text{C}$ of the sediment core BAL16-UPL1-L1 show minimal variation, suggesting a stable depositional environment. Therefore, a hiatus in the sediment is unlikely.

4.1.2 Organic carbon quantity

To estimate future release of greenhouse gases from permafrost soils, the thaw-vulnerable OC pool needs to be identified. The organic carbon quantity is identified by analyzing the TOC and estimating the OC budget. The TOC content of the yedoma deposits is comparable to TOC values reported in previous yedoma studies from across Siberia and Alaska (Figure 5 Figure 6). The thermokarst lake sediments have the highest TOC of the three landscape units, which is likely due to the addition of organic matter from lake primary production, as well as the integration of organic matter from its catchment. The DTLB deposits cover a large range of TOC values, which corroborates the findings from section 4.1.1 that the deposits are a mixture of reworked and new material. The TOC content of the yedoma deposits is comparable to TOC values reported in previous yedoma studies from across Siberia and Alaska (Figure S6) (Schirmer et al., 2008a, 2008b, 2011; Strauss et al., 2012, 2013, 2015). Although the mean TOC content of the yedoma deposits is relatively low compared to that of DTLB deposits and thermokarst lake sediments, the volume of the yedoma deposits is of importance for the OC pool size, due to the large spatial coverage and thickness of the yedoma deposits is are large and thus a significant OC pool is still expected.

The thermokarst lake sediments ($\sim 93 \text{ kg m}^{-3}$) have the highest volumetric OC pool compared to the yedoma deposits and DTLB deposits. The DLTB deposits ($\sim 35 \text{ kg m}^{-3}$) contain four times as much OC by volume as yedoma deposits ($\sim 8 \text{ kg m}^{-3}$)

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and . However, the volumetric OC pool of the DTLB deposits more than two times as much (DTLB: $\sim 37 \text{ kg m}^{-3}$, yedoma: ~ 15) is more than twice as high as the yedoma OC pool ($\sim 15 \text{ kg m}^{-3}$), when excluding WIV. Multiple studies have been carried out to estimate OC pool size of deep yedoma and DTLB deposits. Here, we compare the volumetric OC pool to that of other studies excluding WIV, to compare the OC content of the frozen sediment only. Zimov et al. (2006) found that yedoma deposits contain on average 18.5 kg m^{-3} which is comparable to the yedoma deposits of Baldwin Peninsula. Furthermore, the DTLB OC pool of Baldwin Peninsula is in the range of 35.5 to $86.2 \text{ kg soil OC m}^{-3}$ that Mueller et al. (2015) estimated for DTLB deposits in the Alaskan north slope region. Schirmer et al. (2011) (Northeast Siberia) (DTLB: 53.5 , yedoma: 21.1 - 33.2 kg m^{-3}) and Strauss et al. (2013) (total yedoma region, $\sim 1,387,000 \text{ km}^2$) (DTLB: ~ 33 , yedoma: $\sim 19 \text{ kg m}^{-3}$) both estimated the OC budget of deep permafrost deposits (25 m deep) and found that DTLB deposits contain approximately three-1.5 to 2 times as much OC as yedoma deposits (Figure 7) (including excluding WIV). Shmelev et al. (2017) found a slightly higher volumetric OC budget for thermokarst deposits (14.2 kg m^{-3}) than for yedoma deposits (12.6 kg m^{-3}). Zimov et al. (2006) found that yedoma deposits contain on average 18.5 kg m^{-3} (excluding WIV) which is comparable to the yedoma deposits of Baldwin Peninsula. Furthermore, Mueller et al. (2015) estimated DTLB deposits in the Alaskan north slope region.

On the other hand, Webb et al. (2017) also estimated the OC pool of Siberian deep deposits (0-15 m) and found that the yedoma deposits contained more OC (7.9 to 21.6 kg m^{-3}) than the DTLB deposits (6.9 to 14.5 kg m^{-3}) (excluding WIV). Other studies in Siberia (Fuchs et al., 2018; Siewert et al., 2016) also found more OC stored in yedoma deposits than in DTLB deposits. However, the estimates of these studies were based on near-surface sediments (0-2 m).

Based on our landscape unit map we scaled the total OC pool of the Baldwin Peninsula. About 70% of the area of Baldwin Peninsula is affected by permafrost degradation (e.g. thermokarst processes or coastal erosion) and is therefore classified as thermokarst. Moreover, these thermokarst processes led to more than 10 m of ground subsidence on the Baldwin Peninsula, as suggested by relief differences between yedoma uplands and DTLB. The estimated OC pool of the frozen sediments (yedoma 0-15 m depth, DTLB 0-5 m depth) on Baldwin Peninsula is $\sim 68 \text{ Mt}$, of which roughly three quarters ($\sim 52 \text{ Mt}$) is stored in DTLB deposits and one quarter ($\sim 16 \text{ Mt}$) in yedoma deposits. This high amount of OC stored in an area of approximately 450 km^2 shows the important contribution of these deep thermokarst affected yedoma deposits to pan-arctic soil organic carbon stock estimations.

4.2 Organic carbon quality

Permafrost conditions limit decomposition of OC, thereby preserving the quality of the OC. The OC quality is identified by analyzing the OC composition of the sediments. The source of OC influences both the quantity as well as the quality of OC. The ACL₂₃₋₃₃ for both the yedoma (>28.2) and DTLB (>27.7) deposits suggests that the organic matter is mainly derived from terrestrial higher plants. Additionally, in all samples, the long- and odd-chains dominate, which is also typical for a terrestrial higher plant origin (Eglinton and Hamilton, 1967).

The combination of the C/N ratio and $\delta^{13}\text{C}$ value has been widely used as indicator of OC source. A higher C/N ratio suggests an enhanced input of terrestrial land plants whereas algal-produced matter is generally characterized by a lower C/N ratio. The

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C/N ratio and $\delta^{13}\text{C}$ value of OC furthermore allow to distinguish between marine and lacustrine algae, where marine algae generally have a higher $\delta^{13}\text{C}$ values (e.g. Lenz et al., 2016a; Meyers, 1994, 1997). Moreover, the C/N ratio and $\delta^{13}\text{C}$ value have been used as an indicator of OC decomposition, where a lower C/N ratio and higher $\delta^{13}\text{C}$ value indicate further degraded material (i.e. lower quality) (Gentsch et al., 2015; Gundelwein et al., 2007; Schädel et al., 2014; Weiss et al., 2016) (e.g. Gundelwein et al., 2007; Schädel et al., 2014).

Figure 5 shows a scatter plot of the C/N ratio and $\delta^{13}\text{C}$ for the three landscape units of this study (BP) and other studies from Alaska: yedoma deposits along the Itkillik River (IR) (Lapointe et al., 2017; Strauss et al., 2012), DTLB deposits from the Northern Seward Peninsula (NSP) (Lenz et al., 2016c) and thermokarst lake sediments from lakes in the Kobuk River Delta (KOB) and Central Seward Peninsula (CSP) (Lenz et al., 2018). We found significant statistical differences ($p < 0.05$) in almost all tests attempting to differentiate between OC composition in yedoma, DTLB and thermokarst lake deposits of the Baldwin Peninsula and the other Alaskan studies based on the C/N ratio and the $\delta^{13}\text{C}$ value (Supplement 2.6). A trend exists from a low C/N ratios and high $\delta^{13}\text{C}$ values in yedoma deposits towards a high C/N ratios and low $\delta^{13}\text{C}$ values in the thermokarst lake sediments (Figure 5). Intermediate C/N ratios and $\delta^{13}\text{C}$ values were found for DTLB deposits. The high $\delta^{13}\text{C}$ signal and low C/N ratio for the yedoma exposure (BAL16-B2) is typical (Dutta et al., 2006; Sánchez-García et al., 2014; Vonk et al., 2013), since it is characteristic for stadial periods with decreased productivity and a dry climate (Schirmeister et al., 2011, 2013). The C/N ratio and $\delta^{13}\text{C}$ value of the DTLB deposits show a large range, which reflects the mixed character of the disturbed landscape: these deposits contain on the one hand reworked OC (e.g. from former yedoma deposits) and on the other hand fresh OC (e.g. from thermokarst lake production). OC in thermokarst lake sediments is generally characterized by a low $\delta^{13}\text{C}$ value (Cohen, 2003; Meyers, 1994), which is also the case here (Figure 5). The high C/N ratio of the thermokarst lake sediments likely is an indication that the OC is of high quality due to contribution of fresh OC (Schirmeister et al., 2013; Strauss et al., 2015). Hence, the trends of the C/N ratio and $\delta^{13}\text{C}$ values (Figure 5) between the landscape units can be explained mainly by a combination of the source and quality of the OC.

Comparing the properties of the OC pools per landscape unit at Baldwin Peninsula with the Alaskan study sites in Figure 5 shows that the yedoma from Baldwin Peninsula does not have a significantly different C/N ratio or $\delta^{13}\text{C}$ value than that at Itkillik River ($p > 0.05$; Supplement 2.6) which suggests that the OC in both yedoma deposits has a similar composition. The DTLB deposits of Baldwin Peninsula have a significantly higher C/N ratio ($p < 0.05$, 0.01) compared to Northern Seward Peninsula, but a similar $\delta^{13}\text{C}$ ($p > 0.05$). The lower C/N ratio at Northern Seward Peninsula can be explained by the multiple lake generations this basin went through, leading to generally more degraded OC (Lenz et al., 2016a). Given that the C/N ratios and $\delta^{13}\text{C}$ values among thermokarst lakes at Baldwin Peninsula and Central Seward Peninsula are significantly indistinguishable ($p > 0.05$), we argue that general contributions of lake production and thawed sediments from below are comparable, resulting in a similar OC composition. The thermokarst lake in Kobuk River Delta, however, is significantly different in both proxies compared to Baldwin Peninsula and Central Seward Peninsula ($p < 0.05$, 0.01). The lower $\delta^{13}\text{C}$ value at the Kobuk River Delta can be an indication that the lacustrine contribution to the OC pool is higher than in Baldwin Peninsula and Central Seward Peninsula. Nonetheless, the thermokarst lake sediments from Baldwin Peninsula, Kobuk River Delta and

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Central Seward Peninsula all have a relatively high C/N ratios and low $\delta^{13}\text{C}$ -values, compared to the yedoma and DTLB deposits (Figure 5). The similar composition of the two yedoma study sites suggests a lack of decomposition, thereby implying the undisturbed character of the deposits. Furthermore, the differences of the DTLB study sites are likely caused by further OC composition in the Northern Seward Peninsula basin which could have occurred during multiple lake generations the basin went through. These findings suggest that the range of the C/N and $\delta^{13}\text{C}$ within the study sites can be explained mainly by the quality of the OC.

A significantly higher CPI₂₃₋₃₃ in yedoma compared to DTLB deposits (p-value<0.05) indicates that the OC in the yedoma deposits is less degraded and, therefore, suggests that this pool has a higher potential for future OC decomposition. Furthermore, the decreasing trend in CPI₂₃₋₃₃ in the DTLB deposits indicates progressive degradation with depth, whereas the lack of such a trend in the yedoma deposits indicates that minimal or no decomposition has occurred before or after the organic matter was incorporated into the permafrost, maintaining its high quality (Stapel et al., 2016; Strauss et al., 2015; Weiss et al., 2016). These findings suggest that the OC in yedoma deposits BAL16-B2 is of higher quality than that in the DTLB deposits BAL16-B4. Previous studies reported relatively high carbon turnover rates of Alaskan yedoma deposits (Dutta et al., 2006; Lee et al., 2012; Zimov et al., 2006), which suggest the presence of easily degradable organic matter in these deposits, which corroborates our findings.

Schuur et al. (2009) showed that in thermokarst affected permafrost soils, old OC is released more strongly. Also, it was shown that the formation of mineral-organic associations can be an important factor influencing bioavailability of OC in permafrost soils, because it can improve the stability of OC in soils (Gentsch et al., 2015; Gundelwein et al., 2007; Höfle et al., 2013). Vonk et al. (2010) also showed the presence of this altered mineral bound organic matter component in Arctic river and marine sediments. However, it is not clear yet how large the share of mineral-associated organic matter in permafrost soils is and how it exactly influences the stability in high-latitude soils (Höfle et al., 2013; Mueller et al., 2015).

Regardless, to evaluate implications of permafrost degradation arising from future climate change, it is necessary to assess the vulnerability of the total OC pool. Climate change will increase the frequency and intensity of fires and floods which can lead to soil removal and disturbances of the ground thermal regime which can result in rapid local permafrost degradation (Grosse et al., 2011). Because of the high ice content in the yedoma and DTLB deposits on Baldwin Peninsula (>45 vol%, excluding WIV), the deposits are highly susceptible and vulnerable to deep permafrost thaw which will have a great effect on the topography. With the formation of new, deep lakes, primary productivity is expected to increase on a large scale, which will possibly compensate for increased greenhouse emissions initially. Lindgren et al. (2016) found a net increase in total lake surface area by 3.9% for the Baldwin Peninsula between 1972 and 2014. With the formation of new, deep lakes, primary productivity is expected to increase on a large scale, which will possibly compensate for increased greenhouse emissions initially. However, following the positive permafrost feedback loop, more intensive permafrost degradation will likely lead to an increase of the number of lake drainage events in West-Alaska (Lindgren et al., 2016). This could lead to rapid OC sequestration in new permafrost aggrading in DTLBs and evolving terrestrial peat, but will be followed by ultimate decomposition once this region, which is ~80 km from the discontinuous permafrost boundary (Brown et al., 1997), is affected

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by widespread permafrost near-surface thaw between 2050 and 2100 (Lawrence and Slater, 2005; Walter Anthony et al., 2014). Although the largest share of OC in frozen deposits on Baldwin Peninsula is stored in DTLB deposits (~75%), [we showed that](#) the OC in yedoma deposits is of higher quality and therefore, the OC in the yedoma is especially vulnerable to future microbial degradation and greenhouse gas release which will further enhance climate warming.

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5 Conclusion

This study presents OC characteristics from ice-rich permafrost deposits and thermokarst lake sediments on the Baldwin Peninsula, West-Alaska. Using cryostratigraphical, biogeochemical and biomarker parameters of yedoma deposits and DTLB deposits as well as thermokarst lake sediments, the size and quality of OC pools in ice-rich permafrost were identified. The [first estimate of the](#) OC pool [size](#) of the frozen deposits in a 450 km² study on the Baldwin Peninsula is ~68 Mt, of which three quarters are stored in frozen DTLB deposits. The lake sediments had the highest volumetric OC content of ~93 kg OC m⁻³ compared to yedoma (~8 kg OC m⁻³) and DTLB deposits (~35 kg OC m⁻³). Biogeochemical and biomarker parameters indicated that the OC in the yedoma deposits is best preserved and of higher quality than the OC stored in DTLB deposits and thermokarst lake sediments, demonstrating the higher potential for OC decomposition in yedoma deposits.

Data publishing

15 The data presented in this study [will be](#) available on PANGAEA [after submission](#) (Jongejans et al., 2018).

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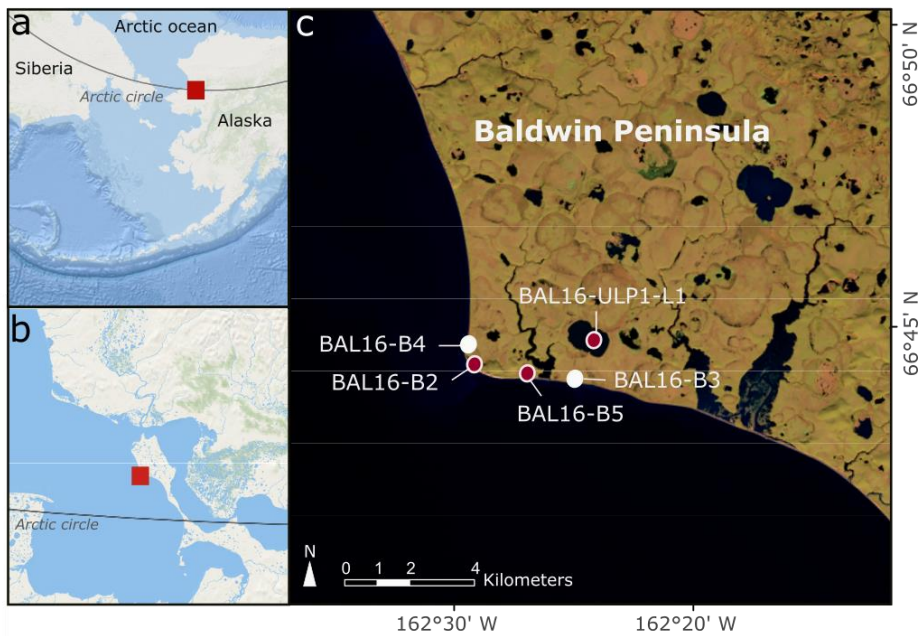


Figure 1: (a) Overview map of Siberia and Alaska with indicated study region (red square). (b) Close-up of Kotzebue Sound with indicated study region (red square). (c) Study sites on Baldwin Peninsula presented in this study (purple filled dots) and additional sites used in the organic carbon pool calculations (white dots); false color image with short-wave infrared, near-infrared and deep blue bands (7-5-1). Source for background map in (a) and (b): World Ocean Base (ESRI); satellite image map (c): Landsat 8 image from 13 Oct. 2016, USGS/NASA.

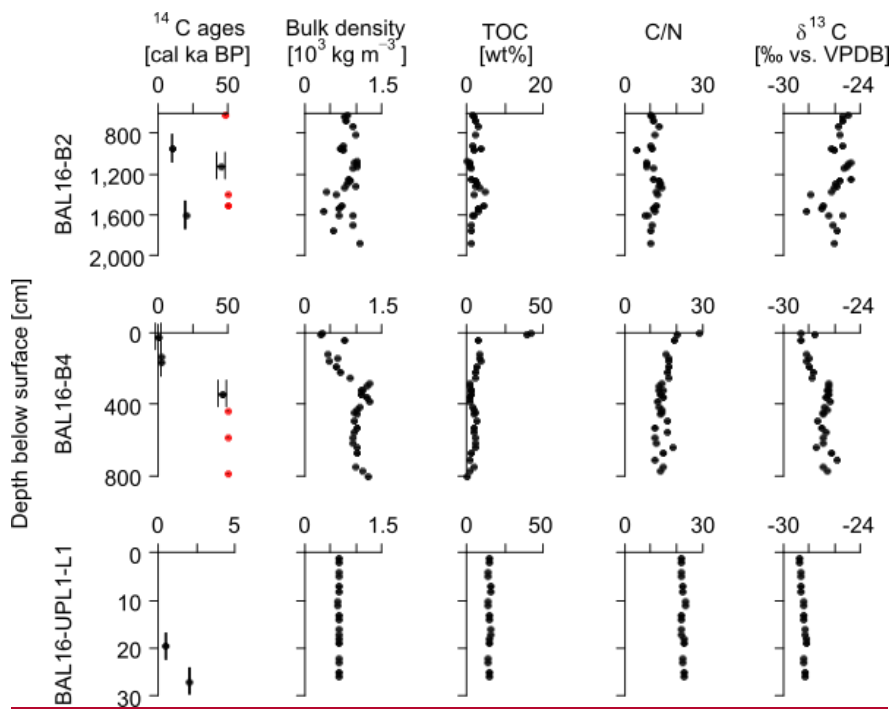
Table 1: Overview of study sites of yedoma exposure, drained thermokarst lake basin (DTLB) exposure and thermokarst lake sediments. BAL: Baldwin Peninsula, 16: expedition in 2016, B: bluff sampling, UPL: upland sampling, L: lake sampling.

<u>Study site ID</u>	<u>Landscape unit</u>	<u>Coordinates</u>	<u>Samples, n</u>
BAL16-B2	Yedoma exposure	66.73262°N, 62.49450°W	18
BAL16-B4	DTLB exposure	66.73644°N, 62.50208°W	31
BAL16-UPL1-L1	Thermokarst lake sediments	66.74220°N, 62.41310°W	9

Table 2: Radiocarbon dates of BAL16-B2 ([vedoma](#)), BAL16-B4 ([drained thermokarst lake basin](#)) and BAL16-UPL1-L1 ([thermokarst lake](#)). Calibrations were performed using CALIB 7.1 software and the IntCal13 calibration curve (Stuiver et al., 2017). \pm : [standard deviation](#), Poz: Poznan Radiocarbon Laboratory, Poland, pMC: percent modern carbon.

Sample ID	Ext. ID	Depth [cm]	¹⁴ C ages [a BP]	\pm	Mean	\pm	Rounded ¹⁴ C ages [cal a BP]	\pm
					eCalibrated ages 2 σ (95.4 %) [a BP]			
BAL16-B2-20	Poz-89527	620	> 48,000					
BAL16-B2-26	Poz-89700	945	8,890	50	10,037.5	153 5307	10,000	150 310
BAL16-B2-31	Poz-89702	1117	40,810	1800	44,747	312 56250	44,700	313 06250
BAL16-B2-39	Poz-89703	1399	> 50,000					
BAL16-B2-1	Poz-89526	1500	> 50,000					
BAL16-B2-5	Poz-89523	1600	16,200	90	19,551.5	287 5575	19,600	290 580
BAL16-B4-2a	Poz-89704	22	105.7	0.33 pMC	239	153 0	240	203 0
BAL16-B4-4a	Poz-89705	132	1,700	30	1,590.5	46 593	1,590	50 90
BAL16-B4-6a	Poz-89706	166	2,125	30	2,079.5	77 5155	2,080	80 160
BAL16-B4-14a	Poz-89707	340	42,800	1,600	46,361.5	2967 55935	46,400	3000 6000
BAL16-B4-18a	Poz-89708	434	> 50,000					
BAL16-B4-24a	Poz-89709	583	> 50,000					
BAL16-B4-31a	Poz-89710	788	> 50,000					
BAL16-UPL1-L1-A	Poz-89349	19-20	425	30	482	438 6	480	409 0
BAL16-UPL1-L1-B	Poz-89351	26.5-27.5	1,940	30	2,014.5	64 5129	2,015	601 30

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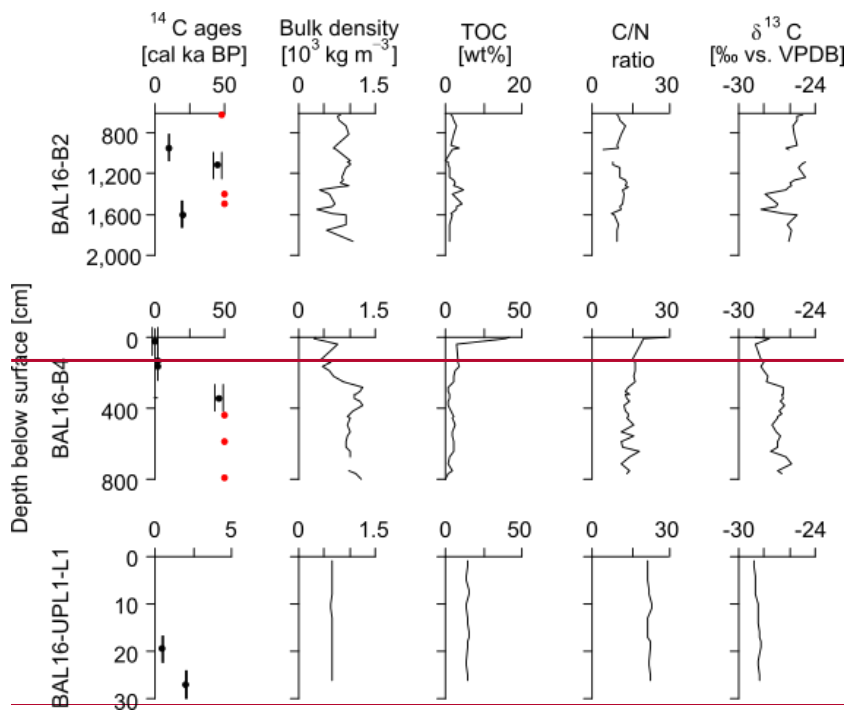


Figure 2: Summary of cryostratigraphical and biogeochemical parameters of BAL16-B2 (yedoma), BAL16-B4 (drained thermokarst lake basin) and BAL16-UPL1-L1 (thermokarst lake): calibrated radiocarbon ages (^{14}C ; infinite ages in red), bulk density, total organic carbon (TOC), total organic carbon-total nitrogen (C/N)-ratio (C/N), stable carbon isotopes ($\delta^{13}\text{C}$). Note: different x-axes for ^{14}C and TOC.

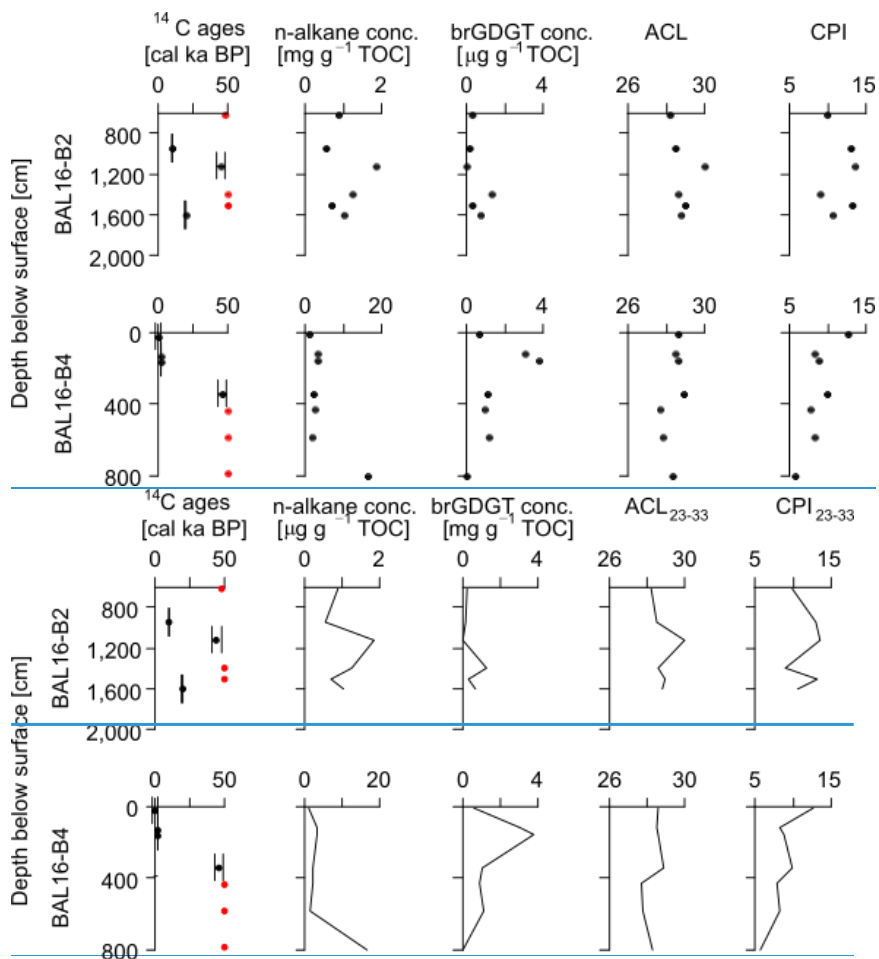


Figure 3: Summary of biomarker parameters of BAL16-B2 (yedoma) and BAL16-B4 (drained thermokarst lake basin): calibrated radiocarbon ages (^{14}C ; infinite ages in red), n-alkane concentration, brGDGT concentration, average chain length (ACL) and carbon preference index (CPI). ACL and CPI were calculated from n-alkane range C_{23-33} . Note: radiocarbon ages are the same as in Figure 2; different x-axes for n-alkane concentration.

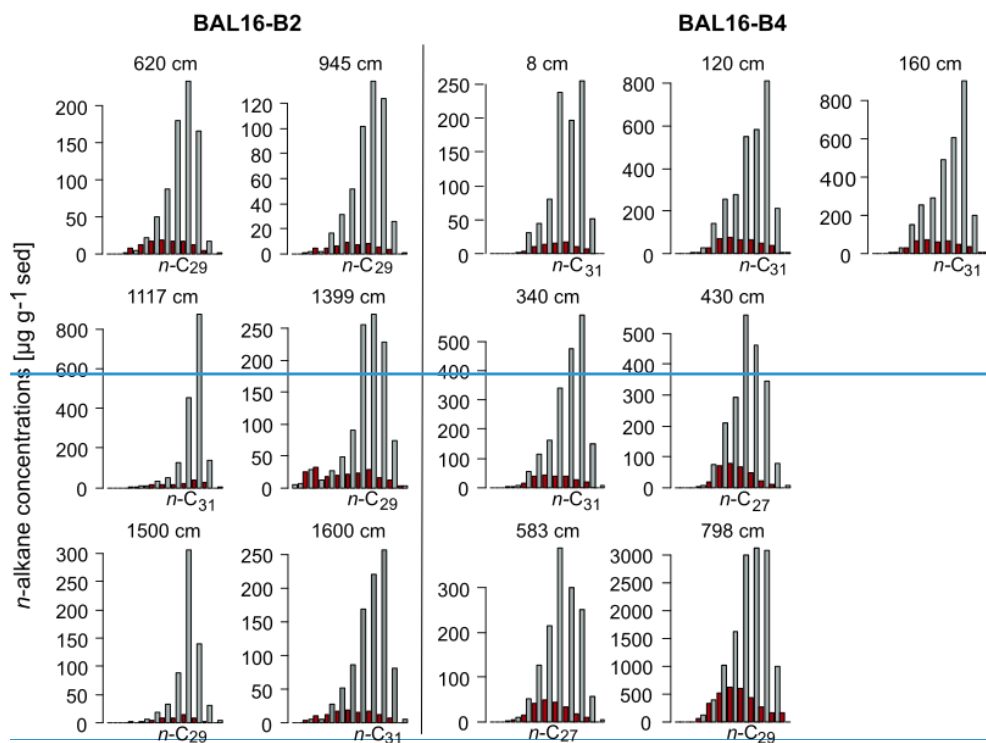


Figure 4: *n*-alkane concentrations of BAL16-B2 (yedoma) and BAL16-B4 (drained thermokarst lake basin) by depth. Odd chains (grey-bars) and even chains (red bars), sample depth and dominating *n*-chain indicated. Note: different y-axes.

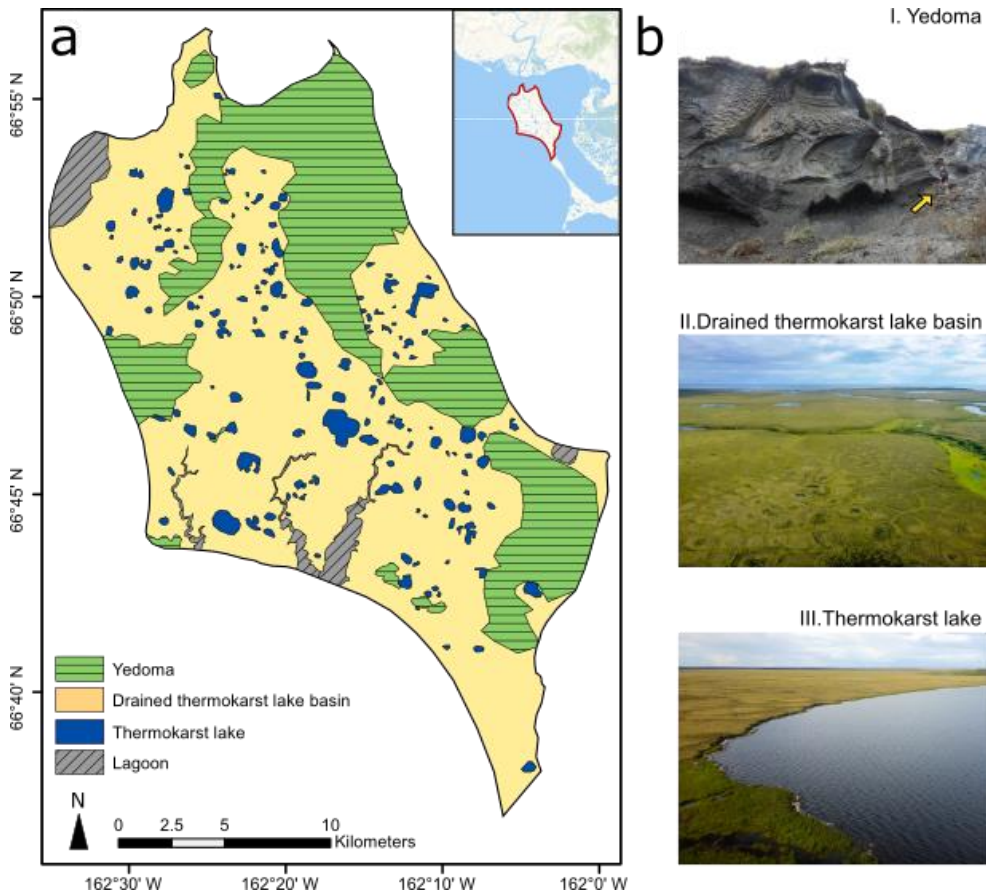


Figure 4: a) Land cover classification map of Baldwin Peninsula (see overview map in upper-right corner) with yedoma (green), drained thermokarst lake basins (yellow), thermokarst lakes (blue) and lagoons (grey). B) Exemplary photos: I. Yedoma (note person on the right indicated by yellow arrow for scale), II. Drained thermokarst lake basin and III. Thermokarst lake. Source for overview map in (a): World Ocean Base (ESRI).

Table 3: Input (left) and output parameters (right) for organic carbon pool calculations for yedoma, drained thermokarst lake basin and thermokarst lake deposits: deposit thickness, landscape coverage, wedge-ice volume (WIV), average weighted bulk density * total organic carbon (BD*TOC). WIV data from Ulrich et al. (2014).

Landscape unit	Thickness [m]	Coverage [m ²]	WIV [vol %]	Volumetric	Total	Volumetric	Total
				OC pool Incl. WIV [kg m ⁻³]	OC pool Incl. WIV [Mt]	OC pool excl. WIV [kg m ⁻³]	OC pool excl. WIV [Mt]
Yedoma	15	136,620,000	48	8.0 ± 0.4 <u>0.8</u>	16.3 ± 0.9 <u>1.7</u>	15.3 ± 0.8 <u>1.6</u>	31.4 ± 1.7 <u>3.3</u>
DTLB	5	296,440,000	7	34.7 ± 1.4 <u>2.9</u>	51.5 ± 2.2 <u>2.3</u>	37.4 ± 1.6 <u>3.1</u>	55.4 ± 2.3 <u>4.6</u>
Thermokarst lake	2	21,180,000	0			92.9 ± 0.4 <u>0.8</u>	3.9 ± 0.0

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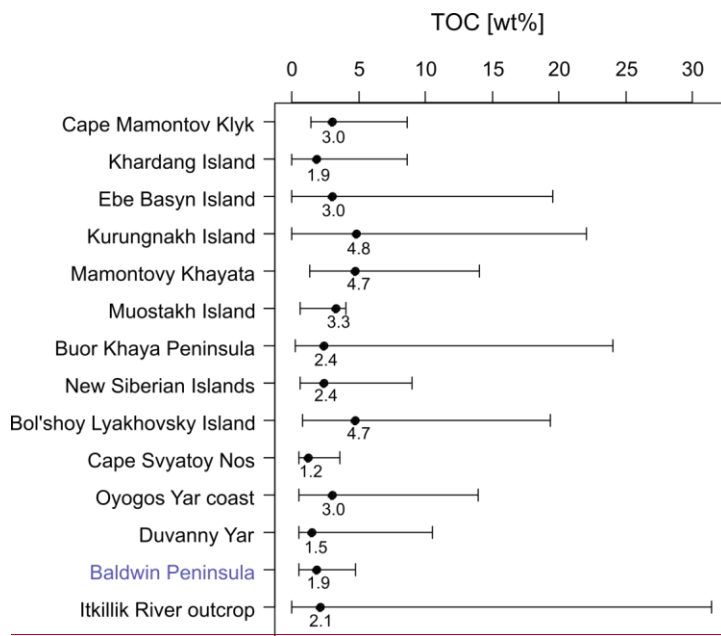


Figure 56: TOC variations from different yedoma study sites in Siberia and Alaska. Sorted from westernmost (Cape Mamontov Klyk, western Laptev Sea) to easternmost (Itkillik River outcrop, Alaskan North Slope) study sites. Data from Schirrmeister et al. (2008a, 2008b, 2011), Strauss et al. (2012, 2013, 2015) and Baldwin Peninsula (this study; blue).

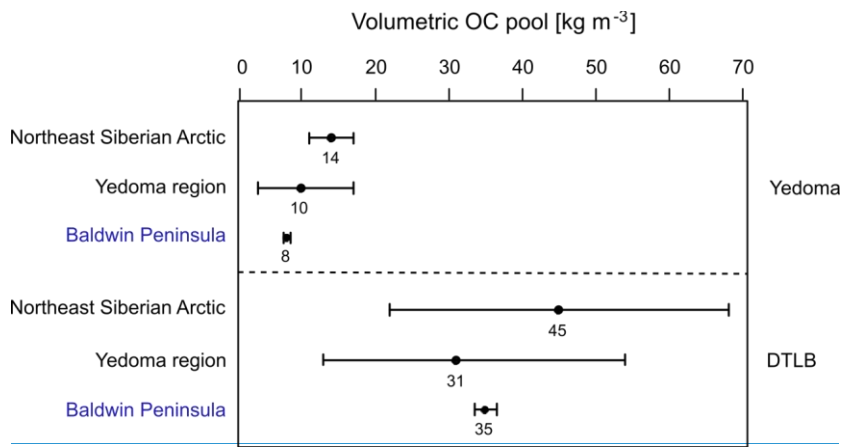


Figure 7: Volumetric OC pool estimates of yedoma deposits (left) and drained thermokarst lake basin (DTLB) deposits (right). Mean values indicated. Data from Northeast Siberian Arctic (Schirmer et al., 2011), the yedoma region in Siberia and Alaska (Strauss et al., 2013) and Baldwin Peninsula (this study; blue).

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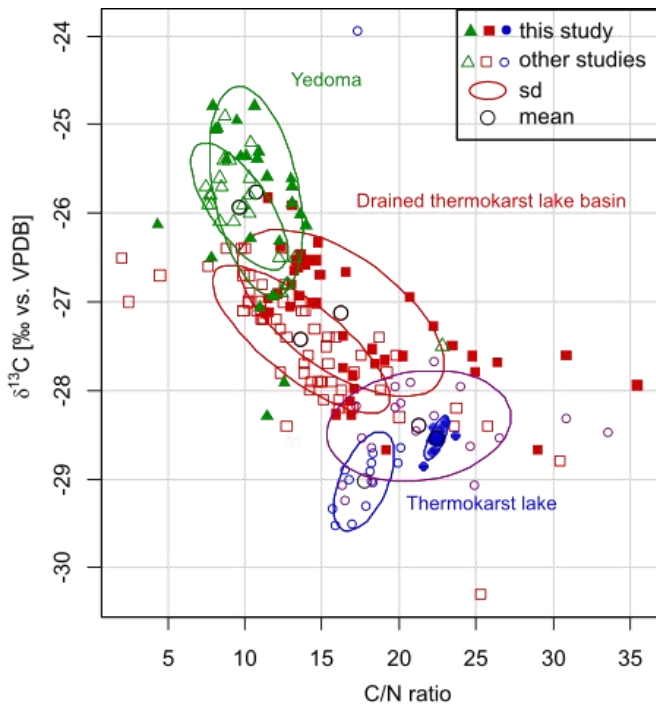


Figure 5: Scatterplot of total organic carbon-total nitrogen (C/N) ratio and stable carbon isotopes ($\delta^{13}\text{C}$) of yedoma deposits (green triangles), drained thermokarst lake basin (DTLB) deposits (red squares) and thermokarst lake sediments (blue and purple dots) in Alaska. Mean value and standard deviation (sd) indicated. Data from this study (filled symbols): Baldwin Peninsula; data from other studies (hollow symbols): Itkillik River (Lapointe et al., 2017; Strauss et al., 2012), Northern Seward Peninsula (Lenz et al., 2016a) and lakes from Kobuk River Delta (blue) and Central Seward Peninsula (purple) (Lenz et al., 2018).

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Supplementary Materials for “Organic carbon characteristics in yedoma and thermokarst deposits on Baldwin Peninsula, West-Alaska”

Loeka L. Jongejans¹, Jens Strauss¹, Josefine Lenz^{1,2}, Francien Peterse³, Kai Mangelsdorf⁴, Matthias Fuchs^{1,5} and Guido Grosse^{1,5}

¹Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research, Periglacial Research Section, Potsdam, Germany

²University of Alaska Fairbanks, Institute of Northern Engineering, Fairbanks, USA

³Utrecht University, Department of Earth Sciences, Utrecht, Netherlands

10 ⁴Helmholtz Centre Potsdam - German Research Centre for Geosciences

⁵University of Potsdam, Institute of Earth and Environmental Sciences, Potsdam, Germany

Correspondence to: Loeka L. Jongejans (loeka.jongejans@awi.de)

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1 Supporting methodology descriptions

1.1 Grain size analysis

The analysis of grain size is important in identifying the origin of sedimentary deposits: grain size distributions of the sedimentary sediments can give insights into the medium of transportation, as well as the depositional mechanism. Grain size was analyzed for yedoma exposure BAL16-B2 and [drained thermokarst lake basinDTLB](#) exposures [BAL16-B3](#), [BAL16-B4](#) and [BAL16-B5](#). In order to only measure the clastic material, organic matter was removed from the samples by treating the samples with hydrogen peroxide (H₂O₂). For this, about 10 ml of sample material was weighed into 400 ml beakers to which 100 ml of 3% H₂O₂ and 4 drops of ammonia were added. The beakers were then placed on a shaker at 60°C for 2-3 weeks. Five times a week, the beakers were cleaned and 10 ml of H₂O₂ was added (once the reaction was less strong 20 ml was added). The pH was kept between 6 and 8 by adding ammonia or acetic acid to allow an optimal reaction. After the complete removal of the organic material, the samples were washed with about one liter of purified water to remove the H₂O₂. The samples were then centrifuged (Cryofuge 8500i for 10 minutes at 5050 RPM, 20°C; Multifuge 3-S Heraeus 2-3 times for 15 minutes at 4000 RPM) and freeze-dried. The samples were manually homogenized and about 1 gram was weighed into 250 ml plastic bottles and a spoon spatula of dispersing agent (tetrasodium pyrophosphate, Na₄P₂O₇) was added. The bottles were filled with an ammonia solution (10 ml NH₄OH in 100 liter of water) and placed in an overhead shaker overnight. As a last step before measuring, the samples were split into 8 homogeneous samples to enable measurement of samples with sediment concentrations between 5-15%. The material was hereby also sieved for >1 mm to avoid damage to the laser. Inorganic particles >1mm were weighed and if the residue was significant compared to the sample, it was included in the distribution afterwards. The grain size was analyzed with the Malvern Mastersizer 3000 laser. The device cleans automatically and measures background scatter. After a sample is inserted into the dispersion unit, it is channeled through the device to the measurement cell where it is exposed to a red laser and blue led (633 nm and 470 nm), of which the scatter is measured by the detectors. The average is calculated of three measurements per split sample (standard deviation <10%). Grain size statistics are calculated using the software GRADISTAT using the grain size scaling from Blott & Pey (2001).

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1.2 Elemental analysis

Total carbon (TC), total nitrogen (TN) and total organic carbon (TOC) are measured based on the principle of combustion chromatography. TC and TN were determined using the Elementar Vario EL III. A subsample of each homogenized sample was weighed into small tin capsules in duplicate. A blank capsule was measured in the beginning for background detection and a calibration run was performed before and after every fifteen samples. The percentage of total carbon and nitrogen was calculated. The determination of TOC was done using the Elementar Vario Max C. Depending on the TC values, fifteen to hundred milligrams of the samples was weighed into crucibles and placed into the machine. TN, TC and TOC are expressed in weight percentage (wt%).

Stable carbon isotopes ($\delta^{13}\text{C}$) could be measured after the removal of carbonates. This was done by treating the samples with 20 ml hydrogen chloride for three hours at 97.7°C. Purified water was added and the samples were decanted and washed three times. When the chloride content was under 500 parts per million, the samples were filtered over a glass microfiber filter (Whatman Grade GF/B, nominal particle retention of 1.0 μm). Afterwards, the residue was dried in a drying cabinet at 50°C. Dry samples were ground manually and weighed into tin capsules. The required sample weight was calculated by dividing 20 by the TOC value. A ThermoFisher Scientific Delta-V-Advantage gas mass spectrometer equipped with a FLASH elemental analyzer EA 2000 and a CONFLO IV gas mixing system was used to determine the $\delta^{13}\text{C}$. In this system, the sample is combusted at 1020°C so that the OC is transferred to CO_2 , after which the isotope ratio is determined relative to a laboratory standard of known isotopic composition. Capsules for control and calibration were run in between. The unit is per mille (‰) and the ratio is compared to the standard established from the Pee Dee Belemnite (VPDB: a limestone formation of which the ratio was set to 0).

1.3 Lipid biomarker analysis

From the GDGT concentration, the branched and isoprenoid tetraethers (BIT) index was calculated following Eq. (1).

$$BIT = \frac{Ia+IIa+IIIa}{Ia+IIa+IIIa+crenarchaeol} \quad (1)$$

The BIT index is the ratio between the mainly terrestrially produced brGDGTs and crenarchaeol (Figure S1), an isoprenoid GDGT which is most abundant in marine and lacustrine environments. Apart from the use of this proxy to distinguish between terrestrial and marine sources (Hopmans et al., 2004), the ratio is correlated to precipitation (Dirghangi et al., 2013). The methylation of branched tetraethers (MBT) index is the ratio between brGDGT-I and -II structures and was calculated according to Peterse et al. (2012) following Eq. (2).

$$MBT = \frac{Ia+Ib+Ic}{Ia+Ib+Ic+IIa+IIb+IIc+IIIa} \quad (2)$$

This ratio can be used in paleoreconstructions. Weijers et al. (2007) found that Arctic soils are generally dominated by brGDGTs with additional methyl branches and suggested that more methyl branches are formed in lower temperatures.

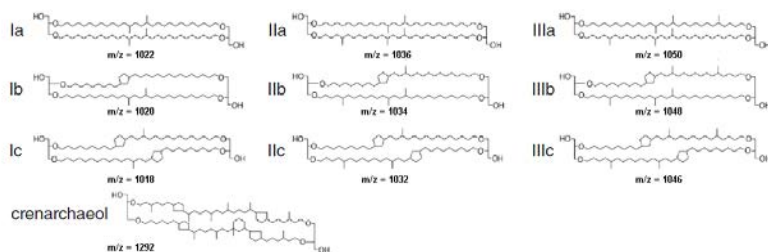


Figure S1: Molecular structures of brGDGTs and crenarchaeol with mass-to-charge ratio (m/z). The structures differ in mass by the presence of cyclopentane moieties (a, b, c) and the number of methyl branches (I, II, III). From Peterse et al. (2014).

2 Supporting figures and tables

2.1 Photographs field sites

Photographs of the field sites are shown for the yedoma exposure BAL16-B2 (Figure S2), [drained thermokarst lake basin](#) exposures BAL16-B3 (Figure S3) and BAL16-B4 (Figure S4) and the thermokarst lake core BAL16-UPL-L1 (Figure S5).

a



b



Figure S2: Yedoma exposure BAL16-B2 from the front (a) and side (b). Photos by J. Strauss, August 2016.



Figure S3: Drained thermokarst lake basin exposure BAL16-B3. Photo by J. Strauss, August 2016.



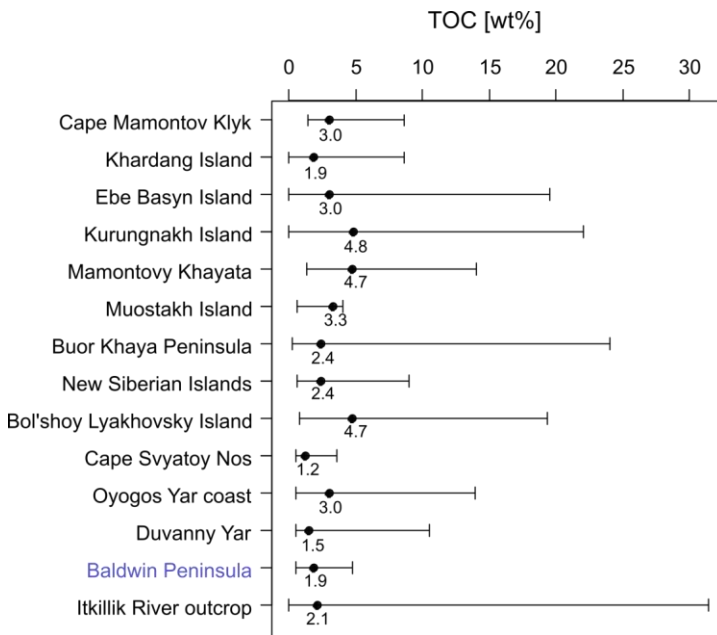
Figure S4: Drained thermokarst lake basin exposure BAL16-B4 from the front (a) and side (b). Photos by J. Strauss, August 2016.



Figure S5: Thermokarst lake core BAL16-UPL1-L1. Photo by J. Lenz, December 2016.

2.2 Total organic carbon

Figure S6 shows variations in total organic carbon of previously studied yedoma deposits in Siberia and Alaska.



5 **Figure S6: Total organic carbon (TOC) variations from different yedoma study sites in Siberia and Alaska. Sorted from westernmost (Cape Mamontov Klyk, western Laptev Sea) to easternmost (Itkillik River outcrop, Alaskan North Slope) study sites. Data from Schirrmeister et al. (2008a, 2008b, 2011), Strauss et al. (2012, 2013, 2015) and Baldwin Peninsula (this study; blue).**

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2.3 Additional profiles

Figure S7 shows the cryostratigraphical and biogeochemical parameters of the additional [drained thermokarst lake basin](#) exposures BAL16-B3 and BAL16-B5 that were used in the organic carbon calculations.

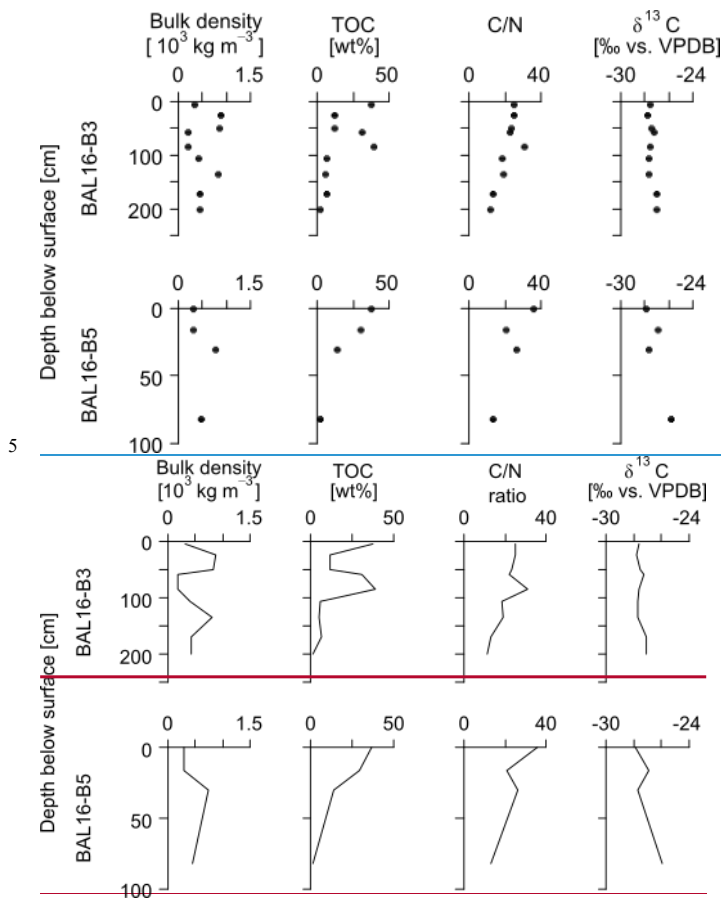


Figure S7: Summary of cryostratigraphical and biogeochemical parameters of BAL16-B3 and BAL16-B5 (drained thermokarst lake basin exposures): bulk density, total organic carbon (TOC), total [organic carbon](#)-total nitrogen ([C/N](#))-ratio ([C/N](#)), stable carbon isotopes ($\delta^{13}\text{C}$).

2.4 *n*-Alkane concentration

The *n*-alkane concentrations per sample are shown in Figure S8. Also, the dominating *n*-C chain is indicated.

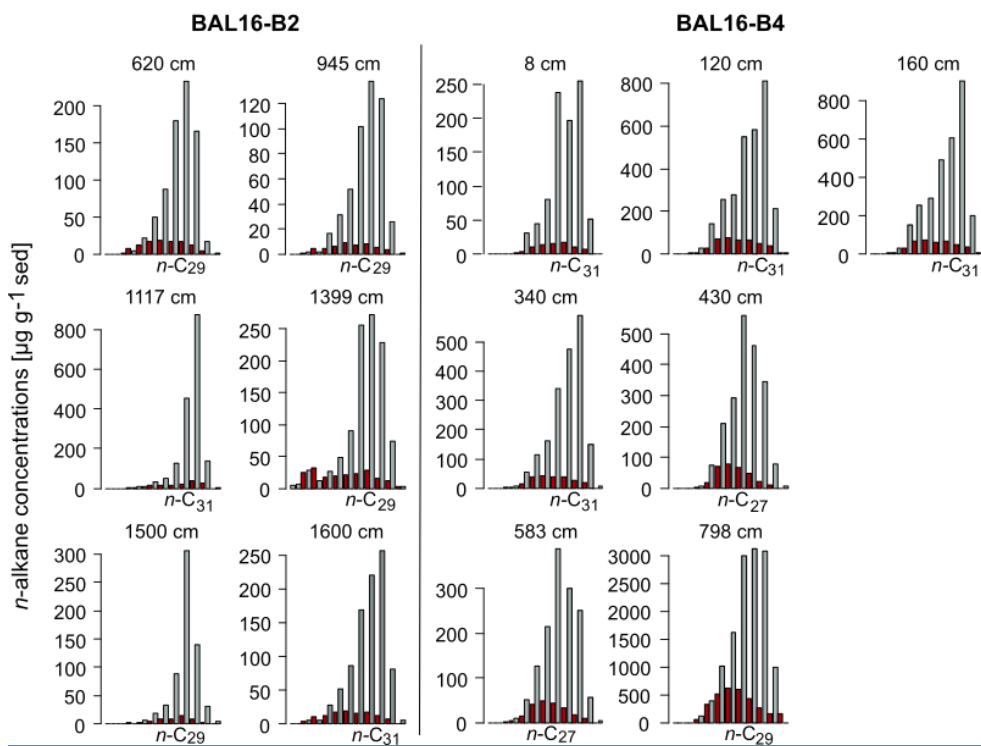
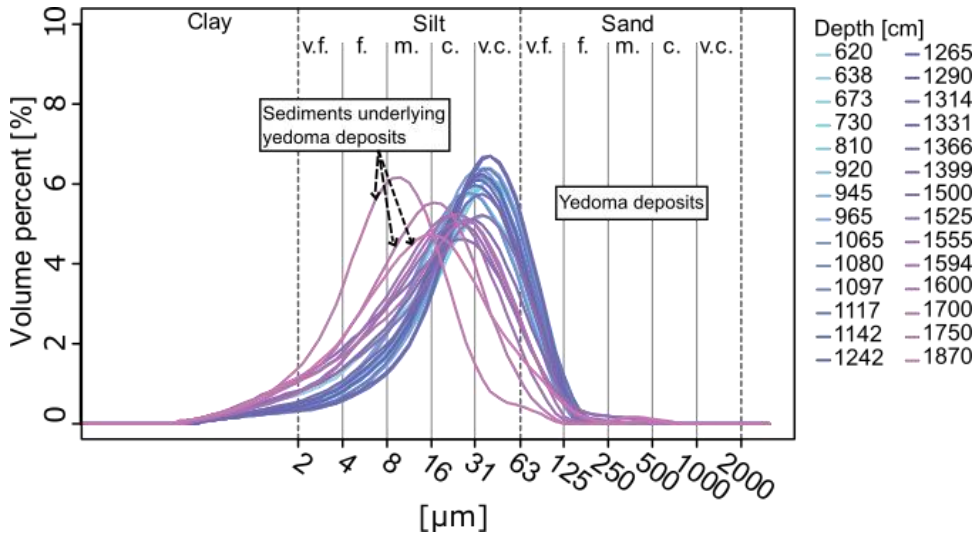


Figure S8: *n*-alkane concentrations of BAL16-B2 (vedoma) and BAL16-B4 (drained thermokarst lake basin) by depth. Odd chains (grey bars) and even chains (red bars), sample depth (above graph) and dominating *n*-chain indicated (below x-axis). Note: different y-axes.

2.4.2.5 Depositional environment

2.4.12.5.1 Grain size distribution

The grain size distributions of yedoma exposure BAL16-B2 and [drained thermokarst lake basin](#) exposure BAL16-B4 are shown in Figure S9 and Figure S10, respectively.



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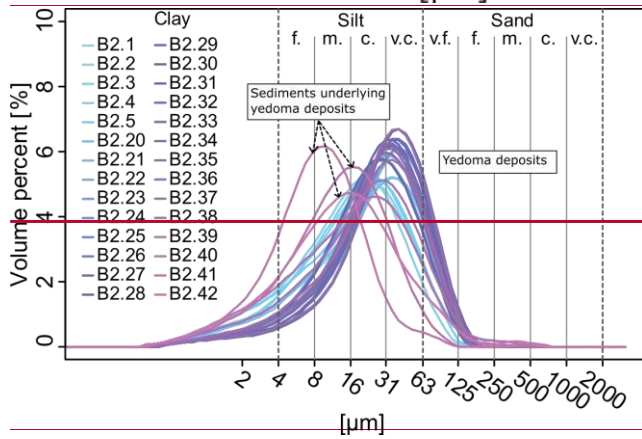


Figure S9: Grain size distribution of ~~yedoma exposure~~ BAL16-B2 (~~yedoma~~). Samples sorted over depth (legend on the right). v.f.: ~~very fine~~, f.: ~~fine~~, m.: ~~medium~~, c.: ~~coarse~~, v.c.: ~~very coarse~~. Sediments from the separate unit underlying the yedoma deposits indicated.

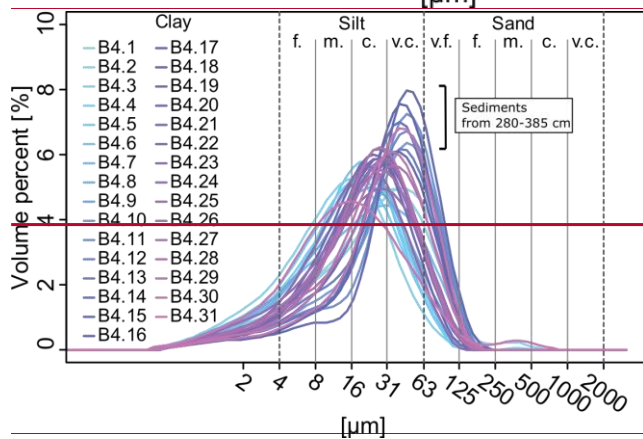
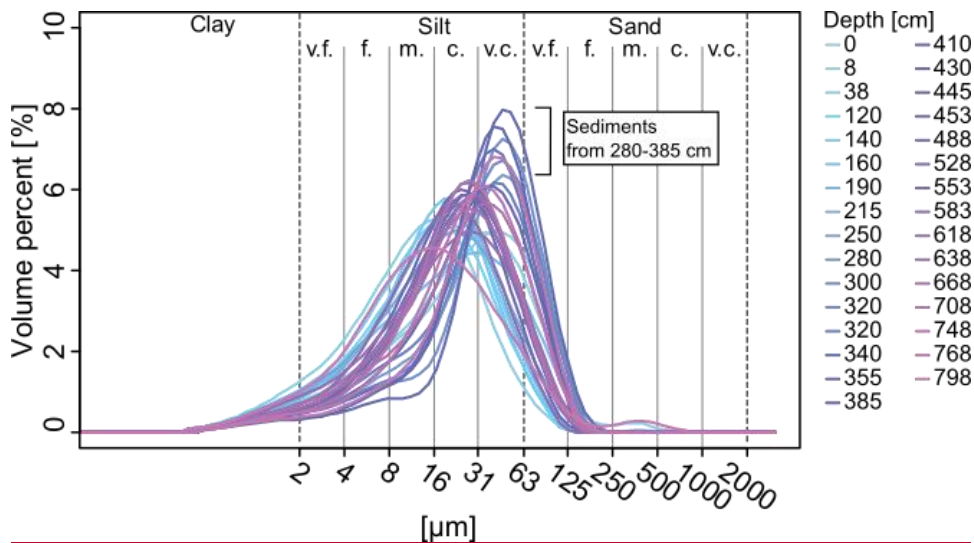


Figure S10: Grain size distribution of ~~drained thermokarst lake basin~~ BAL16-B4 (~~drained thermokarst lake basin~~). Samples sorted over depth (legend on the right). v.f.: ~~very fine~~, f.: ~~fine~~, m.: ~~medium~~, c.: ~~coarse~~, v.c.: ~~very coarse~~. Sediments from depth interval of 280-385 cm indicated.

2.4.22.5.2 Climatic indicators

Table S1 shows the brGDGT derived climatic indices BIT and MBT indices.

5 **Table S1: brGDGT derived climatic indices for yedoma-exposure-BAL16-B2 (yedoma) and DTLB-exposure-BAL16-B4 (drained thermokarst lake basin): branched and isoprenoid tetraethers (BIT) index and methylation of branched tetraethers (MBT) index.**

	Depth	BIT	MBT
	[cm]		
yedoma	620	0.90	0.19
	945	0.96	0.19
	1117	NA	NA
	1399	1.00	0.39
	1500	0.97	0.20
	1600	0.88	0.18
DTLB	8	1.00	0.25
	120	1.00	0.27
	160	1.00	0.30
	340	0.99	0.25
	430	1.00	0.22
	583	1.00	0.16
	798	NA	NA

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2.52.6 Statistical tests

Using the Shapiro-Wilk test, we tested for normality of the data (Table S2). In this test, the null hypothesis states that the data are normally distributed. When the p-value exceeds 0.05, the null hypothesis cannot be rejected. Using Mann-Whitney-Wilcoxon and the Kruskal-Wallis test, a non-parametric test, we tested for significant differences of the biogeochemical parameters (bulk density, total organic carbon, carbon-nitrogen ratio and stable carbon isotopes) between the three stratigraphic landscape units on Baldwin Peninsula (Table S2): yedoma, drained thermokarst lake basin and thermokarst lake sediments. In this test, the null hypothesis states that there is ~~a no~~ statistically significant difference between the samples of the different landscape units. ~~When the p-value exceeds 0.05, the null hypothesis cannot be rejected. A Mann-Whitney-Wilcoxon test was added for pairwise comparisons between the landscape units~~ (Table S2).

The Mann-Whitney-Wilcoxon test was also used to test for significant differences based on the C/N ratio (Table S3) and the $\delta^{13}\text{C}$ value (Table S4) between this study (Baldwin Peninsula) and other studies from Alaska: yedoma deposits along the Itkillik River (IR) (Lapointe et al., 2017; Strauss et al., 2012), DTLB deposits from the Northern Seward Peninsula (NSP) (Lenz et al., 2016) and thermokarst lake sediments from lakes in the Kobuk River Delta (KOB) and Central Seward Peninsula (CSP) (Lenz et al., 2018).

Table S2: Outcome ~~statistical tests Mann-Whitney-Wilcoxon test~~ of bulk density (BD), total organic carbon (TOC), carbon-nitrogen (C/N) ratio and stable carbon isotopes ($\delta^{13}\text{C}$) between ~~yedoma (Y) (BAL16-B2 (yedoma)), BAL16-B4 (drained thermokarst lake basin; (DTLB) (BAL16-B4) and BAL16-UPL1-L1 (thermokarst lake) (TL) (BAL16-UPL1-L1) sediments on Baldwin Peninsula.~~

P-value		BD	TOC	C/N	$\delta^{13}\text{C}$
Shapiro-Wilk Test		<0.05	<0.001	<0.01	<0.001
Kruskal Wallis Test		<0.05	<0.001	<0.05	<0.001
Wilcoxon test	Yedoma – DTLB	<0.05	<0.001	<0.001	<0.001
	Yedoma – Thermokarst lake	<0.001	<0.001	<0.001	<0.001
	DTLB – Thermokarst lake	<0.001	<0.001	<0.001	<0.001

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p-value C/N-ratio	DTLB-BP	TL-BP	Y-IR	DTLB-NSP	TL-KOB	TL-CSP
Y-BP	6.365E-11	5.18E-08	0.1391*	0.000244	5.61E-10	2.50E-12
DTLB-BP		0.001287	9.86E-09	0.002569	0.1215*	0.0003273
TL-BP			4.75E-05	4.71E-05	4.02E-06	0.2634*
Y-IR				2.85E-05	7.43E-06	2.2E-07
DTLB-NSP					0.00029	1.12E-07
TL-KOB						0.003451

Table S3: Outcome Mann-Whitney-Wilcoxon test of C/N ratio between yedoma (Y), drained thermokarst lake basin (DTLB) and thermokarst lake (TL) sediments. BP: Baldwin Peninsula (this study), IT: Itkillik River (Strauss et al., 2012), NSP: Northern Seward Peninsula (Lenz et al., 2016), KOB: Kobuk River Delta and CSP: Central Seward Peninsula (Lenz et al., 2018). P-values that exceed 0.05 are indicated with an asterisk (*).

P-value C/N	DTLB BAL16-B4	Thermokarst lake BAL16-UPL1-L1	Yedoma Itkillik River	DTLB Northern Seward Peninsula	Thermokarst lake Kobuk Delta	Thermokarst lake Central Seward Peninsula
Yedoma BAL16-B2	<0.001	<0.001	>0.05	<0.001	<0.001	<0.001
DTLB BAL16-B4		<0.01	<0.001	<0.01	>0.05	<0.001
Thermokarst lake BAL16-UPL1-L1			<0.001	<0.001	<0.001	>0.05
Yedoma Itkillik River				<0.001	<0.001	<0.001
DTLB Northern Seward Peninsula					<0.001	<0.001
Thermokarst lake Kobuk Peninsula						<0.01

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p-value $\delta^{13}\text{C}$	DTLB-BP	TL-BP	Y-IR	DTLB-NSP	TL-KOB	TL-CSP
Y-BP	5.7E-08	5.19E-08	0.3544*	4.5E-09	3.88E-06	1.67E-10
DTLB-BP		1.95E-05	4.3E-08	0.06033*	3.96E-07	2.21E-09
TL-BP			1.54E-05	8.85E-06	0.002557	0.3568*
Y-IR				1.98E-10	3.27E-05	3.55E-08
DTLB-NSP					4.34E-06	1.98E-07
TL-KOB						0.004546

Table S4: Outcome Mann-Whitney-Wilcoxon test of $\delta^{13}\text{C}$ between yedoma (Y), drained thermokarst lake basin (DTLB) and thermokarst lake (TL) sediments. BP: Baldwin Peninsula (this study), IT: Itkillik River (Strauss et al., 2012), NSP: Northern Seward Peninsula (Lenz et al., 2016), KOB: Kobuk River Delta and CSP: Central Seward Peninsula (Lenz et al., 2018). P-values that exceed 0.05 are indicated with an asterisk (*).

P-value $\delta^{13}\text{C}$	DTLB BAL16-B4	Thermokarst lake BAL16-UPL1-L1	Yedoma Itkillik River	DTLB Northern Seward Peninsula	Thermokarst lake Kobuk Delta	Thermokarst lake Central Seward Peninsula
Yedoma BAL16-B2	<0.001	<0.001	>0.05	<0.001	<0.001	<0.001
DTLB BAL16-B4		<0.001	<0.001	>0.05	<0.001	<0.001
Thermokarst lake BAL16-UPL1-L2			<0.001	<0.001	<0.01	>0.05
Yedoma Itkillik River				<0.001	<0.001	<0.001
DTLB Northern Seward Peninsula					<0.001	<0.001
Thermokarst lake Kobuk Delta						<0.01

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