1 Holocene thermokarst dynamics in Central Yakutia – A multi-core and

2 robust grain-size endmember modeling approach

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16 Highlights

- 17 Small thermokarst lakes and basins grew rapidly during the Holocene Thermal Maximum
- 18 Short-term phases of forcing climate lead to very active thermokarst processes
- 19 Endmember analysis reveals different depositional environments in growing lakes
- 20 Distal and proximal depositional and post-sedimentary conditions are differentiated
- 21 Sedimentological and biogeochemical characteristics are weakly correlated
- 22

23 Abstract:

Differentiating thermokarst basin sediments with respect to the involved processes and environmental conditions is an important tool to understand permafrost landscape dynamics and scenarios and future trajectories in a warming Arctic and Subarctic. Thermokarst basin deposits have complex sedimentary structures due to the variability of Yedoma source sediments, reworking during the Late Glacial to Holocene climate changes, and different stages of thermokarst history. 30 Here we reconstruct the dynamic growth of thermokarst lakes and basins and related changes 31 of depositional conditions preserved in sediment sequences using a combination of 32 biogeochemical data and robust grain-size endmember analysis (rEMMA). This multi-proxy 33 approach is used on 10 sediment cores (each 300-400 cm deep) from two key thermokarst sites 34 to distinguish four time slices that describe the Holocene thermokarst (lake) basin evolution in 35 Central Yakutia (CY). Biogeochemical proxies and rEMMA reveal fine-grained sedimentation 36 with rather high lake levels and/or reducing conditions, and coarse-grained sedimentation with 37 rather shallow lake levels and/or oxidizing (i.e. terrestrial) conditions in relation to distal and 38 proximal depositional and post-sedimentary conditions. Statistical analysis suggests that the 39 biogeochemical parameters are almost independent of thermokarst deposit sedimentology. 40 Thus, the biogeochemical parameters are considered as signals of secondary (post-sedimentary) 41 reworking. The rEMMA results are clearly reflecting grain-size variations and depositional 42 conditions. This indicates small-scale varying depositional environments, frequently changing 43 lake levels, and predominantly lateral expansion at the edges of rapidly growing small 44 thermokarst lakes and basins. These small bodies finally coalesced, forming the large 45 thermokarst basins we see today in CY.

46 Considering previous paleoenvironmental reconstructions in Siberia, we show the initiation of 47 thaw and subsidence during the Late Glacial to Holocene transition between about 11 and 9 cal 48 kyrs BP, intensive and extensive thermokarst activity for the Holocene Thermal Maximum 49 (HTM) at about 7 to 5 cal kyrs BP, severely fluctuating water levels and further lateral basin growth between 3.5 cal kyrs BP and 1.5 cal kyrs BP, and the cessation of thermokarst activity 50 51 and extensive frost-induced processes (i.e. permafrost aggradation) after about 1.5 cal kyrs BP. 52 However, gradual permafrost warming over recent decades, in addition to human impacts, has 53 led to renewed high rates of subsidence and abrupt, rapid CY thermokarst processes.

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55 Keywords:

56 Permafrost; Climate change; Holocene Thermal Maximum; Granulometry; Palaeolimnology;
57 Ice complex; XRF; Eastern Siberia; Russia

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61 **1 Introduction**

62 Global climate change results in higher temperatures and stronger climate variability in the 63 northern high latitudes and permafrost-affected landscapes (Serreze et al., 2000; Serreze and 64 Barry, 2011; IPCC, 2013; Fedorov et al., 2014a). Positive feedbacks are expected as temperatures warm due to greenhouse gas emissions from rapidly evolving thermokarst lakes 65 66 and microbial decomposition of permafrost organic material (Walter Anthony et al., 2018), 67 although during the Holocene, thaw lakes also served as carbon sinks (Walter Anthony et al., 68 2014). Hence, thermokarst lakes and thaw processes in ice-rich permafrost deposits are a crucial 69 part of the global carbon cycle (Grosse et al., 2013; Olefeldt et al., 2016. Strauss et al., 2013, 70 2017). According to e.g. Stendel and Christensen (2002) the active layer (i.e. the seasonal thaw 71 layer) thickness in the Northern Hemisphere will increase by 30 % to 40 % by the end of the 72 21st Century. Regions with high ground-ice content, such as the Lena-Aldan interfluve in 73 Central Yakutia (CY) (Soloviev, 1959, 1973), will therefore be particularly affected by high 74 thaw and subsidence rates and the resulting ecosystem change (Fedorov et al., 2014b; Ulrich et 75 al., 2017a). Thermokarst is the strongest permafrost degradation process in areas with high 76 ground-ice content and results in a dissected landscape consisting of differentially aged 77 thermokarst basins and lakes (Czudek and Demek, 1970; Tomirdiaro, 1982).

78 Thermokarst landscapes cover more than 60 % of the ice-rich permafrost domain (Olefeldt et 79 al., 2016). This coverage makes them a key player in permafrost ecosystems and highlights the 80 importance of understanding these basins as a legacy of permafrost degradation after the Late 81 Glacial Maximum (LGM). Regional thermokarst landscapes have been studied, and formative 82 processes have been hypothesized by Soviet scientists using the example of CY thermokarst 83 landscapes (e.g. Soloviev, 1973; Katasonov et al., 1979; Bosikov, 1998). Nevertheless, a 84 knowledge gap remains and is addressed in this study by using spatially extensive 85 sedimentological analyses to decipher Late Glacial to Holocene small-scale processes as well 86 as related depositional and environmental conditions. The questions of how and when large 87 thermokarst (lake) basins (i.e. alases; dry, grass-covered basins within the Taiga forest) were 88 formed and how they evolved during the Holocene are still the subject of contemporary research 89 (Pestryakova et al., 2012; Nazarova et al., 2013; Ulrich et al., 2017b); it is necessary to answer 90 these questions in order to understand current and to predict future thermokarst processes.

Several studies of Late Glacial to Holocene thermokarst lake sediments have focused on
northern and eastern Siberia (Andreev et al., 1997; Katamura et al., 2006, 2009; Popp et al.,
2006; Biskaborn et al., 2013a,b, 2016; Schleusner et al., 2015; Klemm et al., 2016; see Fig 1a),

94 Alaska (Farquharson et al., 2016; Lenz et al., 2016; Jongejans et al., 2018) and Canada (Lenz 95 et al., 2013; Fritz et al., 2018). The understanding of thermokarst processes is often linked to 96 their spatial distribution (e.g., Morgenstern et al., 2011) as well as to the sediment archives (e.g., 97 Morgenstern et al., 2013). Elucidating the sedimentary characteristics in combination with the 98 elemental composition and deduced biogeochemical properties (Bouchard et al., 2011, 2017; 99 Biskaborn et al., 2013b) is, moreover, important for understanding geomorphological 100 thermokarst processes. However, a statistical (i.e. manual) evaluation of polymodal grain size 101 distribution (GSD) according to e.g. Folk and Ward (1957) only allows limited interpretation 102 and discussion of sediment origin and transport conditions (Hartmann, 2007). But polymodal 103 GSDs are typical for natural sediments, in general, due to different sediment sources as well as 104 different transport and accumulation processes (e.g., Weltje and Prins, 2007), and typical of 105 postsedimentary treatments and ice-rich permafrost deposits of the Yedoma type, in particular, 106 due to their polygenetic origin (Schirrmeister et al., 2011a; 2013). A statistical analysis based 107 on the eigenvalues of a grain size dataset was therefore recognized as a promising approach 108 (e.g., Flemming, 2007). Endmember (EM) modeling is a widely used method for process-109 oriented differentiation of multimodal GSDs (e.g., Weltje and Prins, 2003; Hamann et al., 2008; 110 IJmker et al., 2012; Dietze et al., 2014; Nottebaum et al., 2015) but it has very rarely been 111 applied to permafrost and in particular to thermokarst deposits so far (Strauss et al., 2012; 112 Klemm et al., 2016; Schirrmeister et al., 2017; Macumber et al., 2018). EM modeling uses the 113 eigenvalues of a dataset and reduces the dimensions of the data space by applying a factor 114 analysis method (Veganzones and Grana, 2008; Dietze et al., 2012). Finally, recurring patterns 115 in the source record are grouped into specific classes (i.e. EMs). The statistical method of 116 rEMMA thus offers meaningful genetic results to differentiate GSDs for reconstructing 117 sedimentological and paleoenvironmental conditions (Weltje and Prins, 2003, 2007; Dietze et 118 al., 2014; Dietze and Dietze, 2019).

119 By applying a multi-proxy approach in connection to rEMMA using ten sediment cores from 120 two thermokarst basins with different geocryolithological conditions, we aim to identify and to 121 interpret characteristic patterns in GSD and biogeochemical data of thermokarst deposits in 122 relation to age, location, geomorphology, and main depositional conditions of the different 123 drilling sites. The main objectives of this paper are (i) the lithostratigraphic characterization of 124 different kinds of thermokarst sediments using a multi-proxy approach and rEMMA; (ii) the 125 reconstruction of local geomorphological processes responsible for the individual development 126 of CY thermokarst basins during the Late Glacial and Holocene periods; and (iii) the detection

of regional climatic imprints with respect to similarities in thermokarst development,considering previous paleoenvironmental reconstructions in Siberia.

129

130 2 Regional setting

131 CY is bounded by the Lena and Aldan rivers and located in the east-Siberian Subarctic. The 132 region is a special case due to the lack of lowland glaciation during the late Pleistocene, a strong 133 continental climate setting, and continuous permafrost with depths reaching several hundred 134 meters (Fig. 1, Czudek and Demek, 1970). The active layer reaches depths of about 2.0 m in 135 grassland areas but has much lower depths below Taiga forest (e.g. Fedorov et al., 2014b). 136 Taliks (i.e. bodies of unfrozen ground) usually exist only below beds of major rivers and below 137 lakes where the water is deeper than winter lake-ice thickness. Isotherm subsurface 138 temperatures in the region at 10-15 m depth usually range from about -3°C below dense forest 139 cover to about -2°C below grassland areas (Fedorov et al., 2014b). The region is characterized 140 by low annual precipitation of 223±54 mm and a mean annual air temperature (MAAT) of -141 9.8±1.8 °C (1910 – 2014 average at Yakutsk weather station, NOAA National Climatic Data 142 Center, http://www.ncdc.noaa.gov/, Station ID: GHCND:RSM00024959). With the exception 143 of stable Siberian anticyclones during winter time, the wind and weather conditions in CY are 144 determined by cyclonic weather conditions that run from west to east across the region. During 145 the winter period from September to May wind conditions are relatively calm. Slightly stronger 146 winds occur during the short summer period from June to August (Péwé and Journaux, 1983). 147 A 75-year record at Yakutsk indicates a mean annual wind velocity of 2.4 m/s (Gavrilova, 1973). 148 For our study region, the Global Wind Atlas (2018) indicates an average wind speed of about 149 4 m/s.

150 The Lena-Aldan interfluve region is a low-relief landscape. Geomorphological processes are 151 linked to Holocene and current thermokarst phenomena (Brouchkov et al., 2004; Katamura et 152 al., 2006; Fedorov and Konstantinov, 2009; Iijima et al., 2014; Séjourné et al., 2015; Ulrich et 153 al., 2017a, b). Thermokarst basins (i.e. alases) and thaw lakes are widespread, typical landscape 154 features in this region (e.g., Soloviev, 1973; Fig. 1b). Larch forest with inclusion of pine and 155 birch trees typically dominates the CY vegetation. Thermokarst basins form islands of 156 grasslands within the taiga forest that are dominated by steppe-like to swamp communities, 157 depending on edaphic factors within the basins (Mirkin et al., 1985; Desyatkin, 2008).

158 Major parts of the Lena-Aldan interfluve region are covered by ice-rich fine-grained late 159 Pleistocene sediments containing up to 80 % ice by volume, mainly in the form of huge 160 syngenetic ice wedges. These Yedoma deposits form the forest-covered Yedoma uplands 161 surrounding the thermokarst landscapes. Several terraces above the major rivers are 162 differentiated due to cryolithology and sediment genesis in the Lena-Aldan interfluve region 163 (Soloviev, 1959, 1973; Fig. 1c). Typical well-developed thermokarst basins and lakes in 164 different evolutionary stages are mainly found on the Tyungyulyu and Abalakh terraces. Both 165 terraces are characterized by thick ice-rich deposits (more than 40 m thick in some places) with 166 different cryolithological conditions and characteristics (Soloviev, 1959; Fedorov and Konstantinov, 2003, 2009; Ulrich et al., 2017a). 167

168 Two thermokarst basins were chosen as study sites that were considered representative for the 169 respective terraces. Both are located about 80 km southeast from Yakutsk (Fig. 1c). The Khara 170 Bulgunnyakh (KB) alas (Lat: 61,836680, Long: 130,646150; Fig. 2a) is part of a larger alas 171 system and is located about 130 m a.s.l (above sea level) on the Tyungyuluy Terrace. The KB 172 alas is about 1000 to 1500 m in diameter and about 6 to 10 m deeper than surrounding Yedoma 173 uplands. Three large pingos within the KB alas reach heights of 8 to 12 m and diameters of 50 174 to 100 m. They are surrounded by very shallow lakes. The alas system was investigated by a 175 Russian research group in the 1970s (Katasonov et al., 1979). They suggest that its evolution 176 started around 12-10 thousand years ago because they found stalks of reed grass in the 177 northwest of the alas system below 3.0 m depth that has been dated to a ${}^{14}C$ age of 9120±200 178 yrs BP (10,261 \pm 289 cal yrs. BP).

179 The Yukechi (YU) alas (Lat: 61.764950, Long: 130.465260, Fig. 2b) is located on the Abalakh 180 Terrace at about 202 m a.s.l. The YU alas is about 300 to 500 m in diameter and about 10 to 15 m deep. Two larger lakes and one small shallow pond existed within the alas. The area around 181 182 the YU alas is characterized by many young thermokarst features and lakes, indicating current 183 intensive thermokarst development. In particular, small, young thermokarst lakes are ubiquitous 184 on the Yedoma uplands. Many of these Yedoma lakes were evidently developed 185 anthropogenically (e.g. in former agricultural areas) (Fedorov et al., 2014b; Ulrich et al., 2017a). 186 The YU alas and its surroundings have been monitored for several decades by the Melnikov Permafrost Institute in Yakutsk (Bosikov, 1998; Fedorov and Konstantinov, 2003, 2008, 2009; 187 188 Fedorov et al., 2014b), but the exact timing and rate of YU alas evolution during the Holocene 189 remain unclear.

3 Material and methods

192 3.1 Permafrost coring and sampling

193 At both study sites five cores were drilled in August 2013 from the dry basin ground (Fig. 2).

The drilling and sampling design follows a transect sampling design covering the major alaslandscape units (Fig. 3).

- Thus, for each study site at least one to three cores were taken from the basin centers (KB3, KB7, and YU2, YU3, YU4) and one or two cores from marginal basin areas (KB1, KB4, and YU1). The KB7 core was taken on the top of a small pingo in the basin center. Several analyses of KB7 core content including ostracods, chironomids, and pollen are already published (Ulrich et al., 2017b). Addionally, at each study site one core was taken from the center of a neighboring smaller drained thermokarst basin that is connected to the main basin (KB6, YU5).
- 202 If possible, an active layer pit was dug first and the frozen core was drilled from the permafrost 203 table. This was often necessary due to difficult drilling conditions within the active layer. 204 Sediment samples were then additionally taken from the active layer pit. Hereafter, they are 205 also discussed as part of the sediment cores. Data gaps especially within the active layer are the 206 result of core losses during drilling. All sediment cores were drilled down to 300 - 400 cm 207 below surface (bs). Each core segment (about <20-30 cm long) was cleaned, described, and 208 photographed according to sediment composition, color, and cryolithological properties 209 following French and Shur (2010). Subsequently, the cores were continuously sampled at <10 210 to 20 cm intervals, the samples were packed in plastic bags, and transported to the laboratories.

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212 3.2 Laboratory sedimentological and biogeochemical analyses

213 We analyzed sedimentological and biogeochemical parameters for all sediment samples after 214 freeze-drying. For each analytical method, individual subsamples were used. Grain-size 215 analyses were carried out using carbonate-free (treated with 10% HCL) and organic-free 216 (treated with 35% H₂O₂) subsamples using a laser particle analyzer (Beckmann Coulter LS 200) 217 according to DIN ISO 11277. The proportions of sand, silt, and clay fractions are given as sums 218 between 2 mm and 63 µm, <63 µm and 2 µm, and <2 µm, respectively. Grain-size parameters 219 such as the geometric mean of the respective particle size distribution in μ m and sorting in φ 220 were determined according to Folk and Ward (1957) with the Gradistat software (Version 8.0, 221 Blott and Pye, 2001). The mass-specific magnetic susceptibility (MS) was analyzed using a 222 Bartington MS2B dual-frequency sensor. The values are expressed in SI units (10⁻⁶ m³ kg⁻¹).

223 Total carbon (TC) and total nitrogen (TN) were measured with a Vario EL cube elemental 224 analyzer and are given as weight percent (wt%). The total inorganic carbon (TIC) content was 225 taken from carbonate measurements, which were conducted using the Scheibler method on an 226 Eijkelkamp Calcimeter apparatus by continuously adding 4 N HCL to a subsample until the 227 CO₂ outgassing reaction ceased. TIC is then calculated by dividing the percent calcium 228 carbonate (CaCO3) with 8.33. The total organic carbon (TOC) content was determined by 229 subtracting TIC from TC. The TOC/TN ratio is expressed as the quotient of TOC and TN 230 weight percentages but is given as mass ratio by dividing them by 1.167 (the ratio of atomic 231 weights of N and C; Meyers and Teranes, 2001). Stable carbon isotopes (δ^{13} C) were measured 232 on carbonate-free subsamples with a Delta V Advantage isotope mass spectrometer 233 (ThermoFisher ScientificTM) coupled with a ConFlo IV Interface and a Flash 2000 IRMS Elemental Analyzer. The values are expressed in delta per mil notation (δ ‰) relative to the 234 235 Vienna Pee Dee Belemnite (VPDB) Standard.

Bulk samples from the recovered cores were analyzed according to Zielhofer et al. (2017) with a Spectro Xepos XRF (X-ray fluorescence) device. For XRF sample preparation bulk subsamples (8 g) were homogenized with a vibratory Retsch mill MM 200. Pressed pellets were prepared using a Vaneox press at 20 t for 2 min. Measurements were conducted in a He atmosphere.

According to Davison (1993), Bouchard et al. (2011), and Biskaborn et al. (2013a) only a few
elements can be used for tracking thermokarst processes. We chose the elements Mg, S, Ca,
Mn, Fe and their ratios for detailed discussion of sediment properties and depositional
conditions and dynamics during thermokarst (lake) processes and basin evolution.

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246 3.3 Radiocarbon dating

Radiocarbon dating was performed on 34 sediment samples from eight cores at the Curt-Engelhorn-Centre of Archaeometry (Mannheim, Germany) using the Accelerator Mass Spectrometer (AMS) technique (Tab. 1). The treatment was conducted with the Acid-Base-Acid method and the samples were measured in the MICADAS accelerator. Hand-picked terrestrial plant remains, wood fragments, and charcoal was dated only from 14 sediment samples. For 20 samples soil organic carbon was dated using bulk sediment samples due to the lack of organic macro remains within samples. Finally, the conventional ¹⁴C ages were calibrated using the INTCAL13 data set (Reimer et al., 2013) and SwissCal 1.0 (L. Wacker,

ETH Zürich). The ¹⁴C ages are normalized to $\delta^{13}C = -25\%$ (Stuiver and Polach, 1977).

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257 3.4 Grain-size EM modeling and statistical analysis

258 The GSDs derived for each sample were used in an EM modeling approach to try and unmix 259 the polymodal GSDs into characteristic grain-size subpopulations which can be attributed to 260 sedimentary processes and conditions. EM modeling reduces the dimensions of a data space 261 (the GSDs) using the method of factor analysis, where eigenvalues are used to extract recurring 262 patterns in the initial data set (the EMs) (Veganzones and Grana, 2008). For rEMMA, the open 263 source R-package EMMAgeo following Dietze et al. (2012, 2014, 2016) and Dietze and Dietze 264 (2019) was used, which contains the following steps: (1) Before conducting the actual EM 265 analysis, a grain size data matrix is constructed, containing the grain size distributions 266 (columns) for each sample (rows), which is then rescaled to constant row sums. Then, a weight 267 transformation is applied according to Klovan and Imbrie (1971), using a weight transformation 268 parameter to yield a weight matrix that is not biased by variables with large standard deviations. 269 To allow the extraction of robust EMs, the ranges of the weight transformation parameter and 270 the minimum and maximum number of possible EMs are identified from the measured data set. 271 (2) EM modeling is performed for all combinations of the weight transformation parameter and 272 the number of EMs. Robust EMs (rEMs) are defined as grain-size subpopulations that appear 273 independent of the chosen model parameters and have an overall explained variance >50%. (3) 274 In order to quantify the uncertainties associated with the contribution of each grain size class to 275 each mean rEM, an uncertainty estimate is calculated from the spread of the contributing EM. 276 (4) The scores for each sample, which provide a quantitative estimate of how much a rEM 277 contributes to a sample, are calculated for the mean rEM loadings. An uncertainty estimate for 278 the scores is calculated via a Monte-Carlo simulation. From the rEM loadings and scores, 279 sample and class-wise explained variances are calculated to assess the overall quality of the 280 chosen model.

Finally, a principal component analysis (PCA) was performed on all rEM scores and sample biogeochemical parameters to examine connections and relationships between sedimentology and biogeochemistry of the thermokarst deposits. The PCA was carried out using R (R Core Team, 2014). Because of the very different nature of the scores from the rEMMA and the biogeochemical analyses, all variables were standardized by substracting the mean and scaling to the actual data range of the specific variable. Note that this means that variances are not equal to 1 for all variables. A four-fold cross validation approach was used to assess the number of relevant PCs, using the R package missMDA (Josse and Husson, 2016). The resulting ordination diagram presents standardized metric scores and expresses the relationship among metrics as correlations. Additionally, all core samples are projected onto the ordination graph for interpretation purposes only, because they do not affect calculations; sample scores on the first and second principal component (PC1 and PC2, respectively) were plotted against core depth.

294

295 4 Results

296 4.1 Geochronology

297 The results of dating 34 samples from eight cores shown in Table 1 indicate a clear difference 298 in age between bulk organic material and terrestrial organic macro remains. Overall, macro 299 remain and bulk radiocarbon dates of YU study site samples are older than dates of KB site 300 samples (Fig. 3). While almost all macro remains show middle to late Holocene ages, with the 301 exception of two samples from cores YU3 and YU5, all bulk samples, except one sample from 302 the YU2 core, were dated between Late Glacial and early Holocene ages (Tab. 1). This age 303 discrepancy is attributed to the different carbon sources of the dated material and will be 304 independently considered in further geochronological discussions. Since there is a very low 305 TIC content in all cores (0.1 to 1.7 wt%, Appendix A), carbonate-derived reservoir effects are 306 suggested to be insignificant.

307 For the KB study site, macro remain ages from the central basin have been derived only for the 308 KB7 core. The five KB7 macro remain samples taken between 320 and 60 cm bs show middle 309 Holocene radiocarbon dates between 6582±78 and 5752±102 cal. yrs. BP (see also Ulrich et al., 310 2017b). For the KB1 core at the basin edge, three late Holocene macro remain ages range 311 between 3314±62 and 1812.5±72.5 cal. yrs. BP (between 243 and 125 cm bs, respectively). 312 One sample (92 cm bs) of the KB6 core located centrally within the small drained basin 313 orginated from an organic inclusion with wooden remains and was dated to 3316±61 cal. yrs. 314 BP. The late Pleistocene and Late Glacial to early Holocene bulk ages for the KB study site 315 generally decrease from the basin center (KB3, KB7) to the basin edge (KB1) (Tab. 1). The 316 KB3 core show the highest ages at the KB study site and continuously upward decreasing bulk 317 ages between 38,355±615 and 12,902.5±184.5 cal yrs BP.

318 For all YU study sites, five radiocarbon dates were derived from macro remain samples. Two 319 samples from the basin's center (YU3) show ages of 34,865±235 cal. yrs. BP at 185-170 cm bs 320 and 3975±102 cal. yrs. BP at 40-36 cm bs. One macro remain sample from the YU1 core (110-321 105 cm bs) at the basin edge was dated to 4606±175 cal yrs. BP. Another two samples from the 322 YU5 core of the small drained neighboring basin (Figs. 2b and 3) show strong differences: 323 While the macro remain sample from 222-210 cm bs was dated to the late Pleistocene time 324 period (39,340±650 cal. yrs. BP) and thus represents the oldest macro remain sample from both 325 study sites, another sample from 40-20 cm bs shows a modern age but may have been 326 contaminated during drilling, which is even more probable considering the bulk sample age 327 from same depth (see Tab. 1). The YU bulk ages show a rather unclear division between the 328 central and the marginal parts of the basin (Tab. 1). Late Pleistocene to middle Holocene bulk 329 ages are shown by the basin center cores YU3 and YU2, while the dates from the lower part of 330 the YU1 core at the basin edge show a strong inversion of downward decreasing Late Glacial 331 ages. This probably indicates the strong redeposition of Yedoma deposits during the lateral 332 expansion of the thermokarst basins (see discussion below). The YU5 core from the 333 neighboring small basins, however, shows continuously upward decreasing late Pleistocene 334 bulk ages between 41,625±405 and 27,625±165 cal yrs BP.

335

336 4.2 Cryolithology, Sedimentology, and Biogeochemistry

337 As a result of our laboratory analysis of sedimentological and biogeochemical parameters, we indentify different facies within the thermokarst deposits from the basin edges and from the 338 339 central basin parts, which will be described separately for each study site. Generally, all KB 340 and YU sediment sequences were composed of visibly homogeneous silt-dominated deposits 341 with varying but generally low amounts of organic inclusions and varying ice contents (Fig. 4). 342 Usually, the upper half of all sediment cores was characterized by rust-stained brownish-greyish 343 deposits. These horizons generally continued downwards in the frozen core material with some 344 blackish streaky layers and spots (Fig. 4a-f). The lower frozen parts of the cores were usually 345 dominated by dark grey to dark olive-green sediments, interrupted by blackish spots and streaky 346 layers (Fig. 4g-l), which were less pronounced within the YU cores. Furthermore, sediment 347 layers in the basin edge cores (KB1 and YU1) showed bent or mixed layers in relation to 348 topography changes on the thermoterraces (Fig. 3).

The ground-ice content often increased with depth and was represented by parallel to nonparallel lenticular-layered to reticulate cryostructures, which reached a thickness of up to 6 mm in some deeper core parts (Fig. 4). Exceptions here were represented by the cores from the neighboring small basins (KB6 and YU5). They showed significantly lower ice content and fewer to no cryostructures in their frozen parts. In addition, all cores from the YU basin center (YU2, YU3, YU4) were unfrozen. This is due to an existing talik below the YU alas. Postcryogenic structures in the lower core parts show once-frozen, ice-rich sediments. In particular, the YU2 core is currently strongly influenced by ground water changes due to the nearby lake and showed pronounced redox properties in the sediment (Fig. 4a).

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4.2.1 Sedimentological and biogeochemical results of the KB thermokarst basin

360 The KB thermokarst deposits are composed of uni-, bi-, tri-, and polymodally distributed, 361 poorly to very-poorly sorted, very-fine to fine sandy medium-to-coarse silt. The grain-size 362 characteristics differ between the horizons of a core as well as between the drilling sites (Fig. 363 5). The GSDs and MS values of the KB3 core from the basin center show little variability (Fig. 364 6). The KB3 GSDs are bimodal with main peaks in the coarse silt fraction. The second mode 365 can be found in the coarse clay to very-fine silt. The grain-size mean for the KB3 core is 366 16.7±1.1 μm (see Appendix A, Table S3). GSD peaks of the KB7 core from the small pingo 367 top are located mainly in the coarse-silt fraction, but all samples show high variability (mean 368 25.2±6.8 µm; Tab. S4). Additional minor but distinct peaks in the fine-sand fraction are mainly 369 found in samples from the upper and lower KB7 profile, while minor peaks in the coarse clay 370 to very-fine silt fraction are predominantly found in the middle of the core. The MS decreases upwards (0.3 to 1.3 x 10^{-6} m³ kg⁻¹) with a distinct peak of 2.2 x 10^{-6} m³ kg⁻¹ at 285–258 cm bs. 371 The GSDs of the KB1 and KB4 basin edge cores show main peaks in the coarse silt fraction 372 373 with generally two minor peaks in the fine sand and clay fractions (Fig. 5). The grain-size means 374 for KB1 and KB4 are 23.5±5.8 μm and 18.0±2.0 μm, respectively (Tabs. S1 and S2). Increased 375 amounts of sand in the upper and lower KB1 profile and in the upper KB4 profile correspond 376 to increased MS values. The GSDs of the KB6 core from the neighboring smaller basin also 377 show high variability with a grain-size mean of 23.3±8.7 µm (Tab. S5) and main peaks in the 378 coarse silt fractions. MS values show comparably less variability, slighty decreasing upwards 379 $(1.2 \text{ to } 0.6 \text{ x } 10^{-6} \text{ m}^3 \text{ kg}^{-1}).$

The KB biogeochemical records and elemental composition show some pronounced differences in relation to the sedimentation history and geomorphological location of the individual sediment profiles. With the exception of the KB7 core, the TOC values of all cores are comparably low with hardly any variations. In contrast, the upper half of the KB7 core shows 384 upwardly increasing TOC values that reach a high value of 10.1 % at the active layer bottom 385 (Fig. 6; mean 3.6±2.9 %). The variations of TOC/TN ratios are higher for the KB basin edges 386 (KB1: 7.9 – 14.0, KB4: 6.6 – 15.1) than for the basin centers (KB3: 6.3 – 11.2, KB7: 7.1 – 10.5) and the KB6 core in the small neighboring basin (5.8 – 8.6). The δ^{13} C values are, however, 387 homogeneous for KB3 (-25.4 to -24.4 ‰) and KB1 (-25.1 to -24.1 ‰) and show little variation 388 389 in KB4 (-26.2 to -24.1 ‰), but the values are lower and upwardly decreasing in KB7 (-28.8 to 390 -25.4 ‰) and KB6 (-27.2 to -24.5 ‰). The CaCO₃ content is strongly negatively correlated 391 with the Ca/Mg ratio in all KB core profiles, indicating that Ca is largely bound as CaCO₃. 392 CaCO₃ concentration shows relatively strong variations and several pronounced peaks were 393 found in all cores which ranged between 0.0 and 7.4 % with highest values reached in the lowest 394 parts of KB7 and KB3 (basin center) and the middel core parts of KB4 (basin edge). Strong 395 differences between the coring locations and within the cores can be seen in the S contents of 396 the KB cores (Fig. 6). Usually, peaks in S contents coincide with the above-described black 397 layers and spots in the middle and lower parts of the KB sediment cores. With the exception of 398 KB7, the Fe/S ratio is low in the frozen core parts but increases strongly in the active layer of 399 all KB cores. The KB7 core, however, shows a very narrow Fe/S ratio, but the ratio is higher at 400 the profile bottom and decreases upwards with two small peaks at 258-143 cm bs and at 70-30 401 cm bs. The Fe/Mn ratio shows strong variation in the KB1 and KB4 basin edge cores and the 402 KB3 basin center core. Variations are smaller in KB6 and the ratio increases stepwise in KB7.

403

404 4.2.2 Sedimentological and biogeochemical results from the YU basin

405 The GSDs of YU thermokarst basin deposits are differentiated within and between the cores, 406 similar to the KB results (Fig. 5). The GSDs of the YU2 core from the lowest part of the basin 407 center show variations with bimodal distributions, main peaks in the coarse silt fraction, and 408 secondary modes in the coarse clay to very-fine silt. The grain-size mean is the lowest of all 409 cores at 15.4±1.5 µm (Tab. S7). The grain-size mean and GSD variability is a little higher for 410 the other basin center cores YU3 and YU4; 17.3±3.5 µm and 19.8±3.9 µm, respectively (Tabs. 411 S8 and S9). Both cores, moreover, show main peaks in the coarse silt fraction, with generally 412 minor peaks in the fine sand and clay fractions. However, the YU3 GSD variability is 413 determined by a few sandy samples and mostly stronger secondary modes in the coarse clay 414 fraction, while the YU4 GSDs are characterized by more distinct secondary modes in the fine 415 sand fraction. Besides a strong decrease in the upper part of core YU2, the MS shows little 416 variation in the basin center sediments. The GSDs of the YU1 core from the thermoterrace at 417 the basin edge show high variability from uni- to polymodal distributions with main peaks in 418 the coarse silt fraction and fluctuating secondary peaks in the fine sand and coarse clay to fine 419 silt fractions (Fig. 5). The grain-size mean is 21.1±5.9 µm (Tab. S6). Similar to the KB basin 420 edges, increased amounts of sand in the upper and lower part of YU1 correspond to increasing 421 MS values. Compared to the KB6 core, the YU5 core from the neighboring small drained basin 422 shows little GSD variability. The lower half of the YU5 core is characterized by higher amounts 423 of fine sand while the upper half shows higher clay contents. The grain-size mean is 19.0±3.0 424 μm (Tab. S10).

425 The biogeochemistry of the YU sediments shows some differences from the KB thermokarst 426 basin deposits (Fig. 7). The TOC content in the YU basin is higher on average than in KB. TOC 427 increases upwards from 0.5 % to 4.0 % in the uppermost part within the basin edge core YU1. 428 In the basin center, YU3 shows lower downward increasing values between 0.3 and 1.3 %. The 429 TOC content peaks in the uppermost part of YU2 to 2.3 % and increases slightly downwards in 430 the YU4 core, ranging between 0.3 and 2.6 %. The YU5 core TOC content is more stable, 431 ranging between 1.3 and 2.5 %. The variations of the TOC/TN ratio are generally less at the 432 YU study site than at the KB study site. But similar to KB, variations are larger at the basin 433 edge (YU1: 5.6 - 11.2) than in the center area of the YU basin (YU2: 7.2 - 9.6; YU3: 6.2 - 8.3; YU4: 6.0 - 8.5). The ratio, however, increases slightly upwards in the YU5 profile (6.6 - 10.0). 434 435 The δ^{13} C values are homogeneous in all cores, averaging around -25 ‰. Only the uppermost 436 part of core YU2 shows a strong decrease of δ^{13} C to -29.2 ‰. Similar to KB, the CaCO₃ content 437 in the YU cores is negatively correlated to the Mg/Ca ratio. However, the CaCO₃ content is 438 higher on average in YU cores than in KB cores, ranging between 0.8 and 13.8 % (Fig. 7). With 439 the exception of the YU5 core, the S contents are on average lower in the YU than in the KB 440 sediment sequences and differences between basin center and edge are smaller, but some 441 variations within the YU cores are obvious as well. The higher average TOC content at the YU 442 site and the generally stronger correlation of the S contents with TOC as well as the visible 443 absence of significant black streaks in YU sediments suggest that there is a larger amount of 444 organically bound S within the YU sediment sequences. The Fe/S ratio shows a different 445 behavior within the YU sediment sequences than within the KB cores. Within the basin center, 446 the Fe/S ratio is narrower and shows less variation in the YU3 core but shows an upwards 447 increase and higher ratios in the YU2 and YU4 cores with a strong peak at about 80 to 100 cm 448 bs; the Fe/S decreases again towards the surface. For YU1 at the basin edge, the Fe/S ratio 449 shows a little more variation with wider ratios in the upper middle core part. The Fe/S ratio is 450 small within the YU5 core and shows less variation. The Fe/Mn ratio in the YU sediment 451 sequences shows less variation than in the KB sequences, but variations are generally largest in452 YU1 at the basin edge and smallest in core YU5.

453

454 4.3 rEMMA

455 The four final rEMs account for 79.5% of the total variance of all initial variables (i.e. all input GSDs) with modes in various grain-size fractions between clay and medium sand. The unmixed 456 457 rEMs are characterized by their loadings (Fig. 8). The rEM1 shows its strongest peak in the coarse clay to very-fine silt fraction (mode = $2.9 \,\mu\text{m}$, explained variance (EV) = 24%). rEM1 458 459 exhibits some unmixed residuals in the coarse silt and fine sand fraction. rEM2 has a bimodal 460 distribution with residual mode in the fine sand fraction. The main mode is located in the 461 medium to coarse silt fraction at 27.4 μ m (EV = 18%), but its slightly left-skewed distribution 462 shows some tendency to increased finer grain sizes. rEM3 shows a distinct unimodal 463 distribution with a high peaked mode in the coarse silt fraction at 39.8 μ m (EV = 25%) but it 464 shows some tendency to the coarser grain sizes by a slightly right-skewed distribution. The 465 mode of rEM4 peaks in the fine to medium sand fraction at 176.8 μ m (EV = 32%) with some 466 tendency towards the finer grain sizes. The class-wise explained variance in Fig. 8 shows that 467 in particular the very-fine to fine-grained fractions show highest variances.

468 Calculated mean scores are shown in Fig. 8 with their confidence intervals and in Fig. 9 against 469 depth for each core individually. The scores represent the relative rEM contribution to each 470 sample, i.e. how each sample is composed of a specific rEM. Equal compositions of mostly 471 rEM1, rEM2, and rEM3 with comparatively larger shares of rEM1 and rEM2 over the entire 472 sediment sequences are reflected in particular for the basin centers (i.e. KB3, YU2, YU3). An 473 exception is shown by the KB7 scores, which exhibit strong variations of all rEMs but generally 474 a division of the sediment sequence. While rEM3 and rEM4 dominate the lower half of the KB7 475 core, the shares of rEM1 and rEM2 are significantly higher in the upper half. The basin edge 476 sediment sequences (i.e. KB1, KB4, YU1) show varying compositions including all rEMs. 477 Smaller contribution of rEM1 in KB1 and YU1 emphasize the proximal accumulation 478 conditions of these sites and the increased influence of coarser basin slope sediments. The 479 higher contribution of rEM1 in the KB4 basin edge core are a sign of comparatively lower 480 erosion at the northern edge of the KB basin than at the eastern edge captured by the KB1 core 481 (Figs. 2 and 3). In contrast, the larger shares of rEM3 and rEM4 in the middle part of the YU4 482 basin center core (Fig. 9) are probably related to YU4's location at the edge of a flat hill within 483 the YU bottom. The KB6 and YU5 rEM scores show the individual rEM compositions within

the neighboring small drained basins. KB6 shows a bulge increase and subsequent decrease of rEM1 und rEM2 in the lower half of the sequence. The upper half shows stronger variations of all rEMs. YU5, on the other hand, is dominated by a homogeneous share of rEM1, rEM2, and rEM3, but also shows higher amounts of rEM4 in three samples from the lower part of the core.

- 488
- 489 4.4 PCA

490 The PCA shows two major principal components (PC1 and PC2) that explain 27.9% and 22.3%, 491 respectively, of the total variance of all rEMs and biogeochemical parameters from the KB and 492 YU thermokarst deposit core samples (Fig. 10). PC1 is positively correlated with rEM1, rEM2, 493 and Fe/Mn and negatively correlated with rEM3, rEM4, and MS. PC2 is clearly controlled only 494 by the biogeochemical parameters; it shows positive correlations with S, TOC, Mg/Ca, and 495 TOC/TN, and negative correlations with δ^{13} C, Fe/S, and CaCO₃. The core samples projected 496 into the ordination graph show only rough separations. The basin edge core samples (KB1, KB4, 497 YU1) are scattered throughout the complete PC1 axis but are more common on the negative 498 part of the PC2 axis, while the basin center core samples (KB3, YU3) occur more frequently 499 on the positive part of PC1 but more evenly throughout the complete PC2 axis. The YU2 core 500 samples are clustered in the lower right of the ordination plot and the KB7 core samples are 501 broadly scattered on the positive part of the PC2 axis and across the complete PC1 axis. The 502 core samples from the neighboring small basins, KB6 and YU5, as well as the YU4 core sample 503 are scattered across both PC axes.

While PC3-PC5 are considered relevant due to the results of the four-fold cross validation approach, a reasonable physical interpretation is not possible. Ordination plots for all PCs are included in the supplement material (Appendix B, Figs. S1, S2, and S3).

507

508 5 Discussion

509 5.1 Sedimentary properties and biogeochemical characteristics of CY thermokarst deposits

510 The GSDs and the resulting sorting provide information on the erosion and transport processes

511 as well as on the accumulation conditions of the thermokarst sediments. Assuming that

512 individual transport processes would have led to better fractionation of the grain sizes (e.g.,

513 Folk and Ward, 1957), the poor to very poor sorting of all GSDs indicates short transport paths

514 (see Appendix A) or a mixture/overlay of different formation processes (Blott and Pye, 2001).

The subsequent reconstruction of erosion, transport, and sedimentation conditions is thus difficult to perform (Dietze et al., 2012, 2014; IJmker et al., 2012). In the study region, the GSDs of the source sediments from Yedoma deposits are heterogeneous due to their polygenetic formation, leading to polymodal GSDs (Schirrmeister et al., 2011a, 2017; Strauss et al., 2017). The spatial understanding of the local Yedoma structure is thus of great importance for interpreting GSDs in thermokarst deposits (see section 5.2).

In general, coarser grain sizes (especially the fine sand fraction) within the thermokarst (lake) basins point to shorter transport paths after erosion processes along lake shores. Coarser-grained sediments can also be accumulated via longer high-energy transport or by subsidence and in situ accumulation close to the lake center within small thermokarst lake basins. Finer grain sizes like coarse clay are thought to be indicative of distal (to the lake center) transport in larger lakes (e.g. Biskaborn et al., 2013b).

527 Generally, the MS parameter depends on source rock and/or sediment (Schirrmeister et al., 528 2011b). We found stronger MS signal variations especially in the basin edge sediments from 529 both study sites (KB1, KB4, YU1) and the lower part of the KB7 core. The reworking of 530 Yedoma deposits under lacustrine conditions may have altered the MS signal in the basin 531 centers, since the magnetizable minerals degrade under reducing conditions, resulting in lower 532 values (Evans et al., 1997; Hanesch and Scholger, 2005). The MS signal therefore also indicates 533 an increase in lacustrine finer-grained detritus (lower MS) or increased terrestrial coarser-534 grained input (higher MS) (Matasova et al., 2001). In accordance with Schirrmeister et al. 535 (2011c), we also observe that the MS is inversely proportional to the sediment TOC.

536 The TOC proportion is a main parameter in the analysis of thermokarst sediment developmental 537 history (e.g., Walter et al., 2007; Lenz et al., 2016; Bouchard et al., 2017). Assuming constant 538 input, high TOC values in ice-rich permafrost deposits generally reflect low organic matter 539 degradation during the past and high quality in the sense of future microbial degradation 540 (Strauss et al. 2015). This is generally typical for polar regions, as microbial decomposition of 541 organic matter has limited amount of time before freezing, thus it generally ceases at low 542 temperatures and organic matter is preserved for a long time in the frozen state (Strauss et al., 543 2013, 2017). Generally, we measured comparably low TOC values in the KB and YU alas 544 thermokarst deposits, suggesting low organic carbon input or strong post-sedimentary organic 545 carbon decomposition, at least within the drilled 300 - 400 cm bs. Based on the fact that there 546 is no depth-dependent trend in some cores, we assume a relatively low organic carbon input 547 compared to other thermokarst and Yedoma landscapes (Strauss et al., 2013). The exceptionally

high TOC values in the upper half of the KB7 profile are suggested to be related to high
bioproductivity and a nutrient oversupply within a very fast-growing thermokarst lake during
the mid-Holocene at this specific site (Ulrich et al., 2017b).

551 Organic carbon transport pathways (e.g. lacustrine or terrestrial) into the formerly existing 552 thermokarst lake can be identified by combined analysis of δ^{13} C, the TOC/TN ratio, and TOC 553 (e.g. Meyers and Teranes, 2001). High TOC/TN ratios around 20 are produced by cellulosic, 554 low-protein, vascular land plants. In contrast, narrow TOC/TN ratios between 4 and 10 are 555 produced by phytoplankton or algae (Meyers, 1994; Meyers and Lallier-Vergès, 1999). Ratios 556 between these very high and very low values as shown in our data can be seen as a mixture of 557 different entry paths. However, especially in oligotrophic lakes, the TOC/TN ratio can be 558 reduced by organic matter decomposition under oxic conditions, because carbon is preferred 559 over nitrogen (Meyers and Lallier-Vergès, 1999; Meyers and Teranes, 2001). For the 560 thermokarst deposits studied here, higher TOC/TN values have been interpreted as greater 561 terrestrial input, whereas lower values have been considered as greater lacustrine input and/or 562 post-sedimentary carbon decomposition (Meyers, 1994; Biskaborn et al., 2013b; Lenz et al., 563 2013).

The δ^{13} C values provide information about plant metabolism because sedimentary organic 564 565 matter δ^{13} C values are generally very similar to those of the parent vegetation and post-566 sedimentation of carbon isotopes is therefore negligible (O'Leary, 1981; Melillo et al., 1989). 567 Thus, we interpreted very low values as an indication of increased bioproductivity. This is 568 obvious in the upper part of the KB7 and YU2 profiles. Finally, different sedimentation milieus 569 can be derived from the TOC/TN– δ^{13} C relationship (Fig.11), which reflects organic matter 570 origin (e.g., Meyers and Teranes, 2001). This relationship shows a shift from lacustrine algae 571 to terrestrial plants in several samples from the KB1 and KB4 basin edge cores as well as within 572 the lower and upper part of the YU1 core and with decreasing profile depth in the YU5 core. 573 Otherwise the TOC/TN– δ^{13} C relationship suggests mainly lacustrine sediment origin for great 574 parts of the KB and YU drilling locations (Fig. 11).

575 S together with phosphorus (P) contents can be used as a proxy for organic entry in thermokarst 576 (lake) deposits (Bouchard et al., 2011). The P content, however, shows only weak correlations 577 with the TOC in our drilled sediments and little variations (see Appendix A). The S content 578 show more connections to TOC but higher S contents also often corresponds with black streaks 579 and spots, especially in the KB sediment sequences. These streaks are interpreted as iron sulfide 580 (FeS) precipitation by iron hydroxide reduction. This is bacterially mediated under anoxic 581 conditions in relatively shallow warm waters and can therefore be seen as evidence of reducing 582 conditions (Siegert, 1979, 1987; Biskaborn et al., 2012). Since the streaks and spots do not 583 follow stratigraphic boundaries, the precipitation of this compound is considered to be post-584 sedimentary.

585 CaCO₃ can reach the lacustrine sediment archive via several paths. Generally, biogenic and 586 geogenic carbonate origin can be differentiated. The biogenic part is usually formed by aquatic 587 invertebrates like molluscs or ostracods in thermokarst lakes (e.g., Wetterich et al., 2005, 2008). 588 Geogenic formation in CY thermokarst deposits is probably the result of authigenic origin in 589 Yedoma deposits, which are usually rich in carbonate (Katasonov et al., 1979; Schirrmeister et 590 al., 2011a). Furthermore; the original mineral supply to growing thermokarst lakes in ice-rich 591 areas such as the Lena Aldan interfluve may be greater than in areas with lower ice contents 592 (Pestryakova et al., 2012). The Mg/Ca ratio in connection with CaCO₃ (geo- and biogenic) 593 provides estimations not only about lake level changes (Haberzettl et al., 2007) but also lake 594 temperature, salinity, and photosynthetic activity during summer months. Generally, higher 595 Mg/Ca ratios and lower CaCO₃ values in the studied thermokarst deposits were interpreted to 596 reflect higher lake level and vice versa. For instance, CO₂ is often withdrawn from lake water 597 during summertime due to increased water temperatures and photosynthesis, thus shifting the 598 CO₂ equilibrium at pH values around 8 (typical for current thermokarst lakes in the Lena Aldan 599 interfluve region; Pestryakova et al., 2012) towards insoluble CaCO₃, increasing the Mg/Ca 600 ratio (e.g., Ji et al., 2005; Liu et al, 2008). Opitz et al. (2013) discussed high carbonate content 601 in lacustrine sediments of a thermokarst system on the Tibetan Plateau as an indicator of 602 shallow lake levels with corresponding ostracod associations. However, high sediment 603 carbonate content despite the absence of ostracods suggest also decreasing lake levels and 604 increasing salinity due to high evaporation (Liu et al., 2008; Wetterich et al., 2008; Ulrich et al., 605 2017b).

606 The Fe/Mn ratio is generally used to evaluate redox conditions in thermokarst lakes (Bouchard 607 et al., 2011, 2014; Biskaborn et al., 2013b). Since Mn is mobilized faster than Fe under anoxic 608 conditions, Fe accumulates residually under reducing conditions (e.g. Davison, 1993). During 609 sedimentation, this ratio is stored and thus preserved as an indicator of the redox milieu during 610 sediment deposition into the lacustrine archive. Increasing Fe/Mn values are thus interpreted as 611 indicative of a change to reducing limnic conditions during sediment accumulation. According 612 to Bouchard et al. (2011, 2014) those anoxic hypolimnic conditions are typical in thermokarst 613 lakes. In contrast, narrow Fe/Mn ratios indicate oxidizing conditions. According to Biskaborn 614 et al. (2013b), these are usually present in shallow waters or better-oxygenated marginal areas

615 of thermokarst lakes. Strong variations in the Fe/Mn ratio as seen in particular in the KB 616 sediment sequences thus suggest continuously changing redox conditions during sedimentation 617 at this location, which indicates either a change in lake level or a change between lacustrine and 618 terrestrial depositional conditions.

619 The Fe/S ratio was additionally used for qualitative estimations of thermokarst (lake) basin 620 sediment oxygenation. Lenz et al. (2016) discuss increasing Fe/S ratios in relation to increasing 621 lake water oxygenation as being due to increasing water depth and lake size for a thermokarst 622 lake in northern Alaska. They also suggest a relationship between lake-ice cover during 623 wintertime and decreasing Fe/S ratio; reduced water volume over lake sediments favors anoxic 624 conditions, in particular in the near-shore lake zone. The Fe/S ratio is also higher in relation to 625 terrestrial sediment deposition (Lenz et al., 2016), but in light of the data presented here it also 626 seems to increase after lake drainage or water loss and active layer oxygenation.

627

628 5.2 Grain-size EM modeling analysis of thermokarst deposits

629 Overall, the thermokarst deposits at both study sites have been transported and deposited in situ 630 with the Yedoma deposits as source sediment. In particular, the dating of the bulk organic 631 material suggests that there was no remarkable additional sediment input from other sources 632 than the Yedoma deposits during thermokarst evolution at the study sites. For instance, greater 633 Holocene dust entry into the CY thermokarst (lake) basins has not been proven so far. There is 634 still a knowledge gap concerning past wind pattern in the CY region. This is likely connected 635 to the fact that modern wind conditions are relatively calm throughout the year due to the highly 636 continental climate (see section 2). Similar conditions probably existed in East Siberia 637 throughout the Pleistocene/Holocene (Zimov et al., 1995). However, the polygenetic 638 provenance of the Yedoma source sediments is suggested to be partly influenced by aeolian 639 transport beside mainly proluvial, alluvial, fluvial transport, and lacustrine deposition (Soloviev, 640 1959; Katasonov and Ivanov, 1973; Schirrmeister et al., 2013). Péwé and Journaux (1983) even 641 discuss a dominant eolian component of silty Yedoma deposits in CY.

However, the depositional activities during thermokarst (lake) development overprinted the original depositional processes of the Yedoma source sediments. The final rEMs are therefore the result of the grain size fractionation during thermokarst development. With the rEMMA we were able to detect and to discuss them as single-endmembers, which is hardly possible from the classical measures of grain size properties (Dietze and Dietze, 2019). The rEMMA 647 presented here has yielded four final rEMs (Fig. 8). In general, the final rEMs can be assigned 648 to specific processes, allowing specific sediment transport and accumulation pathways during 649 thermokarst development to be identified (Table 2). The mean scores of the studied core 650 samples (Fig. 9) show the internal variability and thus the sediment history of each sediment 651 sequence taken from different geomorphological sites within the thermokarst basins. Generally, 652 the scores show that the internal variability of basin centers is rather low compared to that of

- basin edges and the small neighboring basins.
- 654

655 5.2.1 Distal transport dynamics

656 The rEM1 shows a very coarse-grained clay to very-fine silt. Thus, rEM1 is assigned to a (long) 657 distal transport or a low-energy sedimentation milieu under generally lacustrine conditions 658 (Table 2). In particular, the pronounced rEM1 and the high class-wise explained variance in the 659 range of the very fine particle sizes (Fig. 8) seem to be characteristic for CY thermokarst 660 deposits, and can therefore be used as a distinctive mark to distinguish these from Siberian 661 Yedoma deposits that show less pronounced or absent EMs in the clay fraction of comparable 662 EM analyses (Strauss et al., 2012; Schirrmeister et al., 2017). The rEM2 and rEM3 show silty 663 GSDs. These are background sedimentation signals, since the initial polymodal GSDs of 664 Yedoma deposits are mainly characterized by silty grain sizes (Soloviev, 1959; Schirrmeister 665 et al., 2011a). By comparison, rEM2 tend to have finer grain sizes, whereas rEM3 tend to have 666 coarser grain sizes. Based on the proportion of rEM2 and rEM3 in an individual thermokarst 667 sediment sequence (Fig. 9), it is possible to deduce the sedimentation milieu; rEM2 indicates 668 more distal transport or lower-energy conditions, whereas rEM3 indicates more proximal 669 transport or higher-energy conditions.

670

671 5.2.2 Local transport and in situ accumulation processes

At both study sites, rEM4 shows a strong influence of fine sand, which is also contained in the CY Yedoma deposits (Strauss et al., 2012; Schirrmeister et al., 2017). Thus, rEM4 (together with rEM3) is interpreted as an indicator of very short transport pathways and/or in situ accumulation of source sediments during thermokarst initiation. The former occurs especially in the immediate vicinity of eroding lake shores (Table 2).

678 5.2.3 Indications for thermokarst dynamics

679 During lake bank erosion, coarser components accumulate at the shores, while the finer grain 680 sizes can be transported to the lake basin center, resulting in grain size fractionation (Biskaborn 681 et al., 2013b). This requires a corresponding lake size, which can be assumed from the rEM 682 score of the KB3, YU2, and YU3 cores (Figs. 2 and 9). With the exception of these core 683 locations, the variations in rEM scores within all sediment cores show alternating homogeneous 684 and heterogeneous fractionation phases, suggesting that a clear sedimentological relationship 685 between a former lake center and corresponding lake shore areas of a single growing 686 thermokarst lake in each basin are difficult to detect at both study sites. This suggests that 687 sedimentary characteristics represent depositional environments of many smaller thermokarst 688 lake basins, which are characterized by changing water levels, poorer sediment fractionation, 689 and changing erosion and thermokarst activities. This is seen in rEM scores from the small 690 neighboring basins (KB6 and YU5) and the small pingo (KB7), in which lacustrine phases that 691 are associated with the dominance of fine-grained sediment accumulation (rEM1 and rEM2) 692 alternate with accumulation phases of coarser sediment (rEM3 and rEM4), probably originating 693 by lateral lake basin expansion and erosive shore processes (Biskaborn et al., 2013b; Lenz et 694 al., 2013). The core locations in the first case became more distal to the sediment source, 695 indicating open water zone enlargement; in the second case the disappearance of the 696 thermokarst lakes and a change to terrestrial depositional conditions is suggested, as seen in the 697 uppermost parts of cores KB6 and KB7. Furthermore, the dominance of coarser rEMs as seen 698 in the scores of the lower parts of these cores is interpreted to represent thermokarst initiation 699 stages, characterized by in situ thaw and subsequent Yedoma deposit subsidence (for e.g. KB7 700 see also Ulrich et al., 2017b).

701

5.3 Relationships between sedimentology and biogeochemistry in PCA

703 The PCA ordination results show that the sedimentological characteristics of the rEMMA 704 results are not or only very weakly correlated to the biogeochemical characteristics of the 705 thermokarst deposits (Fig. 10). Statistical analysis suggests that biogeochemical parameters, 706 with the exception of MS and the Fe/Mn ratio, are independent of thermokarst deposit 707 cryolithology and sedimentology. PC1 is dominated by all rEMs, the MS, and the Fe/Mn ratio, 708 and can thus be considered to largely reflect grain-size variations and depositional conditions. 709 However, while the MS increases with the input of coarser-grained terrestrial source sediments 710 (rEM3 and rEM4), a higher Fe/Mn ratio and thus reducing lacustrine depositional conditions

are related to the sedimentation of finer-grained sediments (rEM1 and rEM2) or less detritic input. PC2 is dominated by the biogeochemical parameters with a general separation of organicindicating and inorganic-indicating parameters. PC2 can thus be considered to indicate postsedimentary biogeochemical conditions, lake level changes, and organic input.

After plotting the sample scores of PC1 and PC2 against each core depth (Figs. 6 and 7), we can distinguish between fine-grained sedimentation along with rather high lake levels and/or reducing conditions (higher PC1 and PC2 scores), and coarse-grained sedimentation with rather shallow lake levels and/or oxidizing (i.e. terrestrial) conditions (lower PC1 and PC2 scores). An exception is shown by the YU2 basin center core with always negative PC2 scores that are likely the result of an exceptionally high Fe/S ratio due to oxidizing conditions in the active

121 layer and unfrozen talik sediments after lake disappearance (Lenz et al., 2016).

722

5.4 Late Glacial to late Holocene environmental conditions and thermokarst basin evolution

724 The sedimentary and biogeochemical composition of thermokarst deposits are complex, but are 725 specifically marked by the alternation of terrestrial and lacustrine phases (Bouchard et al., 2017). 726 The paleoecological and paleolimnological conditions during thermokarst (lake) basin 727 formation are ideally reconstructed by the use of bioindicators such as chironomides (e.g., 728 Nazarova et al., 2013), diatoms (e.g., Pestryakova et al., 2012; Biskaborn et al., 2013a; 729 Bouchard et al., 2013), ostracodes (Wetterich et al., 2008; Ulrich et al., 2017b), pollen (e.g., 730 Katamura et al., 2006; Klemm et al., 2016), cladocera (Frolova et al., 2014, 2017), amoebae 731 (Lenz et al., 2016), and macro remain analyses (Schleusner et al., 2015). Therefore the 732 differentiation of sedimentary facies and palaeoenvironmental changes are usually based on a 733 multi-proxy approach and an adequate geochronology (Bouchard et al., 2017).

734 Deriving clear age-depth relationships for all the thermokarst sediment sequences from the KB 735 and YU study sites is challenging. Generally, care should be taken with the interpretation of 736 dating of thermokarst deposits due to repeated mixing of brought in Yedoma sediments as well 737 as changing bio-ecological conditions and the rearrangement of already deposited material 738 during thermokarst development (e.g., Biskaborn et al., 2013b; Lenz et al., 2016; Jongejans et 739 al., 2018). Having this in mind and due to the small-scale varying depositional environments 740 discussed above, our core locations cannot be compared by the sediment ages and it is not 741 possible to pinpoint a uniform stratigraphy over the studied sediment sequences (see also Fig. 742 3, 6 and 7). However, in particular the individual combination of macro remain and bulk ages

743 as well as our multi-core and -proxy approach led us to discuss the Late Glacial to Holocene 744 evolution of thermokarst basins and geomorphological conditions in relation to literature-745 derived environmental and climatic changes in CY during four time slices (Fig. 12). Following 746 Gaglioti et al. (2014) we assume that our macro remain ages (Tab.1) represent the true 747 sedimentation age of the sediment layer. This make sense because the sampled organic macro 748 remains originate from terrestrial plants (grasses, trees, etc.) that grew in the immediate vicinity 749 of an evolving thermokarst lake and were probably incorporated into thermokarst deposits 750 immediately after they died. Our bulk ages are interpreted as inherited ages of organic matter 751 from the Yedoma deposits, which subsequently subsided or were erosionally redeposited due 752 to thermokarst processes and lake growth (Biskaborn et al., 2013b; Gaglioti et al., 2014).

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754 5.4.1 Late Glacial - Early Holocene thermokarst initiation

755 It is generally assumed that thermokarst development in CY was initiated by the onset of the 756 Holocene (e.g., Katamura et al., 2006). According to Fradkina et al. (2005), thermokarst lakes 757 began to form near the end of the Late Glacial in the Allerød after about 13,300 cal. yrs. BP. 758 Katamura et al. (2009) suggest thermokarst initiation between 11,000 and 9000 cal. yrs. BP, 759 basing their assumption on increased charcoal and specific pollen findings in thermokarst 760 deposits. Biskaborn et al. (2012) discuss enhanced thermokarst processes in relation to a transition to the mid-Holocene warm period after about 9100 cal. yrs. BP. Following former 761 762 research results in CY, lake sediment dating by Katasonov et al. (1979) for the KB study site 763 (Fig. 12a; see also above section 2), and in connection with the thermokarst sediment 764 rearrangement that is dated before the mid-Holocene Thermal Maximum (HTM) (see below 765 and Tab. 1), we assume that the onset of permafrost degradation at both study sites also took 766 place at the transition from Late Glacial to Early Holocene. Taking into account the 767 geomorphology of both studied basins, we assume, however, that the KB alas began to develop 768 earlier than the YU alas. Due to the climatic conditions during the Younger Dryas this likely 769 did not happen with the same intensity as later during the middle Holocene.

The late Pleistocene/Holocene transition in CY (ca. 11,.500 – 11,200 cal. yrs. BP) is characterized by an abrupt increase in tree pollen and a decline in steppe vegetation (Andreev et al., 1997; Fradkina et al., 2005; Andreev and Tarasov, 2013). During the Allerød Interstadial the average temperatures first rose, then fell again during the Younger Dryas (YD; Andreev et al., 2012). During the YD (between about 13,000 – 11,500 cal. yrs. BP), precipitation was about 150 mm per year lower than today (Andreev et al., 1997; Andreev and Tarasov, 2013). A cold, dry phase during the YD was assumed as well by Biskaborn et al. (2012) for eastern Siberia; as
they discussed, the resulting prolonged winter ice layer on thermokarst lakes caused reducing
water conditions. Nazarova et al. (2013) show that colder conditions than today existed between
10,000 and 8000 cal. yrs. BP, but they assume wet summers also.

780 Although clear evidence of the thermokarst initiation timing could not be deduced from the 781 studied sediments, we interpret the different biogeochemical characteristics of the lower basin 782 center core parts (>Fe/Mn, >S, >CaCO3) as indicators for initial thermokarst processes (i.e. 783 surface subsidence, ponding water; e.g. Bouchard et al., 2011; Biskaborn et al., 2013b; 784 Schleusner et al., 2015; Figs. 6 and 7). The evolution of both studied thermokarst basins likely 785 began at the KB3, YU2, and YU3 core locations (Fig. 12a). The late Pleistocene ages at the 786 bottom of these sediment sequences does not show the "true" deposition age but rather indicate 787 in situ thaw and subsequent Yedoma deposit subsidence (Tab. 1). The low TOC level in the 788 KB3, YU2, and YU3 basin center cores also suggests further organic matter decomposition 789 within a talik below the developing thermokarst lakes as described from taberal deposits found 790 below Yedoma thermokarst lakes elsewhere (Schirrmeister et al., 2011a; Wetterich et al., 2012; 791 Farquharson et al., 2016). The rEMs also show continuous low-energy deposition, a conclusion 792 mainly supported by a strong influence of rEM1 and rEM2. However, the base of the 793 thermokarst deposits was probably not drilled, as no grain size changes were found indicating 794 the somewhat sandier facies typical for CY Yedoma deposits (Windirsch, 2018).

795

796 5.4.2 Holocene thermal maximum thermokarst intensification

797 Our results show high sedimentation rates and increased bioproductivity in the thermokarst 798 deposits during the mid-HTM (about 7000 - 5000 cal. yrs. BP). This is especially evident by 799 higher organic carbon contents and decreasing δ^{13} C values in the KB7 core. In addition the 800 YU1 profile, for example, shows increased TOC contents and strong sediment rearrangements 801 during the middle Holocene. The exceptionally high KB7 core sedimentation rates in addition 802 to lacustrine microfossil and palynological data analyses are interpreted by Ulrich et al. (2017b) 803 to indicate rapid, highly dynamic lake evolution during the HTM. In particular, analysis of 804 changes in the species composition of subfossil cladoceras communities made it possible to 805 identify a rapid initiation, growth, and disappearance of an individual thermokarst lake at the 806 KB7 location (Frolova et al., 2017; Ulrich et al., 2017b). Permafrost thaw and extensive 807 thermokarst basin development was probably accelerated especially by rising precipitation 808 (Monserud et al., 1998; Fradkina et al., 2005; Grosse et al., 2013; Nazarova et al., 2013).

809 Terrestrial conditions changed quickly to lacustrine conditions, and thermokarst lakes rapidly 810 emerged and grew larger in a very short time before thermokarst processes declined locally 811 (Fig. 12b). The same can be seen in the KB6 sediment sequence of the small neighboring basin 812 at the KB study site. The grain-size means are higher in the lower and upper core parts but show 813 a significant increase of finer grain sizes (silt and clay) in the middle part. This change is also 814 reflected in the biogeochemical properties. However, the exact timing of lake drainage to the 815 KB basin is uncertain. The sedimentological and biogeochemical parameters from the upper 816 core indicate changing phases of higher and lower lateral erosion and changing lake levels. A 817 piece of wood sampled from about 90 cm bs was dated to 3316±61 cal. yrs. BP and originated 818 from a short, stable, but rather shallow lake phase (Fig. 3; Tab. 1). The basin center core 819 locations were probably already in a distal position to the lake shore as indicated by 820 homogeneous fine-grained sedimentation. However, the fluctuations in the Fe/Mn and Mg/Ca 821 ratios indicate changing redox conditions during lake level changes.

822 The HTM is characterized by rising temperatures and higher precipitation in CY (Fradkina et 823 al., 2005; Nazarova et al., 2013). According to Monserud et al. (1998), winter temperatures in 824 Siberia were about 3.7°C and July means about 0.7°C warmer than today and annual rainfall 825 was about 154 mm higher than today. Postglacial warming reached its maximum between about 826 6000 and 4600 cal. yrs. BP and was characterized by a similar vegetation composition as we 827 see today in Siberia (Monserud et al., 1998). Biskaborn et al. (2012) suggest the warmest period 828 of the mid-Holocene occurred between 7100 and 2800 cal. yrs. BP. At the HTM end, the 829 unification of large thermokarst lake basins took place and alases appeared as they do today. 830 The large pingos in the KB basin are an indication of that. Considering very slow pingo growth 831 rates of a few millimeters per year (Soloviev, 1973), the existence of ≤12-m-high pingos proves 832 that permafrost aggradation and talik refreezing must have started at that time. Furthermore, the 833 lenticular-layered to reticulate cryostructures as described from the basal parts of the sediment 834 sequences (Fig. 4), in particular from the KB alas, indicate the vertical refreezing of subaqueous 835 water-saturated talik deposits during lake disappearance (Katasonov and Ivanov, 1973; French 836 and Shur, 2010).

Macro remain dating of the upper YU1 profile shows an age of 4606±175 cal. yrs. BP and indicates strong sediment rearrangement during the decaying HTM, likely by thermoerosional processes at a lake basin shore. The uppermost sediments of the YU3 basin center core instead reveal a macro remain date of 3975±102 cal. yrs. BP and subaquatic to subaerial conditions at that time.

843 5.4.3 Late Holocene thermokarst cessation

844 Temperatures in eastern Siberia declined again after 4800 to 4500 cal. yrs. BP with minima 845 between 3000 and 2000 cal. yrs. BP (Biskaborn et al., 2012; Nazarova et al., 2013). This was 846 followed by a general warming trend and especially increasing winter temperatures in Siberia 847 but also more short-term temperature fluctuations (Meyer et al., 2015). Sedimentary conditions 848 are generally characterized by low carbon content, higher TOC/TN ratios, and low 849 sedimentation rates, all suggesting a general surface stability and permafrost aggradation below 850 large thermokarst basins during the late Holocene, However, very short favorable periods 851 probably led to higher lake levels and increasing thermoerosional processes at the basin edges. 852 In particular, the KB1 profile shows strong sediment rearrangements under subaquatic 853 conditions, likely via lake shore erosion and slumping processes between 1930±57 and 854 1812±72 cal. yrs. BP. The higher sand content within the KB1 sediment sequence, the very 855 poor sorting of the grain sizes, and the multiple inversions of the radiocarbon dates indicate 856 high geomorphic dynamics at the basin edge. The rEM scores of the KB1 profile also show this 857 trend towards higher proportions of coarser grain sizes. The biogeochemical records like the 858 low TOC and S values as well as high, varying TOC/TN, Mg/Ca, and Fe/Mn ratios suggest low 859 bioproductivity during sediment repositioning and accumulation and indicate increasing 860 subaerial conditions under short-term changing environmental conditions (Fig. 12c) (Biskaborn 861 et al., 2013b). Strong single events, e.g. collapsing lake-shore bluffs during lake expansion, are 862 an explanantion for the high sedimentation rates and comparable sedimentary conditions 863 (Biskaborn et al., 2013b; Lenz et al., 2013; Séjourné et al., 2015). However, we cannot directly 864 demonstrate comparable high geomorphologic dynamics in the YU thermokarst basin for the 865 same time period. Thus, a regional climate-relevant signal from late Holocene sediment 866 rearrangements as seen in the KB1 core cannot be detected. Our data point to the cessation of 867 thermokarst activity and extensive frost-induced processes (i.e. permafrost aggradation) for the 868 time after about 1.5 cal kyrs BP. Kachurin (1962) suggested that thermokarst processes in 869 Siberia were generally weaker during the late Holocene period. This is generally confirmed by 870 a late Holocene cooling trend, which is frequently observed from lake sediments in northeastern 871 Siberia (Popp et al., 2006; Swann et al., 2010; Biskaborn et al., 2012, 2013a; Klemm et al., 872 2016).

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- 874

875 5.4.4 Recent to modern renewed thermokarst dynamics

876 Satellite images from the YU basin show different elevated ground levels of the alas bottom 877 around the YU3 and YU1 core locations, which are characterized by drier vegetation conditions 878 (Figs. 2 and 3; see also Ulrich et al., 2017a and their Fig. 2). Different basin ground levels likely 879 indicate small-scale differences in the ground-ice content of the underlying Yedoma deposits 880 and/or differing strong subsidence of the surface and lake deepening (e.g. Morgenstern et al., 881 2013). They probably also mark the former remnants of Yedoma uplands, which have separated 882 several small thermokarst lakes basin before (Fig. 12b). However, the exact time of small lakes 883 coalescing into a larger lake cannot be determined on the basis of the available data. Also, we 884 believe that coalescence was more of a successive process, in which a lake with a higher water 885 level drained into another when the Yedoma bridge between them was sufficiently thawed and 886 eroded. The same is observed today at the YU study site, as the example of the YU5 location 887 shows. This lake drained into the YU alas in spring 2007 after very rainy years (Ulrich et al., 888 2017a).

889 Contemporary northern latitude warming has been confirmed in numerous studies and it is 890 generally known that related environmental changes in the high, northern latitudes and 891 permafrost landscapes can occur very quickly (Serreze et al., 2000; Serreze and Barry, 2011; 892 Grosse et al., 2016; Walter-Anthony et al., 2018). After about 1500 cal. yrs. BP and a relatively 893 long period of relatively stabile, inactive thermokarst processes (Fig. 12d; e.g., Kachurin, 1962), 894 the gradual warming of permafrost over recent decades in Siberia has led to profound changes 895 in the ecosystem (Fedorov et al., 2014a; Iijima et al., 2016). In CY, the rise in temperature in 896 addition to human impacts is leading to renewed high rates of subsidence and abrupt, rapid 897 thermokarst processes (Brouchkov et al., 2004; Fedorov and Konstantinov, 2009; Fedorov et 898 al., 2014b; Ulrich et al., 2017a). The thermokarst landscape in CY is characterized by a long 899 history of human land use (settlements, agriculture, horse and cattle breeding) since the 900 settling/colonization by the Yakut (Sakha) during the 13th century; these activities occurred on 901 the alases, influenced by thaw lake dynamics (Crate et al., 2017). Above all, the YU5 sediment 902 sequence from the neighboring small basin at the YU study site documents very well this 903 subrecent time period. During the short life of the thermokarst lake which developed in a former 904 agricultural area, strong surface subsidence and lateral lake basin expansion took place (Ulrich 905 et al., 2017a). However, we identified very little accumulation of lacustrine sediments . Thus, 906 we expect very low sedimentation rates in modern rapidly growing thermokarst lakes. Finally, 907 this shows that rapid thermokarst processes can happen during even short-term warm climate

conditions. This is proved for the past, is still comprehensible today in many permafrost areasand, above all, is accelerated by human land use in CY.

910

911 6 Conclusion

912 Our study emphasizes that alas and thermokarst lake evolution in CY Yedoma landscapes were 913 not a continuous process during the Late Glacial to Holocene period. Short-term phases of 914 forcing climatic conditions have led to very active thermokarst processes and rapid but locally 915 variable landscape modification. Similar processes are observed in the study region under 916 current climatic changes.

917 Specific sedimentological and biogeochemical thermokarst deposit characteristics allow the 918 reconstruction of erosion, transport, and sedimentation conditions. In particular, the GSD 919 unmixing procedure of the rEMMA offered us a unique and robust method to detect specific 920 grain-size fractionation and depositional processes during thermokarst development. This is not 921 possible by simple ordination methods (e.g., PCA) that usually cannot handle multimodal data 922 and require data linearity or by manual evaluation of the polymodal GSDs from the rearranged 923 polygenetic Yedoma source sediments. The biogeochemical parameters are statistically 924 separated into organic-dominated (e.g., TOC, TOC/TN, S, Mg/Ca) and inorganic-dominated 925 processes (e.g., CaCO₃, Fe/S, δ 13C). However, their general weak correlation to the 926 sedimentological characteristics represented by the four final rEMs are suggesting post-927 sedimentary biogeochemical conditions, lake level changes, and varying organic input.

928 Considering the rather low time resolution of our study as well as the discussed difficulties in 929 dating of thermokarst sediments and the small-scale sedimentation differences, the results of 930 the radiocarbon dating confirm an extensive deposition of reworked late Pleistocene Yedoma 931 source sediments in peripheral basin zones during thermokarst lake growth. The basin centers 932 are characterized by in situ thaw and subsidence of late Pleistocene sediments during early 933 thermokarst evolution phases, but the subsequent accumulation of clayey and fine silt sediments 934 is assigned to a distal transport or a low-energy sedimentation milieu under increasing lacustrine 935 conditions. Finally, the synopsis of rEMMA and all analyzed proxies in relation to the 936 geomorphological location of the cores reveals small-scale variability of sediment origin in 937 each CY alas. Individual small-scale varying depositional environments of many rapidly 938 growing small thermokarst lakes and basins during the mid-HTM, which finally coalesced, 939 have formed the large thermokarst basins we see today in CY.

940 Authors contribution

941 MU conceptualized the project, acquired the funding, collected the profiles, analyzed the data,

and wrote the manuscript. HM did the rEMMA, the statistical analysis, and wrote parts of and

943 corrected the manuscript. JoS, CS, LS, and AF assisted during field work, organized field work

944 logistics, did laboratory and data analysis, and/or wrote and corrected parts of the paper. BS did

945 the XRF laboratory analysis and corrected the paper. JeS and CZ have been involved in the

946 study and project design and advanced the manuscript with their ideas.

947 **Data availability**

948 All data for this paper are properly cited and referenced in the reference list. Moreover, data 949 may be available upon request to the corresponding author, or are already available as 950 supplementary data.

951 **Conflict of interest**

952 The authors declare that they have no conflict of interest.

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1315 Figures



Fig. 1. Study site overview. (a) Location of the study region and the key study sites (black stars)in the zone of continuous permafrost. Roman letters mark previous paleoenvironmental

1319 reconstructions from thermokarst lake sediments in Siberia; i) Andreev et al. (1997), ii) Biskaborn et al. (2012), iii) Biskaborn et al. (2013a), iv) Biskaborn et al. (2013b) / Schleusner 1320 1321 et al. (2014), v) Biskaborn et al. (2016), vi) Katamura et al. (2009), vii) Klemm et al., 2016, Popp et al. (2006). Map of permafrost distribution modified after Brown et al. (2002). (b) The 1322 1323 Lena-Aldan interfluve region (Landsat 8 closeup, July 2013, USGS). (c) Thermokarst 1324 development and lake properties differ on several Pleistocene accumulative-erosive terraces 1325 following Soloviev (1959). B: Bestyakh terrace, T: Tyungyulyu terrace, A: Abalakh terrace, M: Magan terrace. In particular the Tyungyulyu and the Abalakh terraces are differentiated due to 1326 1327 characteristics and distribution of ice-rich permafrost deposits. The stars are indicating the KB key study site (d) on the Tyungyulyu terrace and the YU key study site (f) on the Abalakh 1328 1329 terraces. For details see Figure 2 (DEM generated using data from the ESA DUE Permafrost 1330 Project (Santoro and Strozzi, 2012)).

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Fig. 2. Satellite images showing the drilling sites within (a) the KB basin and (b) the YU basin

- 1335 (Pleiades-1A subsets, 22 Sept. 2012). The dashed lines mark the topographic profiles illustrated1336 in Fig. 3.
- 1337



Fig. 3. Coring transects of the studied thermokarst basin in relation to geomorphology and topography. Topographic data based on geodetic field surveys conducted in summer 2014 (see Ulrich et al., 2017a). The radiocarbon dates are shown as cal. yrs. BP for each dated core. The dating of bulk material is highlighted by italics and the macro remains are highlighted by the wooden stick symbols.



Fig. 4. Example photos of sediment composition, color, and cryolithological properties as seen
in the field from the different thermokarst deposit cores. (a) YU2, (b) KB7, (c) KB6, (d) YU5,
(e) YU1 (f) KB1, (g) YU3, (h) YU1, (i) KB3, (j) YU1, (k) KB4, (l) YU1. Please see scales for
sample depths.



Fig. 5. GSD curves of all thermokarst deposit cores with the KB study site (left) and the YU study site (right). The gray levels of the GSD curves decrease with core depth. Clay, silt, and sand are divided into fine (f), middle (m), and coarse (c) fractions.



1354

1355 Fig. 6. Sedimentological and biogeochemical results of all KB cores against sample depth with 1356 the macro remain and bulk ages in cal. kyrs. BP, the GSD including contents of clay (dark gray), 1357 silt (white), sand (light gray) and the grain-size mean as MGS, the MS in SI units (10-6 m3 kg-1358 1), TOC, TOC/TN, 813C, CaCO3, and selected elements and element ratios from XRF 1359 measurements. Additionally, the sample scores on PC1 and PC2 are plotted (See section 4.4). 1360 Please note the different scaling of the macro remain and bulk ages as well as TOC, S, and 1361 Mg/Ca values and the PC2 scores of KB7. The dashed horizontal lines mark the observed active 1362 layer depths.



1364 Fig. 7. Sedimentological and biogeochemical results of all YU cores against sample depth with 1365 the macro remain and bulk ages in cal. kyrs. BP, the GSD including contents of clay (dark gray), silt (white), sand (light gray), and the grain-size mean as MGS, the MS in SI units (10-6 m3 kg-1366 1367 1), TOC, TOC/TN, 813C, CaCO3, and selected elements and element ratios from XRF 1368 measurements. Additionally, the sample scores on PC1 and PC2 are plotted (See section 4.4). 1369 Please note the different scaling of the macro remain and bulk ages as well as of the S values 1370 of YU5, and the Fe/S values of YU2. The dashed horizontal lines mark the observed active 1371 layer depths.



Fig. 8. EMMA results from all input data showing final rEMs and their explained variances,
class-wise explained variances, and sample-wise explained variances. The robust scores are
plotted with their confidence intervals for each core in their stratigraphic order, i.e. from top
(left) to bottom (right).



Fig. 9. The mean scores (i.e. the relative contribution of an rEM to each sample) for all studiedcores against depth.



Fig. 10. Ordination plot of the PCA on all rEM scores and biogeochemical parameters for PC1
against PC2. For interpretation purposes, all core samples are additionally projected into the
ordination graph.



1389Fig. 11. TOC/TN- δ 13C relationship for all core samples reflecting organic matter origin. The1390classification of organic matter source follows Meyers and Teranes (2001).



(A) Late Pleistocene / Early Holocene



(B) ~ 7000 - 5000 cal. yrs. BP (Climate optimum)



(C) ~ 3500 - 1500 cal. yrs. BP





- **Fig. 12.** Schematic diagram showing the generalized thermokarst landscape evolution in CY as
- 1394 inferred from the sedimentology, biogeochemistry, and rEMMA analysis of the two studied
- 1395 thermokarst basins. Drawings are not to scale.

1396 Tables

1397	Tab. 1. Radiocarbon dating results used in this study are ordered due to the dated materials and
1398	depth. Dating differences of bulk organic material are additionally highlighted in italics.
1399	Calibrated ages refer to the 2σ range.

Lah ID	Sample name		¹⁴ C ag	ges	δ ¹³ C	Calibrated ages		
MAMS	(including depth in cm bs)	Dated material	yrs BP	(±)	(‰)	cal. yrs. BP	(±)	
		KB Bas	in edge					
21300	KB1 145-125	Wood	1890	19	-32.9	1812.5	72.5	
23115	KB1 222-208	Plant remains	1969	24	-21.0	1930.5	57.5	
23116	KB1 243-232	Plant remains	3091	25	-22.6	3314	62	
21299	KB1 55-35	Bulk	7698	27	-28.4	8482	60	
21301	KB1 145-125 (2)	Bulk	11,317	32	-24.7	13,207.5	91.5	
21302	KB1 250-244	Bulk	9619	28	-24.5	1,0978	189	
		KB Basi	n center					
21303	KB3 63-52	Bulk	11,024	37	-24.2	12,902.5	184.5	
23117	KB3 205-199	Bulk	2,8550	120	-29.1	32,877	527	
21304	KB3 222-204	Bulk	29,660	120	-31.0	34,340	400	
21305	KB3 320-306	Bulk	33,660	160	-31.1	38,355	615	
21306	KB7 60	Plant remains	4984	24	-26.3	5752	102	
21307	KB7 154-137	Wood	5587	25	-34.2	6358	51	
23120	KB7 243-224	Wood	5735	27	-15.5	6542	91	
23121	KB7 285-265	Wood	5767	27	-18.5	6568	75	
23122	KB7 320-306	Plant remains	5790	27	-15.5	6582	78	
21308	KB7 264-258	Bulk	12060	40	-30.0	13910	120	
21309	KB7 382-365	Bulk	22,210	70	-30.7	26,835	645	
		KB Neighboring st	nall drained	d basin				
23118	KB6 92	Wood	3093	25	-29.5	3316	61	
23119	KB6 295-280	Bulk	29,190	140	-26.6	33,925	575	
		YU Bas	in edge					
23123	YU1 110-100	Plant remains	4046	25	-27.3	4606	175	
21310	YU1 50-30	Bulk	9918	33	-12.5	11318.5	84.5	
21311	YU1 219-206	Bulk	32,670	150	-28.7	37,225	585	
23124	YU1 317-301	Bulk	16,930	60	-28.8	20,115	235	
21312	YU1 414-398	Bulk	14,500	50	-28.8	17,595	325	
		YU Basi	n center					
21313	YU2 91-76	Bulk	5318	25	-27.8	6092.5	91.5	
21314	YU2 253-240	Bulk	20,280	60	-17.1	24,185	265	
23125	YU3 40-36	Charcoal	3637	26	-27.8	3975	102	
23126	YU3 185-170	Plant remains	30,300	140	-17.7	34,865	235	
23127	YU3 226-208	Bulk	30,500	140	-15.4	34,955	275	
		YU Neighboring st	nall drained	d basin				
23128	YU5 40-20	Plant remains	-189	21	-22.5	modern		
23129	YU5 222-210	Plant remains	34,280	200	-18.6	39,340	650	
21315	YU5 40-20 (2)	Bulk	23,450	90	-28.9	27,625	165	
21316	YU5 183-170	Bulk	33,100	170	-30.3	37,290	700	
21317	YU5 318-300	Bulk	37,080	240	-28.3	41,625	405	

Final EM	Mode (µm)	Explained variance (%)	Grain size	Source, transport and accumulation pathways
rEM1	2.9	24	coarse clay to very-fine silt	distal, homogenuous fractionation under lacustrine thermokarst lake conditions
rEM2	27.4	18	medium to coarse silt	distal to proximal, heterogenuous fractionation under lacustrine thermokarst lake conditions and Yedoma background signal
rEM3	39.8	25	coarse silt	proximal, heterogenuous fractionation under terrestrial conditions and Yedoma background signal
rEM4	176.8	32	fine to medium sand	in situ, homogenuous fractionation due to eroding lake shores and basin slopes

Tab. 2. Robust grain-size endmembers, their characteristics, local source as well as transport and accumulation pathways.

Supplementary data for

Holocene thermokarst dynamics in Central Yakutia – A multi-core and robust grain-size endmember modeling approach

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Appendix A.

Additional sedimentological parameters, elements, and elemental ratios for all thermokarst deposit cores that were additionally used for interpretation and discussion of thermokarst (lake) processes.

Appendix B.

PCA ordination plots and explained PC variances on all robust EM scores and biogeochemical parameters for all PCs. For interpretation purposes, all core samples are additionally projected into the ordination graphs.

Depth	Grain size	Grain size	TIC	ТС	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
45	26.4	3.9	0.4	1.0	0.0	843.8	27155.0	565.2	28260.0	389,6
95	32.1	4.1	0.4	0.8	0.0	1131.0	29050.0	679.8	27280.0	455,95
135	21.8	4.3	0.0	0.6	0.0	594.5	14825.0	426.7	24835.0	430,95
150	30.7	5.0	0.4	0.9	0.0	1074.5	30480.0	822.0	29280.0	423,15
190	20.9	4.2	0.2	1.2	0.1	836.4	22730.0	619.9	29155.0	426,75
201.5	19.5	4.8	0.3	1.0	0.1	942.0	23445.0	628.4	31570.0	379,5
215	19.0	4.9	0.3	1.0	0.1	926.4	23370.0	601.5	31610.0	372,85
227	18.8	4.4	0.5	1.2	0.1	960.9	30455.0	717.2	30340.0	377,7
237.5	20.1	4.2	0.4	1.1	0.0	857.0	29530.0	619.3	29395.0	395,9
247	18.6	4.3	0.4	1.1	0.0	922.2	28425.0	685.6	30130.0	396,25
256.5	24.8	4.7	0.4	1.0	0.1	892.6	28255.0	683.4	31460.0	366,15
269	19.4	4.5	0.4	1.0	0.1	934.0	26090.0	642.3	31040.0	388,65
280.5	20.2	4.7	0.4	1.2	0.1	988.7	26635.0	650.0	31300.0	369,65
292	21.2	4.9	0.3	1.3	0.1	997.6	24420.0	611.0	30235.0	375,95
302.5	38.5	5.5	0.3	1.0	0.1	961.4	32480.0	633.2	24980.0	345,75
min	18.6	3.9	0.0	0.6	0.0	594.5	14825.0	426.7	24835.0	345,8
max	38.5	5.5	0.5	1.3	0.1	1131.0	32480.0	822.0	31610.0	456,0
mean	23.5	4.5	0.4	1.0	0.1	924.2	26489.7	639.0	29391.3	393,0
SD	5.8	0.4	0.1	0.2	0.0	117.3	4197.6	81.3	2136.9	28,4

Appendix A. Table S2. KB4

Depth	Grain size	Grain size	TIC	ТС	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
69.5	15.0	4.3	0.2	1.0	0.0	875.0	25175.0	536.1	30215.0	342,25
78.5	16.9	4.5	0.2	0.5	0.0	933.4	19905.0	516.1	29980.0	421,5
95	22.5	4.7	0.7	1.1	0.0	959.6	41715.0	609.3	27970.0	436,15
115	17.0	4.5	0.7	1.1	0.0	891.3	34980.0	706.3	31445.0	329,35
128	20.0	4.5	0.4	0.8	0.0	951.9	28215.0	634.0	29775.0	428,55
138	17.0	4.8	0.5	0.8	0.0	971.1	29000.0	613.0	29500.0	430,6
147.5	21.1	4.5	0.5	0.9	0.0	902.6	31050.0	668.3	30070.0	439,5
155	14.5	4.6	0.5	0.8	0.0	868.7	25900.0	646.7	32075.0	350,4
166	18.7	4.4	0.4	0.8	0.0	884.6	25230.0	651.1	31665.0	358,2
176	16.8	4.4	0.5	0.9	0.0	912.1	28100.0	663.1	32190.0	366,05
199	18.2	4.2	0.8	1.4	0.0	1001.5	41050.0	858.1	31565.0	360,95
210	18.1	4.2	0.8	1.4	0.1	1039.5	42955.0	839.2	31490.0	347,55
222.5	21.1	5.2	0.6	1.1	0.1	940.4	34055.0	697.2	31380.0	354,5
237	17.8	4.8	0.4	0.9	0.1	927.4	25760.0	576.1	31325.0	354,8
262	17.9	4.7	0.6	1.1	0.1	962.3	33155.0	697.3	31695.0	355,85
277.5	16.9	4.9	0.4	0.9	0.1	973.0	25260.0	621.4	31470.0	353,95
291.5	18.9	5.2	0.3	0.9	0.1	902.1	20535.0	537.3	29790.0	330,85
304	16.8	5.1	0.3	0.8	0.1	931.4	21095.0	549.7	30570.0	363,85
min	14.5	4.2	0.2	0.5	0.0	868.7	19905.0	516.1	27970.0	329,4
max	22.5	5.2	0.8	1.4	0.1	1039.5	42955.0	858.1	32190.0	439,5
mean	18.0	4.6	0.5	1.0	0.0	934.8	29618.6	645.5	30787.2	373,6
SD	2.0	0.3	0.2	0.2	0.0	44.1	6907.1	91.3	1080.4	37,1

Appendix A. Table S3. KB3

Depth	Grain size	Grain size	TIC	ТС	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
57.5	18.1	4.3	0.8	1.2	0.0	845.0	37855.0	673.3	30770.0	379,6
82	17.1	4.1	0.4	0.9	0.0	887.0	26735.0	662.4	33060.0	373,9
98	15.6	4.3	0.4	0.9	0.0	1018.0	28205.0	795.5	35065.0	365,1
116	15.0	4.6	0.4	0.8	0.0	887.3	24610.0	670.9	33365.0	363,1
132.5	16.1	4.5	0.2	0.6	0.0	855.0	17405.0	517.0	31965.0	369,85
149	17.5	4.5	0.2	0.7	0.0	842.6	18165.0	504.9	32915.0	375,45
163	16.3	4.6	0.2	0.8	0.1	860.6	17015.0	533.9	32645.0	379,35
191	15.5	4.5	0.2	1.3	0.1	895.3	18320.0	687.0	31610.0	385,9
202	17.6	4.2	0.6	1.8	0.1	962.8	34670.0	904.2	32320.0	348,4
213	17.8	4.1	0.3	1.5	0.2	974.8	22295.0	752.1	32230.0	380,1
226	17.7	4.3	0.2	1.3	0.1	924.4	15650.0	672.5	32290.0	369,7
236.5	17.8	4.0	0.2	1.3	0.1	943.5	16425.0	681.2	32025.0	379,6
251	16.3	4.3	0.2	1.8	0.2	1056.5	18060.0	1095.0	35315.0	370,9
264.5	14.2	4.3	0.1	1.4	0.2	967.3	12880.0	739.9	34005.0	374,75
286	17.4	4.3	0.0	1.3	0.2	817.4	11685.0	486.5	30190.0	393,5
299	17.1	4.5	0.6	1.6	0.1	1012.0	33480.0	747.2	32030.0	409,35
313	16.3	4.5	0.4	1.6	0.1	1003.5	26230.0	702.1	31440.0	393,75
min	14.2	4.0	0.0	0.6	0.0	817.4	11685.0	486.5	30190.0	348,4
max	18.1	4.6	0.8	1.8	0.2	1056.5	37855.0	1095.0	35315.0	409,4
mean	16.7	4.4	0.3	1.2	0.1	926.6	22334.4	695.6	32543.5	377,2
SD	1.1	0.2	0.2	0.4	0.1	70.0	7585.7	145.9	1308.8	13,3

Appendix A. Table S4. KB7

Depth	Grain size	Grain size	TIC	TC	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
50	37.1	6.1	0.0	5.1	0.5	2318.5	8891.5	649.6	42975.0	179,25
80	16.1	4.2	0.0	4.7	0.5	1083.5	10810.0	433.7	27995.0	320,3
100	23.3	5.0	0.0	10.1	0.9	1030.5	10970.0	553.9	26085.0	285,1
129	38.5	5.3	0.0	6.9	0.6	1111.0	12860.0	547.4	27570.0	335
146	21.8	4.7	0.0	7.7	0.7	1302.0	11905.0	579.6	29445.0	286,95
159	20.6	4.9	0.0	6.5	0.6	1641.5	11950.0	618.1	29680.0	301,5
175	15.0	4.4	0.0	6.4	0.5	1449.5	13990.0	585.2	28960.0	303,1
195	18.3	5.8	0.1	6.1	0.6	1893.5	12690.0	669.4	34290.0	225,55
210	11.9	4.7	0.3	4.2	0.4	1924.5	21450.0	1024.0	33350.0	247,4
220	28.9	5.1	0.3	2.4	0.2	1543.0	23265.0	875.7	28695.0	386,35
229	25.0	4.6	0.3	1.4	0.1	1245.5	23535.0	758.4	26135.0	447,05
251	31.2	4.6	0.3	1.5	0.1	1096.0	23260.0	811.2	27355.0	365,5
261	25.2	4.6	0.4	1.2	0.1	1147.5	26610.0	1048.0	29585.0	330,35
275	25.9	4.6	0.4	1.4	0.1	1189.0	28885.0	1029.0	30165.0	386
295	26.4	4.8	0.5	1.5	0.1	1086.5	29495.0	879.0	30985.0	372,8
313	27.1	4.9	0.5	1.4	0.1	1236.5	33480.0	1006.0	29980.0	443,45
330	32.6	4.7	0.6	1.5	0.1	1163.0	36095.0	969.1	29885.0	381,9
353	29.1	4.8	0.8	1.5	0.1	1293.5	47205.0	923.0	27740.0	381,2
374	24.7	4.8	0.9	1.5	0.1	1202.0	47790.0	925.3	28515.0	390,6
min	11.9	4.2	0.0	1.2	0.1	1030.5	8891.5	433.7	26085.0	179,3
max	38.5	6.1	0.9	10.1	0.9	2318.5	47790.0	1048.0	42975.0	447,1
mean	25.2	4.9	0.3	3.8	0.3	1366.2	22901.9	783.4	29967.9	335,2
SD	6.8	0.4	0.3	2.7	0.3	340.9	11702.7	192.0	3676.6	69,0

Appendix A. Table S5. KB6

Depth	Grain size	Grain size	TIC	TC	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
39	19.7	4.1	0.5	1.3	0.1	869.3	28645.0	656.9	29540.0	361,65
54	44.0	4.6	0.4	1.0	0.1	1039.0	27295.0	647.5	31680.0	380,1
63	28.4	4.5	0.5	0.9	0.0	996.7	28785.0	661.9	31905.0	364,85
100.5	17.9	4.2	0.5	0.8	0.0	972.9	28910.0	609.2	31725.0	374,95
110	39.3	5.0	0.4	0.8	0.0	933.7	27110.0	589.8	31845.0	376,8
142	28.8	4.5	0.4	1.1	0.1	837.7	24370.0	757.7	33705.0	355,55
151.5	18.3	4.1	0.2	1.1	0.1	873.5	18110.0	671.9	32990.0	368,4
162	29.8	4.6	0.6	1.4	0.1	1009.7	34040.0	704.9	33975.0	366,25
168.5	23.6	4.9	0.5	1.3	0.1	977.1	29415.0	782.9	32385.0	369,65
174.5	18.1	4.2	0.4	1.1	0.1	906.4	26275.0	758.4	32645.0	359,05
179	15.4	4.4	0.3	1.0	0.1	844.9	19965.0	692.2	33055.0	367
191	14.8	4.3	0.2	1.0	0.1	887.9	18020.0	647.5	32605.0	372,05
207.5	16.6	4.3	0.3	1.0	0.1	938.6	20555.0	663.2	31740.0	368,7
224	16.9	4.4	0.4	1.2	0.1	947.0	23740.0	672.6	32085.0	390,85
239.5	15.3	4.4	0.4	1.3	0.1	985.7	24995.0	625.6	32460.0	360,4
252	16.4	4.2	0.5	1.4	0.1	1022.5	28365.0	619.5	32070.0	360,45
261	14.5	4.4	0.5	1.5	0.1	1042.5	27205.0	668.8	32405.0	371,15
274	17.3	4.0	0.4	1.4	0.1	1030.5	23650.0	758.2	33020.0	368,45
287.5	24.9	4.7	0.4	1.3	0.1	1017.1	22625.0	835.9	33575.0	372,3
300	37.0	4.9	0.4	1.0	0.1	951.5	25525.0	709.5	32385.0	359,8
311	32.7	4.6	0.4	0.9	0.1	1005.0	22540.0	646.4	33015.0	355,55
min	14.5	4.0	0.2	0.8	0.0	837.7	18020.0	589.8	29540.0	355,6
max	44.0	5.0	0.6	1.5	0.1	1042.5	34040.0	835.9	33975.0	390,9
mean	23.3	4.4	0.4	1.1	0.1	956.6	25244.8	684.8	32419.5	367,8
SD	8.7	0.3	0.1	0.2	0.0	63.7	3948.9	61.2	914.2	8,4

Depth	Grain size	Grain size	TIC	ТС	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
10	30.2	5.0	0.2	4.2	0.3	1611.0	24100.0	690.3	31245.0	323,8
25	19.0	4.5	1.7	4.1	0.2	1004.3	62715.0	658.9	28580.0	293,05
40	29.3	4.9	0.9	1.5	0.0	781.8	43865.0	855.5	32725.0	334,95
70	23.0	4.0	0.5	1.0	0.1	863.9	29710.0	741.4	32940.0	373,35
95	30.7	5.0	0.1	0.7	0.1	927.4	17340.0	581.3	36140.0	359,45
105	18.3	4.4	0.3	1.3	0.1	937.5	22815.0	586.8	31470.0	341,4
130	17.9	4.5	0.5	1.0	0.1	962.4	29625.0	715.2	36505.0	346,4
157	21.1	4.9	0.5	1.0	0.1	1067.5	28790.0	736.0	37335.0	343,65
170	17.4	4.5	0.4	0.9	0.1	902.3	24010.0	685.6	36620.0	338,45
179	18.5	4.6	0.4	0.9	0.1	924.7	23950.0	672.2	36040.0	349,6
187	21.1	4.1	0.5	1.0	0.1	891.5	25765.0	609.7	34545.0	349,15
197	17.9	4.7	0.4	0.9	0.1	855.5	22155.0	558.9	33880.0	337,35
213	16.6	3.9	0.1	1.5	0.2	828.3	16060.0	661.1	34060.0	347,15
226	17.4	4.5	0.2	1.5	0.2	949.5	16605.0	796.2	36135.0	342,2
242	15.0	4.4	0.3	1.6	0.2	990.8	21445.0	867.6	36320.0	351,1
257	16.4	4.5	0.3	1.4	0.2	951.7	17465.0	842.7	36370.0	355,75
269	16.1	4.3	0.2	1.4	0.2	953.8	17625.0	767.4	35785.0	369,7
282	31.4	5.0	0.3	1.4	0.1	969.5	20200.0	870.9	35865.0	353,7
297	15.2	4.2	0.5	1.3	0.1	883.4	28880.0	798.4	32705.0	355,5
309	19.4	4.4	0.6	1.1	0.1	909.2	32945.0	743.6	33920.0	352,85
321	14.3	3.9	0.7	1.3	0.1	916.3	37190.0	763.1	33820.0	344,8
329	24.1	3.9	0.6	1.1	0.1	892.9	33065.0	760.9	34200.0	348,85
339	32.7	5.0	0.8	1.3	0.1	847.9	38135.0	655.7	32630.0	338,9
354	22.0	4.1	0.9	1.6	0.1	872.4	46820.0	675.7	31200.0	348,35
369	17.8	4.5	0.8	1.4	0.1	872.7	40315.0	666.2	31790.0	332,35
385	15.1	4.1	0.9	1.6	0.1	947.8	47095.0	651.8	31210.0	340,9
406	32.6	4.4	0.4	1.1	0.1	941.8	26645.0	745.0	33760.0	359,25
min	14.3	3.9	0.1	0.7	0.0	781.8	16060.0	558.9	28580.0	293,1
max	32.7	5.0	1.7	4.2	0.3	1611.0	62715.0	870.9	37335.0	373,4
mean	21.1	4.5	0.5	1.4	0.1	942.9	29456.7	716.9	33992.4	345,6
SD	5.9	0.4	0.3	0.8	0.1	143.2	11119.1	85.6	2159.0	14,7

Appendix A. Table S6. YU1

Appendix A. Table S7. YU2

Depth	Grain size	Grain size	TIC	TC	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
10	14.4	4.0	1.3	3.6	0.2	748.3	30250.0	738.4	31525.0	283,45
28	12.7	4.0	0.6	1.4	0.1	671.6	26340.0	613.9	34355.0	273,85
44	15.9	4.2	0.4	0.9	0.1	737.9	25090.0	627.4	31570.0	367,4
65	14.1	4.5	0.2	0.9	0.1	817.5	18890.0	659.1	31555.0	370,6
82	16.7	4.1	0.2	0.7	0.1	936.5	18685.0	707.2	31000.0	377,8
152	15.8	4.3	0.6	1.1	0.0	875.4	30080.0	699.7	32520.0	351,55
168	19.0	4.0	0.5	1.0	0.0	928.4	28085.0	631.7	32930.0	363,75
194	15.7	4.3	0.5	1.0	0.1	885.9	26045.0	601.4	31860.0	363,55
207	15.9	4.2	0.7	1.2	0.1	851.5	31760.0	652.2	32045.0	358,75
230	16.7	4.2	0.6	1.2	0.1	878.0	28100.0	653.0	32365.0	358
247	13.8	4.5	0.9	1.7	0.1	876.9	39645.0	796.7	34000.0	327,2
277	14.8	4.3	0.9	1.7	0.1	924.9	41290.0	730.9	33750.0	328,1
293	14.9	4.2	0.8	1.6	0.1	886.0	36900.0	752.7	33975.0	326,2
min	12.7	4.0	0.2	0.7	0.0	671.6	18685.0	601.4	31000.0	273,9
max	19.0	4.5	1.3	3.6	0.2	936.5	41290.0	796.7	34355.0	377,8
mean	15.4	4.2	0.6	1.4	0.1	847.6	29320.0	681.8	32573.1	342,3
SD	1.5	0.2	0.3	0.7	0.0	78.3	6663.0	57.9	1079.1	31,7

Appendix A. Table S8. YU3

Depth	Grain size	Grain size	TIC	ТС	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
30	18.4		1.2	1.9	0.1	637.0	38950.0	642.9	31140.0	329,7
38	17.4	4.3	0.5	0.9	0.0	709.9	29600.0	633.5	30535.0	391,05
81	15.6		0.8	1.4	0.1	819.8	36405.0	783.9	34515.0	340,2
92	17.3	4.4	0.8	1.4	0.1	882.0	35215.0	874.9	33890.0	329,95
100	16.1	4.3	0.5	1.2	0.1	1021.0	26270.0	800.5	36135.0	352,2
113	16.7	4.5	0.6	1.2	0.1	1081.0	27845.0	693.1	35505.0	343,75
149	13.7	4.4	0.3	1.3	0.1	914.4	21225.0	646.8	35730.0	344,85
178	14.4	4.5	0.2	1.6	0.2	963.9	19950.0	693.5	34530.0	356,95
190	14.3	4.2	0.5	1.4	0.1	1047.0	27220.0	785.8	34770.0	358,85
202	28.9	5.6	0.5	1.4	0.1	956.6	29920.0	773.2	35065.0	358,05
217	16.0	4.4	0.9	1.9	0.1	999.8	46155.0	760.5	32500.0	349,1
235	18.5	4.3	0.9	1.9	0.1	1021.0	45090.0	729.3	32225.0	337,8
255	18.6	4.2	0.7	1.8	0.1	905.7	37435.0	749.1	33130.0	348,9
267	18.6	4.5	0.9	2.1	0.1	1032.5	44200.0	764.8	31325.0	343,1
277	15.8	4.5	0.7	1.8	0.1	932.8	38025.0	768.9	33860.0	352,6
min	12 7	4.2	0.2	0.0	0.0	637.0	10050.0	633 5	30535.0	320.7
111111	13.7	4.2	0.2	0.9	0.0	1091.0	19930.0	033.3 874.0	26125.0	329,7
max	20.9	3.0 4.5	1.2	2.1 1 5	0.2	1081.0	40155.0	0/4.9 740.0	30133.0	391,1
mean	17.3	4.5	0.7	1.5	0.1	928.3	33567.0	/40.0	33657.0	349,1
SD	3.5	0.3	0.2	0.3	0.0	121.1	8049.6	64.7	1709.9	14,3

Appendix A. Table S9. YU4

Depth	Grain size	Grain size	TIC	TC	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
23	14.4	4.5	1.3	2.0	0.1	677.0	36995.0	690.1	32260.0	319,5
47	17.0	4.3	0.8	1.2	0.0	710.6	32730.0	722.6	32785.0	346,4
80	25.2	5.2	0.6	0.9	0.0	714.5	31385.0	655.6	32370.0	350,8
90	22.2	4.6	0.5	0.8	0.0	848.6	28270.0	604.9	32820.0	356,9
115	20.5	4.0	0.5	0.8	0.0	899.9	26610.0	604.8	33915.0	356,35
127	22.0	5.1	0.5	0.9	0.0	894.5	29430.0	657.1	33985.0	358,2
150	15.6	4.5	0.6	0.9	0.0	890.2	27415.0	646.8	34365.0	343,15
175	29.1	5.0	0.7	1.5	0.1	922.3	35160.0	821.5	33665.0	371
185	25.2	4.9	0.5	1.4	0.1	910.7	26405.0	748.9	34840.0	356,25
200	16.2	4.5	0.2	1.6	0.2	956.9	18890.0	829.0	36210.0	360,45
219	18.9	4.7	0.3	2.0	0.2	1081.5	22180.0	858.5	35925.0	344,35
228	19.3	4.7	0.1	2.1	0.2	1061.0	17730.0	702.9	35315.0	356,1
256	16.6		0.1	2.7	0.3	1057.5	15670.0	729.1	35630.0	337,65
274	18.2	4.3	0.5	2.1	0.2	1131.5	31125.0	723.9	34850.0	338,55
290	19.6	4.7	0.6	2.5	0.2	1215.0	32620.0	761.7	35050.0	354,45
316	15.8	4.4	0.3	2.2	0.2	1055.5	22070.0	785.9	36665.0	349,85
324	20.8	4.7	0.4	1.9	0.2	1124.5	26370.0	772.8	35460.0	350,25
min	14.4	4.0	0.1	0.8	0.0	677.0	15670.0	604.8	32260.0	319,5
max	29.1	5.2	1.3	2.7	0.3	1215.0	36995.0	858.5	36665.0	371,0
mean	19.8	4.6	0.5	1.6	0.1	950.1	27120.9	724.5	34477.1	350,0
SD	3.9	0.3	0.3	0.6	0.1	152.4	5956.2	73.6	1321.3	11,1

Appendix A. Table S10. YU5

Depth	Grain size	Grain size	TIC	ТС	TN	Р	Ca	Mn	Fe	Zr
[cm bs]	mean [µm]	sorting	[wt%]	[wt%]	[wt%]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]	[mg/kg]
10	17.8	4.7	0.3	1.7	0.1	814.5	29665.0	750.3	34070.0	344,3
30	18.2	4.9	0.2	2.3	0.2	874.9	26335.0	737.7	34095.0	306,2
50	16.0	4.4	0.4	2.2	0.2	897.4	24500.0	678.4	33935.0	328,3
80	16.1	4.3	0.4	2.9	0.3	962.0	24695.0	738.2	34535.0	324,6
104	16.9	4.5	0.4	2.8	0.2	1028.5	29620.0	741.4	33950.0	339,4
150	16.4	4.6	0.1	2.6	0.3	1036.5	16080.0	692.5	33845.0	346,95
160	17.3	4.9	0.3	2.3	0.2	971.8	31205.0	730.3	33470.0	336,4
176	17.0	4.4	0.2	1.9	0.2	935.0	22195.0	752.3	34290.0	345,9
188	21.0	5.1	0.1	1.8	0.2	966.3	17915.0	695.4	33840.0	337,45
205	24.4	5.3	0.2	2.5	0.3	1001.2	19815.0	751.3	34380.0	358,1
216	19.2	4.8	0.1	2.6	0.3	988.3	18655.0	772.8	34020.0	358,05
232	17.7	4.8	0.5	2.2	0.2	963.5	35010.0	811.4	33230.0	369,15
242	27.5	4.9	0.5	2.2	0.2	920.5	30330.0	853.0	33295.0	377,2
252	17.8	4.4	0.5	2.1	0.2	951.1	28755.0	768.6	33560.0	364,45
272	21.2	4.8	0.3	2.0	0.2	938.3	21745.0	797.3	34605.0	357,3
291	19.4	4.9	0.3	2.1	0.2	896.9	22195.0	737.0	33480.0	363,3
309	19.0	4.8	0.5	1.9	0.2	870.4	29625.0	772.9	34000.0	345,05
min	16.0	4.3	0.1	1.7	0.1	814.5	16080.0	678.4	33230.0	306,2
max	27.5	5.3	0.5	2.9	0.3	1036.5	35010.0	853.0	34605.0	377,2
mean	19.0	4.7	0.3	2.2	0.2	942.2	25196.5	751.8	33917.6	347,2
SD	3.0	0.3	0.1	0.3	0.0	56.8	5260.4	42.2	396.4	17,3







Appendix B. Figure S2. Ordination plot of PC3 against PC4



Appendix B. Figure S3. Ordination plot of PC4 against PC5