



1 **Glycerol dialkyl glycerol tetraether variations in the northern Chukchi Sea,**

2 **Arctic Ocean, during the Holocene**

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18

19 **Abstract**

20 Glycerol dialkyl glycerol tetraethers (GDGTs) have become a useful tool in

21 paleoclimate research in ocean environments, but their applications in the Arctic are yet to be

22 developed. GDGTs were analyzed in three sediment cores from the northern/northeastern



23 margin of the Chukchi Sea to test the applicability of GDGT proxies for reconstructing sea
24 surface temperature and sea-ice variability in the Holocene. Interpretation was enabled by an
25 earlier investigation of GDGT composition in surface sediments from the study area. Low
26 GDGT concentrations and high BIT and CBT values in core sediments older than ca. 8 ka
27 probably indicate heavy sea-ice conditions in combination with terrestrial inputs during
28 deglaciation and incomplete sea-level rise. Higher concentrations of isoprenoid GDGTs after
29 ca. 8 ka, consistent with an increase in total organic carbon and some other biogenic proxies,
30 are interpreted to represent increased primary production combined with elevated
31 sedimentation rates. These patterns were likely controlled by sea-ice conditions and
32 variations in Pacific water inflow. Geographic heterogeneity in these processes is indicated
33 by differences in GDGTs distribution patterns between cores across the Chukchi margin.
34 TEX_{86} and TEX_{86}^L indices potentially indicative of sea surface temperatures appear to show
35 millennial-scale variability, but the controls on these fluctuations are yet to be understood.

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37 Keywords: GDGT, TEX_{86} , TEX_{86}^L , BIT, MBT', CBT, Holocene, sediment records, Arctic
38 Ocean, Chukchi Sea

39

40 **1. Introduction**

41 The Arctic Ocean currently experiences fast environmental changes due to its high
42 sensitivity to global warming on various time scales (Screen and Simmonds, 2010; Miller et
43 al., 2010). In particular, the Chukchi Sea (Fig. 1) is a region of dramatic changes in sea-ice
44 and ocean current conditions due to its proximity to the North Pacific (e.g., Shimada et al.,
45 2006). As the observational period of these changes covers only the last 2-3 decades



46 (Woodgate et al., 2005 a,b; Shimada et al., 2006), reconstruction of sea-ice cover and ocean
47 circulation on time scales beyond this period is important to comprehend the ongoing
48 processes and future climate. A number of paleo-proxy records has been obtained recently on
49 sediment cores recovering Holocene deposits from the Chukchi region (e.g., Belicka et al.,
50 2002; de Vernal et al., 2005, 2013; McKay et al., 2008; Darby et al., 2009, 2012; Ortiz et al.,
51 2009; Faux et al., 2011; Polyak et al., 2009, 2016). This data suggests that the Holocene
52 paleoceanography in the Chukchi Sea was considerably different from other Arctic margins.
53 For example, some of the proxies indicate that while the Arctic was overall warmer in the
54 early Holocene owing to higher summer insolation (CAPE Project Members, 2001), the
55 Chukchi region may have had expanded sea ice, possibly related to a diminutive inflow of
56 warm Pacific water via the Bering Strait (e.g., de Vernal et al., 2005, 2013; Polyak et al.,
57 2016). There is also evidence that the Pacific inflow intensified and peaked in the middle
58 Holocene (Ortiz et al., 2009; Polyak et al., 2016; Yamamoto et al., 2016). In addition to these
59 long-term changes, higher frequency variabilities have also been identified in records with
60 enhanced resolution (Polyak et al., 2016; Yamamoto et al., 2016).

61 These results pose numerous questions to the responses of the Chukchi Sea current
62 system, sea ice, and biota to climatic changes, which can be addressed by more detailed and
63 multifaceted proxy studies. Biomarker research, including glycerol dialkyl glycerol
64 tetraethers (GDGTs), has a potential to augment paleoclimatic data as an independent proxy
65 approach to paleoproductivity and hydrographic environments. We have analyzed GDGTs in
66 three sediment cores from the northern/northeastern Chukchi Sea to evaluate changes in their
67 abundance and composition during the Holocene (last ~10 ka) in the context of regional
68 hydrographic and sea-ice environments. Some of the basic GDGT data on two of the studied



69 cores have been used by Polyak et al. (2016) to corroborate biomarker-based sea-ice
70 reconstructions, while regional distribution of GDGTs in surficial sediments has been
71 reported in Park et al. (2014). In this paper we provide a comprehensive investigation of the
72 GDGT distribution in the Holocene sediments under study. We also evaluate the applicability
73 of GDGTs for paleoclimatic reconstructions in the Arctic seas by comparing them to other
74 proxies in cores under study and data from other marine sites in the Chukchi-Alaskan region.

75

76 **2. Material and methods**

77 *2.1 GDGT proxies*

78 Glycerol dialkyl glycerol tetraethers (GDGTs) are increasingly used as proxies to trace
79 environmental changes such as contribution of soil organic matter, sea surface temperature
80 (SST), air temperature, and soil pH in the source areas (e.g., Sinninghe Damsté et al., 2000;
81 Schouten et al., 2002; Hopmans et al., 2004; Weijers et al., 2007), but their application to
82 Arctic marine sediments has been limited. Park et al. (2014) discussed the production,
83 advection, and preservation of both isoprenoid and branched GDGTs as key processes
84 determining the distribution of GDGTs and derived indices in surface sediments of the
85 Chukchi Sea and adjacent areas of the Arctic Ocean (Fig. 2). In particular, this study
86 demonstrates that GDGT composition in the Arctic Ocean north of the Chukchi margin
87 (approximately 75°N) is strongly affected by allochthonous soil bacteria, while GDGTs on the
88 Chukchi shelf have a higher marine component, and thus, a better potential for characterizing
89 their sources and related paleoceanographic environments (Park et al., 2014). This knowledge
90 can be now used for interpreting GDGT records in marine sediment cores from the Chukchi
91 margin.



92 In this study, in addition to measured isoprenoid and branched GDGT, we employed
93 TEX₈₆ (TetraEther indeX of tetraethers consisting of 86 carbon atoms), MI (methane index),
94 MBT' and CBT indices (methylation index and cyclization ratio of branched tetraethers), and
95 BIT (Branched and isoprenoid tetraether). TEX₈₆ used to reconstruct sea surface temperature
96 (Schouten et al., 2002; Kim et al., 2010) is based on an initial global core-top dataset
97 (Schouten et al., 2002), whereas, TEX₈₆^L is based on an expanded dataset including polar
98 waters (Kim et al., 2010). MI indicate a degree of anaerobic oxidation by Euryarchaeota
99 (Zhang et al., 2011). High MI (> 0.4), representing high contribution of Euryarchaeota, is
100 associated with bias of TEX₈₆ values. MBT' and CBT have been proposed as proxies of soil
101 pH and mean annual air temperature (Peterse et al., 2012; Weijers et al., 2007), and BIT as a
102 proxy of soil organic matter contribution (Hopmans et al., 2004; Kim et al., 2006). These
103 indices have been shown useful for identifying the sources of branched GDGT in the study
104 region (Park et al., 2014).

105

106 2.2 Samples and age constraints

107 Sediment cores ARA02B 01A-GC (gravity core) and HLY0501-05TC/JPC, -06JPC and
108 -08TC/JPC (trigger/ jumbo piston cores), hereafter referred to as 01A-GC, 05JPC, 06JPC and
109 08JPC, were collected from the northern to northeastern margin of the Chukchi shelf during
110 the 2011 cruise of the RV Araon and 2005 cruise of USCG Healy (Table 1; Fig. 1). Trigger
111 cores are used for a better representation of the uppermost soft sediments that may have been
112 missed by respective piston cores due to overpenetration. Cores 01A-GC and 08JPC are sited
113 on the outer shelf in water depths of 111 and 90 m, which were above the sea level at the time
114 of the last glaciation and inundated during the postglacial transgression since about 15 ka



115 (e.g., Keigwin et al., 2006). In contrast, cores 05JPC and 06JPC were raised from the
116 continental slope at 462 m and 673 m depth, where sediment deposition was not interrupted
117 by sea-level changes. Only a few samples from the bottom part of stratigraphically most
118 extensive 06JPC are used in this study to augment the 05JPC record.

119 Various stratigraphic and sedimentological data on one or several of the cores under
120 study have been reported in a number of papers (Barletta et al., 2008; McKay et al., 2008;
121 Darby et al., 2009, 2012; Polyak et al., 2009, 2016; Lisé-Pronovost et al., 2009; Brachfeld et
122 al., 2009; Faux et al., 2011; Kim et al., 2016). Sediments in cores 08JPC and 01A-GC consist
123 predominantly of homogenous clayey silts indicative of open-marine environments, with a
124 more sandy composition near the core bottom, possibly related to shallow-water erosion and
125 redeposition during the shelf flooding (Darby et al., 2009). In cores 05JPC and 06JPC the
126 homogenous, fine-grained marine unit is underlain by a more complex lithostratigraphy with
127 laminations and coarse ice-rafted debris indicative of glaciomarine environments affected by
128 glacial/deglacial processes (McKay et al., 2008; Lisé-Pronovost et al., 2009; Polyak et al.,
129 2009).

130 In total 110, 47, 2 and 34 samples were collected for the GDGT analysis from cores
131 01A-GC, 05JPC, 06JPC, and 08JPC, respectively. Samples were mostly taken from the
132 Holocene marine sediments at intervals providing a multidecadal- to multicentury-scale
133 resolution. In addition, several samples from the lower part of cores 05JPC and 06JPC span
134 the pre-Holocene sedimentary sequence. Samples were stored in a refrigerator since
135 collection, subsampled and freeze-dried for further processing.

136 Age constraints were provided by seven, six, and ten accelerator mass spectrometry
137 (AMS) ^{14}C ages of mollusc shells from cores 01A-GC (Kim et al., 2016), 05JPC, and 08JPC



138 (Darby et al., 2009), respectively (Supplementary Table S1; Fig. 3). ^{14}C ages were converted
139 to calendar ages using the CALIB7.0 program and marine13 dataset (Reimer et al., 2013).
140 Local reservoir corrections (ΔR) were taken as 500 years for 01A-GC and 08JPC washed by
141 surface waters and 0 years for 05JPC washed by subsurface Atlantic waters (McNeely et al.,
142 2006; Darby et al., 2012).

143 The age model was constructed by linear interpolation between the dating points, which
144 fall within the interval of ca. 1.5–8.6 ka (Fig. 3), as well as the assumed modern age of the
145 core tops. Ages below the dated interval were extrapolated to the bottom of cores 01A-GC
146 and 08JPC and to the bottom of marine unit in stratigraphically longer core 05JPC. Core
147 05JPC was further expanded by the addition of two samples from the nearby core 06JPC.
148 Rough age constraints for older sediments in cores 05JPC and 06JPC were estimated by
149 correlation with cores from the adjacent western Arctic Ocean (Polyak et al., 2009), where
150 samples from core 05JPC span the last deglaciation, and the two samples from core 06JPC
151 possibly represent pre-LGM ($>$ ca. 25 ka) environments. Due to inevitable inaccuracies in the
152 age estimation beyond the dated interval, the bottom of marine sediments in the 01A-GC,
153 05JPC and 08JPC came out with slightly different ages between ca. 8.5–9.5 ka, so we assume
154 that the actual age of this stratigraphic boundary is close to 9 ka. The distribution of linear
155 sedimentation rates shows maximal values in all studied cores around 5–6 ka, with especially
156 high rates in core 08JPC (Fig. 3). The synchronicity of this peak corroborates the validity of
157 the difference in ΔR used for cores from different water masses (01A-GC and 08JPC vs.
158 05JPC).

159 Sedimentation rates estimated from ^{210}Pb measurements in the upper 15 cm in 05TC
160 suggest somewhat younger ages than those derived from available ^{14}C datings (McKay et al.,



161 2008). While comparing these two approaches require more precise chronostratigraphic
162 constraints, in this study we used the ^{14}C -based age model because of the uncertainty with
163 extrapolating ^{210}Pb -based age estimates related to potential variability in sedimentation rates.
164 We note that the difference applies only to the uppermost part of the stratigraphy and does not
165 have a considerable effect on the conclusions of this study.

166

167 *2.3 GDGT analysis*

168 Freeze-dried and homogenized sediments were extracted using accelerated solvent
169 extractor (DIONEX ASE-200) with 11 ml of mixture of dichloromethane:methanol (6:4, v:v)
170 at 100 °C and 1000 psi for 10 minutes ($\times 3$). The extract was concentrated by a rotary
171 evaporation and then separated into four fractions (F1 to F4) depending on polarity of lipid
172 by silica gel column chromatography. F4 fraction including GDGTs, treated according to
173 procedure of Yamamoto and Polyak (2009), was analysed using HPLC/MS (high
174 performance liquid chromatography/mass spectrometry) connected to a Bruker Daltonics
175 micrOTOF-HS time-of-flight MS. GDGTs were separated using an Alltech Prevail Cyano
176 column (2.1 \times 150 mm, 3 μm) at 30 °C in HPLC and identified according to Hopmans et al.
177 (2000) and Schouten et al. (2007). GDGTs investigated for this study are shown in the
178 Appendix. TEX_{86} and $\text{TEX}_{86}^{\text{L}}$ were calculated according to Schouten et al. (2002) and Kim et
179 al. (2010), respectively, and converted to temperature using calibration suggested by Kim et
180 al. (2010; $T(^{\circ}\text{C}) = 81.5 \text{TEX}_{86} - 26.6$; $T(^{\circ}\text{C}) = 67.5 \text{TEX}_{86}^{\text{L}} + 46.9$). The standard deviations
181 (SD) of TEX_{86} and $\text{TEX}_{86}^{\text{L}}$ derived temperatures were 0.8 °C and 1.5 °C, respectively. MI,
182 BIT, CBT indexes were calculated according to Zhang et al. (2011), Hopmans et al. (2004)
183 and Weijers et al. (2007), respectively. The SD of MI, BIT, CBT were averaged 0.002, 0.002,



184 0.009, respectively. MBT' was calculated according to Peterse et al. (2012), and the SD was
185 0.006.

186

187 **3. Results**

188 Isoprenoid GDGTs detected include GDGT-0, GDGT-1, GDGT-2, GDGT-3,
189 crenarchaeol, and crenarchaeol regioisomer (Structures are shown in the Appendix).
190 Crenarchaeol and GDGT-0 are the most abundant isoprenoid GDGTs in the studied samples
191 (Fig. 4). In the averaged fractional abundance of isoprenoid GDGTs, crenarchaeol in cores
192 01A-GC and 08JPC is most abundant during middle and late Holocene, comparable to GDGT
193 values in surface sediment from the shelf edge of the Chukchi Sea (Fig. 4). Fractional
194 abundances of isoprenoid GDGTs show a considerable variability at the transition from early
195 to middle Holocene, especially in core 05JPC.

196 Total concentrations of isoprenoid GDGT have highest values in core 01A-GC, from
197 2.6 to 31.6 $\mu\text{g/g}$, and do not exceed 18.4 $\mu\text{g/g}$ in cores 05JPC and 08JPC (Fig. 5).
198 Concentrations vary between the cores but show similar, stratigraphically consistent
199 downcore patterns, especially for cores 01A-GC and 05JPC. In the late deglacial interval to
200 early Holocene (until ca. 9 ka) concentrations were low in all three cores, then increased
201 markedly to ca. 7–8 ka, and reached a maximum around ca. 5–6 ka in cores 01A-GC and
202 05JPC. In the late Holocene, isoprenoid GDGTs had overall high but variable concentrations,
203 with a distinct maximum around 3 ka in 08JPC. Near the core top in 01A-GC, 05JPC and
204 08JPC concentrations show a decrease. In the deglacial unit studied in core 05JPC,
205 isoprenoid GDGT get relatively more abundant towards the bottom. In the yet older (possibly
206 pre-LGM) sediment recovered in core 06JPC, the concentrations of isoprenoid GDGT were



207 low.

208 Branched GDGTs detected include I, Ib, Ic, II, IIb, IIc, III, IIIb, and IIIc (Structures are
209 shown in the Appendix). Acyclic GDGTs such as I, II and III are the most abundant in the
210 studied sediments (Fig. 4). Whereas acyclic GDGTs are abundant in core 05JPC and 08JPC
211 during the entire Holocene, GDGT-IIb is also abundant in core 01A-GC, especially in the
212 middle and late Holocene, with values comparable to surface sediment from the shelf edge of
213 the Chukchi Sea (Fig. 4). Branched GDGTs show a considerable difference in fractional
214 abundances between middle and early Holocene, especially in core 05JPC, similar to
215 isoprenoid GDGTs (Fig. 4). The total concentrations of branched GDGTs reach 1.3 $\mu\text{g/g}$, 1.1
216 $\mu\text{g/g}$, and 1.9 $\mu\text{g/g}$ in cores 01A-GC, 05JPC, and 08JPC, respectively (Fig. 5). Like
217 isoprenoid GDGTs, the branched GDGTs concentrations show a similar downcore
218 distribution pattern. In all studied cores, concentrations were low until ca. 9 ka, and then
219 increased to around 8 ka. The peak at ca. 5–6 ka is well expressed in cores 01A-GC and
220 05JPC, followed by variable concentrations with another, somewhat lower maximum at 1–2
221 ka. In core 08JPC maximal values were reached around 7 ka and 3 ka. An increase in
222 branched GDGTs is also evident at the bottom of the deglacial unit in core 05JPC, whereas,
223 low concentrations characterize the two older samples from core 06JPC (Fig. 5).

224 The BIT index shows highest levels of >0.5 in the deglacial unit, decreases to very low
225 levels by ca. 7–8 ka, and stays consistently low since then in cores 01A-GC and 05JPC (Fig.
226 5). In core 08JPC, BIT decreases with some fluctuations from >0.3 at 9–10 ka to 0.1 by 3 ka,
227 and then slightly increases towards the core top. In the early deglacial and older sediments in
228 cores 05JPC/06JPC, BIT values are lower than later in deglaciation to early Holocene, but
229 somewhat higher than in the late Holocene (Fig. 5).



230 The ratio of GDGT-0 to crenarchaeol (GDGT-0/cren) and the MI are overall low except
231 for somewhat elevated values in the early Holocene until ca. 8 ka in cores 05JPC and 08JPC
232 (Fig. 5).

233 The distribution of the CBT index is similar to that of the BIT (Fig. 5). Maximal CBT
234 values characterize the early Holocene and decrease to low levels by 7–8 ka in cores 01A-GC
235 and 05JPC. In 08JPC the Holocene CBT is somewhat higher and shows an overall decrease
236 towards ca. 3 ka and a slight increase thereafter. In the early deglacial and older section in
237 cores 05JPC/06JPC, CBT values are relatively elevated, but not as much as in later deglacial
238 to early Holocene sediments.

239 The MBT' index also shows relatively high values in the early Holocene with a peak
240 around 10 ka in core 05JPC and another peak around 8 ka (Fig. 5). In core 08JPC MBT' is
241 variable with peaks at ca. 6, 7 and 8 ka. In core 01A-GC the MBT' shows less variability.

242 TEX_{86} -derived temperatures strongly fluctuate, ranging mostly between 5 and 15 °C
243 with a slight general increase throughout the Holocene in cores 01A-GC and 05JPC (Fig. 5).
244 No such trend occurs in core 08JPC. TEX_{86}^L -derived temperatures are overall lower by up to
245 10 °C than TEX_{86} -derived temperatures and show no trend in their distribution. In core 01A-
246 GC, the amplitude of TEX_{86}^L fluctuations increases noticeably after ca. 6 ka.

247

248 **4. Discussion**

249 *4.1. Changes in production and sources of GDGTs during the Holocene*

250 GDGT distribution in surface sediments from the Chukchi Sea and the adjacent western
251 Arctic Ocean and northern Bering Sea shows abundant isoprenoid GDGTs on the outer shelf
252 and slope of the Chukchi Sea and the upper slope of the Bering Sea (Fig.2; Park et al., 2014).



253 The higher abundances are attributed to a combination of higher production of marine
254 Archaea (Thaumarchaeota) at the shelf edge, redeposition of GDGT-carrying fine sediment
255 particles from the shelf, and better preservation of GDGTs at sites with higher sedimentation
256 rates.

257 The fast increase in isoprenoid GDGTs is observed in all three studied cores at the
258 bottom of marine sedimentary unit, from ca. 9 to 8 ka (Fig. 6). This change is consistent with
259 an increase in total organic carbon and some other biogenic proxies, such as silica, in these
260 and nearby cores (Darby et al., 2001; Lundeen, 2005; McKay et al., 2008; Currie, 2009, and
261 unpublished data for 01A-GC). This correspondence suggests that the increase in isoprenoid
262 GDGT concentrations was driven by increasing bioproduction with the establishment of
263 marine environments in the Chukchi Sea after the end of deglaciation and opening of
264 sufficient inflow through the Bering Strait.

265 Further changes in isoprenoid GDGT concentrations are more complex. The peak
266 values around 5-6 ka in cores 01A-GC and 05JPC correspond to maximal sedimentation rates
267 (Fig. 5) suggesting that preservation may have been a factor. However, no isoprenoid GDGT
268 peak occurs in core 08JPC at this time despite a well expressed sedimentation rate maximum.
269 This discrepancy indicates that isoprenoid GDGT concentrations were primarily controlled
270 by factors other than preservation, like primary production, which could have varied spatially
271 due to differing sea- ice conditions. A comparison with the distribution of sea-ice related
272 biomarker IP₂₅ shows that concentrations of isoprenoid GDGTs in both cores increased after
273 the decline of IP₂₅ values peaking at ca. 5–6 ka in 5JPC and ca. 3 ka in 8JPC (Fig. 5; Polyak
274 et al., 2016). This offset of isoprenoid GDGT peaks relative to IP₂₅ is consistent with the
275 inferred negative effect of sea ice on local GDGT production (Park et al., 2014). Another



276 possibility is that the GDGT peak in cores 01A-GC and 05JPC was related to an increase in
277 the Bering Strait inflow in the middle Holocene (Ortiz et al., 2009; Polyak et al., 2016),
278 which may have had different effects in the northern and eastern parts of the Chukchi Sea
279 washed by different branches of the Bering Strait inflow (Fig. 1). In the pre-Holocene section,
280 a relative increase in isoprenoid GDGTs near the bottom of deglacial sediments in core
281 05JPC (Fig. 5) might represent a post-LGM warming and resultant sea ice retreat and
282 enhanced primary production, such as during the Bølling/Allerød period, but the age control
283 is insufficient to constrain this interval.

284 Branched GDGTs in surface sediments are abundant on the Chukchi shelf and in the
285 Yukon and Mackenzie River estuaries (Fig.2; Park et al., 2014). A concerted abundance of
286 both branched and isoprenoid GDGTs at the Chukchi shelf edge indicates common
287 concentration processes, such as sediment redeposition and enhanced preservation at sites
288 with high sedimentation rates. High cyclization ratios of branched tetraethers (CBT)
289 characterize sediments from the Arctic Ocean north of the Chukchi margin (~75 °N), as well
290 as the Yukon and Mackenzie River estuaries, in contrast to lower CBT in sediments from the
291 Chukchi and Bering seas. This difference indicates two principal sources of branched GDGTs
292 tentatively interpreted as soil bacteria and in situ marine bacteria, respectively.

293 High BIT and CBT vs. MBT' values peaking in the deglacial sediments and extending
294 into lower Holocene until ca. 8 ka, as expressed especially clearly in cores 01A-GC and
295 05JPC (Fig. 6), are similar to these indices in surface sediments of the study region north of
296 75 °N (Park et al., 2014). Fig. 4 also shows similarity in fractional distribution of branched
297 GDGTs between the early Holocene interval in the studied cores and offshore areas of
298 Chukchi Sea. These high BIT and CBT values along with low GDGT concentrations north of



299 75 °N were interpreted as a result of very low marine production and/or severe degradation
300 under multi-year ice, and thus relatively high content of imported terrestrial GDGTs. We infer
301 that similar conditions prevailed in the study area in the early Holocene before ca. 8 ka. This
302 conclusion is consistent with the dinocyst-based proxy record from core 05JPC indicative of
303 high sea-ice concentration in the early Holocene (McKay et al., 2008). Other dinocyst studies
304 from this region also show a generally similar pattern (de Vernal et al., 2005; 2008; 2013;
305 Farmer et al., 2011), but lack resolution or stratigraphic recovery for a comprehensive
306 characterization of the early Holocene. In addition to the effect of high sea-ice concentration
307 in the early Holocene, low Bering Strait inflow and elevated freshwater inputs due to
308 incomplete sea-level rise and deglacial processes (Darby et al., 2001; Lundeen, 2005;
309 Yamamoto et al., in review) could inhibit marine production and enhance import of terrestrial
310 material in the northern Chukchi Sea.

311 BIT and CBT vs. MBT' values in the early deglacial and older sediments in cores
312 05JPC/06JPC show intermediate levels between the deglacial to early Holocene and middle-
313 late Holocene data (Figs. 4–6). This suggests the possibility of either relatively high organic
314 production (low ice concentrations) during those times or redeposition of organic material
315 from stratigraphically older deposits. The latter may be especially applicable to 06JPC
316 samples that show very low isoprenoid GDGT concentrations.

317

318 4.2. Variations in TEX_{86} and TEX_{86}^L

319 Based on GDGT distribution in surface sediments from the study region Park et al.
320 (2014) concluded that TEX_{86} and TEX_{86}^L indices are not applicable for SST reconstructions
321 north of roughly 73 °N. Further south in the Chukchi Sea, these indices show a more



322 reasonable relation to SST, although still off the global core top calibration curve (Kim et al.,
323 2010). Therefore, one must be cautious about translating TEX_{86} and $\text{TEX}_{86}^{\text{L}}$ data into
324 absolute SST values in the study area. Nevertheless, relative downcore changes in TEX_{86} and
325 $\text{TEX}_{86}^{\text{L}}$ may be indicative of SST variability, especially in full-marine environments. GDGT
326 distribution in surficial shallow-water sediments shows that high BIT, GDGT-0/cren, and MI
327 indices relate to high TEX_{86} and $\text{TEX}_{86}^{\text{L}}$ values, indicating their terrestrial bias (Park et al.,
328 2014). However, in full-marine sediments deposited in cores under study after ca. 8-9 ka,
329 these indices are consistently low (Fig. 5), suggesting that variation in TEX_{86} and $\text{TEX}_{86}^{\text{L}}$
330 here is not related to terrestrial contribution or appreciable methanotrophic euryarchaea,
331 which can produce GDGT-1, GDGT-2 and GDGT-3.

332 A millennial- to multcentury-scale variability in TEX_{86} - and $\text{TEX}_{86}^{\text{L}}$ -derived SST is
333 well expressed in the investigated Holocene records, especially in core 01A-GC studied with
334 the highest resolution (Fig. 5). This variability appears to increase in amplitude, along with a
335 slight increase in TEX_{86} - and $\text{TEX}_{86}^{\text{L}}$ -derived SST values, during the Holocene towards the
336 core top. This variation does not resemble the record of chlorite abundance reflecting the
337 strength of Bering Strait inflow (Ortiz et al., 2009) nor the proxy records of sea-ice cover (de
338 Vernal et al., 2013; Polyak et al., 2016). This differing pattern suggests that neither Pacific
339 water advection nor sea-ice cover had a major control on TEX_{86} and $\text{TEX}_{86}^{\text{L}}$ variability in the
340 northern Chukchi Sea. More studies are needed to understand the controls on these indices
341 and their applicability for SST reconstructions in the Arctic.

342

343 5. Conclusions

344 The analysis of GDGTs in three sediment cores from the northern/northeastern Chukchi



345 Sea margin provides insights into GDGTs production and sources in this region of the Arctic
346 during the Holocene. Concentrations of isoprenoid GDGTs reached high values by ca. 8 ka
347 with the establishment of marine conditions after deglaciation and sea-level rise. Low
348 GDGTs concentrations combined with high BIT and CBT values prior to ca. 8 ka may
349 suggest high concentrations of sea-ice in the northern Chukchi Sea, with overall milder sea-
350 ice conditions later in the Holocene. Higher inputs of terrestrial material were also likely
351 during the deglaciation extending into the early Holocene. After ca. 8 ka, GDGTs distribution
352 was variable and probably controlled by a combination of sea-ice conditions and Bering
353 Strait inflow that affected primary production and sediment transport and deposition.
354 Different patterns in GDGTs distribution between cores from the northern and northeastern
355 sites may indicate spatial differences in the pathways of Pacific waters and sea-ice extent.
356 TEX_{86} and TEX_{86}^L indices potentially useful for SST reconstruction show millennial-scale
357 variability, but the controls are not well understood. More investigations using multiple
358 proxies are needed to comprehend sea-ice, temperature, and circulation history in the
359 Chukchi Sea, a critical region for the Arctic climate change.

360

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- 575



576 Table 1. Information of sediment cores used in this study

Cruise name	Core name	Latitude (°N)	Longitude (°W)	Water depth (m)	Core length (cm)
ARA02B	01A-GC	73.63	166.52	119	563
HLY0501	05TC/JPC	72.70	157.45	462	1648
HLY0501	06JPC	72.69	157.03	673	1554
HLY0501	08TC/JPC	71.63	156.84	90	1396

577



578 Figure captions

579 Fig. 1. Index map with location of sediment cores under study and a generalized bathymetry.

580 Arrows indicate major currents in the Chukchi Sea: Siberian Coastal Current (brown color)

581 and three branches of Pacific water flowing through the Bering Strait (orange color). The

582 blue-sky and red dashed lines indicate summer sea-ice margin (15% concentration) for the

583 late 20th century average and the all-time observational 2012 minimum, respectively (data

584 from the National Snow and Ice Data Center).

585

586 Fig. 2. GDGT distribution in surface sediments of the Chukchi Sea and adjacent western

587 Arctic Ocean and northern Bering Sea: concentrations of (A) isoprenoid GDGTs and (B)

588 branched GDGTs, (C) BIT indices, (D) TEX_{86}^L , (E) TEX_{86} , (F) CBT and (G) MBT' indices

589 (Park et al., 2014).

590

591 Fig. 3. Age-depth distribution of calibrated ^{14}C ages in cores under study

592

593 Fig. 4. The average fractional abundance of isoprenoid and branched GDGTs, respectively, in

594 (a) surface sediments from the Chukchi Sea and the adjacent western Arctic Ocean and

595 northern Bering Sea (Park et al., 2014) grouped by geographic environment type, and (b-d)

596 cores in this study. The core data is shown in major stratigraphic intervals: late, middle, and

597 early Holocene, deglacial, and pre-LGM.

598

599 Fig. 5. Downcore plots of linear sedimentation rates (LSR) and distribution of GDGT (this

600 study) and IP_{25} (Polyak et al., 2016). Dashed lines indicate the lower boundary of the full

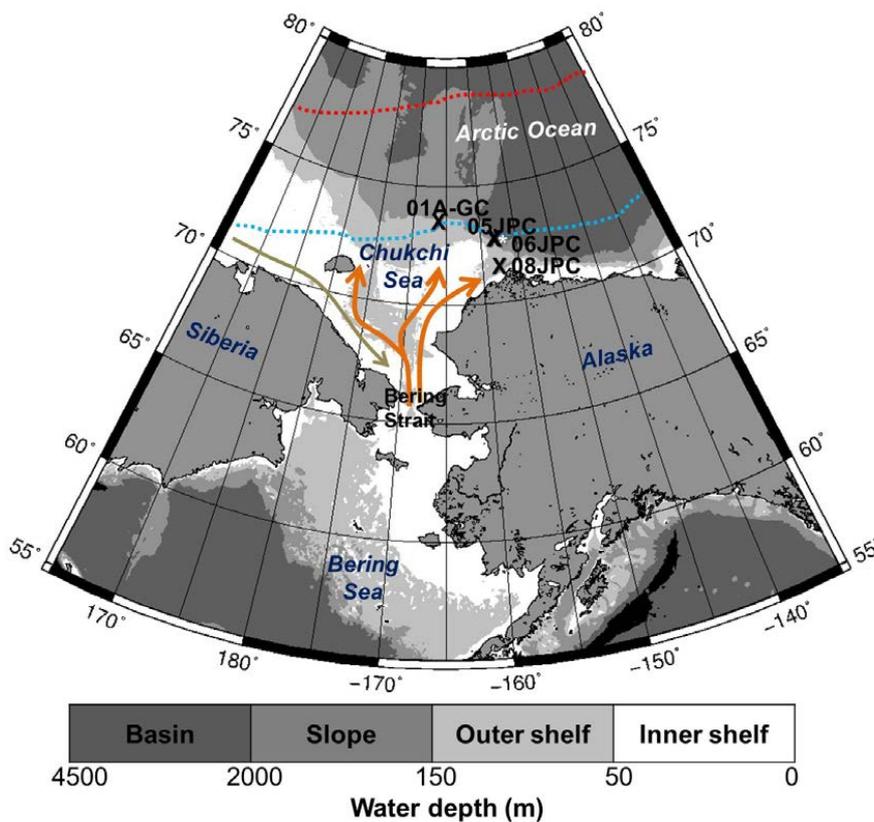


601 marine sedimentary unit around ca. 9 ka, with some variance probably due to imperfect age
602 control. Note a break in the age scale between 05JPC and 06JPC samples.

603

604 Fig. 6. MBT' vs. CBT plots. Values above the dotted line correspond to surface-sediment data
605 from Arctic rivers and from perennially sea-ice covered Arctic Ocean (Park et al., 2014). Data
606 points are color-coded by major paleoenvironmental intervals identified in cores under study
607 (approximate ages with some variance due to imperfect age control).

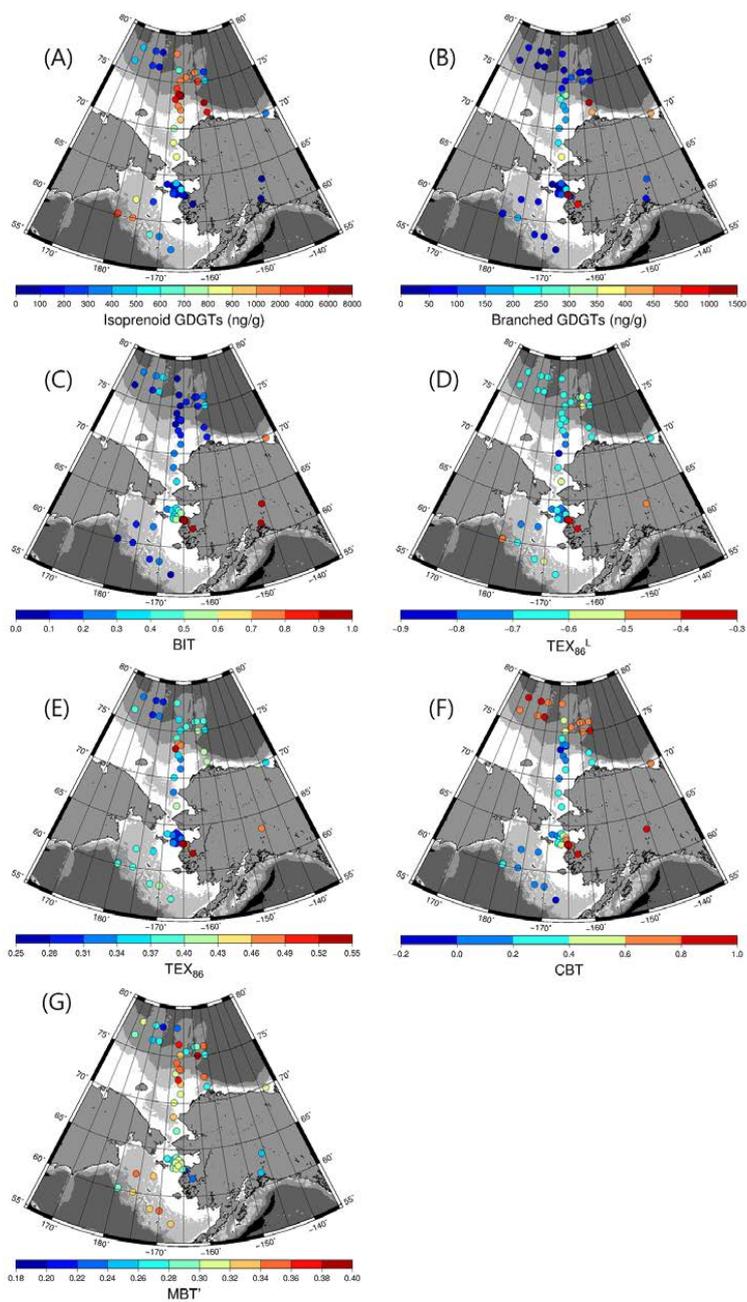
608



609

610 Fig. 1.

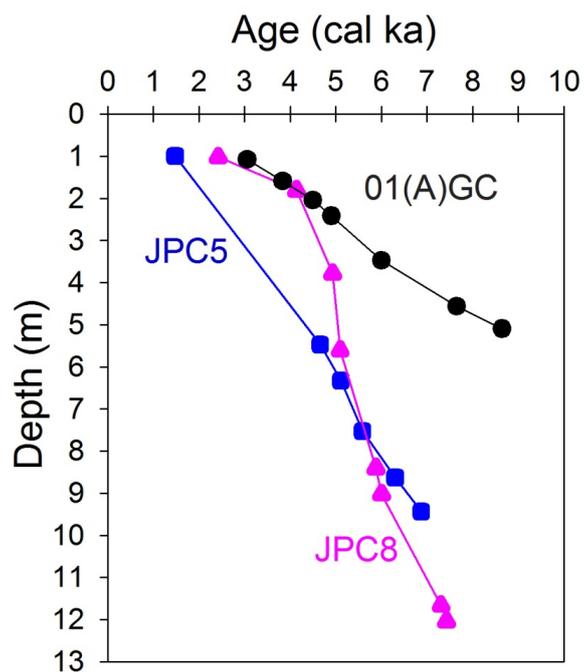
611



612

613 Fig. 2.

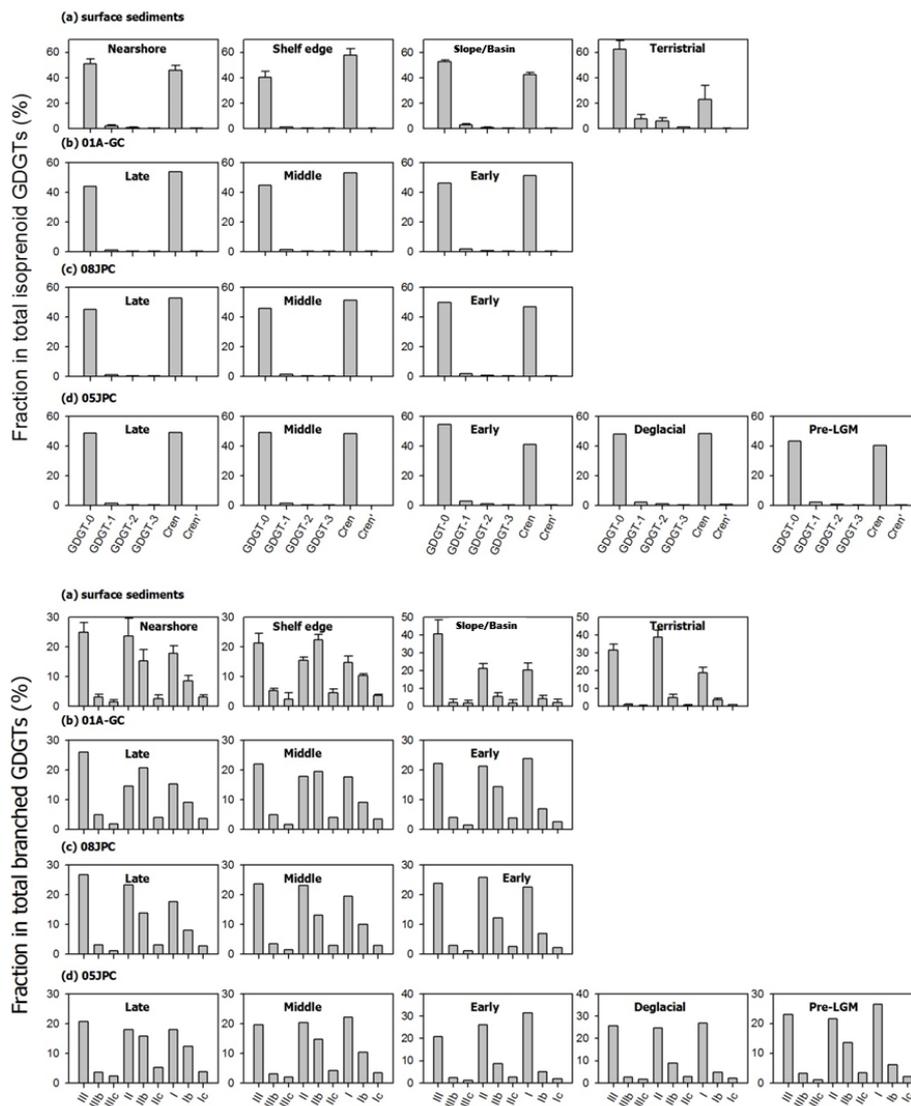
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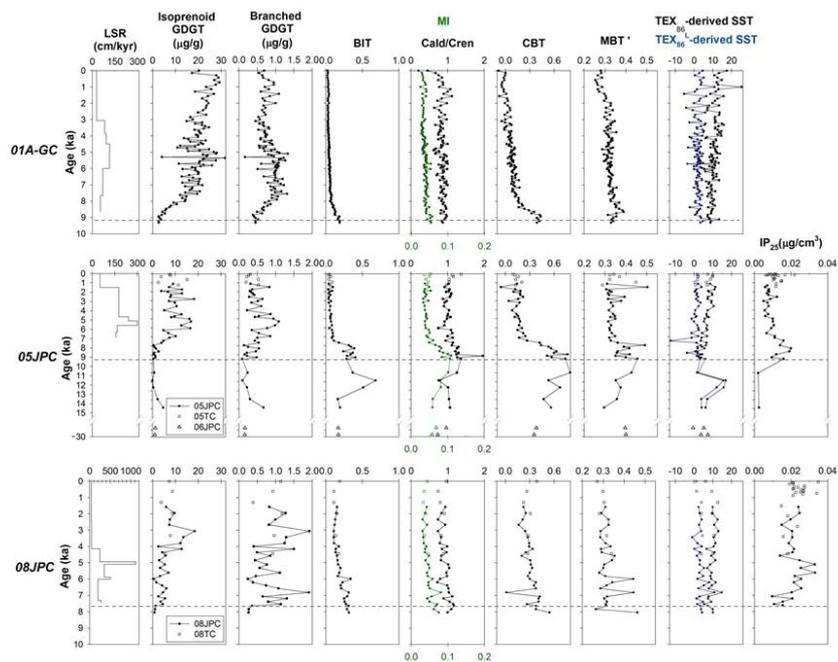
616 Fig. 3.

617



618

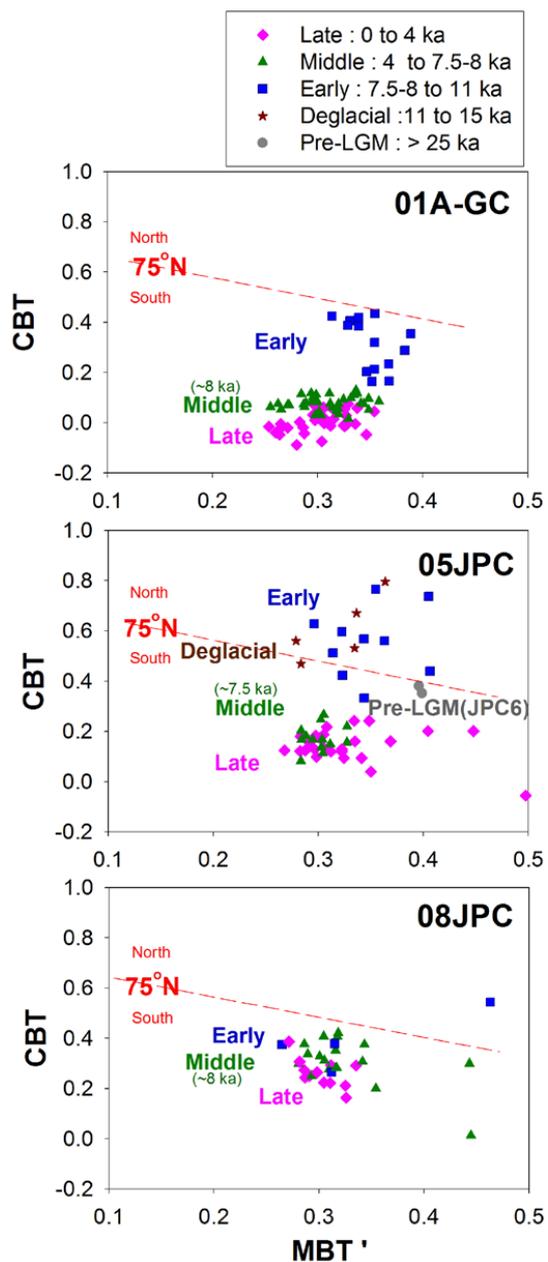
619 Fig. 4.



620

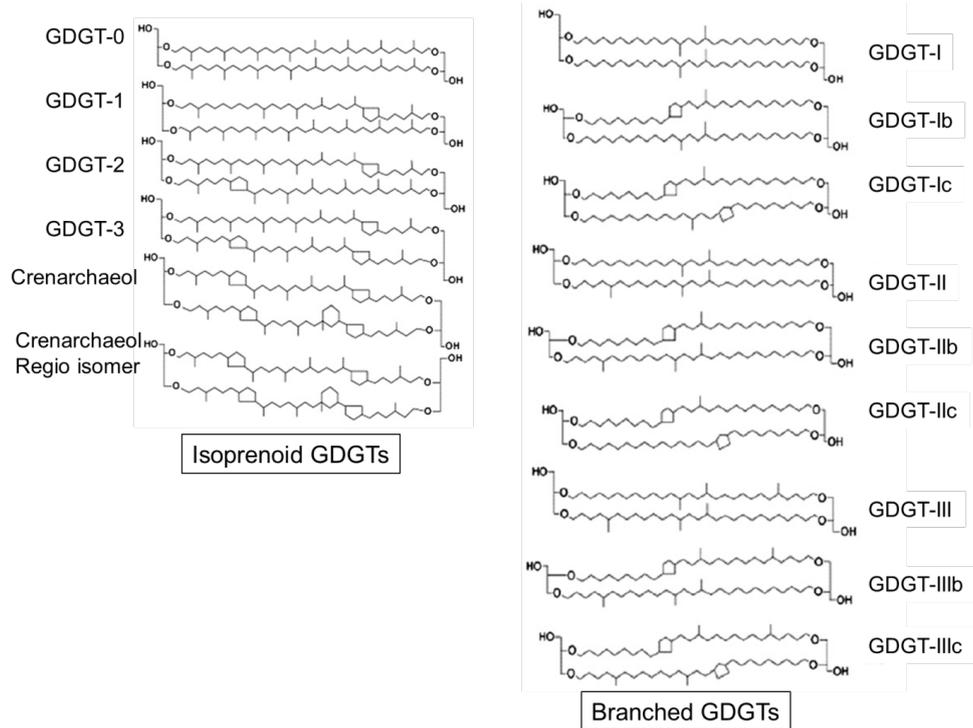
621 Fig. 5.

622



623

624 Fig. 6.



625

626 Appendix

627

628