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1 **Centennial-scale variability of sea-ice cover in the Chukchi Sea since**
2 **AD 1850 based on biomarker reconstruction**

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Abstract

Paleo-climate proxy records documenting sea-ice extent are important sources of information to assess the time of emergence and magnitude of on-going changes in the Arctic Ocean and better predict future climate and environmental evolution in that region. In this study, a suite of geochemical tracers including total organic carbon (TOC), total nitrogen (TN), carbon/nitrogen ratio (C/N), stable isotope composition of organic carbon and nitrogen ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$), and phytoplankton biomarkers (highly branched isoprenoids (HBIs) and sterols) were measured in a marine sediment core to document the sea-ice variability in the Chukchi Sea since the beginning of the Industrial Era. The downcore profile of the sea-ice proxy HBIs suggests a transition from extensive sea ice in the late 19th century to Marginal Ice Zone (MIZ) in AD 1930-1990s and then moderate sea-ice cover since 1990s. Rising of all HBI abundances between AD 1865-1875 indicate a transient retreat of summer ice edge off the shelf followed by a return to near-perennial sea ice till 1920-1930 as revealed by the absence of HBIs and brassicasterol. Sea ice retreat occurred again in AD 1920-1930 and followed by colder decades in 1940s-1960s before a sustained decline since the 1990s. The downcore profile of C/N, $\delta^{13}\text{C}$ of organic matter and sterols indicates a gradual increase of terrigenous inputs accelerating during the most recent decades likely due to enhanced fluvial run-off and sediment transport by sediment-laden sea ice. Concomitantly, increasing $\delta^{15}\text{N}$ values suggest limited nutrient utilization due to enhanced stratification of the surface ocean caused by increased freshening. The role of the Arctic Oscillation (AO), the Pacific Decadal Oscillation (PDO) and Atlantic Multidecadal Oscillation (AMO) are discussed to explore potential drivers of the observed sea-ice changes.

Key words: Sea ice, biomarkers, IP₂₅, HBI III, PIP₂₅ index, Chukchi Sea

1. Introduction

Over the past decades, the Arctic Ocean has experienced dramatic climate and environmental changes including warming and rapid sea-ice retreat (Steele et al., 2008; Onarheim et al., 2018; Screen et al., 2018; Stroeve and Notz, 2018), increased inflow of Pacific Water (PW) (Woodgate, 2018; Woodgate and Peralta-Ferriz, 2021), enhanced freshening of surface waters (Yamamoto et al., 2009; Giles et al., 2012) and increased ocean acidification (Qi et al., 2017). These changes are having a strong impact on primary production (Arrigo et al., 2008; Arrigo and Van Dijken, 2015; Renaut et al., 2018) and marine ecosystems (Grebmeier, 2012; Wassmann et al., 2011), especially in the western Arctic Ocean where warming is more pronounced (Li et al., 2009; Coupel et al., 2012, 2015; Lee et al., 2019; Zhuang et al., 2020).

Sea ice is an essential component of Arctic climate and a key player in Arctic amplification (Thomas, 2017). The loss of sea ice affects heat and moisture exchanges between the ocean and the atmosphere and modifies deep-water formation through brine formation and ice melting (Petrich and Eicken, 2017) thereby altering the Arctic Ocean circulation (Aagaard and Carmack, 1989). Since the mid-20th century, thinning of sea ice combined with the lengthening of the melt season (Markus et al., 2009; Stroeve et al., 2014) and the loss of perennial multiyear ice have resulted in larger ice-free areas in summer (Holland et al., 2006), and in some regions, earlier melting (Wang and Overland, 2009, 2012). The western Arctic Ocean has undergone some of the most rapid and persistent decay of sea ice (Cavaliere and Parkinson, 2012; Serreze et al., 2016; Simmonds and Li., 2021). Climate projections predict a reduction of sea-ice cover duration of 20-36 days before 2050 in the southern Chukchi Sea, and up to 60 days in the East Siberian, Chukchi, and Beaufort Seas (Wang et al., 2018).

Sea-ice reconstructions provide a retrospective time frame to help decipher processes driving the long-term decline as well as decadal to centennial-scale variability of sea ice. Several biogenic and sedimentary proxies have been developed to obtain information on past sea ice conditions (Polyak et al., 2010; Kolling et al., 2020; Zhang and Xiao, 2021). Among them the molecular proxy, IP₂₅ (Ice Proxy with 25 carbon atoms), a mono-unsaturated highly-branched isoprenoid (HBI) produced by few sea-ice diatoms has proven to be a valuable tool to assess seasonal sea ice cover from marine sediments (Belt et al., 2007; Sicre et al., 2013; Brown et al., 2014). Since the first reconstruction of Massé et al. (2008) in the North Atlantic, numerous studies have used

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4 101 this biomarker in (sub)-polar oceanic regions (mostly of the northern hemisphere), often
5 102 in combination with phytoplankton biomarkers (e.g., brassicasterol or dinosterol), in
6 103 the so called 'PIP₂₅ (Phytoplankton-IP₂₅) index' to produce semi-quantitatively
7 104 seasonal sea-ice estimates (see reviews of Belt and Müller, 2013; Belt, 2018). More
8 105 recently, a tri-unsaturated HBI (HBI III) has been proposed as an alternative to
9 106 represent pelagic phytoplankton in order to improve the PIP₂₅ index (Bai et al., 2019;
10 107 Belt et al., 2015; Koch et al., 2020; Smik et al., 2016; Su et al., 2022) while Belt (2018)
11 108 hypothesized that HBI II, also associated with sympagic algae, might provide an even
12 109 better seasonal sea ice proxy than IP₂₅.

13 110 Historical data and high-resolution reconstructions of Arctic seasonal sea ice are still
14 111 scarce (Kinnard et al., 2011; Walsh et al., 2016) and only a few paleo-records exist in
15 112 the western Arctic Ocean (Pieńkowski et al., 2017; Stein et al., 2017; Astakhov et al.,
16 113 2019, 2020; Kim et al., 2019; Koch et al., 2020). In the present study, we report on the
17 114 downcore profiles of HBIs including IP₂₅, HBI II and HBI III as well of phytosterols,
18 115 namely brassicasterol (24-methyl-5 α -cholest-5,22E-dien-3 β -ol) and dinosterol (4 α , 23,
19 116 24-trimethyl-5 α -cholest-22E-en-3 β -ol) from a core retrieved in the northern Chukchi
20 117 Sea. These biomarkers were analyzed to derive quantitative information on seasonal
21 118 sea-ice evolution since AD 1850 and to explore causes of observed changes. Terrestrial
22 119 biomarkers were also investigated as a complementary environmental information
23 120 source of a changing Arctic Ocean.

21 121 **2. Regional setting and material**

22 122 The Chukchi Sea is one of the largest marginal seas in the world (Jakobsson, 2002).
23 123 This region is strongly influenced by the northward PW inflow driven by salinity
24 124 gradients between the Pacific and the Arctic Ocean (Coachman et al., 1975) (Fig. 1). In
25 125 the Bering Strait, the PW splits into three main water masses i.e., the warmer, fresher
26 126 and nutrient-depleted seasonal Alaska Coastal Water to the East (ACW; Hunt et al.,
27 127 2013), the saline and nutrient-rich Anadyr Water (AW) to the West, and the Bering
28 128 Shelf Water (BSW) in the central channel with moderate nutrient and salinity values
29 129 (Weingartner et al., 2005). The surface hydrology of the region is also influenced by
30 130 the Beaufort Gyre (BG) clockwise circulation driven by the Beaufort High winds
31 131 (Proshutinsky and Johnson, 1997). In the past two decades, strong and more effective
32 132 spin-up of the BG circulation has resulted in enhanced freshening in the western Arctic
33 133 Ocean (Giles et al., 2012) with consequences on phytoplankton productivity and Arctic

ecosystem (Coupel et al., 2015; Ren et al., 2020). With the steepened decline of Arctic sea ice over the last four decades, the extent and thickness of sea ice in the Chukchi Sea has decreased more than any other marginal seas of the Arctic Ocean (Serreze and Stroeve, 2015; Onarheim et al., 2018). According to satellite observations, sea ice today is present from November to July and retreats during summer to reach a minimum over the northern Chukchi Sea in September (Fig. 1, <https://nsidc.org>, from Cavalieri et al., 1996).

The sediment core ARC4-C07 (72.54°N, 165.33°W, 51 m water depth) used in this study was retrieved from the shelf of the Chukchi Sea using a multi-corer during the 4th Chinese National Arctic Research Expedition (CHINARE) on board the R/V Xue Long in summer 2010 (Fig. 1; Table S1). The ARC4-C07 core (hereafter named C07) is located in an area where sea-ice cover is high in spring but close to the sea-ice edge in summer (Fig. 2). The 20-cm long sediment core was subsampled on board at 1 cm sampling interval from 0 to 10 cm and every 2 cm below 10 cm depth, and stored at 4°C until further treatment (See supplementary material for methods on the core chronology and biomarker analysis).

3. Results

3.1 Bulk parameter downcore profile

Fig. 3 shows the downcore profile of TOC (%), biogenic silica (%), TN (%), C/N molar ratio, $\delta^{13}\text{C}$ (‰) and $\delta^{15}\text{N}$ (‰) in the C07 core since AD 1850. TOC values range from 1.46 to 2.93% at the core top, with a mean value of 1.98%, and show a gradual increase towards present except for lower values from the late 1970s and early 1980s (Fig. 3a, Table S2). Biogenic silica shows broadly similar trends as TOC except for a sharp decay in the uppermost layer after a maximum of 17.3% at AD 1990 (Fig. 3b). TN content increases in a narrow range (0.15 to 0.34%) with lower values in the 1970s and at the end of the 19th century, similar to TOC. The C/N ratio show more variability between 7.8 and 11.5 with a mean value of 9.6. The $\delta^{13}\text{C}$ values of organic matter span from -22.3‰ to -23.6‰ at the core-top with a mean value of -22.7‰ . The $\delta^{15}\text{N}$ values vary between 7.7‰ and 8.1‰ at the core-top with little changes downcore (Fig. 3c-f).

3.2 Biomarkers downcore profile

Generally, all biomarker concentrations show a broad increase towards present (Fig. 4, Table S2). IP₂₅ is generally low until AD 1930, except for a brief increase around AD

166 1870 (1.4 $\mu\text{g/g}$ TOC) coincident with decreasing TOC and biogenic silica values,
167 followed by a gradual increase accelerating in the 1960s to reach highest values in the
168 1990s (4.2 $\mu\text{g/g}$ TOC) (Fig. 4a). The downcore profile and range of concentrations of
169 HBI II and IP₂₅ are very similar, while HBI III is an order of magnitude lower, ranging
170 from 0.01 to 0.47 $\mu\text{g/g}$ TOC (Fig. 4b, c). Brassicasterol abundances are low until the
171 abrupt increase in the 1990s to a maximum of 27.9 $\mu\text{g/g}$ TOC (Fig. 4d). The dinosterol
172 profile is quite different and shows strong decadal-scale variability that share
173 resemblance with terrigenous biomarkers (campesterol and β -sitosterol) (Fig. 4e, f).

174 4. Discussion

175 4.1 Organic matter sources and trends

176 The high TOC content of the core top (2.93%) falls in the upper range of TOC values
177 reported in surface sediments across the Chukchi Sea shelf (1.25~2.56%) by Goñi et al.
178 (2013) (Table S2). TN values range from 0.15 to 0.34% (Table S2) and show significant
179 positive correlation with TOC ($r^2=0.68$, $p<0.01$, Fig. 5) suggesting similar sources and
180 fate of C and N. The C/N ratio has commonly been used to distinguish between algal
181 and terrigenous organic matter (OM) (Hedges et al., 1986). Fresh marine algae typically
182 exhibit C/N ratios around 4-6 whereas in vascular plants these values are above 20
183 (Meyers, 1994). C/N ratios > 10 typically indicate significant terrestrial influence
184 (Grebmeier et al., 1989). Downcore C/N values at C07 vary between 7.8 and 11.5
185 (mean value of 9.5, Table S2) indicating notable and increasing contribution of land-
186 derived inputs since the late 1970s likely due to enhanced river run-off resulting
187 thawing permafrost and ground ice melting induced by Arctic warming. The SCC
188 flowing eastward towards the Chukchi shelf where it is diverted offshore by PW could
189 occasionally contribute to the transport of terrestrial material to the core site (green
190 arrows in Fig. 1). In addition, the central current BSW (blue arrow in Fig.1) is likely to
191 carry fine sediment particles from the Bering Strait and shallow southern Chukchi shelf
192 to C07 site (Weingartner et al., 2005; Darby et al., 2009; Yamamoto et al., 2017). A
193 substantial amount of terrestrial organic matter to marine sediments may have be
194 delivered via the melting of sea ice (Eicken et al., 2005). Lalande et al. (2007) also
195 underlined commonly found high concentrations of lithogenic material in the lower
196 layers of sea ice implying substantial contribution to vertical fluxes.

197 The $\delta^{13}\text{C}$ provides complementary information on organic carbon sources. Previous
198 evaluation of isotopic end-members in the northern Bering and Chukchi Seas indicates
199 distinctly $\delta^{13}\text{C}$ values for marine phytoplankton (-21.2‰) and terrigenous C_3 plant
200 material (-27.0‰) (Naidu et al., 2000). However, it is important to note that the $\delta^{13}\text{C}$
201 values of OM from sea ice algae can vary between -15 to -18‰ (Schubert and Calvert,
202 2001). Belt et al. (2008) further reported that the mean $\delta^{13}\text{C}$ value for IP_{25} (-19.3‰) lies
203 at the heavily ^{13}C -enriched marine end-member for Arctic OM. Therefore, $\delta^{13}\text{C}$
204 downcore values of -22.3‰ to -23.6‰ are likely indicative of marine sources with
205 some contribution of isotopically lighter terrestrial organic material. More depleted
206 $\delta^{13}\text{C}$ values towards present are also indicative of enhanced inputs from the land over
207 the last decades as also inferred from C/N ratios (Fig. 3d) and higher amounts of
208 campesterol and β -sitosterol, in particular since the 1960s (Fig. 4f).

209 The downcore profile of $\delta^{15}\text{N}$ values, spanning from 7.7 to 8.1‰, indicate a long-
210 term increase. The $\delta^{15}\text{N}$ values of OM produced by plankton range between 7‰ and
211 10‰, whereas in C_3 higher plants they are comprised between 0‰ and 2‰ (Meyers,
212 1997). The $\delta^{15}\text{N}$ trend may feature enhanced nutrient utilization by phytoplankton due
213 limited nutrient availability under stratified conditions triggered by sea ice melting as
214 postulated by Lee et al. (2012) based on N and C uptake rates. Increasing riverine
215 freshwater inputs also contribute to ocean stratification and freshening of the Arctic
216 Ocean (Li et al., 2009). A recent modeling study has shown that riverine water
217 discharge would also impact significantly on the heat budget of the Arctic Ocean and
218 consequently contribute to the sea-ice decline (Park et al., 2020).

219 **4.2 Downcore profiles of biomarkers**

220 As can be seen from Figure 4, sympagic HBIs (IP_{25} and HBI II) and pelagic
221 biomarkers (HBI III, brassicasterol and dinosterol) display similar trends but at
222 different concentration levels. The presence of IP_{25} and pelagic biomarkers throughout
223 the core indicates seasonal sea-ice conditions at C07 since AD 1850 (Bai et al., 2019;
224 Hu et al., 2020). All HBIs occur at extremely low levels in the older part of the record
225 pointing out extensive sea-ice cover, except for a short interval of higher values
226 between AD 1865 and AD 1875 (Fig. 4a-c). These results are consistent with historical
227 sea ice data from whaling ship logbooks throughout the northern Bering Sea reporting
228 more severe sea-ice conditions in the 1850s to 1860s, comparable to the 1970s taken as
229 a reference for recent cold conditions (Mahoney et al., 2011). Indeed, while winter sea-

230 ice extent was similar during both periods, summer sea-ice extended further South in
231 the Bering Strait in the 1850s to 1860s due to thicker sea ice and slower melting
232 (Mahoney et al., 2011). The logbooks also describe a period of sea-ice retreat to the
233 northern Bering Strait, which corresponds to sympagic HBIs and HBI III peaks found
234 at AD 1865-1875 in our core (Mahoney et al., 2011; Fig. 4). Subsequent decades of the
235 early 1900s are characterized by lower sympagic HBIs caused by a temporary return to
236 colder conditions until a first stepwise increase at AD 1930 and second one in the 1990s
237 (Fig. 4a-b). Stable HBI values in the 1940-60s are consistent with temporarily cold
238 conditions reported by Mahoney et al. (2011).

239 The IP_{25} profile of the neighboring core ARA01B-03MUC (73.52°N, 168.94°W,
240 72.5m water depth; Fig. 1) located northwest of C07 shows similar features (Kim et al.,
241 2019) (Fig. 6a). However, HBI III and brassicasterol decrease earlier in the upper
242 ARA01B-03MUC, while IP_{25} declines synchronously in both cores. High brassicasterol
243 together with high IP_{25} in ARA01B-03MUC since AD 1950 are indicative of sea-ice
244 edge conditions. This is further supported by the co-eval lower and decreasing
245 abundance of HBI III attesting the retreat of sea ice (0.15 to 0.07 $\mu\text{g g}^{-1}$ TOC for
246 ARA01B-03MUC versus 0.2 to 0.5 $\mu\text{g g}^{-1}$ TOC for C07) (Fig. 6b).

247 The downcore profile of dinosterol is different from that of sympagic HBIs and
248 brassicasterol (Fig. 4d-e). Low brassicasterol and HBI III from the late 19th century
249 until AD 1920-1930 are coherent with extensive sea ice in the Chukchi Sea (Fig. 4).
250 However, high amounts of dinosterol (i.e., >10 $\mu\text{g/g}$ TOC) are surprising and could
251 reflect other source organisms such as sea-ice microalgae as reported by Nichols et al.
252 (1990) in sea ice samples from Antarctica. Note that the downcore profile of
253 brassicasterol and dinosterol in ARA01B-03MUC show similar differences (Kim et al.,
254 2019). Another common feature in C07 and ARA01B-03MUC is the recent decline of
255 IP_{25} and brassicasterol, supporting accelerating summer sea ice retreat in the Chukchi
256 Sea driven by climate change in recent decades (Fig. 6a, c).

257 Increased terrigenous sterols (campesterol and β -sitosterol) are consistent with C/N
258 and $\delta^{13}\text{C}$ data and further evidence high and increasing river run-off (Fig. 4f). With the
259 decline of sea ice, airborne continentally-derived material from adjacent continents
260 could also contribute more to Arctic sediments. Indeed, in addition to fluvial deposits,
261 terrestrial constituents transported over long distances from their emission sources as
262 aerosols accumulate in sea ice and get released during the melting season. This is

263 evidenced by terrigenous *n*-alkanes at station DM (Fig. 1) showing high export fluxes
264 as sea ice breaks-up in summer (Bai et al, 2019). Ren et al. (2020) also reported similar
265 results with high terrigenous campesterol and β -sitosterol fluxes in summer contrasting
266 with no detectable export in winter.

267 **4.3 HBIs and phytosterols as proxies of seasonal sea ice**

268 Semi-quantitative estimates of sea ice can be calculated from the PIP_{25} index by
269 combining sympagic IP_{25} with pelagic phytoplankton biomarkers using the following
270 expression:

$$271 \quad PIP_{25} = \frac{[IP_{25}]}{[IP_{25}] + [\text{phytoplankton biomarker}] * c}$$

$$272 \quad \text{with } c = \frac{\text{mean } IP_{25} \text{ concentration}}{\text{mean phytoplankton biomarker concentration}}$$

273 where *c* is a balance factor accounting for the different magnitude concentrations
274 between IP_{25} and phytoplankton biomarkers, such as brassicasterol and dinosterol that
275 were used as the phytoplankton end-member to derive P_BIP_{25} , P_DIP_{25} . More recently,
276 HBI III has also been proposed as an alternative pelagic phytoplankton end-member to
277 calculate $P_{III}IP_{25}$ index.

278 Indeed, surface sediments from the Barents and Chukchi seas have shown elevated
279 abundances of HBI III in the vicinity of the sea-ice edge or in the MIZ (Belt et al., 2015;
280 Smik et al., 2016; Ribeiro et al., 2017; Belt, 2018; Bai et al., 2019). Yet, different
281 relationships of this compound to sea ice have been postulated. For example, maximum
282 HBI III in surface sediments from the western Barents Sea has been associated with
283 winter sea-ice extent and/or the MIZ (Belt et al., 2015) while Ribeiro et al. (2017)
284 correlated HBI III in East Greenland fjord sediments to mid-July MIZ conditions.
285 Uncertainties still remain as to the HBI III producers and habitat mainly because of the
286 lack of modern ocean observations. Knowledge on HBI III has been essentially inferred
287 from correlations between concentrations in surface sediments and SIC at different
288 seasons. The only sediment trap data on HBI III in the Arctic Ocean are from Bai et al.
289 (2019) at station DM (Fig. 1) that shows high fluxes at moderate sea-ice cover (< 50%)
290 and their decrease towards ice-free conditions that are consistent with production by
291 sea-ice edge thriving diatoms. Furthermore, cross-plots of HBI III against IP_{25}
292 downcore concentrations exhibit a stronger positive relationship than brassicasterol
293 against IP_{25} (Fig. S2) in agreement with HBI III production being closely associated

294 with the sea-ice edge. This interpretation is also supported by co-eval higher fluxes of
295 the IP₂₅ and HBI III from end of July to early August at the station DM (Bai et al., 2019).
296 Synchronous changes of IP₂₅ and HBI III have been further interpreted by Belt (2018)
297 as reflecting rapidly changing sea ice (advancing and retreating) such as for example
298 the two-step concomitant rise of HBI III and IP₂₅ in the 1970s and early 1990s in our
299 core (Fig. 6a, b; Fig. 7; Smik et al., 2016; Belt, 2018; Bai et al., 2019).

300 HBI III, as well as brassicasterol and dinosterol were thus used to calculate PIP₂₅
301 indexes and derive semi-quantitatively sea ice changes since AD 1850. On average,
302 concentrations of brassicasterol were about five times higher, dinosterol about
303 twelfold higher and HBI III about twelve times lower than IP₂₅, resulting in balance
304 factor *c* of 0.20, 0.08 and 11.74, respectively. Downcore P_BIP₂₅ values vary between
305 0.19 and 0.79, which according to Müller et al. (2011), correspond to low sea ice to
306 MIZ conditions, with only one value above the permanent sea-ice cover threshold (i.e.
307 P_BIP₂₅ > 0.75) (Fig. 6d). The sharp drop in the 1920s -1930s suggests a transient period
308 of sea-ice retreat (Fig. 7a). Since AD 1930, P_BIP₂₅ values increase from 0.54 to 0.74,
309 indicating proximal MIZ conditions except for the last decade of accelerated decline
310 (Fig. 6d, Table S2). Indeed, P_BIP₂₅ shows a decreasing trend towards less/variable sea-
311 ice cover since the late 1990s as expected from the recent unprecedented decline of sea
312 ice in the northern Chukchi Sea (Walsh et al., 2016). P_{III}IP₂₅ values show different
313 trends and variability that share more resemblance with the sea ice extent in the Chukchi
314 Sea of Walsh et al. (2016) with relatively high and stable values from the 1880s to the
315 1920s followed by a continuing decrease reflecting the progressive and long-term
316 retreat of seasonal sea ice extent (Fig. 6d, Table S2). Yet, the drastic reduction of the
317 last decades is not reproduced by the P_{III}IP₂₅ index. This result can be explained by HBI
318 III production tending to zero as surface waters become ice-free during summer as
319 opposed to brassicasterol (Bai et al., 2019). The P_DIP₂₅ values show similar features as
320 P_BIP₂₅, despite discrepancies between dinosterol and brassicasterol profiles, except
321 since the 1980s when P_DIP₂₅ indicates increased sea ice contrary to observations (Walsh
322 et al., 2016) (Fig. 4d, e and 6d). Diverging spatial distribution of these two sterols has
323 also been underlined by Su et al. (2022) in the Eastern Siberian Sea. It has been
324 speculated that dinosterol in the Arctic Ocean might be produced by other organisms
325 than dinoflagellates (Nichols et al., 1990). Due to remaining source uncertainties of this
326 sterol in the Arctic Ocean and inconsistent trends at moderate to low sea ice, P_DIP₂₅,
327 was not further considered for sea-ice reconstruction.

328 Finally, the tri-unsaturated HBI ratio (HBI TR₂₅) was also calculated using the
329 following equation of Belt et al. (2019) (HBI TR₂₅ = [Z-triene] / ([Z-triene] + [E-triene]))
330 to explore this new biomarker as a potential proxy of spring phytoplankton bloom in
331 the Arctic and sub-Arctic. The downcore values of HBI TR₂₅ are all below the spring
332 phytoplankton bloom threshold of 0.62 from which we can conclude that primary
333 production did not reach high levels as suggested by the $\delta^{15}\text{N}$ values pointing out
334 limited nutrient availability of surface waters (Fig. 6e).

335 **4.4. Centennial-scale variability of sea-ice and potential key physical drivers**

336 **SIC reconstruction.** In order to translate the downcore profiles of P_{III}IP₂₅ and P_BIP₂₅
337 into SIC, we performed sensitivity tests using the surface sediment dataset from the
338 Chukchi Sea of Bai et al. (2019) and the monthly SIC values from satellite data for
339 June, July, August and September as well as for the summer months (July-September,
340 JAS); SuSIC) (Fig. S4 and S5). P_{III}IP₂₅ in surface sediments shows a stronger
341 correlation with SuSIC ($r^2=0.47$, $p<0.01$) than P_BIP₂₅ ($r^2=0.09$, $p<0.01$) (Fig. S3; Bai et
342 al., 2019). Furthermore, downcore P_BIP₂₅ derived SuSIC yield negative values at low
343 sea ice and values exceeding 100% at high sea ice (Fig. S6). The SuSIC obtained from
344 P_{III}IP₂₅ leads to generally lower estimates and negative values over the most recent
345 decades. Reconstructions obtained from P_{III}IP₂₅ and P_BIP₂₅ for June, July, August and
346 September (Fig. S4 and S5) show similar trends as for SuSIC but differ in absolute
347 values (Fig. S6). As found for SuSIC, correlations between monthly SIC and P_{III}IP₂₅
348 were stronger than with P_BIP₂₅. June seems to provide the best downcore SIC estimates
349 and range of variation, and P_{III}IP₂₅ to perform better than P_BIP₂₅ (Fig. S6). Yet the steep
350 decline of the last decades (Fig. 7g) is not reproduced by our reconstruction possibly
351 because of smoothing effect due to the temporal resolution (on average of 10 years) or
352 low HBI III production as highlighted earlier. In contrast, P_BIP₂₅ still overestimate SIC
353 at high sea ice and underestimate SIC at low SIC (Fig. S6).

354 **Key physical drivers.** Important parameters affecting Arctic sea ice include winds
355 and ice motion, cloud cover, ice albedo feedback and ocean heat advection (Perovich,
356 2011). Modes of climate variability such as the Arctic Oscillation (AO, Thompson and
357 Wallace, 1998), the Pacific Decadal Oscillation (PDO, Mantua et al., 1997), and the
358 Atlantic Multidecadal Oscillation (AMO, Enfield, 2001) can have an impact on sea ice
359 variability on decadal time scales (Frey et al., 2015; Mahajan et al., 2011). In order to

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3 360 evaluate the role of these factors in the northern Chukchi Sea, our data were compared
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5 361 to AO, PDO, and AMO indices as well as to the Arctic surface air temperature (Fig. 7).

6 362 **Links with the AO.** The AO is a major atmospheric circulation feature of the mid-
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8 363 to high latitudes of the Northern Hemisphere. Several studies have recognized the
9
10 364 complex relationship between Arctic sea ice and AO or its expression in the North
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12 365 Atlantic, the North Atlantic Oscillation (NAO) (Rigor et al., 2002; Danielson et al.,
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14 366 2011; Stroeve et al., 2011; Yang et al., 2016; Luo et al., 2017). In the Arctic, the AO is
15
16 367 known to affect sea ice motion through surface winds thereby altering sea ice thickness
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18 368 and concentration. A positive and significant correlation was demonstrated between
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20 369 summer AO and sea ice in the Chukchi Sea (see Fig. 13d from Frey et al. 2015). Indeed,
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22 370 during summer season, negative AO promotes southeasterly winds bringing warm air
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24 371 over the Chukchi Sea decreasing the sea ice cover, a mechanism that could account for
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26 372 the long-term AO weakening and associated sea-ice decline from AD 1920-1930s to
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28 373 the 1970s (Fig. 7f, g). On the other hand, during positive AO the transport and piling
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30 374 up of old sea ice pushed by northeasterly winds result in extensive summer sea ice in
31
32 375 the Chukchi and Beaufort seas such as found in the late 1800s – early 1900s (Frey et
33
34 376 al., 2015). However, in the recent decade of positive AO, warmer surface ocean would
35
36 377 have favored sea ice melting preventing its accumulation (Wang et al, 2009; Stroeve et
37
38 378 al., 2011), leading instead to sea ice decline at unprecedented rate as illustrated by the
39
40 379 ice-free period reconstruction from the nearby 14R07 of Astakhov et al. (2019) (Fig.
41
42 380 7e).

43 381 **Links with the PDO.** The PDO is another factor of sea-ice variability at decadal
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45 382 timescales. The PDO is defined as the leading empirical orthogonal function of monthly
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47 383 sea surface temperature in the North Pacific, North of 20°N (Mantua et al., 1997).
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49 384 Instrumental data show that the PDO was predominantly positive for the 1922-1944
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51 385 period, negative between 1945 and 1976, and positive again between 1977 and 1990
52
53 386 (Rodionov et al., 2007; Wendler et al., 2014; Fig. 7c). Negative PDO in the early 20th
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55 387 century (AD 1910-1920) corresponds to high sea-ice cover while the subsequent
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57 388 positive PDO years (AD 1922-1944) are associated with decreasing sea ice in our
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59 389 record. These results are in accordance with the negative correlation between summer
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390 PDO and sea ice in the Chukchi Sea although not statistically significant (see Fig.13c
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392 in Frey et al., 2015). However, negative PDO years since 1940 seem to roughly coincide
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20th century. Furthermore, negative PDO phases coincide with colder years in the

394 Arctic air temperature record except for the last warmer decades possibly reflecting
395 overprinting of the global warming trend as suggested for AO (Fig. 7b, Connolly et al.,
396 2017).

397 **Links with the AMO and AMOC.** The AMO, which is thought to be mainly driven
398 by the Atlantic Meridional Overturning Circulation (AMOC), has been investigated as
399 an additional possible source of natural variability of sea ice. In fact, the AMO also has
400 been found to have an impact on Eurasian surface air temperature (Luo et al., 2017).
401 Control simulations using the GFDL CM2.1 model have shown a strong negative
402 correlation between both AMO and AMOC and Arctic sea ice at all seasons (Mahajan
403 et al., 2011). Comparison of AMO with our data broadly shows opposite sign evolution
404 with extensive sea ice until 1910s-1920s at negative AMO, followed by decreasing sea
405 ice until the 1960s as AMO increases (Fig. 7d, e). Thereafter, this relationship seems to
406 attenuate. According to Mahajan et al. (2011), the influence of AMO on the recent
407 summer sea ice decline would have been weak compared to the response of sea ice to
408 AO. This finding is further supported by CMIP3 coupled general circulation model
409 simulations using pre-industrial forcing of Day et al. (2012) showing that the AMO
410 shift from negative to positive phase between 1979 and 2010 accounted for only 0.5–
411 3.1%/decade of the 10.1%/decade of September sea ice decline. If confirmed, this result
412 would suggest that the North Atlantic heat transported to the Arctic would not be as
413 determinant in sea ice changes in the Chukchi Sea. Very recently Luo et al. (2022)
414 further highlighted the joint effect of AMO and PDO on the decadal variability of Arctic
415 air temperatures. The loss of summer Arctic sea ice and flushing of multi-layer sea ice
416 out of basin in the past decade has already changed the atmospheric circulation. The
417 overprinting effect of global warming in the Arctic may have caused different
418 environmental mechanisms and feedbacks that could explain changes in the
419 relationship between sea ice, air temperature and climatic drivers.

420

421 5. Conclusions

422 The downcore record of sea ice proxies since AD 1850 allowed to improve semi-
423 quantitative assessment of sea ice and gain insights into past sea ice and environmental
424 changes in the Chukchi Sea. Bulk parameters and sterol data along the core indicate
425 increase terrestrial material to the sediments due to enhanced continental run-off,
426 sediment transport by sediment-laden sea ice and airborne inputs. The $\delta^{15}\text{N}$ data
427 indicate gradual nutrient limitation of phytoplankton growth due to enhance

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3 428 stratification induced by surface freshening.

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5 429 Among PIP₂₅ indexes, P_{III}IP₂₅ appears to best reproduce trends and variability of sea
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7 430 ice in the Chukchi Sea. Our data also question the use of dinosterol as an end-member
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9 431 for pelagic phytoplankton in the Arctic Ocean due to potential bias of sea ice microalgae
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11 432 producing dinosterol that would deserve further investigation on sea ice samples. The
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13 433 sea ice reconstruction based on P_{III}IP₂₅ shows variations that are coherent with the
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15 434 positive and statistically significant relationship with the summer AO evidenced by
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17 435 Frey et al. (2015). Notably, the sea-ice retreat in the 1865s-1875s and from the 1920s
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19 436 to the 1970-1980s are co-eval with negative AO phases, while extensive sea-ice cover
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21 437 in the 1910-1920s occurs during positive AO. However, this negative relationship
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23 438 between AO and seasonal sea ice seems to have been altered by global warming
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25 439 overprinting effect over the last decades. Our results also suggest a minor role of PDO
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27 440 and AMO on seasonal sea ice as compared to AO since the 20th century in agreement
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29 441 with model experiments.

30 442

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45 457 **Data availability statement**

46 458 All data that support the findings of this study are included within the article (and
47 459 supplementary information files).

48 460 **Author contributions**

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3 461 Y.B. designed the study and wrote the manuscript with contribution of M.-A.S., J.R., B.J., J.C.,
4 462 H.L. and H.J. V.K. and Z.J. contributed the biomarker analyses. L.L. retrieved the
5 463 environmental data from different database. Q.Z. contributed the biogenic silica and L.S.
6 464 performed the determination of TOC. All authors contributed to the final version of the
7 465 manuscript.

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11 466 **Declarations of interest:** None
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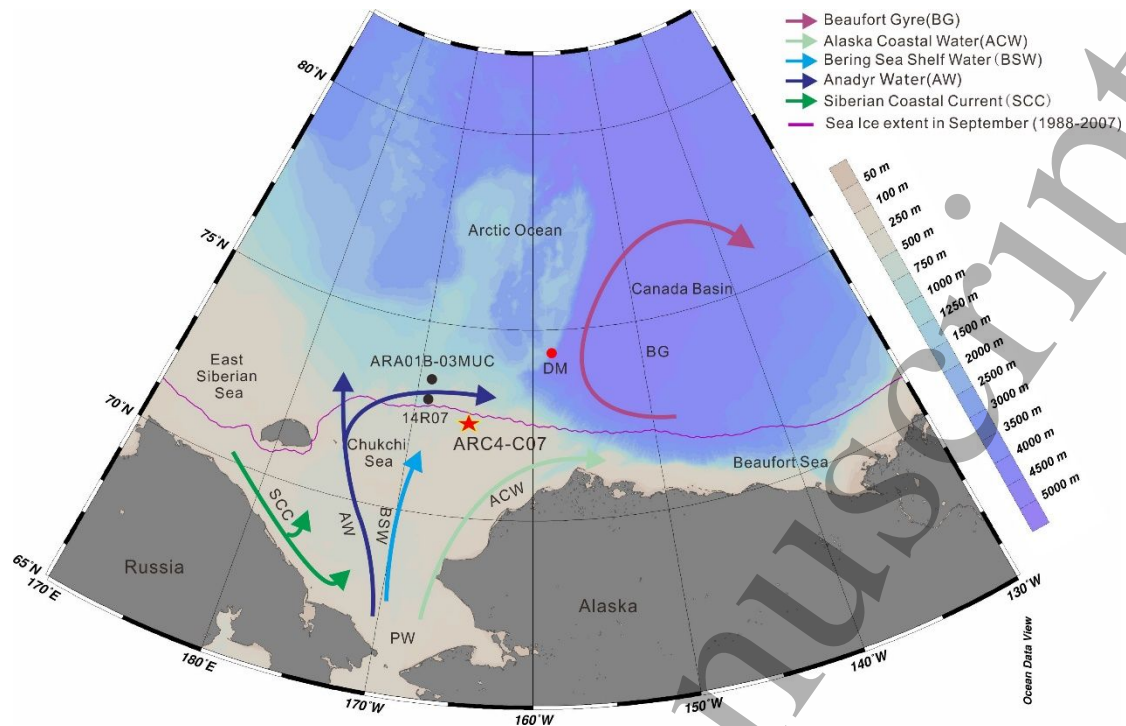
1 **Centennial-scale variability of sea-ice cover in the Chukchi Sea since**
2 **AD 1850 based on biomarker reconstruction**

5 **Youcheng Bai *et al.***

8 Figures and captions

10 Figure 1-7

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33 **Figure 1**

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35 **Figure 1.** Map of the western Arctic Ocean and oceanographic setting (after Danielson
 36 et al., 2011). The red star indicates the locations of core ARC4-C07. The black dots
 37 show other sediment cores discussed in the text (Astakhov et al., 2019; Kim et al., 2019;
 38 more information can be found in Table S1). The red dot indicates the mooring site
 39 (Station DM) (Bai et al., 2019). The purple line features the mean September sea-ice
 40 extent for the 1988-2007 period (15% of sea ice concentration) (from Cavalieri et al.,
 41 1996, <https://nsidc.org/>). Current systems; PW-Pacific Water; SCC-Siberia Coastal
 42 Current; AW-Anadyr Water; BSW-Bering Shelf Water; ACW-Alaska Coastal Water;
 43 BG-Beaufort Gyre.

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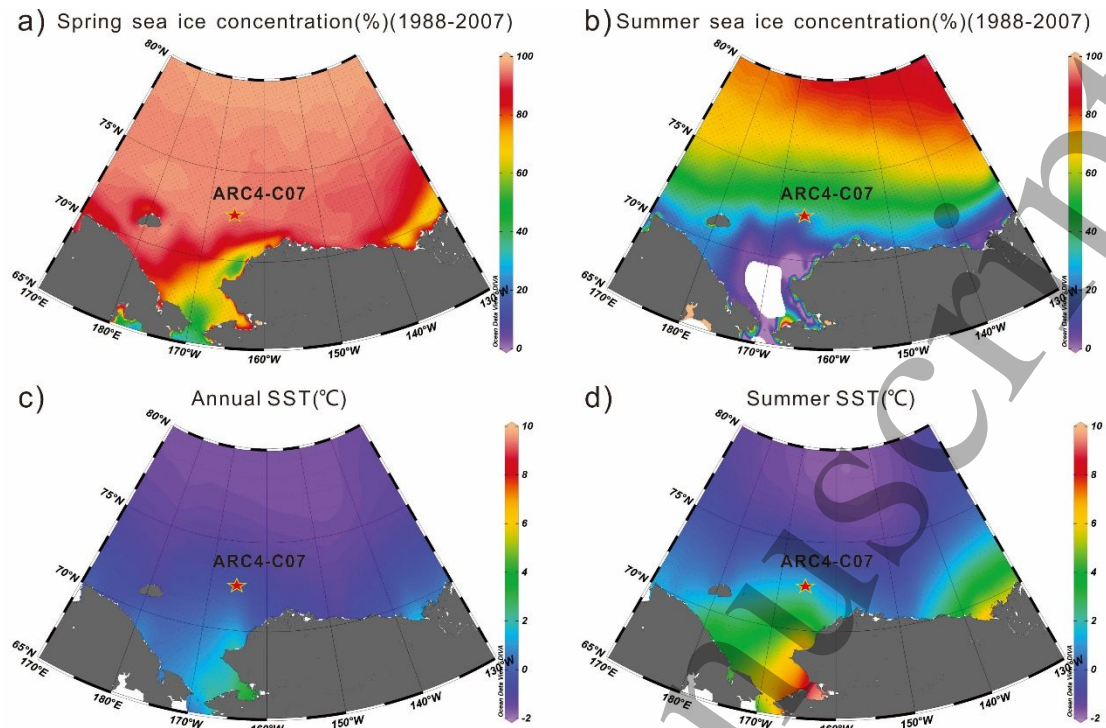
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54 **Figure 2**

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56 **Figure 2.** Averaged sea-ice concentrations from 1988-2007 for (a) spring (April, May
 57 and June) and (b) summer (July, August and September) (data source, <http://nsidc.org>).
 58 Averaged (c) annual Sea Surface Temperature (SST) and (d) summer SST (data source:
 59 Word Ocean Atlas 2013). The red star indicates location of core ARC4-C07.

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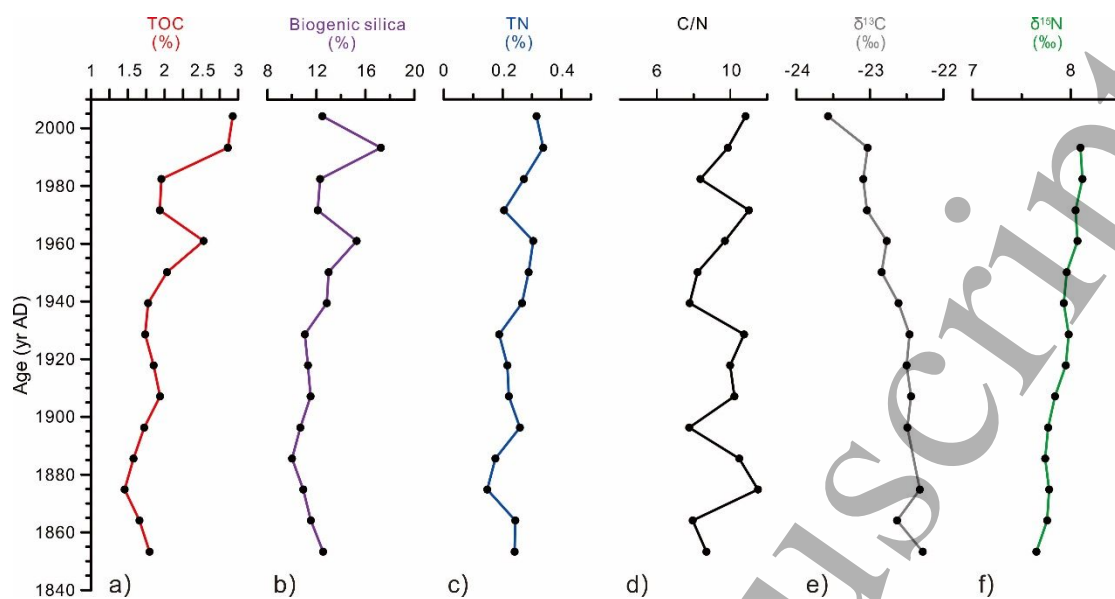
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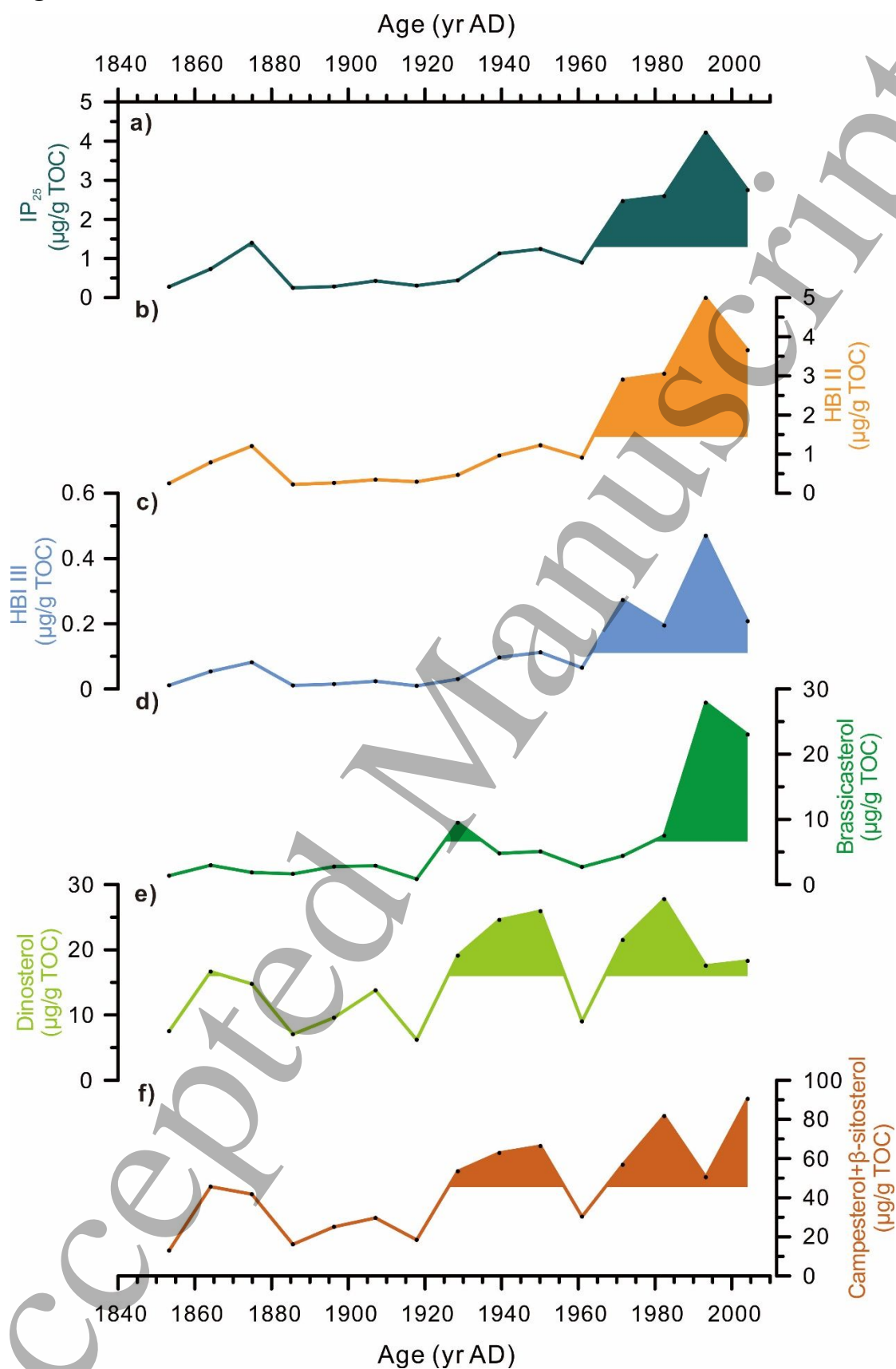
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71 **Figure 3**

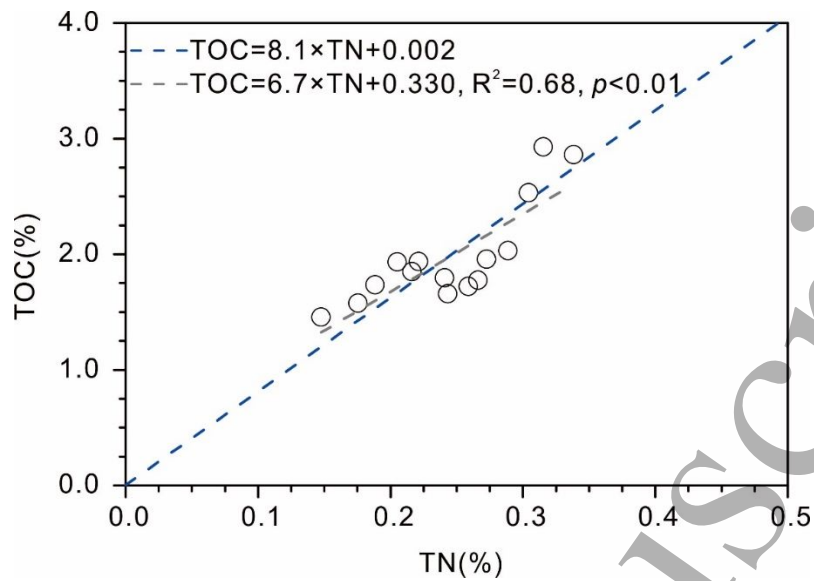
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73 **Figure 3.** Downcore profiles of a) Total Organic Carbon, TOC in %; b) Biogenic silica
74 in %; c) Total Nitrogen, TN in %; d) C/N ratio; e) carbon isotopic composition ($\delta^{13}\text{C}$)
75 of the TOC in ‰ and f) nitrogen isotopic composition ($\delta^{15}\text{N}$) in ‰ from core ARC4-
76 C07.

89 **Figure 4**

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91 **Figure 4.** Biomarker concentrations in core ARC4-C07. a) Abundances relative to
 92 internal standard and normalized to TOC of IP_{25} in $\mu\text{g/g TOC}$; b) HBI II in $\mu\text{g/g TOC}$;

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4 93 c) and HBI III in $\mu\text{g/g}$ TOC; Concentrations normalized to TOC of d) Brassicasterol in
5 94 $\mu\text{g/g}$ TOC; e) Dinosterol in $\mu\text{g/g}$ TOC; f) terrigenous sterols campesterol+ β -sitosterol
6 95 in $\mu\text{g/g}$ TOC. Colored shading highlights values above the downcore mean.
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117 **Figure 5**

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119 **Figure 5.** Cross-plot of downcore values of TOC (in %) and TN (in %) for core ARC4-
120 C07 (in grey). The blue dashed line shows the linear from Goñi et al. (2013) derived
121 from surface sediments across the North American Arctic margin and showing the slope
122 of 8.1.

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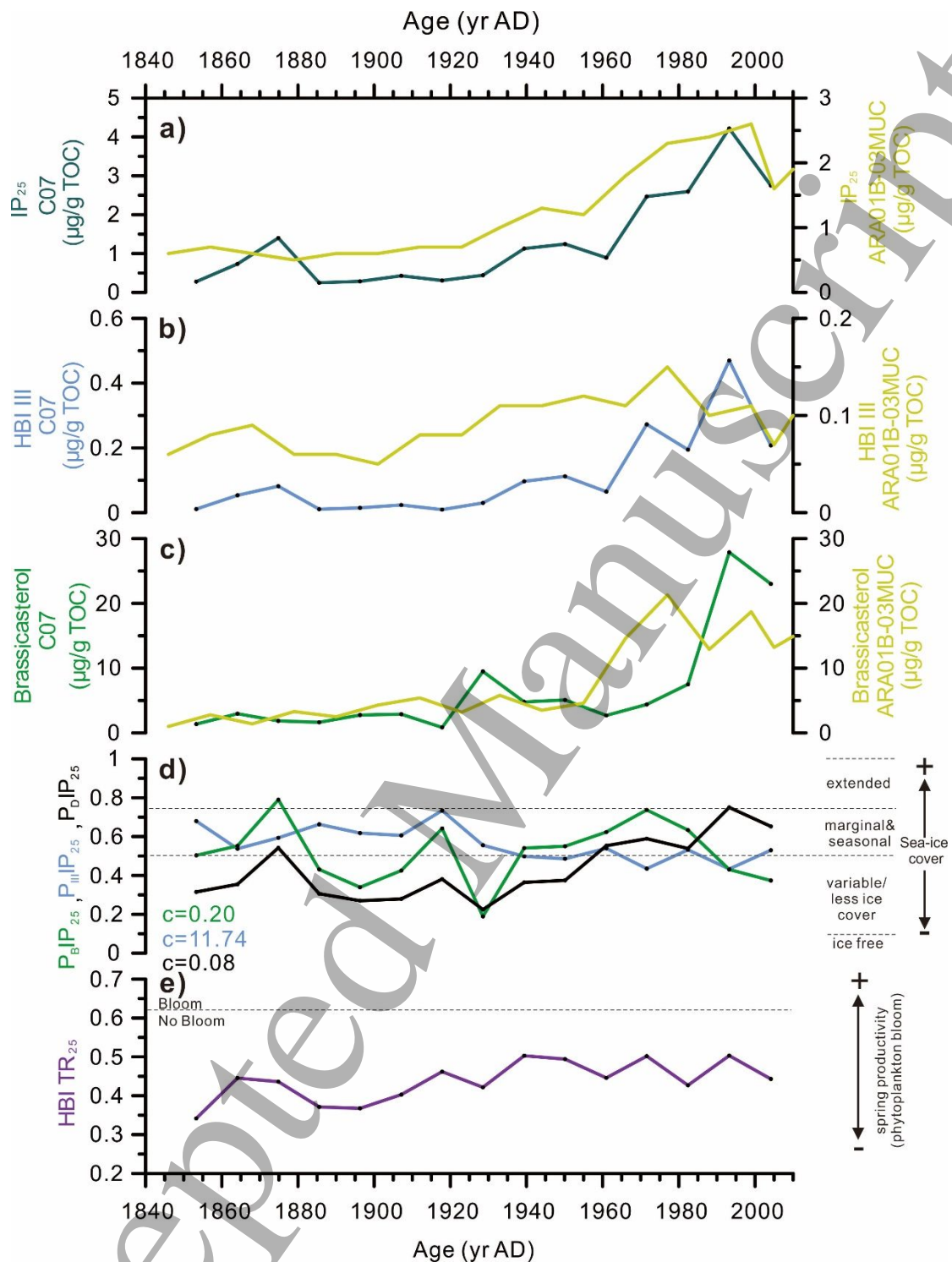
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137 **Figure 6**

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139 **Figure 6.** Downcore biomarker profiles of a) IP₂₅; b) HBI III; c) brassicasterol from
 140 core ARC4-C07; d) PIP₂₅ index using brassicasterol (P_BIP₂₅, $c=0.20$ as calculated from
 141 the current core dataset), dinosterol (P_DIP₂₅, $c=0.08$ as calculated from the current core
 142 dataset), and HBI III (P_{III}IP₂₅, $c=11.74$ as calculated from the current core
 143 dataset); e) HBI TR₂₅ calculated according to Belt et al. (2019). The dotted horizontal line ~0.62
 144 indicates the threshold for the spring phytoplankton as established by Belt et al. (2019).
 145 The yellow lines show the IP₂₅, HBI III and brassicasterol measured in core ARA01B-

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146 03MUC (Kim et al., 2019). Values can be found in Supplementary Table 2.

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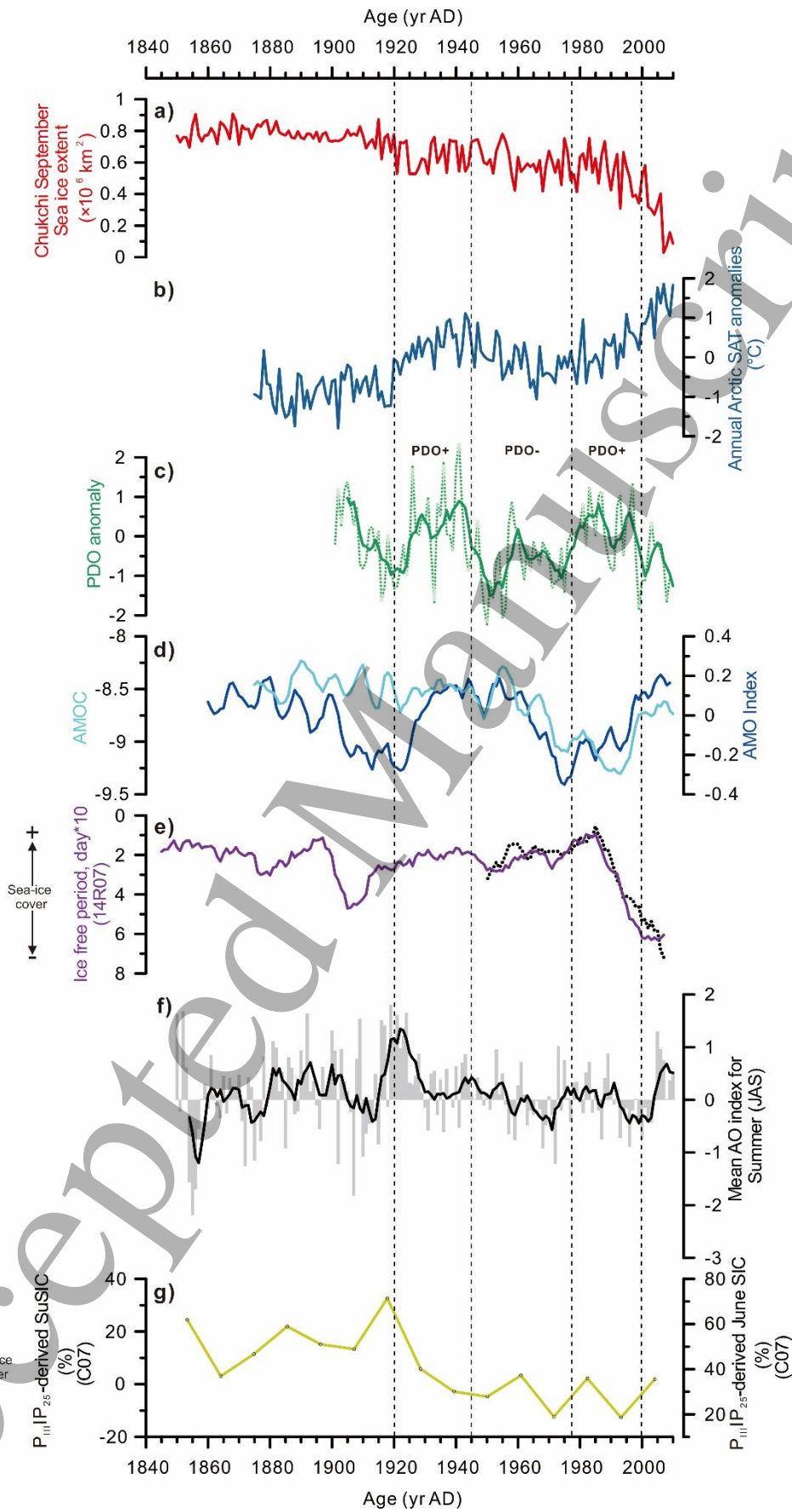
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169 **Figure 7**



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4 171 **Figure 7.** Comparison of P_{III}IP₂₅-derived summer sea ice (SuSIC) and June sea ice
5 172 (June-SIC) of core ARC4-C07 with climatic and paleoceanographic records. a)
6 173 September sea-ice extent record for the Chukchi Sea (Walsh et al., 2016); b) Composite
7 174 time series of the annual Arctic surface air temperature (SAT) anomalies (°C) relative
8 175 to 1961-90 Arctic temperature anomalies (Polyakov et al., 2002); c) Pacific Decadal
9 176 Oscillation (PDO) Index (from Halfar et al., 2011); d) AMO (Atlantic Multidecadal
10 177 Oscillation) Index (www.psl.noaa.gov/data/timeseries/AMO/) and AMOC (Atlantic
11 178 Meridional Overturning Circulation) (from Caesar et al., 2018); e) Reconstruction of
12 179 ice-free period at core 14R07 (in days). The black dotted line shows the observed
13 180 duration of the ice-free period based on the elemental composition of sediments (from
14 181 Astakhov et al., 2019); f) Arctic Oscillation (AO) mean index for summer (JAS) (Li
15 182 and Wang, 2003); g) P_{III}IP₂₅-derived summer sea ice calculated using the relationship
16 183 in Fig. S3a and for June-SIC using the relationship in Fig. S4a (see discussion). The
17 184 bold lines in c and f represent the five-point running means. The vertical dashed lines
18 185 indicate division of PDO phases as defined by Rodionov et al. (2007).