Holocene changes in deep-water circulation inferred from authigenic Nd and Hf isotopes in sediment records from the Chukchi-Alaskan and Canadian Beaufort margins

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Key points

1. The first authigenic Nd and Hf isotope records in Holocene sediment cores from the Canadian Beaufort and Chukchi-Alaskan margins are presented.

2. Unradiogenic Nd-Hf isotopic values between 11 and 4 ka cal BP are due to major weathering in the drainage basin of the Yukon/Mackenzie Rivers.

3. Radiogenic Nd-Hf isotopic compositions after 4 ka cal BP reveal an increase in the Atlantic/Pacific water inflows through the Arctic Ocean.

Abstract

The rare earth element (REE) concentrations and radiogenic isotope (Sr-Nd-Hf) compositions measured in bulk sediment leachates, together with bulk and clay mineralogical data, from two piston cores recovered in the Canadian Beaufort (AMD0214-02PC) and Chukchi-Alaskan (HLY0501-01JPC) margins were studied to investigate changes in the weathering regimes and deep-water circulation during the Holocene. The coupled evolutions of the Nd and Hf isotopic compositions (expressed in epsilon units: εNd and εHf, respectively) are in good agreement with modern seawater and bulk sediment leachate data from Pacific water, Atlantic water and the Mackenzie River. This agreement supports the idea that boundary exchange and brine formation likely play a significant role in the εNd and εHf values of the bottom waters in the western Arctic Ocean. The εNd and εHf records from the Canadian Beaufort and Chukchi-Alaskan margins reveal changes towards more radiogenic values from the early to late Holocene. Based on the εNd and εHf records, we suggest that the unradiogenic values are not controlled by water mass provenance and mixing but rather by provenance and a change in the weathering regime in the Mackenzie and Yukon drainage basins during the early to mid-Holocene. In contrast, the more radiogenic εNd and εHf values in the Chukchi-Alaskan margin and the mineralogical records in the late Holocene have primarily been controlled by an increase in the contributions of seawater and detrital particles from the Bering Sea via the Bering Strait inflow, which is likely related to major changes in the Pacific Ocean–atmospheric dynamics.

1 Introduction

The Arctic Ocean plays an important role in regulating Earth's climate because (1) its perennial sea ice cover modulates the atmospheric and oceanic heat budget since it reflects a large part of the incoming solar radiation during the summer (albedo) and acts as an insulating shield during the winter (Serreze et al., 2007) and (2) the export of freshwater into the North Atlantic affects the Atlantic meridional overturning circulation by changing the deep-water convection (Dickson et al., 2007). The northward flows of Atlantic and Pacific waters (hereinafter referred to as AW and PW, respectively) are the major sources of heat advection towards the Arctic Ocean and strongly affect sea ice distribution (Kinnard et al., 2011; Polyakov et al., 2017). For example, the increase in warm AW to the Arctic over the past 2,000 years seems to be the main factor in sea ice decline (Kinnard et al., 2011). Likewise, the advection of warm PW into the Arctic Ocean induces a greater supply of heat in the western Arctic Ocean and acts as a trigger for sea ice decline in the Chukchi Sea (Shimada et al., 2006; Stein et al., 2017). In this context, paleoceanographic and paleoclimate proxy records from marine sediment cores can provide evidence for the large-scale natural variability in the Arctic deep-water circulation during the late Quaternary, against which recent changes can be compared. A better understanding of the past variation in the deep-water circulation may help to decipher the processes controlling Arctic climate and sea ice variability.

The neodymium (Nd) and hafnium (Hf) isotope compositions of rocks largely depend on the lithology and crustal age. The Nd and Hf isotope compositions are denoted in epsilon units (εNd and εHf), which reflect the normalization of the ^{143}Nd/^{144}Nd and ^{176}Hf/^{177}Hf ratios to the chondritic uniform reservoir (Jacobsen and Wasserburg, 1980; Nowell et al., 1998). Hence, low ^{143}Nd/^{144}Nd values (εNd of -40) reflect old continental crust, while high ^{143}Nd/^{144}Nd values (εNd

68   of +20) reflect young mantle-derived rocks (Frank, 2002). The variability in the Hf isotope

69   composition in terrestrial rocks is larger than that in Nd, ranging from the most unradiogenic

70   values of εHf ~ -50 in Archean rocks to values as high as +25 in mid-ocean ridge basalt

71   (Zimmermann et al., 2009a). Water masses from different origins therefore acquire distinct

72   regional εNd and εHf signatures derived primarily from riverine continental input, particle-

73   dissolved exchange processes (a process commonly referred to as boundary exchange) and/or

74   benthic sources (e.g., Frank, 2002; Jeandel et al., 2007; Rickli et al., 2009; Zimmermann et al.,

75   2009a,b; Wilson et al., 2013; Abbott et al., 2016; Haley et al., 2017). Consequently, Nd and Hf

76   can be used as sensitive tracers for both (1) water mass mixing and provenance (Chen et al.,

77   2012; Rickli et al., 2009; Stichel et al., 2012), because both elements' oceanic residence times are

78   shorter than the oceanic mixing time (approximately 1500 yr; Frank, 2002), and (2) detrital

79   provenance changes and continental weathering intensity (Gutjahr et al., 2014; Rickli et al.,

80   2010). Because dissolved trace elements are incorporated by coprecipitation processes during

81   early burial in the top few centimeters of the sediments (Bayon et al., 2004; Haley et al., 2004),

82   the authigenic Nd and Hf signatures can be extracted from ferromanganese (Fe–Mn)

83   oxyhydroxide coatings on marine sediment samples (Bayon et al., 2004; Gutjahr et al., 2007;

84   Chen et al., 2012). Based on these findings, the analysis of εNd and εHf signatures of authigenic

85   fractions in marine sediments is a powerful tool for assessing late Quaternary changes in water

86   mass provenance and in pathways of weathering inputs.

87   Taking this into account, a number of studies have investigated the seawater Nd and Hf

88   isotope compositions of past Arctic Intermediate Water extracted from the authigenic Fe–Mn

89   oxyhydroxide fraction of late Tertiary (mid-Miocene) to Quaternary sedimentary records to

90   decipher changes in the weathering regimes and water mass mixing (e.g., Haley et al., 2008;

91   Horikawa et al., 2015; Chen et al., 2012; Maccali et al., 2013; Meinhardt et al., 2016a; Jang et al.,

92 2017). Likewise, modern Nd and Hf isotope compositions of seawater in Arctic Ocean basins

93 (Porcelli et al., 2009; Zimmermann et al., 2009a) and pre-modern authigenic Nd isotope

94 signatures of surface sediments from the Arctic Ocean seafloor (Haley & Polyak, 2013) have

95 been investigated to better present the Arctic Ocean's circulation patterns. Nonetheless, the

96 Holocene variability in the contributions of different water masses to the deeper parts of the

97 western Arctic Ocean is not yet completely understood. Thus, authigenic Nd and Hf isotopic

98 compositions retrieved from sediment cores may provide new clues concerning the evolution of

99 the Holocene deep circulation and climate in the western Arctic Ocean, which may then help to

100 place modern environmental changes in perspective.

101   In this context, the Nd and Hf isotope signatures and the rare earth element (REE)

102 concentrations obtained from the authigenic Fe–Mn oxyhydroxide fractions, together with the

103 bulk and clay mineralogical analysis, of two sediment piston cores recovered from the Chukchi-

104 Alaskan (core HLY0501-01JPC) and Canadian Beaufort (core AMD0214-02PC) margins are

105 used here to (1) assess changes in the provenance of deep-water masses, (2) interpret variations in

106 the dynamics of deep-water circulation in terms of paleoenvironmental changes since the last

107 deglaciation, and (3) provide new insights into the potential relationships between changes in

108 erosional inputs and oceanic circulation variations in the western Arctic Ocean since the last

109 deglaciation.

110 ## 2   Regional setting

111 **2.1. Oceanic circulation**

112   The Arctic surface oceanic circulation is related to two main wind-driven circulation

113 systems, which are the anticyclonic Beaufort Gyre (BG) in the western Arctic and the Transpolar

114 Drift (TPD; Darby & Bischof, 2004). On the Beaufort Shelf, the anticyclonic BG pushes both

115   surface currents and sea ice westward at the shelf break. Conversely, closer to shore around the

116   50-m isobath, the Beaufort Undercurrent transports both PW and AW eastward along the

117   continental margin and into the Amundsen Gulf (Forest et al., 2011). The AW flows through the

118   Fram Strait and the Barents Sea. The depth of the AW is between 200 and 1000 m and has a

119   temperature above 0 °C (Rudels et al., 2004). The AW circulation is counterclockwise along the

120   margins of the Arctic Ocean. It begins along the Eurasian margin and then separates at the level

121   of the Lomonosov Ridge. A branch of the AW circulation diverges towards the eastern face of

122   the Lomonosov Ridge, and the other branch continues in the Canadian basin (Figure 1; Poirier et

123   al., 2012). The modern Nd and Hf isotope signatures of the AW in Arctic Ocean basins have been

124   studied in Porcelli et al. (2009) and Zimmermann et al. (2009a), and the results showed εNd and

125   εHf values of -9 and 1.6, respectively.

126   The Chukchi Shelf circulation is controlled by an inflow of PW via the Bering Strait

127   (referred to as the Bering Strait inflow or BSI), the Siberian coastal current, and the Atlantic

128   Intermediate Water affecting the northern margin (Figure 1a; Pickart, 2004; Weingartner et al.,

129   2005). PW from the Bering Sea flows into the Chukchi Sea in three major branches (Figure 1a;

130   Pickart et al., 2004, 2005; Weingartner et al., 2005; Grebmeier et al., 2006; Okkonen et al., 2009;

131   Danielson et al., 2014; Corlett & Pickart, 2017). The first branch, composed of cold, salty (>32.5)

132   and nutrient-rich Anadyr Water, flows through Hope Valley into Herald Canyon. The third

133   branch, composed of warm, fresh (<31.8) and nutrient-poor Alaskan Coastal Water, flows into

134   Barrow Canyon following the northwestern Alaskan coast in the eastern Chukchi Sea (known as

135   the Alaska Coastal Current). The second branch, composed of intermediate saline (31.8–32.5)

136   and lower nutrient Bering Shelf Water, flows between the first and the third branches through the

137   Central Channel. In general, within the Chukchi Sea, the Anadyr and Bering Shelf waters are

138   both named Bering Sea Water (e.g., Grebmeier et al., 2006; Stein et al., 2017). At interannual

139 time scales, the intensity of the BSI is controlled mainly by the Aleutian Low pressure center's

140 strength and position (Yamamoto et al., 2017). Periods of strengthening of the Aleutian Low

141 pressure center, located over the eastern North Pacific, induce a decrease in the BSI into the

142 Arctic Ocean (Danielson et al., 2014). According to modern hydrographic observations, dense

143 waters (brines) generated at the Chukchi-Alaskan margin during fall/winter sea ice formation can

144 descend to a pycnocline depth of up to 200 m (Pickart et al., 2005; Woodgate et al., 2005).

145 Corlett & Pickart (2017) have shown that PW flows into the Barrow Canyon and forms a slope

146 current called the Chukchi Slope current. The Chukchi Slope current can transport 0.50 Sv of PW

147 westward of Barrow Canyon and can extend into the Atlantic layer (Corlett & Pickart, 2017). The

148 isotopic signature of the PW before entering the Chukchi Sea has been described in Zimmermann

149 et al. (2009b) and Asahara et al. (2012), with εNd values of approximately -2 to -3 and εHf values

150 ranging from 3.5 to 8.6 with a mean value of 6.8. The isotopic signature of the PW throughflow

151 water in the Chukchi Sea is εNd= -5 and εHf= 5.8 (Haley & Polyak, 2013; Porcelli et al., 2009;

152 Zimmermann et al., 2009a). The change in terms of the isotopic signature of the PW before and

153 after the Bering Strait is possibly due to the influence of runoff from the Yukon and Anadyr

154 rivers and/or so-called boundary exchange with northeastern Bering Sea sediments (Haley &

155 Polyak, 2013; Porcelli et al., 2009). The εNd signature of the Yukon River is εNd= -8 to -9

156 (VanLaningham et al., 2009), and the Hf isotopic signature is still unknown (Horikawa et al.,

157 2010). An increase in the BSI into the Chukchi Sea generally leads to reduced sea ice cover and

158 an increase in sea surface temperature (e.g., McKay et al., 2008; Polyak et al., 2016; Stein et al.,

159 2017). However, as the BSI flows into the Chukchi Sea in three main branches (Figure 1a), the

160 sea ice cover in this region show considerable spatial and temporal variability (Polyak et al.,

161 2016).

162

2.2. Sedimentation

On the Canadian Beaufort Shelf and the Chukchi Sea, most of the surficial seabed sediments are predominantly composed of Holocene bioturbated gray to olive-gray marine silts and clays (Gamboa et al., 2017; Kobayashi et al., 2016). The modern sedimentation in the Chukchi Sea is believed to be mainly derived from northeastern Siberia, the northeastern Bering Sea (mainly from the Yukon River and to lesser extents from the Aleutian arc and Anadyr River) and the Mackenzie River, whereas the Canadian Beaufort margin sediment originates primarily from the Mackenzie River basin (Nelson and Creager, 1977; Darby et al., 2011; Asahara et al., 2012; Horikawa et al., 2015; Kobayashi et al., 2016; Gamboa et al., 2017; Deschamps et al., 2018a). Smaller Alaskan rivers have a more local impact but may have been a more important sediment source during the early stages of the last transgression (Hill & Driscoll, 2008). During deglaciation and the early Holocene, sediment inputs to the Chukchi-Alaskan and Beaufort margins were presumably higher due to the rising sea level associated with meltwater and iceberg discharge from the retreat of large ice sheets (Deschamps et al., 2018a).

3 Material and methods

3.1 Sample and chronology

The sediment core HLY0501-01JPC (hereinafter referred to as core 01JPC; water depth: 1163 m; location: 72°90'N, 158°42'W) was recovered at the Chukchi-Alaskan margin on board the USCGC Healy as part of the 2005 Healy-Oden Trans-Arctic Expedition (Figure 1a,b). Core AMD0214-02PC (hereinafter referred as core 02PC; water depth: 998 m; location: 71°22.910'N, 133°34.040'W) was collected on the Canadian Beaufort margin on board the CCGS Amundsen during the 2014 ArcticNet expedition (Figure 1a,b). Age models and physical properties of the

185 01JPC and 02PC sediment cores have been described in Deschamps et al. (2018b). The

186 authigenic Fe–Mn oxyhydroxide coatings preserved within these sediment cores likely records

187 past bottom water REE signatures, as reductive diagenesis processes is seem to be negligible

188 (Figure S1; Deschamps et al., 2018b). The sedimentation rate for core 02PC ranged from 2-20

189 $cm.ka^{-1}$ in the postglacial units, whereas the sedimentation rate in core 01JPC is constant in the

190 postglacial unit (60 $cm.ka^{-1}$). Core 01JPC is characterized by a hiatus in the sedimentary

191 sequence at approximately 6 ka cal BP and the deglacial section of the core (Deschamps et al.,

192 2018b). For this reason, only the Holocene units of core 01JPC have been sampled (n=11,

193 resolution of 500 years). Core 02PC spans the last 13.5 ka cal BP and is characterized by two ice-

194 rafted debris (IRD) intervals between 140 and 160 cm (IRD1) and 320 and 360 cm (IRD2)

195 (Deschamps et al., 2018b). In this study, we focus on the last 11.5 ka cal BP (n= 22, resolution of

196 500 years).

197

198 **3.2   Radiogenic isotopes and REE analyses**

199 **3.2.1   Bulk sediment leaching**

200   Seawater Sr, Nd and Hf isotopic signatures from authigenic Fe-Mn coatings of the bulk

201 sediment were extracted applying the leaching protocol of Chen et al. (2012). Briefly, 1 g of dried

202 and powdered sediments was rinsed three times with Milli-Q water. Next, Sr, Nd and Hf

203 contained in the sediment oxyhydroxide fraction were leached for approximately 1 h in a single

204 step using a dilute reducing and complexing solution consisting of 0.005 M hydroxylamine

205 hydrochloride (HH), 1.5% acetic acid, and 0.03 M Na-EDTA, buffered to pH = 4 with suprapur®

206 NaOH. A buffered acetic acid leach step was omitted since biogenic carbonates are negligible in

207 all sediment samples (Deschamps et al., 2018a). The hydroxylamine hydrochloride and acetic

208 acid mixture was 10-fold diluted compared with the method of Gutjahr et al. (2007) to avoid any

209 potential contamination caused by the leaching of clay minerals. During treatment, the sediment

210 samples were gently shaken to enhance the reaction. Leaching method was applied on two

211 sediments sample, one for the REE concentration analyses and the other one for the Nd and Sr

212 chromatographic extraction. The aliquot for REE concentration analysis was evaporated almost

213 to dryness and the residue was re-dissolved in 1 mL of concentrated HNO_3 and subsequently

214 diluted with Milli-Q water to a total volume of 5 mL.

215

216 **3.2.2   Sr, Nd and Hf separation: column chemistry**

217   The Sr, Nd and Hf were separated from the other elements by applying a single-step ion

218 chromatographic separation (Li et al., 2014). Briefly, the leaching solutions obtained from the

219 previous steps were centrifuged at 5000 rpm for 8 min. Then, 1 mL of the supernatant solution

220 was passed through a two-layered mixed resin column (70 mm length, 6 mm diameter) with the

221 upper layer containing 1.5 mL of Biorad® AG50W-X12 (200–400 mesh) resin and the bottom

222 layer containing 0.45 mL of Eichrom® LN Spec resin (100–150 µm). Before sample loading for

223 the separation of Sr–Nd–Hf from the sample matrix, the mixed resin column was pre-washed

224 with 18 mL of 6 M HCl, 8 mL of 3 M HF, and 4 mL of H_2O in turn. After sample loading and

225 rinsing four times with 0.5 mL of 2.5 M HCl, the column was washed with 13.5 mL of 2.5 M

226 HCl. Most matrix elements (K, Ca, Na, Mg, Al, Fe, Mn, Ti) and Rb were removed during this

227 step. Then, the Sr fraction was stripped with 5.5 mL of 2.5 M HCl. Part of the HREE (Dy, Ho,

228 Er, Tm, Yb, Lu) and Ba were then washed out with 3 mL of 2.5 M HCl. Next, the Nd was then

229 isolated from the other REE with 8 mL of 6 M HCl. Finally, the Hf was isolated with 5 mL of 3

230 M Hf. Then, the Sr, Nd, and Hf fractions were dried on a hotplate at 120°C to dryness and

231 prepared for isotope measurements.

3.2.3 REE concentrations and Sr-Nd-Hf analysis

233 The REE concentrations (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu)

234 were determined using an inductively coupled plasma-quadrupole mass spectrometer (ICP-QMS

235 Agilent 7500c) at ISMER. Procedural blanks (chemistry and mass spectrometry) always

236 accounted for less than 1% of the lowest concentrations measured in the samples. Multi-element

237 stock standard solution containing all REE (multi-element solution 1, CLMS-1, Spex Certiprep

238 Inc., Quebec, Canada) was used to prepare external calibration and a quality control standard

239 solution containing 4 ng/mL of each REE. ICP-QMS external reproducibility, based on replicate

240 analysis of this control standard solution, was <11% relative standard deviation (RSD, 1σ) for all

241 REE. The REE abundances were normalized to Post-Archean Australian Shale (PAAS; Taylor &

242 McClennan, 1985) in order to evaluate the REE patterns as given in (Maccali et al., 2013) and Du

243 et al. (2016). The fractionation between the light REE (LREE: La–Nd), medium REE (MREE:

244 Sm–Dy) and heavy REE (HREE: Tm–Lu) was investigated using the following indices:

245 HREE/LREE ([Yb + Lu]/[Pr + Nd] and MREE* (2[Tb + Dy]/[Pr + Nd + Yb + Lu]) to investigate

246 the fractionation between LREE, MREE, and HREE (Du et al., 2016; Molina-Kescher et al.,

247 2014).

248 The Sr isotopic ratios ($^{88}Sr/^{86}Sr$) were measured in dynamic mode on a Thermo Scientific

249 Triton Plus™ multicollector thermal ionization mass spectrometer (TIMS) at GEOTOP

250 (Montreal, Canada). The Sr samples were loaded and analyzed on a single outgassed zone-refined

251 Re filament, layered with a tantalum activator solution (Birck, 1986). Repeated analyses of the

252 NIST-987 standard (n=6) yielded values of 0.710276 (±0.000021, 2σ reproducibility). This mean

253 value compares well to its certified value of 0.710248 (Weis et al., 2006). The total procedural

254   blanks for Sr were less than 0.5 ng, which is considered negligible compared to the sample yields

255   (> 100 ng).

256   The Nd and Hf isotopic ratios ($^{143}Nd/^{144}Nd$ and $^{176}Hf/^{177}Hf$) were analyzed on a Nu

257   Plasma II instrument, a Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MC-

258   ICP-MS), also at GEOTOP, in ''dry-plasma'' conditions using an Aridus II desolvating

259   membrane as the introduction device. The mass-bias correction was made by monitoring

260    $^{146}Nd/^{144}Nd$ (taken to be equal to 0.7219) and $^{176}Hf/^{177}Hf$ (taken to be equal to 0.7325) and by

261   applying an exponential beta-factor correction to the other ratios. Replicate analyses of the

262   standard JNdi-1 and JMC 475 yielded a mean value of $^{143}Nd/^{144}Nd$ = 0.512108 ±0.000020 (2σ;

263   n=20) and $^{176}Hf/^{177}Hf$ = 0.282159 ±0.000009 (2σ; n=20) which are within the uncertainty of its

264   certified values of 0.512115 ±0.000007 (Tanaka et al., 2000) and 0.282160 ±0.000032 (Nowell et

265   al., 1998), respectively. Hence, no correction has been applied to the Nd and Hf isotope data. The

266   external reproducibility was provided by the repeated measurements of the JNdi-1 (from 0.1 to

267   0.3 ε units, 2σ; n=31) and JMC 475 (0.5 to 1.3 ε units, 2σ; n=31) standards at the same

268   concentration as the samples. Thus, the analytical error for each sample analysis is taken as the

269   external reproducibility of the JNdi-1 and JMC 475 standard in each analytical session. Some

270   samples have higher uncertainties (up to 0.4 ε units for Nd and up to 9 ε units for Hf; Table S2)

271   because of poorer counting statistics of samples with low Nd and Hf concentrations.The

272   procedural blank values were < 0.5 ng for Nd and for Hf and were therefore neglected as they

273   represented less than 0.1% of the Nd and Hf analyzed per sample, respectively. The $^{143}Nd/^{144}Nd$

274   and $^{176}Hf/^{177}Hf$ isotopic ratios are expressed in ε units (εNd and εHf).

3.3 Bulk and clay mineralogical analyses

Complementary bulk mineral associations were studied by quantitative X-ray diffraction (qXRD) following the method developed by Eberl (2003). Briefly, ~1 g of each sample was spiked with 0.25 g of corundum, and the powder samples were scanned from 5° to 65° two-theta in steps of 0.02° two-theta on a PANalytical X'Pert Powder diffractometer. For the quantification of the major mineralogical components, sediment XRD scans obtained were converted into mineral weight percent (wt. %) using the Excel macro program ROCKJOCK v11 (Eberl, 2003). Then, we used the non-linear unmixing Excel macro program SedUnMixMC (Andrews & Eberl, 2012) to gain a quantitative understanding of the downcore changes in bulk sediment provenance. In addition, clay mineral associations were studied using XRD following established protocols (Bout-Roumazeilles et al., 1999). The separated clay-sized fraction was concentrated by centrifugation and oriented by wet smearing on glass slides. The analyses were run from 2.49° to 32.49° two-theta on a PANalytical X'Pert Powder diffractometer. Three X-ray diagrams were performed, and after the sample was air-dried, ethylene glycol vapor saturation was completed for 12 h, followed by heating at 490 °C for 2 h. A semi-quantitative estimation of clay mineral abundances (smectite, illite, chlorite, kaolinite, vermiculite and a chlorite/smectite mixed layer) based on peak areas was performed using the MacDiff® 4.2.5 software (Petschick, 2000). Similar to other Arctic clay mineral studies (Schoster et al., 2000; Wahsner et al., 1999), the clay mineral contents were calculated by using the weighting factors introduced by Biscaye (1965) and calculated to a sum of 100%. Note that bulk and clay mineralogical analyses on sediments for the core 02PC were reported previously (Deschamps et al., 2018a). In this study, based on a previous sediment provenance study in the western Arctic Ocean (Deschamps et al., 2018a), we used the proportion of sediments derived from the Bering Strait and Mackenzie River (SedUnMixMC

298 results), as well as the amorphous silica contents and Log(illite+kaolinite/chlorite+vermiculite)

299 ratio or Log(I+K/C+V) to trace sediment provenance changes over time. The high amorphous

300 silica concentrations in the Chukchi Sea sediments can be interpreted as a major inflow of

301 biosilica-rich PW through the Bering Strait (Jakobsson et al., 2017; Stein et al., 2017). Likewise,

302 the Log(I+K/C+V) ratio allowed us to discriminate between sediments from the Bering Strait

303 (rich in chlorite and vermiculite) and sediments from the Mackenzie River (rich in illite and

304 kaolinite).

305 4   Results

306 4.1   Authigenic REE distribution

307 The REE concentrations are presented in Table S1. The PAAS-normalized REE of the bulk

308 sediment leachates from both cores reveal an MREE bulge-type pattern (Figure 2), with an

309 enrichment in MREEs compared to HREE and LREE, which is a common pattern in leachates

310 and authigenic material (Gutjahr et al., 2007; Du et al., 2016; Abbott et al., 2016). Likewise, to

311 further evaluate the efficiency of our procedure for extracting the authigenic phase, we compared

312 the HREE/LREE ratios to the MREE* values (Figure 3b). The HREE/LREE-MREE* cross-plots

313 reveal that all our bulk sediment leachates plot on the Fe–Mn leachate array (Du et al., 2016;

314 Gutjahr et al., 2010). This finding indicates that authigenic Fe–Mn oxyhydroxide coatings control

315 the Nd and Hf signals in our bulk sediment leachates. Furthermore, the significant negative

316 correlation observed between the ΣREE content and εNd values in cores 01JPC (r= -0.75) and

317 02PC (r= -0.59) suggests ΣREE enrichment towards more unradiogenic values (Figures 3b).

4.2 Sr, Nd and Hf isotope signatures

The Sr, Nd, and Hf isotope data obtained from the leachates are provided in Table S2. The $^{87}Sr/^{86}Sr$ values obtained from the bulk sediment leachates in cores 01JPC and 02PC range from 0.70922 to 0.70940, with a mean value of 0.70929 ± 0.00004 (Figure 3a). These values are characteristic of the present-day sea water values recorded in the western Arctic Ocean (0.70920; Asahara et al., 2012). In core 01JPC, the εNd values range from -3.7 to -5.7, while the εHf values range from 4.1 to 9.2 (Figures 3d and 4a). In general, the Nd and Hf isotopic signatures in core 02PC are less radiogenic than those in core 01JPC. The εNd values range from -7.6 to -16.8, while the εHf values range from 4.8 to -10.4 (Figures 3d and 4b,c). As shown in Figure 3d, the Nd-Hf isotope values of core 01JPC fall into the sea water array, whereas the data from core 02PC range from the sea water array to the detrital array.

4.3 Bulk and clay mineralogical data

The stratigraphic distributions of the bulk and clay mineralogical data from core 02PC are shown in Deschamps et al. (2018a). The mineralogy of the bulk sediment fraction of core 02PC is dominated by quartz (~22%) and total clays (72%). The clay mineral assemblage of core 01JPC consists of illite (60%), kaolinite (14%), chlorite (14%), vermiculite (8%) and mixed-layer chlorite/smectite (2%). Based on the SedUnMixMC results (Figure 5g), the major source of sediment for core 02PC is related to the Mackenzie River (> 80%), and the secondary source is the Canadian Arctic Archipelago (up to 60%). The bulk and clay minerals concentrations of the core 01JPC are presented in Tables S3 and S4, respectively. The mineralogy of the bulk sediment fraction of core 01JPC is dominated by quartz (~20%), total clays (52%), plagioclase (11%), K-feldspar (5%) and amorphous silica (3-14%), and the clay mineral assemblage of core 01JPC consists of illite (27-45%), kaolinite (5-10%), chlorite (10-15%), vermiculite (7-50%) and mixed-

341 layer chlorite/smectite (0-40%). Based on the SedUnMixMC results (Figure 6f; Table S5), the

342 major source of sediment for core 01JPC is related to the northeastern Bering Sea (40-60%), and

343 the secondary source is the Mackenzie River (10-30%).

5 Discussion

5.1 Nd and Hf isotopic compositions of cores 02PC and 01JPC

346 As shown in Figures 3d and 4b,c, the Nd and Hf isotopic composition of core 02PC

347 displays high variations and ranges between the signatures of the AW (εNd: -9, εHf: 1.6;

348 Zimmermann et al., 2009a) and the modern Mackenzie River (εNd: -12.9, εHf: -7.1;

349 Zimmermann et al., 2009a). From 12 to 6 ka cal BP, the εNd values in core 02PC are clearly

350 similar to those of the Mackenzie River, whereas between 6 and 2 ka cal BP, the Nd isotopic

351 values become more radiogenic and reflect a mixture between those of the AW and the

352 Mackenzie River (Figure 4b,c). After 2 ka cal BP, the εNd values match those of the AW. Similar

353 to the Nd isotopic compositions, the εHf isotopic values in core 02PC transition from a clear

354 Mackenzie River signature (12-6 ka cal BP) to a mixed signature between the Mackenzie River

355 and the AW (6-2 ka cal BP) to predominantly an AW signature after 2 ka cal BP (Figure 4b,c).

356 As shown in Figures 3d and 4a, the Nd isotopic values of core 01JPC between 6 and 4 ka

357 cal BP (εNd ~ -5.5) are very similar to those of the PW recorded in the Chukchi Sea region

358 (Haley & Polyak 2013; Zimmermann et al., 2009a). This εNd signature likely represents a

359 mixture dominated mainly by unradiogenic Nd isotope compositions from the Yukon River (εNd

360 ~ -8 to -9) and a minor proportion of more radiogenic Nd isotope compositions from the Aleutian

361 arc (εNd ~ +6 to +10; Asahara et al., 2012; Jang et al., 2017). From 1 to 4 ka cal BP, the Nd

362 isotopic values become more radiogenic (εNd ~ -4), shifting towards the eastern Bering Sea

363   Water signatures (εNd= -2 to -3), as estimated from the εNd values of the Fe–Mn oxide fraction

364   (Asahara et al., 2012). Despite the large uncertainties in the Hf isotopic composition, the overall

365   values in core 01JPC match the overall Hf isotopic signature of the PW (Zimmermann et al.,

366   2009a,b; Figure 3d). However, the large external uncertainties associated with the Hf isotopic

367   values in core 01JPC do not allow the determination of Holocene changes in the PW values

368   (Figure S2 and Table S2).

369   Overall, the εNd and εHf values from the studied sediment leachates represent the bottom

370   seawater values in the Chukchi and Beaufort seas, and the PW (including Bering Sea and

371   Alaskan Coastal waters), AW and Mackenzie River end members are clearly distinguishable

372   from one another (Figure 3d and 4). The long-term Nd-Hf isotope variations observed in our bulk

373   sediment leachates, the proportion of sediments derived from the northeastern Bering Sea and

374   Mackenzie River (SedUnMixMC results), the amorphous silica contents and the Log(I+K/C+V)

375   ratio are discussed below in terms of changes in water sources, shelf-seawater interaction, brine

376   formation, continental input, and possible relationships with both deglacial/Holocene climate

377   variability and relative sea level variations.

378   **5.2   Role of weathering regime changes in the Nd and Hf isotopic evolution**

379   Seawater εNd and εHf values are essentially determined by the mixing of different water

380   masses in the open ocean, whereas the interaction between dissolved and detrital fractions is

381   significant near river mouths and continental margins (Chen et al., 2012). Several studies (Lacan

382   & Jeandel, 2005; Pearce et al., 2013; Tachikawa et al., 1999) have discussed the exchange of

383   material between lithogenic particles and seawater along continental margins, a process

384   commonly referred to as boundary exchange, which is thought to play a significant role in

385   controlling the Nd and Hf isotopic and REE compositions of the oceans. In the Arctic Ocean,

386 continental weathering plays an important role in the εNd and εHf values. Indeed, the authigenic

387 Nd and Hf isotopic values of a core recovered on the Lomonosov Ridge that spans the last 14

388 Myr were more radiogenic than those of the AW, and this difference was greater during glacial

389 periods than during interglacial periods (Chen et al., 2012; Haley et al., 2008). These authors

390 concluded that enhanced continental weathering together with reduced AW inflow during glacial

391 periods was responsible for the more radiogenic values recorded on the Lomonosov Ridge. On

392 the other hand, as described in Porcelli et al. (2009), compared to Atlantic-sourced waters, deep

393 waters in the Canada Basin are enriched in dissolved Nd, apparently through the addition of

394 dissolved Nd from the shelves via brine rejection. Similarly, the seawater Hf concentrations in

395 the Canada Basin are highest at the surface and lowest in the deeper waters, suggesting the

396 addition of river-derived Hf (notably from the Mackenzie River; Zimmermann et al., 2009a).

397 The detrital εNd values in the northeastern Bering Sea are relatively unradiogenic (~ -7),

398 suggesting that sediments are mainly derived from the Yukon River (εNd ~ -8 to -9) and to a

399 lesser degree from the Aleutian arc (εNd ~ +6 to +10) (Asahara et al., 2012; Horikawa et al.,

400 2015; Jang et al., 2017). In contrast, the detrital εNd values in the Mackenzie area are highly

401 unradiogenic (~ -15), reflecting material from the North American Craton (Maccali et al., 2018).

402 Thus, the ΣREE concentrations derived from the bulk sediment leachates in cores 02PC and

403 01JPC increase towards more unradiogenic εNd and εHf values between 12 and 4 ka cal BP,

404 probably reflecting major inputs of both suspended and dissolved loads from the Mackenzie

405 River and Yukon rivers, respectively (Figure 4b,c). We hypothesize that the major Nd and Hf

406 inputs associated with enhanced weathering of the Mackenzie (εNd ~ -12.9; εHf ~ -8.1) and

407 Yukon (εNd ~ -8 to -9) river watersheds likely provided a higher contribution of unradiogenic

408 material to cores 02PC and 01JPC during the early to mid-Holocene. Assuming that the addition

409 of Nd and Hf by the rivers could be applied to all REEs, a period of intense weathering in the

410   Mackenzie and Yukon drainage basin may have increased the input of the dissolved ΣREE load

411   and the release of material with unradiogenic Nd and Hf isotopic values into the Beaufort and

412   Chukchi seas (Figures 3c,d and 4). Conversely, the ΣREE concentrations in cores 02PC and

413   01JPC decreased during the last 4 ka cal BP, and the Hf and Nd isotopic compositions reflect

414   more radiogenic values. These shifts are probably related to a decrease in the weathering rates in

415   the Mackenzie and Yukon basins and enhanced inflow of PW into the Arctic Ocean (Figures 3c,d

416   and 4).

417   **5.3   Causes of deglacial-Holocene seawater εNd and εHf variations**

418   **5.3.1   Canadian Beaufort margin**

419   In the Canadian Beaufort margin, the Nd and Hf isotopic values from core 02PC (located

420   at a depth of ~1000 m) exhibit a large range, from -16 to -8 for εNd and from -10 to 5 for εHf,

421   implying major changes in the seawater εNd and εHf since the deglaciation (Figures 5h,i). Based

422   on previous radiogenic isotope studies (Chen et al., 2012; Porcelli et al., 2009; Zimmermann et

423   al., 2009a), we suggest that seawater εNd and εHf variation records in core 02PC can be

424   interpreted mainly as the mixing of two dominant isotopic end members, namely, the Mackenzie

425   River and AW (Figures 5h,i). The clear Mackenzie River signature recorded in core 02PC during

426   the early Holocene (εNd ~ -13) suggests that enhanced Mackenzie River discharge during this

427   period induced an increase in downslope sediment transfer, resulting in a change in the bottom

428   water Nd-Hf isotope values by particle-dissolved exchange processes (boundary exchange; Haley

429   & Polyak, 2013; Pearce et al., 2013; Tachikawa et al., 1999). Alternatively, we cannot rule out

430   that brine rejection resulting from sea ice formation also plays a significant role in the

431   redistribution of dissolved Nd and Hf within the water column (Haley & Polyak, 2013).

432   Therefore, we speculate that the higher release of dissolved and suspended loads with

433   unradiogenic Nd-Hf signatures from the Mackenzie River during the early Holocene, in

434   conjunction with slope convection of brine-enriched shelf waters to the deeper waters,

435   significantly influenced the bottom water Nd and Hf isotope values on the Canadian Beaufort

436   margin. However, as there is no observational evidence for slope convection under 300 m in the

437   Canadian Beaufort Sea (Forest et al., 2015), further investigations are needed to validate this

438   hypothesis. In addition, the most unradiogenic Nd values (εNd ~ -16) at approximately 11 ka cal

439   BP coincide with the dolomite-rich IRD originating from the Canadian Arctic Archipelago

440   (Deschamps et al., 2018a; Figure 5g,h). The geological terrain in the Canadian Arctic

441   Archipelago are characterize by very unradiogenic Nd isotope values (-14 to -16; Maccali et al.,

442   2018). Thus, the most unradiogenic Nd values recorded in core 02PC during the early Holocene

443   could be explained by (1) the slight dissolution of detrital dolomite during the leaching procedure

444   and/or (2) enhanced dolomite-rich detrital input from the Canadian Arctic Archipelago at this

445   time (Figure 7a) and subsequent particle-dissolved exchange processes (Pearce et al., 2013).

446   Any major modification in detrital inputs from the Mackenzie River that occurred during

447   the early to middle Holocene could not have been driven by the Laurentide Ice Sheet (LIS),

448   which was considerably reduced in size by this time (Dyke, 2004). Wickert (2016) suggested that

449   meltwater inputs to the Mackenzie River ended no later than 11 ka cal BP, when its eastern

450   tributaries were temporarily rerouted eastward due to a combination of ice retreat and glacial

451   isostatic depression. Based on permafrost studies in the Canadian Arctic (Burn, 1997; Dallimore

452   et al., 1996), we hypothesize that the very unradiogenic εNd and εHf values recorded in core

453   02PC during the early to middle Holocene are more likely the result of a major remobilization of

454   readily erodible rock flour and unconsolidated sediments derived from glacially deformed terrain

455   in the Mackenzie River watershed. The permafrost degradation in the western Canadian Arctic

456 correlates with an enhanced boreal summer insolation during the early Holocene (Figure 5a),

457 which would have increased soil moisture storage and facilitated erosion (Burn, 1997).

458 Furthermore, sedimentary processes in the Beaufort Sea during the early to middle Holocene

459 were strongly influenced by regional sea level variations (Deschamps et al., 2018a; 2018b).

460 Recently, Cornuault et al. (2018) showed that the εNd record can be influenced by sea level

461 variations. Similarly, the unradiogenic Nd and Hf isotopic values recorded in core 02PC showed

462 a parallel evolution together with the relative sea level variations during the early to middle

463 Holocene (Figure 5b). In this context, we hypothesize that low relative sea level conditions

464 enhanced the relative influence of the Mackenzie River and Canadian Arctic Archipelago (and

465 hence that of isotopic exchange with particles; Pearce et al., 2013) on the Canadian Beaufort

466 Shelf during the early to middle Holocene, leading to more unradiogenic Nd and Hf isotopic

467 values (Figures 5h,I and 7a).

468 The variability in the weathering regime on the Mackenzie River basin during the mid- to

469 late Holocene is likely linked to changes in the precipitation patterns over the western North

470 American continent. Indeed, previously investigations on the regional hydrologic responses to

471 atmospheric circulation patterns during the Holocene (e.g., Barron & Anderson, 2011; Anderson

472 et al., 2016) suggest that variations in precipitations on western North America could be

473 controlled by changes in the large-scale atmospheric climate modes, similar to the modern El

474 Niño Southern Oscillation (ENSO) and the Pacific Decadal Oscillation (PDO). An increased sea

475 surface temperature in the North Pacific during a positive PDO (PDO+) phase leads to enhanced

476 water vapor transport over the continent, thus increasing winter precipitation in western North

477 America (Anderson et al., 2016). These conditions are reversed during a negative PDO (PDO-)

478 phases. The transition between the mid- to late Holocene (4 ka cal BP) was characterized by a

479 decrease in the boreal summer insolation (Figures 5a), which affected the Northern Hemisphere

480   climate system (Wanner et al., 2008). Several paleoclimate studies based on Alaskan terrestrial

481   records (Anderson et al., 2005; Anderson et al., 2016; Barron & Anderson, 2011) and Bering

482   Shelf marine records (Harada et al., 2014; Katsuki et al., 2009) suggest that this transition was

483   characterized by (1) changes from a PDO+ state to a strong PDO- state (Figure 5d) and (2) a

484   decrease in the amount of precipitation linked to a major change in the Aleutian Low intensity

485   and position over the North Pacific. These changes are inferred to have been caused by Pacific

486   Ocean–atmospheric dynamics attributed to an increase in El Niño frequency and a warm eastern

487   tropical Pacific sea surface (Anderson et al., 2016; Liu et al., 2014).

488   In this context, numerical models of the regional hydrologic responses of the Mackenzie

489   River to large-scale atmospheric circulation patterns suggest a decreasing trend in the freshwater

490   discharges to the Canadian Beaufort margin associated with greatly reduced moisture transport

491   over North America during the mid- to late Holocene (Figure 5e), likely driven by a reduction in

492   the boreal summer insolation (Figure 5a) and a predominantly negative PDO-like conditions

493   (Figure 5d; Anderson et al., 2005; 2016; Barron & Anderson, 2011; Wagner et al., 2011; Wanner

494   et al., 2008). Thus, we suggest that this decreased discharge of the Mackenzie River also reduced

495   the inputs of dissolved ΣREE and detrital material with highly unradiogenic Nd and Hf isotopic

496   values to the Canadian Beaufort Shelf (Figure 3c). From 8 to 1 ka cal BP, the isotopic record of

497   core 02PC increases progressively towards more radiogenic values (εNd: -9.5; εHf: 2), similar to

498   the modern values of the AW recorded in the Canada Basin (Figures 5h and 7b,c; Porcelli et al.,

499   2009; Zimmermann et al., 2009a). However, the εNd values observed in the Canada Basin are

500   slightly higher than those of the water entering the North Atlantic (εNd of ~ -10.7; Porcelli et al.,

501   2009). This difference in εNd values probably reflects the influence of the PW (Figure 7c). A

502   εNd value between -9.6 and -9.1 can be obtained by mixing 20% PW and 80% AW (Porcelli et

503   al., 2009). Similar trends have been observed in the Nd isotopic values of a core from the Fram

504   Strait and have been attributed to PW influence (Figure 5c; Maccali et al., 2013). Thus, the

505   changes observed in the authigenic Nd and Hf isotope signatures in core 02PC from 8 to 1 ka cal

506   BP may likely reflect a combination of a decrease in the weathering rates within the Mackenzie

507   River catchment and a relative increase in the influence of the AW and PW masses (Figures 5h,I

508   and 7b,c). The enhanced PW inflows is supported by quantitative mineralogical data from the

509   Chukchi Sea, which suggest a gradual increase in sediments delivered from the northeastern

510   Bering Sea by the BSI after 8 ka cal BP (Deschamps et al., 2018a). Furthermore, quantitative

511   reconstructions of past sea surface conditions (temperature, salinity, and the duration of sea ice

512   cover), based on dinoflagellate cyst assemblages and transfer functions (Figure 5f), reveal

513   relatively long-term stable oceanographic conditions during the late Holocene (Bringué &

514   Rochon, 2012). However, the low resolution of our geochemical and mineralogical records

515   prevents any linkage with the short-term variation in past sea surface conditions in the Beaufort

516   Sea. Overall, these results suggest that the modern oceanographic conditions in the Canadian

517   Beaufort Shelf were established during the late Holocene with the concomitant dominance of AW

518   and PW.

519   **5.3.2   Chukchi-Alaskan margin**

520   In the Chukchi-Alaskan margin, our bulk sediment leachate data from core 01JPC

521   (located at a depth of > 1000 m) show εNd values between -6 and -4 (Figure 6e), similar to the

522   North Pacific throughflow water signatures measured in the Bering Strait (εNd ~ -6 to -4; Porcelli

523   et al., 2009). Note that regional sea level variations did not influence the geochemical record in

524   the Chukchi-Alaskan margin during the mid- to late Holocene, as these has been relatively stable

525   during the last 6 ka cal BP (Figure 5b; Lambeck et al., 2014; Deschamps et al., 2018a). However,

526   core 01JPC is located in an area where winter hypersaline polynya waters form through

527   additional salt input from brine rejection along the Alaska coast (Hirano et al., 2018). Based on

528   hydrographic and satellite-derived sea ice production data obtained over the eastern Chukchi

529   shelf and southeastern Chukchi borderland in conjunction with numerical modeling, Hirano et al.

530   (2018) suggest that winter hypersaline polynya waters over the eastern Chukchi shelf can

531   potentially intrude (via the Barrow Canyon) to depths comparable to or deeper than the AW layer

532   in the Canada Basin. In this context, we hypothesize that particle-dissolved exchange processes

533   with northeastern Bering Sea sediments and/or downflow of brine-enriched Chukchi Sea shelf

534   waters may play a significant role in the distribution of Nd throughout the water column in the

535   Chukchi-Alaskan margin (Haley & Polyak, 2013; Porcelli et al., 2009). These interpretations are

536   in agreement with observations by Haley & Polyak (2013) on the surface εNd distribution in bulk

537   sediment leachates from the Chukchi margin. These authors speculate that modern to pre-modern

538   distinct radiogenic εNd signals (-6 to -4) observed on the slopes of the Chukchi margin and

539   adjacent borderland could be indicative of PW convection (e.g., via brine rejection) and/or

540   persistent sediment redistribution from the Chukchi shelf. Overall, although we acknowledge that

541   direct observations are needed to validate the deep-water convection driven by brines in the

542   Chukchi-Alaskan margin, our results support the hypothesis that both boundary exchange and

543   brine rejection during sea ice formation have probably influenced the distribution of radiogenic

544   isotope (such as Nd and Hf) compositions in western Arctic seawater for the last 6 ka cal BP.

545   The authigenic εNd record obtained in core 01JPC allows a close look at the changes in

546   the relative contributions between the two main components of the BIS (Figures 6e and 7b,c): (1)

547   Alaskan Coastal Water characterized by more unradiogenic Nd isotope compositions (εNd ~ -8 to

548   -9), which reflect the dissolved Nd inputs from the Yukon drainage basin (e.g., VanLaningham et

549   al., 2009; Horikawa et al., 2010, 2015), and (2) Bering Sea Water, which has more radiogenic Nd

550   isotope compositions (εNd ~ -2 to -3) mainly derived from the western Bering Sea (i.e., coastal

551   water adjacent to the Anadyr region and Bering Shelf; Asahara et al., 2012; Jang et al., 2017).

552   Thus, the more radiogenic εNd values (~-2) found on the Chukchi shelf can be explained by a

553   high contribution of Bering Sea Water to the BSI and/or less influence of the Yukon River

554   discharge to the Alaskan Coastal waters (Figure 7c). In this context, and as previously discussed,

555   large-scale atmospheric climate modes, such as the PDO, provide a potential mechanistic

556   explanation for correlations between changes in the precipitation patterns over North America

557   and the position of the Aleutian Low in the North Pacific, as well as explaining changes in the

558   BSI (Anderson et al., 2016; Yamamoto et al., 2017). The winter precipitation patterns result from

559   the strength and position of the Aleutian Low, which is strengthened and/or located farther to the

560   east of the North Pacific during a PDO+ phase and weakened and/or located more to the west of

561   the North Pacific during a PDO- phase (Barron & Anderson, 2011). In addition, modern data

562   from the Yukon River suggest a positive trend in the annual flow during PDO+ phases, perhaps

563   reflecting the increases in annual precipitation in the interior of Alaska (Brabets and Walvoord,

564   2009).

565   Within this context, we hypothesize that the wetter conditions associated with the PDO+

566   state in conjunction with an enhanced boreal summer insolation during the middle Holocene

567   (Figure 6a,b; Anderson et al., 2016; Liu et al., 2014) may have promoted higher weathering rates

568   in the Yukon drainage basin, which is consistent with the high unradiogenic εNd values recorded

569   in core 01JPC (Figures 6e and 7b). Conversely, during the late Holocene, the drier negative PDO-

570   like conditions in western North America (Figure 6b), together with an long-term decrease in

571   boreal summer insolation (Figure 6a), likely reduced the weathering rates in the Yukon drainage

572   basin and therefore produced a relative increase in the contribution of dissolved and detrital loads

573   with more radiogenic values from the Bering Sea Water to the BSI (Figure 7c). Likewise, the

574   decrease in the Log(I+K/C+V) ratio and the relative increase in the proportions of the

575   northeastern Bering Sea sediments (SedUnMixMC results) and amorphous silica (proxy for the

576   BSI intensity; Jakobsson et al., 2017; Stein et al., 2017) in core 01JPC are also in agreement with

577   increases in the BSI during the late Holocene (Figure 6f-h).

578   The strength and distribution of BSI water between different branches (Figure 7b,c)

579   influences the sea surface temperatures and the spatial and temporal variability of the sea ice

580   cover in the Chukchi Sea (e.g., Shimada et al., 2006; McKay et al., 2008; Polyak et al., 2016;

581   Stein et al., 2017). For example, the results from transfer functions based on dinoflagellate cyst

582   assemblages from the nearby core 05JPC (McKay et al., 2008) suggest a decrease in the duration

583   of sea ice cover and an increase in summer sea surface temperatures after 4 ka cal BP relative to

584   the middle Holocene (between 6 and 4 ka cal BP) in the northeastern Chukchi Sea (Figure 6d). In

585   contrast, based on sea ice biomarker proxy records from two sediment cores from the

586   northwestern Chukchi Sea (ARA2B-1A) and East Siberian Sea (PS72/350-2), Stein et al. (2017)

587   suggested a significantly increased sea ice extent during the last 4.5 ka cal BP relative to the

588   middle Holocene (Figure 6c). These differences in the sea ice extent in the Chukchi Sea seem to

589   be related to changes in the circulation of the BSI across the Chukchi shelf (Figure 7b,c), which is

590   itself modulated by changes in the PDO phase (e.g., Screen and Francis, 2016). Based on

591   numerical models, Winsor and Chapman (2004) suggest that predominantly northeasterly to

592   easterly winds in the northernmost Pacific, usually dominant during PDO+ phases (Zhang et al.,

593   2015), produce an overall reduction in the BSI (induced by an enhanced Aleutian Low) and

594   induce a more northwestward direction to the BSI across the Chukchi shelf edge (Figure 7b).

595   These PDO-like ocean-atmosphere conditions were likely responsible for the sea ice reduction

596   observed in the northwestern Chukchi Sea and East Siberian Sea during the middle Holocene

597   (Stein et al., 2017). Conversely, sustained westerly winds from the northernmost Pacific, usually

598   dominant during PDO- (Zhang et al., 2015), produce an overall intensified BSI (induced by a

599   weakened Aleutian Low) and favor north-northeast diversion of the BSI through the Central

600   Channel and along the Chukchi-Alaskan coast (Figure 7c; Winsor and Chapman, 2004). These

601   atmospheric and oceanographic conditions probably promoted reduced sea ice cover and an

602   increase in sea surface temperatures in the northeastern Chukchi Sea during the last 4 ka cal BP

603   (McKay et al., 2008). Overall, this spatial and temporal variability in the sea ice cover in the

604   Chukchi Sea (Figure 6c,d) supports our interpretation of not only an increase in the relative

605   proportion of the Bering Sea Water component of the BSI (Figures 6e-h) but also major

606   northeastward diversion of the BSI in the Chukchi Sea during the late Holocene (Figure 7c).

607   Although we recognize that the links with PDO-like ocean-atmosphere interactions

608   warrant further study, evidence of enhanced PDO expression along the northeastern Pacific

609   margins during the late Holocene relative to the middle Holocene (Figure 6b; Barron &

610   Anderson, 2011) is a sufficient explanation of the long-term variability that we observe in our

611   paleoceanographic records. A study with a higher temporal (e.g., centennial to millennial scale)

612   resolution that couples quantitative mineralogy and radiogenic isotope data during this period

613   needs to be performed to provide a better understanding of the relationships among the

614   atmospheric climate mode (e.g., PDO), continental weathering and the BSI.

615   **6   Conclusion**

616   Using the combined Nd and Hf isotopic record from bulk sediment leachates in two piston

617   cores recovered from the Canadian Beaufort (02PC) and Chukchi-Alaskan (01JPC) margins, we

618   investigated changes in weathering regimes and deep-water circulation during the Holocene.

619   Overall, our mineralogical and Nd-Hf isotopic data, together with modeled Holocene Arctic river

620   discharges (Wagner et al., 2011), quantitative reconstructions of past sea surface conditions

621 (McKay et al., 2008; Stein et al., 2017), and hypothesized changes in atmospheric circulation

622 (Winsor and Chapman, 2004; Barron & Anderson, 2011), reveal the following:

623 (1) A clear Mackenzie River and PW isotopic signature at coring sites 02PC and 01JPC at

624 approximately 1000 m supports the hypothesis that brine rejection during sea ice formation

625 and/or persistent sediment redistribution from the shelf probably plays significant roles in

626 controlling the εNd and εHf values of the bottom waters of the Canadian Beaufort and Chukchi-

627 Alaskan margins.

628 (2) Since the last deglacial period, the Nd and Hf isotopic compositions of bulk sediment

629 leachates from the Beaufort Sea core transition from resembling those of the Mackenzie River

630 end member to resembling those of the AW end member. The Mackenzie River-like values are

631 linked to the remobilization of rock flour from glacially deformed terrain in the Mackenzie River

632 watershed and lower sea level conditions during the early Holocene. In the middle to late

633 Holocene, the shift towards AW-like isotopic values is inferred to be the result of a decrease in

634 the Mackenzie River discharge, likely associated with predominantly negative PDO-like

635 conditions.

636 (3) The Nd and Hf isotopic compositions of bulk sediment leachates in the Chukchi-Alaskan

637 margin were controlled by (1) more intense precipitation and weathering in the drainage basin of

638 the Yukon River during the middle Holocene and (2) drier conditions and an increase in the

639 relative contribution of the Bering Sea Water component to the BSI during the late Holocene.

640 This transition seems to have resulted from major changes in atmospheric climate modes induced

641 by a PDO/ENSO-like forcing.

642

643 **Acknowledgments**

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1032

1033 **Figure captions**

1034 **Figure 1**. (a) Schematic map of Atlantic water (AW), Pacific water (PW), Transpolar Drift (TPD)
1035 and Beaufort Gyre (BG) circulation in the Arctic Ocean and locations of cores 01JPC and 02PC
1036 (black circles). The PW drifts eastward in the Beaufort Sea and is known as the Alaskan Coastal
1037 Current (ACC). The εNd and εHf values are shown in the map. The Laurentide Ice Sheet at 11.5
1038 ka cal BP is also shown (Dyke, 2004). (b) East-west mean annual temperature profile across the
1039 Beaufort-Chukchi slope (transect 1-2 in gray). Core sites are marked by black circles.
1040 Temperature data are from Polar Science Center Hydrographic Climatology (PHC,
1041 http://psc.apl.washington.edu/Climatology.html).
1042

1043 **Figure 2**. REE patterns normalized to PAAS (Taylor & McClennan, 1985) for the bulk sediment
1044 leachate samples from (a) core 01JPC (Chukchi-Alaskan margin) and (b) core 02PC (Canadian
1045 Beaufort margin).

1046

1047 **Figure 3**. (a) $^{87}Sr/^{86}Sr$ isotope ratios for the bulk sediment leachate samples from cores 01JPC
1048 and 02PC; sea water values in the Arctic ocean are shown by the black lines (Asahara et al.,
1049 2012). (b) Cross-plot of HREE/LREE vs. MREE. A mixing line between the most
1050 MREE-enriched leachates and the most HREE-enriched oxic pore waters reflects the authigenic-
1051 pore water array (Gutjahr et al., 2010). Detrital REE composition of cores 02PC and 05JPC are
1052 from Deschamps (2018). (c) Spearman correlation between ΣREE contents and the authigenic
1053 εNd values from cores 01JPC and 02PC. (d) Hafnium–neodymium isotope systematics of the
1054 bulk sediment leachates obtained in this study together with previously published data and εNd-
1055 εHf correlation lines from the literature. Terrestrial and seawater arrays are from Vervoort et al.
1056 (1999) and Albarède et al. (1998), respectively. Pacific water, Mackenzie River and Arctic Sea
1057 water values are from Zimmermann et al. (2009a,b), and leachate and detrital values are from
1058 Chen et al. (2012).

1059

1060 **Figure 4**. Nd and Hf isotopic evolution of the Arctic deep-waters obtained from cores 02PC and
1061 01JPC. Horizontal blue and red lines illustrate modern values of the Bering Sea Water
1062 (Zimmermann et al., 2009a,b; Asahara et al., 2012; Jang et al., 2017), Atlantic water and
1063 Mackenzie River (Porcelli et al., 2009; Zimmermann et al., 2009a). The green arrows indicate the
1064 more unradiogenic Nd isotope compositions from the Alaskan Coastal Water (VanLaningham et
1065 al., 2009; Horikawa et al., 2010, 2015) and the geological terrains of the Canadian Arctic
1066 Archipelago (CAA; Maccali et al., 2018).

1067

1068 **Figure 5**. On the left: (a) Mean summer insolation at 70°N (Berger & Loutre, 1991); (b) Global
1069 sea level curve (RSL; Lambeck et al., 2014); (c) Authigenic εNd records of a sediment core from
1070 the Fram Strait (Maccali et al., 2013); (d) Holocene $\delta^{18}O$ records from Jellybean Lake used as a
1071 PDO index (Barron & Anderson, 2011); (e) Holocene Mackenzie River discharge based on
1072 numerical models (Wagner et al., 2011); (f) Dinocyst-based reconstructions of sea ice cover from
1073 the Canadian Beaufort Sea (Bringué and Rochon, 2012); (g) Proportion of sediment from the
1074 Mackenzie River (black) and the Canadian Arctic Archipelago (CAA; red) in core 02PC
1075 (Deschamps et al., 2018a); (h-i) Authigenic εNd and εHf evolution for core 02PC (this study). On
1076 the right: schematic illustrations depicting the changes in authigenic εNd and εHf variations in
1077 core 02PC between (j) 12 and 8 ka cal BP (light blue), (k) 8 and 4 ka cal BP (white), and (l) after
1078 4 cal ka BP (light red). Spirals indicate sediment resuspension on the Canadian Beaufort slope
1079 (Osborne and Forest, 2016).

1080

1081 **Figure 6**. On the left: (a) Mean summer insolation at 70°N (Berger & Loutre, 1991); (b)
1082 Holocene $\delta^{18}O$ records from Jellybean Lake used as a PDO index (Barron & Anderson, 2011); (c)
1083 sea ice proxy PIP25 (based on brassicasterol) from core ARA2B-1A (Stein et al., 2017); (d)
1084 Quantitative reconstruction of past sea surface temperature (red) and sea-ice cover (blue) in the
1085 core 05JPC based on dinoflagellate cyst assemblages (McKay et al., 2008); (e) Authigenic εNd
1086 evolution for core 01JPC (this study); (f) Proportion of northeastern Bering Sea sediments (this
1087 study); (g) Log(I+K/C+V) ratio in core 01JPC; (h) Amorphous silica content in core 01JPC (this

study). On the right: (i-j) schematic illustrations depicting the changes in authigenic εNd variations in core 01JPC before 4 cal ka BP (light blue) and after 4 ka cal BP (light red). Spirals indicate possible sediment resuspension on the Chukchi-Alaskan slope (Darby et al., 2009).

Figure 7. Generalized reconstructions showing the possible changes in the Pacific Water inflow into the western Arctic Ocean during the Holocene (modified from Grebmeier et al., 2006) inferred from the authigenic εNd records of cores 01JPC and 02PC. The Laurentide Ice Sheet position in (a) is about of 11.5 ka cal BP (Dyke, 2004). The Aleutian Low (the bold AL in b and c) patterns according to the PDO-like conditions are also show (Anderson et al., 2016). AR: Anadyr River, YR:Yukon River, MR: Mackenzie River, BG: Beaufort Gyre, AG: Amundsen Gulf.

Supplementary material

Figure S1. Fe/k_{LF} and Mn/Ti ratios for cores 01JPC and 02PC (data from Deschamps et al., 2018b). The mean Fe/kLF ratio in cores 01JPC and 02PC is <40 Mcps, suggesting weak reductive diagenesis (Funk, 2004; Hofmann et al., 2005; Hofmann & Fabian, 2009). Moreover, the vertical distribution of the Mn/Ti ratio shows little variability within the studied cores, with elevated ratios recorded towards the top of the cores. These elevated values demonstrate scavenging of Mn/Fe oxyhydroxides and associated trace metals from the water column (e.g., Macdonald and Gobeil, 2012; Meinhardt et al., 2016b).

Figure S2. εHf signature of core 01JPC. Horizontal blue and red lines illustrate modern values of the North Pacific water. Due to the large uncertainties, the εHf signature of core 01JPC cannot be used to observe Holocene changes in the εHf Pacific signature.

Table S1. REE and trace element concentrations (ppm) from bulk sediment leachates of cores 01JPC and 02PC.

Table S2. Nd-Hf-Sr isotopes from bulk sediment leachates of cores 01JPC and 02PC.

Table S3. Bulk minerals data from core 01JPC used in this study (all values given in %).

Table S4. Clay minerals data from core 01JPC used in this study (all values given in %). ND: not detected.

Table S5. SedUnMixMC results from core 01JPC. Source 1: Mackenzie River plume (Gamboa et al., 2017); Source 2: North Alaska (Darby et al., 2011); Source 3: Banks and Victoria Island (Gamboa et al., 2017; this study); Source 4: Bearing Strait (Stein et al., 2017); Source 5: East Siberian Sea & Laptev Sea (Darby et al., 2011); Source 6: Kara Sea (Andrews et al. 2016).
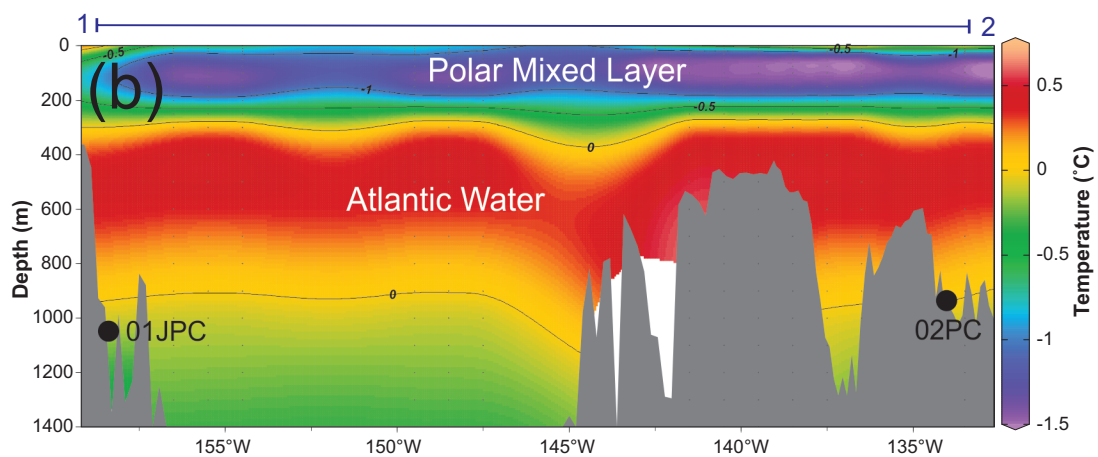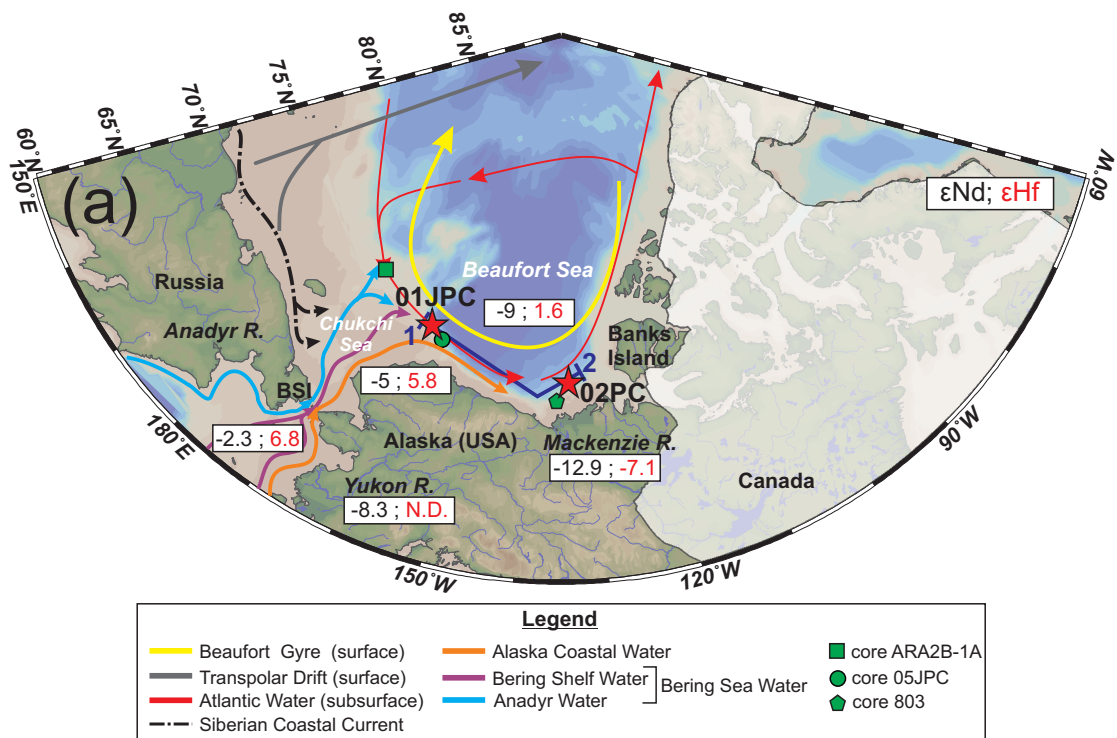
Figure 1.

(a)

εNd; εHf

Russia

Anadyr R.

Beaufort Sea

-9 ; 1.6

01JPC

Chukchi Sea

1

-5 ; 5.8

BSI

-2.3 ; 6.8

Banks Island

2

02PC

Alaska (USA)

Mackenzie R.

-12.9 ; -7.1

Canada

Yukon R.

-8.3 ; N.D.

Legend

— Beaufort Gyre (surface)	— Alaska Coastal Water	■ core ARA2B-1A
— Transpolar Drift (surface)	— Bering Shelf Water ⎤ Bering Sea Water	● core 05JPC
— Atlantic Water (subsurface)	— Anadyr Water ⎦	⬟ core 803
—·— Siberian Coastal Current		

(b)

Polar Mixed Layer

Atlantic Water

01JPC

02PC

Depth (m)

Temperature (°C)

Figure 2.

(a) 02PC
- ■ > 2 ka cal BP
- ■ < 2 ka cal BP

(b) 01JPC
- ■ > 2 ka cal BP
- ■ < 2 ka cal BP

Figure 3.

Figure 4.

(a) 01JPC

Bering Sea Water

Pacific Water
(Chukchi Sea)

Alaskan Coastal
Water

(b) 02PC

Atlantic Water (Canada Basin)

Mackenzie River

CAA

(c) 02PC

Atlantic Water (Canada Basin)

Mackenzie River

Age (cal ka BP)

Figure 5.

Figure panels (a)–(i) left column plotted against Age (ka cal BP); panels (j)–(l) right column schematic diagrams.

(a) June Insolation 70°N (W.m⁻²): 480, 500, 520, 540

(b) RSL (m): 0, -30, -60

(c) εNd (leachate): -9, -10, -11 — core MC16

(d) Z-score δ¹⁸O Jellybean Lake — ENSO-like forcing; West/weak AL (PDO-); East/strong AL (PDO+): 1.5, 0.0, -1.5

(e) Mackenzie River discharge (km³.yr⁻¹): 405, 370

(f) Sea-ice cover (months.year⁻¹): 10, 9, 8, 7 — core 803

(g) Canadian Arctic Archipelago: 0.1 0.3 0.5 0.7; Mackenzie River; Core 02PC

(h) εNd: -6, -8, -10, -12, -14, -16; Atlantic Water (Canada Basin); Mackenzie River; Canadian Arctic Archipelago

(i) εHf: 5, 0, -5, -10; Atlantic Water (Canada Basin); Mackenzie River; IRD

(j) 12-8 ka cal BP: (+)MR runoff; (+)Weathering; (-)Sea level; Polynia; Brine; IRD CAA; Atlantic inflow; Coring Site

(k) 8-4 ka cal BP (PDO+): (+)Precipitation; (+)MR runoff; (-)BSI; (+)Weathering; (+)Sea level; Polynia; Brine; Atlantic inflow; Coring Site

(l) 4-1 ka cal BP (PDO-): (-)Precipitation; (-)MR runoff; (+)BSI; (-)Weathering; (+)Sea level; Polynia; Brine; Atlantic inflow; Coring Site

Figure 6.

(a)

June Insolation 70°N (W.m^{-2})

(b) Jellybean Lake (Z-score δ^{18}O)

ENSO-like forcing

West/weak AL (PDO-)

East/strong AL (PDO+)

(c) core ARA2B-1A

(+)

(-)

PIP$_{25}$ (sea-ice cover)

(d) core 05JPC

Sea-ice > 50% months.a^{-1}

(-) sea ice (+)

Summer sea-surface temperature (°C)

core 01JPC

BSW

ACW

εNd

(e)

(f) Bering Strait source 01JC

BSI

(g)

Bering Strait

Mackenzie River

Log(I+K/C+V)clay 01JPC

(h) Amorphous silica (wt. %) 01JPC

BSW

Age (ka cal BP)

(i)

6-4 ka cal BP (PDO+)

(+)Precipitation

(+)YR runoff / (-)BSW

(+)Weathering

Polynia

Brine

(-)BSI

Atlantic inflow

Coring Site

(j)

4-1 ka cal BP (PDO-)

(-)Precipitation

(-)YR runoff / (+)BSW

(-)Weathering

Polynia

Brine

(+)BSI

Atlantic inflow

Coring Site

Figure 7.

(a) 12-10 ka cal BP

BG

εNd ?

εNd ~-16

BS opening
11ka cal BP

AR

YR

MR

AG

Aleutian Islands

(b) 8-4 ka cal BP (PDO+)

BG

εNd ~-6

εNd ~-11

(-)BSI

AR

Wet

MR

YR

AL

Aleutian Islands

(c) 4-1 ka cal BP (PDO-)

BG

εNd ~-4

εNd ~-9.5

(+)BSI

AR

Dry

MR

AL

YR

Aleutian Islands

Legend

────	Beaufort Gyre (surface)
────	Atlantic Water (subsurface)
─·─·─	Siberian Coastal Current
────	Alaska Coastal Water
────	Bering Shelf Water ⎤ Bering Sea Water
────	Anadyr Water ⎦