

## Surface nitrate utilization in the Bering sea since 180 ka BP: Insight from sedimentary nitrogen isotopes

Riethdorf Jan-Rainer <sup>1,2</sup>, Thibodeau Benoit <sup>2,3</sup>, Ikehara Minoru <sup>4</sup>, Nuernberg Dirk <sup>2</sup>, Max Lars <sup>5</sup>,  
Tiedemann Ralf <sup>5</sup>, Yokoyama Yusuke <sup>1,6,\*</sup>

<sup>1</sup> Univ Tokyo, Atmosphere & Ocean Res Inst, Dept Ocean Floor Geosci, 5-1-5 Kashiwanoha, Kashiwa, Chiba 2778564, Japan.

<sup>2</sup> Helmholtz Ctr Ocean Res Kiel GEOMAR, Wischhofstr 1-3, D-24148 Kiel, Germany.

<sup>3</sup> Univ Tokyo, Atmosphere & Ocean Res Inst, Dept Chem Oceanog, 5-1-5 Kashiwanoha, Kashiwa, Chiba 2778564, Japan.

<sup>4</sup> Kochi Univ, Ctr Adv Marine Core Res, B200 Monobe Nankoku, Kochi 7838502, Japan.

<sup>5</sup> Alfred Wegener Inst Polar & Marine Res, Handelshafen 12, D-27570 Bremerhaven, Germany.

<sup>6</sup> Japan Agcy Marine Earth Sci & Technol, Inst Biogeosci, 2-15 Natsushimacho, Yokosuka, Kanagawa 2370061, Japan.

\* Corresponding author : Yusuke Yokoyama, email address : [yokoyama@ori.u-tokyo.ac.jp](mailto:yokoyama@ori.u-tokyo.ac.jp)

[jan.riethdorf@gmail.com](mailto:jan.riethdorf@gmail.com) ; [bthibodeau@geomar.de](mailto:bthibodeau@geomar.de) ; [ikehara@kochi-u.ac.jp](mailto:ikehara@kochi-u.ac.jp) ; [dnuernberg@geomar.de](mailto:dnuernberg@geomar.de) ; [Lars.Max@awi.de](mailto:Lars.Max@awi.de) ; [Ralf.Tiedemann@awi.de](mailto:Ralf.Tiedemann@awi.de)

### Abstract :

We present high-resolution records of sedimentary nitrogen ( $\delta N-15(\text{bulk})$ ) and carbon isotope ratios ( $\delta C-13(\text{bulk})$ ) from piston core S0201-2-85KL located in the western Bering Sea. The records reflect changes in surface nitrate utilization and terrestrial organic matter contribution in submillennial resolution that span the last 180 kyr. The  $\delta N-15(\text{bulk})$  record is characterized by a minimum during the penultimate interglacial indicating low nitrate utilization (similar to 62-80%) despite the relatively high export production inferred from opal concentrations along with a significant reduction in the terrestrial organic matter fraction ( $m(\text{terr})$ ). This suggests that the consumption of the nitrate pool at our site was incomplete and even more reduced than today (similar to 84%).  $\delta N-15(\text{bulk})$  increases from Marine Isotope Stage (MIS) 5.4 and culminates during the Last Glacial Maximum, which indicates that nitrate utilization in the Bering Sea was raised during cold intervals (MIS 5.4, 5.2, 4) and almost complete during MIS 3 and 2 (similar to 93-100%). This is in agreement with previous hypotheses suggesting that stronger glacial stratification reduced the nutrient supply from the subeuphotic zone, thereby increasing the iron-to-nutrient ratio and therefore the nitrate utilization in the mixed surface layer. Large variations in  $\delta N-15(\text{bulk})$  were also recorded from 180 to 130 ka BP (MIS 6), indicating a potential link to insolation and sea-level forcing and its related feedbacks. Millennial-scale oscillations were observed in  $\delta N-15(\text{bulk})$  and  $\delta C-13(\text{bulk})$  that might be related to Greenland interstadials.

**Keywords :** Bering Sea, Quaternary, Nitrogen isotopes, Nitrate utilization

## 52 **1. Introduction**

53 The polar oceans are thought to have been more stratified during past glacial periods  
54 and the breakdown of stratification in the Southern Ocean during interglacials has  
55 been suggested as a potential control mechanism for the glacial-interglacial cycles in  
56 atmospheric carbon dioxide (CO<sub>2</sub>) (for a review see Sigman et al., 2010). Recent  
57 studies have found supporting evidence that past variations in stratification/ventilation  
58 also occurred in the subarctic North Pacific with implications for ocean-atmosphere

59 gas exchange (Jaccard et al., 2005, 2010; Brunelle et al., 2007, 2010; Galbraith et  
60 al., 2008a; Okazaki et al., 2010, 2012; Chikamoto et al., 2012; Menviel et al., 2012;  
61 Rella et al., 2012; Jaccard and Galbraith, 2013; Max et al., 2014).

62 The modern subarctic North Pacific is characterized by a permanent halocline due to  
63 a low-salinity surface layer that limits the exchange of nutrients between the surface  
64 and subsurface and prevents the formation of deep water masses (e.g., Warren,  
65 1983; Haug et al., 1999; Emile-Geay et al., 2003). At the same time high marine  
66 productivity makes this area a net sink for atmospheric CO<sub>2</sub> (Honda et al., 2002;  
67 Takahashi et al., 2002b). However, the efficiency of the biological pump in the high-  
68 nutrient, low-chlorophyll (HNLC) regions of the subarctic North Pacific is reduced due  
69 to iron limitation (e.g., Tsuda et al., 2003), which results in incomplete nitrate  
70 utilization.

71 Sedimentary records from the North Pacific and its marginal seas consistently show  
72 reduced contents of biogenic opal and barium, CaCO<sub>3</sub>, and organic carbon during  
73 past glacial periods, indicating reduced biological export production (e.g.,  
74 Gorbarenko et al., 2002; Narita et al., 2002; Kienast et al., 2004; Nürnberg and  
75 Tiedemann, 2004; Jaccard et al., 2005, 2010; Okazaki et al., 2005a; Shigemitsu et  
76 al., 2007, 2008; Riethdorf et al., 2013a). Restricted marine productivity both in the  
77 North Pacific and in the Antarctic sector of the Southern Ocean is attributed to (i) light  
78 limitation due to extensive sea-ice coverage (Elderfield and Rickaby, 2000), or (ii) to  
79 enhanced stratification of the upper water column that suppressed nutrient supply to  
80 the euphotic zone (Francois et al., 1997).

81 Studies investigating these hypotheses applied stable nitrogen isotope ratios ( $\delta^{15}\text{N}$ )  
82 to provide a link to the marine nutrient cycle, but they have been mostly focused on  
83 regions not influenced by seasonal sea-ice, which has the potential to modulate  
84 biological and terrigenous fluxes. In the NW Pacific the  $\delta^{15}\text{N}$  signal can be used as a  
85 proxy of surface nitrate utilization, whereas in the NE Pacific, it reflects variations in  
86 the composition of the subsurface nitrate pool (Brunelle et al., 2007; Galbraith et al.,  
87 2008a). The available reconstructions of surface nitrate utilization in the Okhotsk and  
88 Bering seas indicate that in both marginal seas enhanced stratification during glacial  
89 intervals resulted in a reduced supply of nitrate to the surface and a more complete

90 utilization of surface nitrate because of the continued iron supply from atmospheric  
91 deposition (i.e., higher iron-to-nitrate ratio), thereby explaining the low glacial  
92 productivity (e.g., Brunelle et al., 2007, 2010; Kim et al., 2011; Khim et al., 2012).

93 For the Bering Sea, recent studies have found indications for millennial-scale  
94 oscillations in export production and terrigenous matter supply, which might be  
95 connected to changes in stratification and/or sea-ice influence (Gorbarenko et al.,  
96 2005, 2010b; Kim et al., 2011; Riethdorf et al., 2013a; Schlung et al., 2013).  
97 However, most of the available records of  $\delta^{15}\text{N}$  are located in the southern Bering  
98 Sea and therefore not necessarily influenced by seasonal sea-ice. Moreover, the  
99 records are restricted to ~120 ka BP and do not allow for a comparison between the  
100 glacial terminations, which due to the large amplitude of climate and environmental  
101 changes are considered important periods for the understanding of the carbon cycle  
102 (e.g., Yokoyama and Esat, 2011). Especially for Marine Isotope Stage 6 (MIS 6),  
103 which in the Okhotsk Sea environment is characterized by rather extreme glacial ice  
104 conditions with significantly increased accumulation rates of ice-rafted debris (IRD)  
105 (Nürnberg et al., 2011), no records of  $\delta^{15}\text{N}$  are available.

106 Here, we present isotope geochemical records from a supposedly sea-ice influenced  
107 site in the poorly studied western Bering Sea for the last 180 kyr in high-resolution  
108 employing sedimentary carbon and nitrogen isotope ratios to reconstruct changes in  
109 the contribution of terrestrial organic matter and surface nitrate utilization,  
110 respectively. Our results, for the first time, provide information on nitrate utilization in  
111 the Bering Sea beyond 120 ka BP and expand the hypothesis of glacial-interglacial  
112 stratification changes to cold and warm intervals. Moreover, our record suggests that  
113 millennial-scale climate oscillations occurred in the Bering Sea which might be  
114 connected to Greenland interstadials.

115

## 116 **2. Study area**

117 The Bering and Okhotsk seas are marginal seas of the North Pacific, separated from  
118 by the Aleutian and Kurile islands, respectively. They are bounded by the coasts of  
119 eastern Siberia, the Kamchatka Peninsula and/or western Alaska. Wide and shallow

120 continental shelf areas are found in the northern Okhotsk and in the northern and  
121 eastern Bering Sea (Figure 1).

122 With respect to surface circulation, waters from the North Pacific are transported  
123 westward along the Aleutian islands by the Alaskan Stream and enter the Bering Sea  
124 via the Aleutian passes. There, the Bering Slope Current (BSC) and the East  
125 Kamchatka Current (EKC) form boundary currents (Stabeno et al., 1999). Surface  
126 outflow is directed into the Arctic Ocean through the shallow (~50 m) Bering Strait,  
127 whereas surface and deeper waters are transported back into the NW Pacific through  
128 the deeper straits, mainly Kamchatka Strait (Figure 1). The EKC and the Oyashio  
129 current flow southward and represent western boundary currents of the North Pacific  
130 subpolar gyre. The Kurile straits provide entrance and exit pathways to the Okhotsk  
131 Sea.

132 Major climatic and oceanographic characteristics of the North Pacific realm are the  
133 strong seasonality in sea surface temperatures (SST) and sea-ice formation, the  
134 permanent halocline, and a pronounced oxygen minimum zone (OMZ). In the Bering  
135 Sea sea-ice is present from September until July reaching its maximum distribution  
136 during March/April (Tomczak and Godfrey, 1994; Niebauer et al., 1999). The sea-ice  
137 formation is related to the interaction of the Siberian High and the Aleutian Low,  
138 which results in the advection of cold Arctic air masses, subsequent cooling of the  
139 sea surface, and strong winter mixing (Stabeno et al., 1999). Sea-ice is considered  
140 as an important transport agent of terrigenous matter in the Okhotsk and Bering seas  
141 (Nürnberg and Tiedemann, 2004; Nürnberg et al., 2011; Riethdorf et al., 2013a).  
142 Geochemical results indicate that sediments on the eastern Bering Sea shelf and in  
143 the Meiji Drift in the NW Pacific are supplied from Yukon–Bering Sea sources  
144 (VanLaningham et al., 2009; Asahara et al., 2012; Nagashima et al., 2012). Based on  
145 these results Riethdorf et al. (2013a) proposed that terrigenous matter entrained into  
146 sea-ice by tidal pumping, suspension freezing, and beach-ice formation, was  
147 transported from the eastern Bering Sea shelf to the location studied in this paper,  
148 although a contribution by suspension load carried by the BSC could not be  
149 excluded.

150 Although sea-ice formation and according brine rejection in the northern Okhotsk Sea  
151 drive the modern ventilation of North Pacific Intermediate Water (NPIW) (e.g.,  
152 Yasuda, 1997; Yamamoto et al., 2001), the source of NPIW might have shifted to the  
153 Bering Sea in the past (Matsumoto et al., 2002; Ohkushi et al., 2003; Tanaka and  
154 Takahashi, 2005; Rella et al., 2012), where it nowadays resides at the depth of the  
155 26.8 potential density ( $\sigma_\theta$ ) surface in ~200-400 m (Roden, 1995; Macdonald et al.,  
156 2001). The OMZ is found beneath the NPIW with minimum dissolved oxygen  
157 concentrations of ~15-20  $\mu\text{mol kg}^{-1}$  at ~900-1100 m (Roden, 2000; Lehmann et al.,  
158 2005).

159 In the Bering Sea, high marine productivity is observed, which is mainly associated  
160 with shelf areas (e.g., Springer et al., 1996; Stabeno et al., 1999) and dominated by  
161 diatoms. Major biological fluxes occur during spring/summer (mainly diatoms) and  
162 late summer/early fall (coccolithophores and planktonic foraminifera) (Takahashi et  
163 al., 2002a). Nutrients are consumed during the productive seasons and returned from  
164 the subsurface by winter mixing. Although winter mixing supplies nutrients from the  
165 subsurface into the euphotic zone, near-surface nutrients are not completely  
166 consumed by phytoplankton during the productive seasons. Therefore, the western  
167 Bering Sea studied here, as well as the central eastern and western parts of the  
168 subarctic North Pacific are HNLC regions with perennially high surface nitrate  
169 concentrations (e.g., Tyrrell et al., 2005) (Figure 1). As extensively discussed in  
170 Brunelle et al. (2007, 2010) changes in the extent of surface nitrate utilization can be  
171 reconstructed using records of  $\delta^{15}\text{N}$ , if the underlying assumptions include a constant  
172 isotope effect for nitrate assimilation and little or no changes in the  $\delta^{15}\text{N}$  of the source  
173 nitrate. There is evidence for the northward propagation of  $^{15}\text{N}$ -enriched nitrate from  
174 the eastern tropical North Pacific along coastal North America (Liu and Kaplan, 1989;  
175 Altabet et al., 1999; Kienast et al., 2002; Sigman et al., 2003). Hence,  
176 paleoceanographic interpretations of sedimentary  $\delta^{15}\text{N}$  have to consider changes in  
177 the  $\delta^{15}\text{N}$  of the subsurface nitrate pool.

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180

### 181 3. Material and methods

182 This study is based on 18.13 m-long piston core SO201-2-85KL (referred to as 85KL  
183 hereafter) recovered during R/V Sonne expedition SO201 KALMAR Leg 2 in 2009  
184 (Dullo et al., 2009) from Shirshov Ridge, western Bering Sea (57°30.30'N,  
185 170°24.77'E, 968 m water depth; Figure 1). Sediments from this core mainly consist  
186 of terrigenous siliciclastic material bound to the clay- and silt-fractions, but layers of  
187 diatomaceous ooze are repeatedly intercalated. Carbonate preservation is poor and  
188 at best sporadic, and no sediments younger than 7.5 ka BP were recovered.

189

#### 190 3.1 Bulk sedimentary analyses (TOC, TN, $\delta^{13}\text{C}_{\text{bulk}}$ , $\delta^{15}\text{N}_{\text{bulk}}$ )

191 Total organic carbon (TOC) and total nitrogen (TN) concentrations, as well as  
192 sedimentary stable carbon ( $\delta^{13}\text{C}_{\text{bulk}}$ ) and nitrogen ( $\delta^{15}\text{N}_{\text{bulk}}$ ) isotope ratios were  
193 determined downcore (>9.1 ka BP) every 5 cm from a total of 357 bulk sediment  
194 samples. About 25 mg of freeze-dried, hand-ground (agate mortar) sediment was  
195 weighed into Ag capsules, acidified with 100  $\mu\text{l}$  hydrochloric acid (3N) to remove  
196 inorganic carbon, and dried in a desiccator filled with phosphorus pentoxide and  
197 sodium hydroxide. To ensure complete combustion, the Ag capsules were  
198 subsequently wrapped into Sn capsules. TOC, TN,  $\delta^{13}\text{C}_{\text{bulk}}$ , and  $\delta^{15}\text{N}_{\text{bulk}}$  were  
199 determined at the Center for Advanced Marine Core Research, Kochi University,  
200 using a Flash EA 1112 Series elemental analyzer (EA; Thermo Fisher Scientific,  
201 USA) coupled with a Delta Plus Advantage isotope ratio mass spectrometer (IRMS;  
202 Thermo Fisher Scientific, USA) via a Conflo III interface (He carrier). Stable isotope  
203 results are reported in conventional  $\delta$ -notation and referenced to the Vienna PeeDee  
204 Belemnite (VPDB) standard and to atmospheric nitrogen. Molar TN/TOC ratios were  
205 corrected for inorganic nitrogen compounds based on a linear regression between  
206 TOC and TN following Goñi et al. (1998) (referred to as molar N/C ratios hereafter;  
207 Figure 2). Analytical precision ( $1\sigma$ ) was determined from two different standards (L-  
208 Alanine,  $n = 58$ ; Sulfanilamide,  $n = 60$ ) and was <2% RSD (relative standard  
209 deviation) for TOC, <5% RSD for TN,  $\pm 0.01$  for molar N/C ratio,  $\pm 0.14\text{‰}$  for  $\delta^{13}\text{C}_{\text{bulk}}$ ,  
210 and  $\pm 0.29\text{‰}$  for  $\delta^{15}\text{N}_{\text{bulk}}$ . Reproducibility of the samples, determined from replicates  
211 ( $1\sigma$ ,  $n = 14$ ), was  $\pm 0.01$  wt.% for TOC and TN,  $\pm 0.01$  for molar N/C ratio,  $\pm 0.11\text{‰}$  for

212  $\delta^{13}\text{C}_{\text{bulk}}$ , and  $\pm 0.86\text{‰}$  for  $\delta^{15}\text{N}_{\text{bulk}}$ . This rather high value for  $\delta^{15}\text{N}_{\text{bulk}}$ , being significantly  
213 higher than instrumental precision, might be related to sample inhomogeneity and  
214 potential alteration of the sedimentary organic matter during pre-analysis acid  
215 treatment (Brodie et al., 2011a, 2011b).

216

### 217 **3.2 Age model**

218 The stratigraphic framework of 85KL is described in detail in Max et al. (2012) and  
219 Riethdorf et al. (2013a). Briefly, X-ray fluorescence (XRF) and spectrophotometric  
220 (color  $b^*$ ) core logging data were correlated to the  $\delta^{18}\text{O}$  records of the NGRIP ice  
221 core (NGRIP members, 2004; GICC05 timescale, Rasmussen et al., 2006) and the  
222 Sanbao stalagmites (Wang et al., 2008). This approach was validated by benthic  
223  $\delta^{18}\text{O}$  stratigraphy, magnetostratigraphy, accelerator mass spectrometry (AMS)  
224 radiocarbon dating of planktonic foraminifera, and intercore correlations to  
225 neighbouring sediment cores. Linear sedimentation rates vary between 4 and 23 cm  
226  $\text{kyr}^{-1}$  (average of  $\sim 12 \text{ cm kyr}^{-1}$ ), which translates into a submillennial time-resolution  
227 for our reconstructions.

228

### 229 **3.3 Existing data**

230 For comparison we used logging data and geochemical results reflecting changes in  
231 export production and terrigenous matter supply already available for core 85KL.  
232 Method details are given elsewhere (Max et al., 2012; Riethdorf et al., 2013a,  
233 2013b). In summary, light and color reflectance were measured directly after core  
234 recovery every 1 cm using a Minolta CM 508d hand-held spectrophotometer and  
235 converted into *Commission Internationale de l'Éclairage* (CIE)  $L^*$ ,  $a^*$ , and  $b^*$  color  
236 space. XRF scanning for elements Al through to Ba was performed at 1 cm sampling  
237 resolution using the Avaatech XRF core scanner at Alfred Wegener Institute for Polar  
238 and Marine Research, Bremerhaven. Molybdate-blue spectrophotometry was used to  
239 determine biogenic opal concentrations (after Müller and Schneider, 1993), and  
240 concentrations of  $\text{CaCO}_3$  were calculated from the difference of total carbon (TC) and  
241 TOC previously determined using a Carlo Erba CNS analyzer (model NA-1500) at



242 GEOMAR, Kiel. The relative amount of siliciclastics was calculated by subtracting the  
243 sum of  $\text{CaCO}_3$ , TOC, and opal concentrations from a total of 100 wt.%. Records of  
244 XRF Ca/Ti log-ratios, XRF Br count rates (in counts per second, cps), and color  $b^*$   
245 correlated with  $\text{CaCO}_3$  ( $R^2 = 0.65$ ), TOC ( $R^2 = 0.64$ ), and opal ( $R^2 = 0.61$ ),  
246 respectively. This finding is in agreement with other studies linking biogenic  $\text{CaCO}_3$   
247 with normalized XRF records of Ca (Jaccard et al., 2005), TOC with biophilic Br  
248 (Ziegler et al., 2008), and opal and organic matter content with color  $b^*$  (Debret et al.,  
249 2006).

250

### 251 **3.4 Reconstruction of export production**

252 The use of  $\text{CaCO}_3$ , TOC, and opal to reconstruct changes in export production is  
253 subject to specific restrictions.  $\text{CaCO}_3$ , especially in the North Pacific, is influenced  
254 by carbonate dissolution and might be more indicative of changes in the bottom  
255 water calcite saturation state (e.g. Jaccard et al., 2005). With respect to TOC, it is  
256 necessary to discriminate between marine and terrestrial carbon sources. The most  
257 often used proxy for reconstructions of paleo-export production in the North Pacific  
258 realm is biogenic opal (e.g. Kienast et al., 2004).

259 In paleoceanography, fluxes are usually reconstructed using accumulation rates  
260 rather than proxy concentrations. However, for the North Pacific and Bering Sea  
261 several studies provide evidence for the similar evolution of concentration and  
262 accumulation records of biogenic components (e.g. Crusius et al., 2004; Brunelle et  
263 al., 2007, 2010). Here, we used dry bulk density measurements (Riethdorf et al.,  
264 2013a) to calculate bulk mass accumulation rates (AR Bulk, in  $\text{g cm}^{-2} \text{ kyr}^{-1}$ ), as well  
265 as proxy accumulation rates for  $\text{CaCO}_3$ , TOC, opal, and siliciclastics. The result is  
266 shown in Figure 3, clearly demonstrating that sedimentation at Site 85KL is  
267 dominated by siliciclastic input. Overall, concentrations of the biogenic components  
268 are low, but opal concentrations and opal accumulation rates show a positive linear  
269 relationship ( $R^2 = 0.49$ ). We therefore assume in this paper that at Site 85KL  
270 concentrations of opal and color  $b^*$  logging data are related to export production.

271

272

## 273 4. Results

### 274 4.1 Export production and terrigenous matter supply

275 In general, concentrations of  $\text{CaCO}_3$ , TOC, and opal, as well as their approximating  
276 logging data show increased values during warm intervals (MIS 5.5, 5.3, 5.1, and 1)  
277 and Greenland interstadials (GI), but low values during cold intervals (MIS 6, 5.4, 5.2,  
278 4, and 2) and Greenland stadials (GS) (Figure 4; Riethdorf et al., 2013a). Because of  
279 sedimentary dilution, the proxy records reflecting terrigenous matter supply  
280 (%Siliciclastics, XRF data of Al) have an inversed shape with respect to the records  
281 reflecting export production. Our EA-IRMS-based TOC results are in excellent  
282 agreement with those of Riethdorf et al. (2013a) and in higher temporal resolution  
283 extend the respective record by ~30 kyr into MIS 6. The temporal evolution of TOC  
284 recorded during MIS 6 strongly corresponds to that observed in color  $b^*$ , and, to a  
285 lesser degree, in XRF Ca/Ti log-ratios and Br count rates (Figure 4). With respect to  
286 TOC concentrations, MIS 6 is characterized by a strong variability within the range of  
287 ~0.4 to ~1.7 wt.%, showing several short-lived oscillations and highest  
288 concentrations during ~156-137 ka BP.

289

### 290 4.2 $\delta^{13}\text{C}_{\text{bulk}}$ , $\delta^{15}\text{N}_{\text{bulk}}$ , molar N/C ratios, and estimation of the terrestrial organic 291 matter fraction ( $m_{\text{terr}}$ )

292 Values for  $\delta^{13}\text{C}_{\text{bulk}}$  (of TOC) and for  $\delta^{15}\text{N}_{\text{bulk}}$  (of TN) ranged from -25.4 to -21.9‰ and  
293 from 1.7 to 7.5‰, respectively, and molar N/C ratios varied between 0.04 and 0.11  
294 (Table 1; Figures 5 and 6). In general,  $\delta^{13}\text{C}_{\text{bulk}}$  and molar N/C ratios are more positive  
295 during warm intervals (MIS 5.5, 5.3, 5.1, 3, and 1) and show pronounced, but short-  
296 lived maxima during GI, which especially for  $\delta^{13}\text{C}_{\text{bulk}}$  exceed analytical precision and  
297 reproducibility. The cold intervals (MIS 5.4, 5.2, and 4) and some GS are  
298 characterized by decreases in both proxies. A different temporal evolution is  
299 recorded for  $\delta^{15}\text{N}_{\text{bulk}}$ . The base of core 85KL shows  $\delta^{15}\text{N}_{\text{bulk}}$  values of ~5-6‰, which  
300 is followed by a sharp decrease at ~172 ka BP to values of ~2-3‰ (Figure 6).  
301 Subsequently,  $\delta^{15}\text{N}_{\text{bulk}}$  again increases until another sudden drop of ~3‰  
302 characterizes the transition from MIS 6 to 5.5 (Termination II). During the last

303 interglacial  $\delta^{15}\text{N}_{\text{bulk}}$  remained low at  $\sim 3\text{‰}$  but it decreased even further to minimum  
304 values of  $1.7\text{‰}$  at the beginning of cold MIS 5.4. After MIS 5.4, a long-term trend  
305 toward higher values that continues into the early Holocene is observed. In between,  
306 increasing values were recorded during cold MIS 5.2 and 4, whereas  $\delta^{15}\text{N}_{\text{bulk}}$   
307 decreased (MIS 5.1) or remained almost constant (MIS 5.3 and 3) during warm  
308 stages. The transition from MIS 2 to 1 (Termination I) is characterized by local  
309 minima ( $\sim 4\text{-}5\text{‰}$ ) during the cold phases of Heinrich Stadial 1 (HS1; 18.0-14.7 ka BP,  
310 Sarnthein et al., 2001) and the Younger Dryas (YD; 12.9-11.7 ka BP, Blockley et al.,  
311 2012), and by an intercalated pronounced maximum during the Bølling-Allerød warm  
312 phase (B/A; 14.7-12.9 ka BP, Blockley et al., 2012) (up to  $6.8\text{‰}$ ). Subsequent to the  
313 YD,  $\delta^{15}\text{N}_{\text{bulk}}$  continuously increased until highest values of  $7.5\text{‰}$  were recorded  
314 during the early Holocene ( $\sim 10.1$  ka BP). Notably, the  $\delta^{15}\text{N}_{\text{bulk}}$  record also features  
315 short-lived maxima during GI, which, however, must be considered insignificant with  
316 respect to reproducibility.

317 Assuming that the geochemical and isotopic sedimentary composition represents a  
318 mixture of marine and terrestrial organic matter we applied a linear mixing model to  
319 estimate the fraction of terrestrial-derived organic matter ( $m_{\text{terr}}$ ) using hypothetical  
320 endmember compositions. We therefore followed the results of Walinsky et al.  
321 (2009). This approach has recently been applied to a sediment core from the  
322 northern Gulf of Alaska (Addison et al., 2012). Our Holocene samples lie within the  
323 ranges reported by Smith et al. (2002) for surface sediment samples from the  
324 southeastern Bering Sea shelf, whereas our glacial samples compare to terrigenous  
325 particulate organic matter (POM) from the Yukon River (Guo and Macdonald, 2006)  
326 (Figure 5). Accordingly, we assume that the most likely organic matter sources for  
327 our site are marine phytoplankton ( $\delta^{13}\text{C}$ :  $-22$  to  $-20\text{‰}$ ;  $\delta^{15}\text{N}$ :  $>5\text{‰}$ ; molar N/C ratio:  
328  $0.10$  to  $0.15$ ), soil ( $\delta^{13}\text{C}$ :  $-26.5$  to  $-25.5\text{‰}$ ;  $\delta^{15}\text{N}$ :  $0$  to  $1\text{‰}$ ; molar N/C ratio:  $0.08$  to  
329  $0.10$ ), and vascular plant detritus (VPD;  $\delta^{13}\text{C}$ :  $-27$  to  $-25\text{‰}$ ;  $\delta^{15}\text{N}$ :  $0$  to  $1\text{‰}$ ; molar N/C  
330 ratio:  $0$  to  $0.05$ ) (Meyers, 1994; McQuoid et al., 2001; Geider and La Roche, 2002;  
331 Smith et al., 2002; Guo et al., 2004; Gaye-Haake et al., 2005; Guo and Macdonald,  
332 2006; Walsh et al., 2008, and references therein) (Figure 5; Table 2). With the  
333 influence of soil considered insignificant,  $m_{\text{terr}}$  was calculated as:

$$m_{\text{terr}} = (A_{\text{sample}} - A_{\text{mar}}) / (A_{\text{terr}} - A_{\text{mar}}) \quad (\text{Eq. 1})$$

334  
 335 In Equation 1  $A$  refers to the  $\delta^{13}\text{C}_{\text{bulk}}$  or molar N/C ratio of the sample and the  
 336 respective average values for the assumed marine and terrestrial endmember  
 337 composition summarized in Table 2. We preferred using molar N/C over C/N ratios,  
 338 because mixing lines based on C/N are reported to underestimate the fraction of  
 339 terrestrially derived organic carbon (Perdue and Koprivnjak, 2007). This approach  
 340 resulted in similar values but in part different temporal evolutions of the respective  
 341  $m_{\text{terr}}$  records, which is attributed to the low variability in molar N/C ratios.  $m_{\text{terr}}$  varied  
 342 between ~10% and ~90% with average values of ~40-50% (Table 1). It was lowest  
 343 during warm intervals (MIS 5.5, 5.3, 5.1, 3, and 1) and GI, when marine productivity  
 344 was high. Reductions of up to 50% occurred during the transitions from cold to warm  
 345 intervals, whereas pronounced but short-lived increases of ~20% seem to  
 346 correspond to GS (Figure 6). In the discussion we refer to  $m_{\text{terr}}$  derived from  $\delta^{13}\text{C}_{\text{bulk}}$ .

347

## 348 5. Discussion

### 349 5.1 The $\delta^{15}\text{N}_{\text{bulk}}$ signal and potential alteration

350  $\delta^{15}\text{N}_{\text{bulk}}$  values reflect the isotopic signature of the export flux of organic matter plus  
 351 any secondary alteration of this signal during sinking and burial (e.g., Galbraith et al.,  
 352 2008b; Robinson et al., 2012, and references therein). Hence, interpretation of  
 353 variations in the  $\delta^{15}\text{N}_{\text{bulk}}$  record must consider changes in (i) the isotopic composition  
 354 of the subsurface nitrate pool, which is controlled by nitrogen fixation by diazotrophic  
 355 bacteria and by denitrification, (ii) the degree of nitrification and surface nitrate  
 356 utilization, and (iii) secondary alteration.

357 Nitrate is the primary nitrogen source for marine phytoplankton, which preferentially  
 358 incorporates isotopically light ( $^{14}\text{N}$ -enriched) nitrate (Pennock et al., 1996; Waser et  
 359 al., 1998). In the Bering Sea the source nitrate is supplied to the surface from below  
 360 the euphotic zone with a modern value ( $\delta^{15}\text{N}_{\text{nitrate}}$ ) of ~5.5‰ (Lehmann et al., 2005),  
 361 which is slightly higher than the global deep ocean average of ~5‰ because of  
 362 denitrification in the North Pacific (Sigman et al., 2000, Brunelle et al., 2007).  
 363 Nitrogen fixation results in  $\delta^{15}\text{N}_{\text{nitrate}}$  values that are isotopically light and close to that

364 of air (0‰; Carpenter et al., 1997), and it is the main reason for the low  $\delta^{15}\text{N}$  values  
365 of nitrate and sinking detritus in the tropical/subtropical ocean basins (e.g., Some et  
366 al., 2010). Water column denitrification occurs under low dissolved oxygen  
367 concentrations ( $<5 \mu\text{mol l}^{-1}$ ; Codispoti et al., 2001) and results in a  $^{15}\text{N}$ -enriched  
368 nitrate pool (Barford et al., 1999). In this respect,  $\delta^{15}\text{N}$  might also reflect redox  
369 conditions in the past with higher values during bottom water suboxia (Galbraith et  
370 al., 2004; Kashiyama et al., 2008; Jaccard and Galbraith, 2012; Robinson et al.,  
371 2012). Today, water column denitrification is mainly observed in the Arabian Sea, the  
372 eastern tropical North Pacific, and in the eastern tropical South Pacific. Thus, the  
373 export of  $^{15}\text{N}$ -enriched waters might result in a shift toward higher  $\delta^{15}\text{N}_{\text{nitrate}}$ , as  
374 observed along coastal North America in the subarctic NE Pacific (Liu and Kaplan,  
375 1989; Altabet et al., 1999; Kienast et al., 2002; Sigman et al., 2003). However,  
376 modern dissolved oxygen concentrations at Site 85KL lie above the denitrification  
377 threshold.

378 Results from benthic foraminiferal assemblages from the same site (Ovsepyan et al.,  
379 2013) suggest oxidizing conditions in the surface sediment layer from MIS 3 to the  
380 Last Glacial Maximum (LGM), but oxygen-depleted conditions during the mid-B/A  
381 and early Holocene. This is in agreement with Kim et al. (2011) who for Site PC23A  
382 reported on a dominance of oxic benthic foraminiferal species during MIS 2 and 3,  
383 but on dominantly dysoxic species during the B/A, early Holocene, and GI. These  
384 observations and our proxy records for export production indicate the presence of  
385 mostly oxic bottom waters and strongly reduced export of organic matter during most  
386 of the past 180 kyr, arguing against a significant impact of water column  
387 denitrification on  $\delta^{15}\text{N}_{\text{bulk}}$ . Over the last 180 kyr, one can expect a slightly higher  $\delta^{15}\text{N}$   
388 of nitrate during warm stages because of the greater extent of denitrification in the  
389 North Pacific. The isotopic impact of such increased denitrification during warm  
390 stages is thought to be relatively equal to the one observed today ( $\sim 0.5\%$ ; Lehmann  
391 et al., 2005). This shift toward heavier  $\delta^{15}\text{N}$  is opposite to the expected isotopic effect  
392 of decreased nitrate utilization during interglacials. Thus, the relatively small increase  
393 in  $\delta^{15}\text{N}_{\text{bulk}}$  during warm stages ( $\sim 0.5\%$ ) should not mask the larger isotopic variation  
394 expected from the change in nitrate utilization (Brunelle et al., 2007, and references  
395 therein). Thus, we are confident that the observed variations in our  $\delta^{15}\text{N}_{\text{bulk}}$  can be

396 used to assess relative changes in the utilization of nitrate in the Bering Sea surface  
 397 water, except for periods where dysoxia were locally present. During phases of  
 398 locally enhanced export production and oxygen-depleted bottom water conditions, as  
 399 recorded during the B/A and the early Holocene, as well as during GI, denitrification  
 400 might have resulted in a shift toward heavier  $\delta^{15}\text{N}_{\text{bulk}}$  values. Because we can not  
 401 isolate this signal in our record, our nitrate utilization estimates for these periods  
 402 might not be accurate due to potential changes in the  $\delta^{15}\text{N}$  signature of the source  
 403 nitrate

404 Nitrate utilization is incomplete in the modern subarctic North Pacific and in the  
 405 Bering Sea. Accordingly, the isotopic value of the export flux of organic matter  
 406 ( $\delta^{15}\text{N}_{\text{export}}$ ) is lighter than that of  $\delta^{15}\text{N}_{\text{nitrate}}$  (Altabet and Francois, 1994; Sigman et al.,  
 407 1999; Needoba et al., 2003; Galbraith et al., 2008a). The difference between  
 408  $\delta^{15}\text{N}_{\text{nitrate}}$  and  $\delta^{15}\text{N}_{\text{export}}$  is primarily controlled by the nitrate utilization and decreases  
 409 as it becomes more complete and in most of the global ocean the difference is zero  
 410 due to almost complete utilization (Altabet et al., 1999; Thunell et al., 2004). We can  
 411 calculate  $\delta^{15}\text{N}_{\text{export}}$  for the expected integrated organic nitrogen export at Site 85KL  
 412 assuming Rayleigh fractionation kinetics (Altabet and Francois, 1994; Mariotti et al.,  
 413 1981) after:

$$414 \quad \delta^{15}\text{N}_{\text{export}} = \delta^{15}\text{N}_{\text{nitrate}} + f / (1 - f) \varepsilon \ln (f) \quad (\text{Eq. 2})$$

415 In Equation 2  $f$  is the fraction of unutilized nitrate (i.e.,  $[\text{NO}_3^-]_{\text{summer}} / [\text{NO}_3^-]_{\text{winter}}$ ) and  $\varepsilon$   
 416 is the isotope effect for nitrate incorporation by phytoplankton, which was assumed to  
 417 be constant at  $\sim 5\text{‰}$  for simplification (Brunelle et al., 2007, 2010). Using a modern  
 418 average for  $f$  of 0.16 (84% utilization), estimated from WOA 2009 surface nitrate  
 419 concentrations (Garcia et al., 2010), gives  $\delta^{15}\text{N}_{\text{export}} = \sim 3.8\text{‰}$  for our site, which is  
 420  $1.7\text{‰}$  lower than the modern  $\delta^{15}\text{N}_{\text{nitrate}}$ . Unfortunately, no coretop  $\delta^{15}\text{N}_{\text{bulk}}$  results are  
 421 available for verification at our site.

422 Alternatively, secondary alteration (preferential loss of organic nitrogen, leakage of  
 423 ammonium into pore waters, ammonium absorption into clay minerals,  
 424 winnowing/size fractionation) might have raised the modern coretop  $\delta^{15}\text{N}$  value. In  
 425 general, however, alteration of the  $\delta^{15}\text{N}_{\text{bulk}}$  from that of the sinking flux is not

426 considered to have a considerable influence in organic-rich sediments from high-  
427 accumulation regions with low contributions of inorganic nitrogen compounds (for a  
428 review see Robinson et al., 2012). Core 85KL has rather high average TOC  
429 concentrations (~0.9 wt.%), whereas TIN is low at ~0.017 wt.% (Figure 2a), and the  
430 relationship between TN and  $\delta^{15}\text{N}_{\text{bulk}}$  is only weak ( $R^2 = 0.24$ ; Figure 2b), which  
431 argues against alteration. Moreover, in comparison with other Bering Sea records  
432 using  $\delta^{15}\text{N}$  as a proxy for nitrate utilization (Brunelle et al., 2007, 2010; Kim et al.,  
433 2011; Schlung et al., 2013) our  $\delta^{15}\text{N}_{\text{bulk}}$  record generally shows a similar evolution  
434 (Figure 7). Notably, at our study site  $\delta^{15}\text{N}_{\text{bulk}}$  values  $>5.5\text{‰}$  were mainly recorded  
435 during Termination I, which, in accordance with the previously mentioned studies is  
436 more likely attributed to enhanced deglacial water column denitrification.

437 Finally, there might have been influence from terrestrial nitrogen, since our estimates  
438 for  $m_{\text{terr}}$  suggest significant average contributions (~40-50%) of terrestrial organic  
439 matter in Shirshov Ridge sediments. In this respect, variations in  $\delta^{15}\text{N}_{\text{bulk}}$  might reflect  
440 changes in the supply of terrestrial nitrogen. However, for the estimation of  $m_{\text{terr}}$  we  
441 assumed that the  $\delta^{15}\text{N}_{\text{bulk}}$  of terrestrial organic matter is lower than that of marine  
442 organic matter. If there was considerable influence from terrestrial nitrogen we would  
443 expect a strong positive relationship between molar N/C ratios and  $\delta^{15}\text{N}_{\text{bulk}}$ , which is  
444 not observed ( $R^2 = 0.10$ ). Accordingly, although Site 85KL is characterized by overall  
445 high  $m_{\text{terr}}$  values with a strong downcore variability, there seems to have been no  
446 significant influence of  $m_{\text{terr}}$  on the  $\delta^{15}\text{N}_{\text{bulk}}$  signal, which might be explained by an  
447 only low fraction of terrestrial nitrogen. We therefore consider the influence of  
448 secondary alteration and contamination from terrestrial nitrogen on the  $\delta^{15}\text{N}_{\text{bulk}}$  signal  
449 as insignificant and in the following discuss variations in  $\delta^{15}\text{N}_{\text{bulk}}$  by means of  
450 changes in surface nitrate utilization.

451

## 452 **5.2 Cold and warm intervals of the past 180 kyr**

453 At Site 85KL, glacial periods, specifically cold intervals (MIS 6, 5.4, 5.2, and 4 to 2),  
454 were characterized by significantly reduced export production and enhanced  
455 terrigenous matter supply (Riethdorf et al., 2013a; Figure 4), which is in agreement

456 with other studies from the subarctic North Pacific (e.g., Kienast et al., 2004; Jaccard  
457 et al., 2005), the Okhotsk Sea (e.g., Narita et al., 2002; Nürnberg and Tiedemann,  
458 2004; Okazaki et al., 2005b; Nürnberg et al., 2011), and the Bering Sea (e.g.,  
459 Okazaki et al., 2005a; Brunelle et al., 2007; Kim et al., 2011). This is supported by  
460 our reconstruction of  $m_{\text{terr}}$  indicating an average terrestrial organic matter fraction of  
461 ~40-50%, which is significantly reduced only during warm intervals. Today, the  
462 organic matter in Bering Sea sediments is dominantly of marine origin (Méheust et  
463 al., 2013). The glacial terrigenous matter source of Bering Sea sediments is under  
464 debate, but there are indications that they originate from source rocks drained by the  
465 Yukon River (VanLaningham et al., 2009), and/or from sea-ice rafting in the NE  
466 Bering Sea (Riethdorf et al., 2013a).

467 For the Bering and Okhotsk seas the outlined observations were explained by  
468 enhanced sea-ice influence and stronger stratification of the upper water column  
469 during cold climate conditions restricting marine productivity (Nürnberg and  
470 Tiedemann, 2004; Brunelle et al., 2007, 2010; Kim et al., 2011; Khim et al., 2012;  
471 Riethdorf et al., 2013a). This restriction results from the extended sea-ice season and  
472 coverage and the subsequent limitation of light availability and vertical mixing  
473 (nutrient supply), but temperature limitation is likely to have played an additional role.  
474 An extended Bering Sea sea-ice coverage during cold phases is supported by  
475 reconstructions from diatom assemblages (Katsuki and Takahashi, 2005) and from  
476 the diatom-derived, highly branched isoprenoid sea ice biomarker (IP<sub>25</sub>) (Max et al.,  
477 2012).

478 Records of sedimentary and diatom-bound  $\delta^{15}\text{N}$  imply enhanced surface nitrate  
479 utilization as a result of stronger upper water column stratification in the Bering Sea,  
480 especially during MIS 3 and 2 (Brunelle et al., 2007, 2010; Kim et al., 2011). Similar  
481 observations are reported for the Okhotsk Sea (Brunelle et al., 2010; Khim et al.,  
482 2012) and the subarctic NW Pacific (Galbraith et al., 2008a; Brunelle et al., 2010),  
483 indicating that these regions were not always HNLC. This shift towards higher  $\delta^{15}\text{N}$  in  
484 MIS 3 is, however, not observed at IODP Site U1340 at the northeastern flank of  
485 Bowers Ridge (Schlung et al., 2013). Instead, a sharp decrease in  $\delta^{15}\text{N}_{\text{bulk}}$  was  
486 recorded at this site at ~55 ka BP, which might be related to local stratification  
487 changes, or to the influence of turbidites that are reported to compromise records



488 recovered from that area (Nakatsuka et al., 1995). Our  $\delta^{15}\text{N}_{\text{bulk}}$  results are in  
489 accordance with the former Bering Sea studies confirming enhanced nitrate  
490 utilization during cold intervals, but they seem to reveal a more complex development  
491 of stratification and for the first time provide information for MIS 6.

492 At the base of core 85KL (~180-173 ka BP), early MIS 6 is characterized by high  
493  $\delta^{15}\text{N}_{\text{bulk}}$  values indicating almost complete nitrate utilization, when export production  
494 was reduced, but maintained. Relatively high  $\delta^{15}\text{N}_{\text{bulk}}$  during MIS 6 were also  
495 recorded at Okhotsk Sea sites GGC27 (Brunelle et al., 2010) and GC9A (Khim et al.,  
496 2012), as well as at ODP Site 882 and at Site MD01-2416 (Galbraith et al., 2008a).  
497 Hence, low insolation and weak seasonal contrasts most probably caused a  
498 prolonged sea-ice season, extended sea-ice coverage, and suppressed vertical  
499 mixing. At ~172 ka BP, when Northern Hemisphere summer insolation had a local  
500 maximum, a sharp decrease in  $\delta^{15}\text{N}_{\text{bulk}}$  implies a sudden drop in nitrate utilization  
501 (from ~100% to ~50-70%) in the Bering Sea (Figure 6). At the same time export  
502 production was increased, whereas  $m_{\text{terr}}$  decreased. This might be explained by a  
503 shortened sea-ice season, reduced sea-ice coverage, and enhanced winter mixing  
504 due to stronger seasonal contrasts, which increased the nutrient supply from the  
505 subeuphotic zone.

506 Accordingly, we speculate that changes in nitrate utilization are strongly affected by  
507 insolation forcing and a feedback by sea-ice processes that drive the extent of  
508 vertical mixing during winter, as well as the input of terrestrial organic matter.  
509 Dominant climate control via insolation has already been proposed for the Okhotsk  
510 Sea (Gorbarenko et al., 2010a, 2012). The long-term increase in nitrate utilization  
511 after ~172 ka BP until Termination II, which is also observed at the Okhotsk Sea and  
512 NW Pacific sites (Figure 7), as well as the long-term increase from MIS 5.4 until  
513 Termination I might be explained by increasingly fostered stratification (i.e. a  
514 reduction in the supply of nutrients into the euphotic zone), which is finally subject to  
515 a 'breakdown' during the deglaciations. It is beyond the scope of this paper to  
516 decipher the underlying causes of this deglacial breakdown, but increasing insolation  
517 resulting in a reduced sea-ice season and strengthened winter mixing, is a likely  
518 contributing factor.

519 The observation that the strongest maxima in Northern Hemisphere summer  
520 insolation, mainly those of warm intervals (MIS 5.5, 5.3, 5.1, and 1), are reflected by  
521 maxima in export production (Figure 4), minima in  $m_{\text{terr}}$ , and decreasing or constant  
522  $\delta^{15}\text{N}_{\text{bulk}}$  (Figure 6), supports the view that insolation changes affect nutrient-limited  
523 marine productivity by a feedback in sea-ice processes and winter mixing. It is also in  
524 agreement with previously published concepts proposed to explain glacial-interglacial  
525 changes in the Okhotsk (Seki et al., 2004; Okazaki et al., 2005b; Khim et al., 2012)  
526 and Bering seas (Nakatsuka et al., 1995; Kim et al., 2011). When applying Equation  
527 2, our  $\delta^{15}\text{N}_{\text{bulk}}$  record indicates increasing nitrate utilization during cold MIS 5.4 (from  
528 ~50 to ~90%), MIS 5.2 (from ~90 to ~100%), and MIS 4 (from ~80 to ~93%), and  
529 almost complete utilization during MIS 3 and 2 (~93-100%). On the other hand,  
530 decreasing or constant nitrate utilization was recorded during warm MIS 5.5 (from  
531 ~97 to ~62%), MIS 5.3 (~90%), and MIS 5.1 (from ~100 to ~80%). This suggests that  
532 stratification was fostered during cold intervals, but weakened during warm intervals  
533 due to the processes outlined above.

534 The low glacial  $\delta^{15}\text{N}_{\text{bulk}}$  values of ~2-3‰ at the beginning of MIS 5.4, of ~3.5-4.5‰ at  
535 the beginning of MIS 4, and the concurrent increases in  $m_{\text{terr}}$  (Figure 6) might be  
536 explained (i) by a higher contribution of ( $^{15}\text{N}$ -depleted) terrestrial organic matter, or (ii)  
537 by stronger vertical mixing. We disregard the first possibility, because we already  
538 discarded the potential effect of terrestrial nitrogen on the  $\delta^{15}\text{N}_{\text{bulk}}$  signal (Section  
539 5.1). Stronger vertical mixing in the Bering Sea during MIS 5.4 and 4 might be related  
540 to the increased formation and/or ventilation of intermediate waters as inferred from  
541 neodymium isotope ratios by Horikawa et al. (2010). The authors suggested that sea-  
542 ice formation and according brine rejection led to the subduction of surface waters to  
543 intermediate depths. Enhanced formation of sea-ice, acting as the transport agent for  
544 terrestrial organic matter would be in accordance with this assumption and explain  
545 the higher  $m_{\text{terr}}$  values. Other studies support the idea of well-ventilated intermediate  
546 waters in the Bering Sea and North Pacific during glacial times (Ohkushi et al., 2003;  
547 Itaki et al., 2009; Kim et al., 2011) and during severe stadial episodes (Rella et al.,  
548 2012). The enhanced formation and/or ventilation of intermediate waters at the end  
549 of MIS 6 and during the LGM implied by the record of Horikawa et al. (2010) is not  
550 reflected in our  $\delta^{15}\text{N}_{\text{bulk}}$  record, which rather suggests strong stratification during that

551 time. However, these observations are not necessarily contradictory, since  
552 intermediate waters could have been formed outside the still-stratified Bering Sea. In  
553 fact, recent reconstructions of past ventilation changes in the subarctic North Pacific  
554 using radiocarbon-derived ventilation ages in combination with epibenthic stable  
555 carbon isotope ratios point to the Okhotsk Sea as the source region of intermediate  
556 waters during HS1 and the YD (Max et al., 2014).

557 In addition to insolation forcing, sea-level changes might have influenced the extent  
558 of stratification in the Bering Sea. Today, the only shallow (~50 m) Bering Strait  
559 allows for oceanic communication between the North Pacific and the N Atlantic.  
560 During glacial times the closed Bering Strait prevented the flux of relatively fresh  
561 waters into the Atlantic, which is thought to have affected the Atlantic meridional  
562 overturning circulation (Hu et al., 2010). Lower glacial sea-level is also likely to have  
563 reduced the inflow of Alaskan Stream waters into the Bering Sea (Gorbarenko et al.,  
564 2005; Tanaka and Takahashi, 2005). As suggested by relative sea-level  
565 reconstructions (e.g., Waelbroeck et al., 2002; Yokoyama and Esat, 2011), the  
566 Bering Strait was closed during MIS 6 and in between MIS 4 to 2 until ~12-11 ka BP  
567 (Keigwin et al., 2006) (Figure 6). Our  $\delta^{15}\text{N}_{\text{bulk}}$  values indicate almost complete nitrate  
568 utilization during late MIS 6 (~150-130 ka BP) and during MIS 3 and 2 as a result of  
569 strong stratification. During this time a closed Bering Strait is likely to have fostered  
570 stratification due to the pooling of the relatively fresh waters within the Bering Sea,  
571 which would have resulted in a strengthened pycnocline. Support for this view and for  
572 fresher glacial conditions in the Bering Sea comes from diatom and radiolarian  
573 assemblages (Sancetta, 1983; Katsuki and Takahashi, 2005; Tanaka and Takahashi,  
574 2005). Notably, during MIS 3 and 2 the  $\delta^{15}\text{N}_{\text{bulk}}$  values recorded at Shirshov Ridge  
575 are on average ~1‰ lower than at Bowers Ridge Site 17JPC and ~0.5‰ lower than  
576 at Site PC24A (Figure 7). This might indicate that stratification in the Bering Sea was  
577 regionally different and more pronounced in the South, or that influence from  
578 denitrification resulted in the heavier  $\delta^{15}\text{N}$  values.

579

580

581

### 582 5.3 Deglacial and interglacial conditions

583 In our records, the deglaciations are characterized by the transition from the glacial  
584 situation of pronounced stratification with almost complete nitrate utilization and low  
585 export production toward the interglacial situation of reduced stratification, high  
586 marine productivity, and reduced terrestrial input. Yet, Termination II and Termination  
587 I show some notable differences. During Termination I our  $\delta^{15}\text{N}_{\text{bulk}}$  record is  
588 characterized by an initial decrease, which might correspond to the HS1 cold phase,  
589 subsequent local maxima during the B/A and the early Holocene warm phases, and  
590 an intercalated minimum during the YD (Figure 6). The same temporal evolution was  
591 reported for sedimentary and diatom-bound  $\delta^{15}\text{N}$  at Bering Sea sites 17JPC (Brunelle  
592 et al., 2007, 2010) and PC24A (Kim et al., 2011). The B/A-peak, occurring  
593 simultaneously with a rise in export production, is found in several other records from  
594 the North Pacific realm and related to enhanced denitrification (Keigwin et al., 1992;  
595 Emmer and Thunell, 2000; Ternois et al., 2001; Kienast et al., 2002; Galbraith et al.,  
596 2008a; Kao et al., 2008; Brunelle et al., 2007, 2010; Addison et al., 2012; Khim et al.,  
597 2012; Schlung et al., 2013).

598 Notably,  $m_{\text{terr}}$  shows a local maximum during HS1 at Site 85KL. Hence, the initial  
599 decrease in  $\delta^{15}\text{N}_{\text{bulk}}$  might be related to higher terrestrial input or to lower nitrate  
600 utilization due to weakened stratification. However, the latter should have resulted in  
601 higher export production, which is not observed at our site. This drop is not fully  
602 understood and alternative explanations include changes in  $\delta^{15}\text{N}_{\text{nitrate}}$ , iron limitation,  
603 and light limitation (Brunelle et al., 2007, 2010; Lam et al., 2013). Light limitation by  
604 expanded sea-ice coverage is supported by the qualitative detection of  $\text{IP}_{25}$  in  
605 western Bering Sea sediments during HS1 and the YD (Max et al., 2012).

606 During the B/A and the early Holocene our  $\delta^{15}\text{N}_{\text{bulk}}$  values exceeded the modern  
607  $\delta^{15}\text{N}_{\text{nitrate}}$  value, supporting an increase in denitrification. At the same time a shift  
608 toward oxygen-depleted bottom water conditions is inferred from benthic foraminiferal  
609 assemblages (Kim et al., 2011; Ovsepyan et al., 2013), which is in agreement with  
610 the proposed expansion of the OMZ and the occurrence of laminated sediments  
611 during warm intervals (e.g., Zheng et al., 2000; van Geen et al., 2003; Cook et al.,  
612 2005; Kuehn et al., 2014). A recent comparison between alkenone- and Mg/Ca-

613 based paleotemperature estimates suggests enhanced thermal mixed-layer  
614 stratification in the western Bering Sea during the B/A (Riethdorf et al., 2013b),  
615 implying that at least some of the recorded  $\delta^{15}\text{N}_{\text{bulk}}$  increase is due to stronger  
616 surface nitrate utilization. Recently, Lam et al. (2013) suggested two stepwise events  
617 starting with deep convection initialized at  $\sim 18$  ka BP increasing the nutrient supply  
618 but inducing light limitation, and subsequent meltwater-induced stratification resulting  
619 in bloom conditions and leaving surface waters enriched in nutrients. The drop in  
620  $\delta^{15}\text{N}_{\text{bulk}}$  observed during the YD in hand with decreasing SSTs and the presence of  
621  $\text{IP}_{25}$  (Max et al., 2012) argues for a similar situation as recorded during HS1.

622 Termination II differs from Termination I at Site 85KL in such that the  $\delta^{15}\text{N}_{\text{bulk}}$  values  
623 are lower and presumably not affected by denitrification. An early deglacial  $\delta^{15}\text{N}_{\text{bulk}}$   
624 minimum at  $\sim 133$  ka BP, followed by a local maximum at  $\sim 131$  ka BP might hint  
625 toward analogs of the HS1 and the B/A, respectively. The subsequent drop in  $\delta^{15}\text{N}_{\text{bulk}}$   
626 into MIS 5.5 reflecting the 'breakdown' of glacial stratification is sudden and  
627 accompanied by the decrease in both,  $m_{\text{terr}}$  and bottom water oxygenation then  
628 prevailing during the penultimate interglacial (Figure 6). Notably, this drop from  
629 relatively high MIS 6 values occurred  $\sim 5$  kyr before the maximum in insolation was  
630 reached, but its timing is comparable to that recorded in the NW Pacific. In the  
631 Okhotsk Sea it seems to have occurred significantly earlier at  $\sim 147$ - $141$  ka BP  
632 (Figure 7). A respective drop in  $\delta^{15}\text{N}_{\text{bulk}}$  during Termination I was not recorded at Site  
633 85KL until  $\sim 9.1$  ka BP, while at Bowers Ridge sites U1340 (Schlung et al., 2013) and  
634 17JPC (Brunelle et al., 2007) it occurred directly after the B/A maximum.

635

#### 636 **5.4 Millennial-scale oscillations**

637 Riethdorf et al. (2013a) reported on millennial-scale oscillations in core 85KL, thought  
638 to reflect increased export production and sudden sea-ice melt, which might be  
639 connected to GI (Dansgaard-Oeschger events; e.g., Dansgaard et al., 1993). Similar  
640 observations are reported for other sediment cores from the Bering Sea (Gorbarenko  
641 et al., 2005, 2010b; Kim et al., 2011; Rella et al., 2012; Schlung et al., 2013) and the  
642 Okhotsk Sea (Gorbarenko et al., 2007, 2010a, 2012), indicating warmer SSTs,  
643 enhanced marine productivity, weak ventilation of intermediate waters, and poor

644 (dysoxic) dissolved oxygen conditions during interstadials. NE Pacific sediments  
645 related to GI are in part laminated and suggested to reflect phases of weak  
646 ventilation of NPIW and fluctuations in the strength of the OMZ (Behl and Kennett,  
647 1996; Cannariato and Kennett, 1999; Hendy and Kennett, 2000, 2003). Results of  
648 Ortiz et al. (2004) from a core off Baja California implied that elevated marine  
649 productivity was caused by enhanced nutrient flux to surface waters.

650 In the Bering Sea higher interstadial  $\delta^{15}\text{N}_{\text{bulk}}$  values were explained by Kim et al.  
651 (2011) by increased marine productivity as a result of reduced sea-ice influence and  
652 a strengthened BSC. They also suggested that stronger inflow of water masses from  
653 the Gulf of Alaska (Gorbarenko et al., 2005) resulted in enhanced nutrient supply to  
654 Bering Sea surface waters. Schlung et al. (2013) attributed higher  $\delta^{15}\text{N}_{\text{bulk}}$  and  
655 concurrent minima in planktonic  $\delta^{13}\text{C}$  to amplified local upwelling of subsurface  
656 nitrate rather than to increased nitrate utilization. Our data show short-lived maxima  
657 in  $\delta^{15}\text{N}_{\text{bulk}}$  and concurrent minima in  $m_{\text{terr}}$  during some, but not all, GI (1, 7, 8, 12, 17-  
658 20), and during MIS 6 (at ~133, ~148, and ~173 ka BP) when export production was  
659 high (Figure 6). The opposite pattern was recorded when export production was low  
660 during some GS (2, 7, 18, 20) and also during MIS 6 (at ~151, ~157, and ~170 ka  
661 BP). Despite the low reproducibility of our  $\delta^{15}\text{N}_{\text{bulk}}$  results our data support the view of  
662 increased interstadial marine productivity which led to stronger utilization of the  
663 available nitrate in a still stratified upper water column. During interstadials warmer  
664 SSTs most probably resulted in less sea-ice influence and reduced supply of  
665 terrestrial organic matter. Conversely, during stadials strengthened sea-ice formation  
666 and coverage is likely to have restricted marine productivity, led to enhanced  
667 terrestrial organic matter supply to Bering Sea sediments, and resulted in better  
668 ventilation of NPIW, potentially making the Bering Sea a proximate source of this  
669 water mass as suggested by Rella et al. (2012).

670

## 671 **6. Summary**

672 We determined TN, TOC,  $\delta^{13}\text{C}_{\text{bulk}}$  and  $\delta^{15}\text{N}_{\text{bulk}}$  in a core from the western Bering Sea  
673 in high-resolution to reconstruct changes in surface nitrate utilization (stratification)  
674 over the last 180 kyr. A linear endmember model was applied to assess the

675 contributions of marine- and terrestrial-derived organic matter. Besides the expected  
676 difference between glacial and interglacial conditions reported for the subarctic NW  
677 Pacific and its marginal seas, our results suggest a more complex evolution of  
678 stratification with enhanced vertical mixing during warm intervals (MIS 5.5, 5.3, 5.1,  
679 1), and stratification becoming fostered during cold intervals (MIS 6, 5.4, 5.2, 4-2).  
680 This development is explained by insolation forcing and a feedback in sea-ice  
681 formation and the strength of winter mixing. In addition, sea-level changes might  
682 have further influenced the extent of stratification when the Bering Strait was closed  
683 and relatively fresh waters pooled in the Bering Sea. During warm intervals,  
684 variations in seasonal contrasts, sea-ice influence, and stratification resulted in  
685 enhanced export production and dominantly marine-derived organic matter, but less  
686 nitrate utilization due to better vertical mixing. Conversely, enhanced terrestrial-  
687 derived organic matter, most probably associated with sea-ice formation, low export  
688 production, and enhanced stratification characterized cold intervals of the past 180  
689 kyr. Moreover, we present supporting evidence that millennial-scale climate  
690 oscillations connected with Greenland interstadials occurred in the Bering Sea  
691 environment, and that sea-ice formation there influenced the ventilation of North  
692 Pacific Intermediate Water.

693

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1187

#### 1188 **Figure captions**

1189 Figure 1: (a) Surface circulation pattern (red arrows; after Tomczak and Godfrey,  
1190 1994; Stabeno et al., 1999) and bathymetry of the subarctic North Pacific realm. The  
1191 red dot marks the location of sediment core SO201-2-85KL studied here. Published  
1192 reference records are marked by yellow dots. Bering Sea: MR06-04-PC24A (Kim et  
1193 al., 2011), KH99-3-BOW-8A (Horikawa et al., 2010), HLY02-02-17JPC (Brunelle et  
1194 al., 2007, 2010), IODP Site U1340 (Schlung et al., 2013). Okhotsk Sea: YK0712-  
1195 GC9A (Khim et al., 2012), GGC27 (Brunelle et al., 2010). NW Pacific: MD01-2416  
1196 (Galbraith et al., 2008a), PC13 (Brunelle et al., 2010). NE Pacific: ODP Site 887  
1197 (Galbraith et al., 2008a). The modern average maximum sea-ice extent during March  
1198 is indicated by the dashed black line (after Niebauer et al., 1999; Zhang et al., 2010;  
1199 IRI/LDEO Climate Data Library, <http://iridl.ldeo.columbia.edu/>). Surface currents:  
1200 ANSC = Aleutian North Slope Current, BSC = Bering Slope Current, EKC = East  
1201 Kamchatka Current, ESC = East Sakhalin Current, NOC = North Okhotsk Current,  
1202 SC = Soya Current, WKC = West Kamchatka Current. Straits: bus = Bussol Strait,  
1203 kss = Kruzenshtern Strait, ks = Kamchatka Strait, ns = Near Strait, bp = Buldir Pass,  
1204 as = Amchitka Strait, ap = Amukta Pass, up = Unimak Pass, bs = Bering Strait. (b)



1205 Surface nitrate concentration during modern summer (July-September; in  $\mu\text{mol l}^{-1}$ )  
1206 from World Ocean Atlas 2009 data (Garcia et al., 2010). Maps produced with "Ocean  
1207 Data View" (Schlitzer, 2013).

1208

1209 Figure 2: (a) Relationship between concentrations of total nitrogen (TN) and total  
1210 organic carbon (TOC) in samples from core SO201-2-85KL. For the calculation of  
1211 molar N/C ratios a linear regression between TOC and TN was used to assess the  
1212 fraction of inorganic nitrogen, represented by the intercept of the regression line at  
1213  $\text{TOC} = 0$ . (b) Comparison with  $\delta^{15}\text{N}_{\text{bulk}}$  indicates that there is only a weak linear  
1214 relationship between the isotopic signal and TN concentrations ( $R^2 = 0.24$ ;  $p < 10^{-4}$ ).

1215

1216 Figure 3: Linear sedimentation rate (LSR) vs. bulk accumulation rate (AR Bulk), and  
1217 comparison of concentration and accumulation rate (AR) records of Siliciclastics,  
1218  $\text{CaCO}_3$ , TOC and opal for core SO201-2-85KL.

1219

1220 Figure 4: Records reflecting changes in export production and terrigenous matter  
1221 supply in core SO201-2-85KL over the past 180 kyr in comparison with Northern  
1222 Hemisphere summer (July-September) insolation at  $65^\circ\text{N}$  (after Laskar et al., 2004).  
1223 Logging data (underlying grey lines), %Siliciclastics, as well as  $\text{CaCO}_3$  and opal  
1224 concentrations are from Max et al. (2012) and Riethdorf et al. (2013a, 2013b). Note  
1225 inverted axes of %Siliciclastics and XRF Al count rates. The  $\delta^{18}\text{O}$  records from the  
1226 NGRIP ice core in Greenland (NGRIP members, 2004; GICC05 timescale,  
1227 Rasmussen et al., 2006) and from the Sanbao stalagmites in China (Wang et al.,  
1228 2008) are shown for reference. Greenland interstadials (GI) are highlighted by pale  
1229 red vertical bars. Boundaries of Marine Isotope Stages (MIS) after Lisiecki and  
1230 Raymo (2005).

1231

1232 Figure 5: Comparison of  $\delta^{13}\text{C}_{\text{bulk}}$  with (a) molar N/C ratios and (b)  $\delta^{15}\text{N}_{\text{bulk}}$  for core  
1233 SO201-2-85KL. Samples from warm intervals (MIS 5.5, 5.3, 5.1, 3, and 1) are  
1234 marked by red dots, while blue dots mark those from cold intervals (MIS 6, 5.4, 5.2,

1235 4, 2), and green triangles indicate Holocene (<11.7 ka BP) samples. Grey-shaded  
1236 boxes represent geochemical provenances (after literature data; see text for  
1237 references). The dashed lines indicate the applied linear mixing model for the  
1238 estimation of  $m_{\text{terr}}$ . Marine phytoplankton and vascular plant detritus (VPD) are  
1239 considered as potential marine and terrestrial organic matter sources (endmembers).

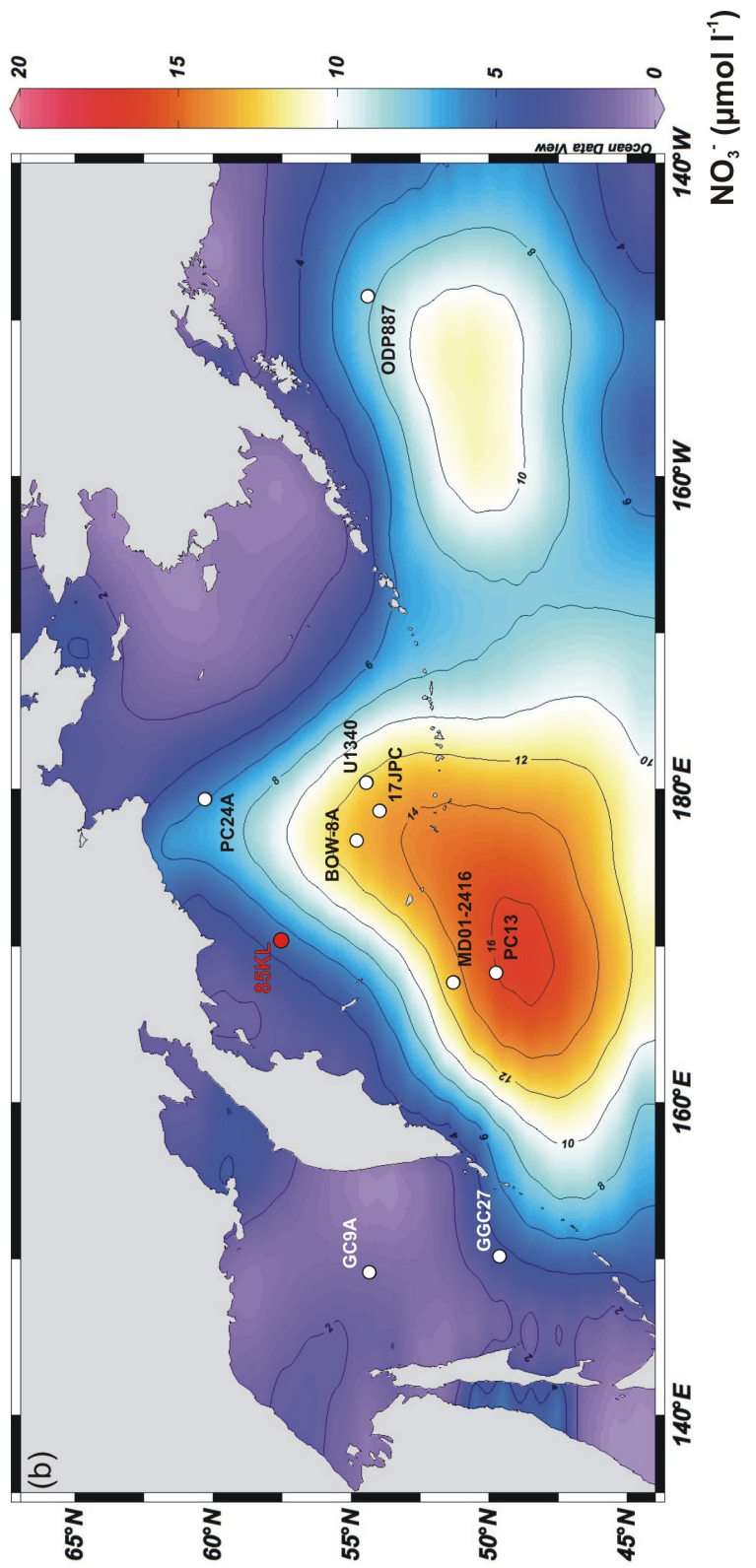
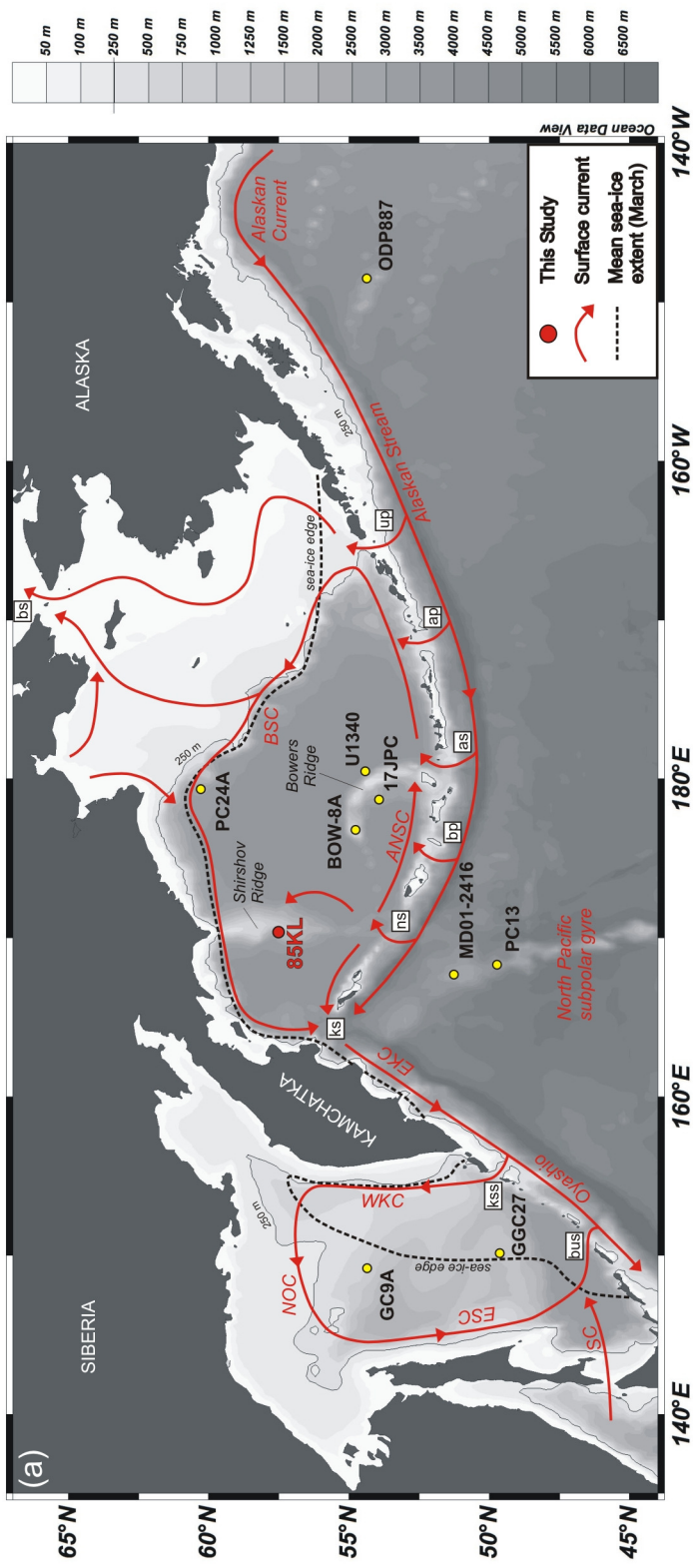
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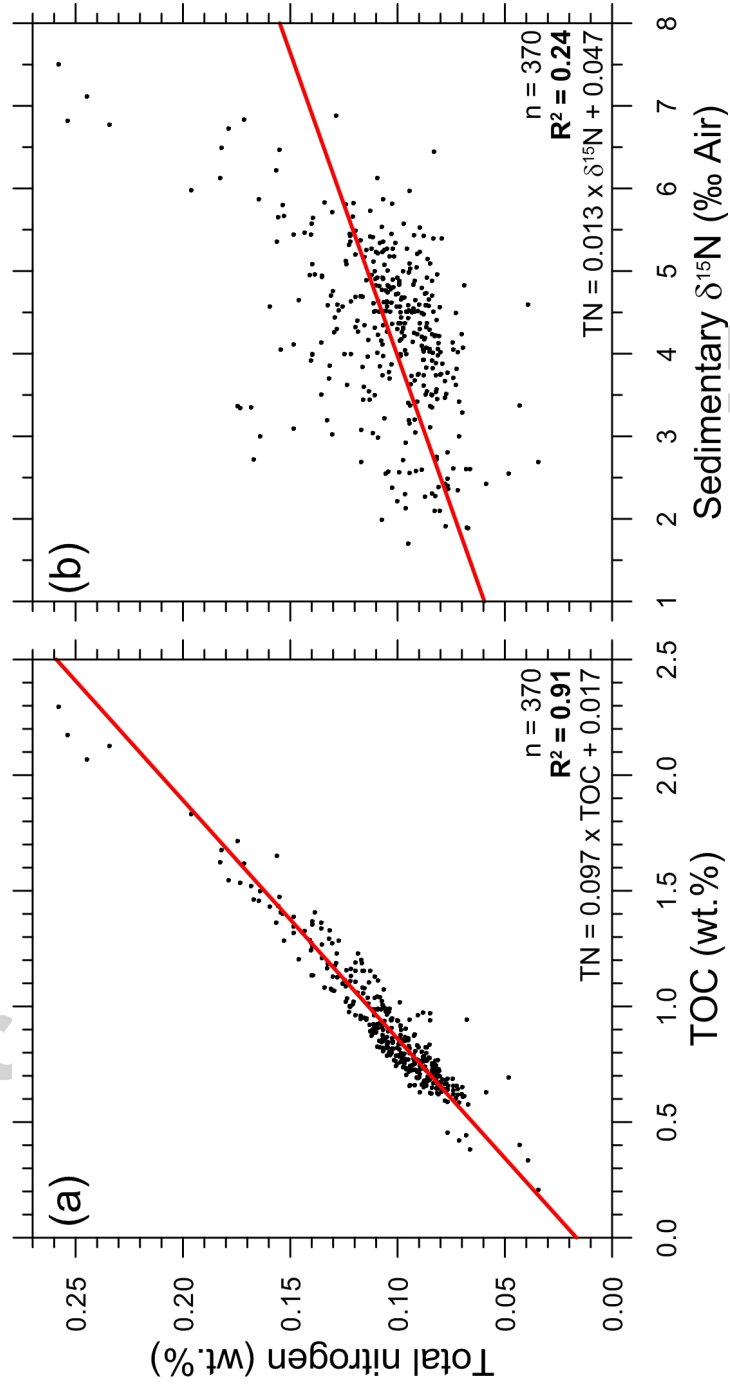
1241 Figure 6: Proxy records from core SO201-2-85KL in comparison with published  
1242 reference records covering the last 180 kyr: (a) Northern Hemisphere summer (65°;  
1243 July-September) insolation after Laskar et al. (2004), (b) sedimentary  $\delta^{13}\text{C}$  and (c)  
1244 molar N/C ratios used to estimate the fraction of terrestrial organic matter ( $m_{\text{terr}}$ ;  
1245 respective axes apply to those of  $\delta^{13}\text{C}$  and N/C), (d) color  $b^*$  assumed to reflect  
1246 export production, (e)  $\delta^{15}\text{N}_{\text{bulk}}$  reflecting surface nitrate utilization, (f) neodymium  
1247 isotope ratios from core KH99-3-BOW-8A (Horikawa et al., 2010; cf. Figure 1)  
1248 considered to approximate intermediate water formation, and (g) relative sea-level  
1249 (Waelbroeck et al., 2002) normalized to the sill depth (~50 m; dashed line) of the  
1250 Bering Strait ( $\Delta\text{RSL}$ ). MIS boundaries after Lisiecki and Raymo (2005), GI highlighted  
1251 by pale red vertical bars.

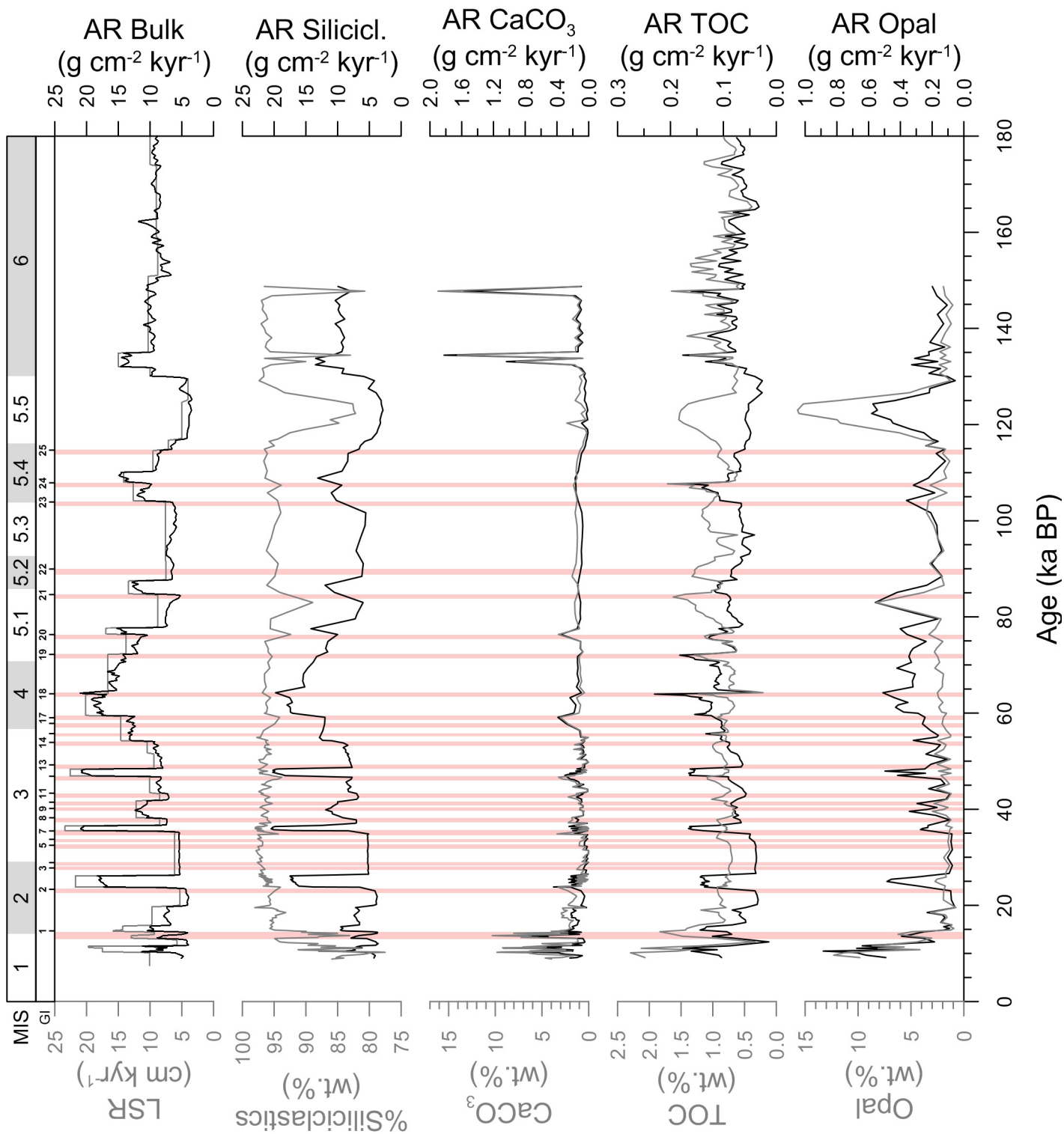
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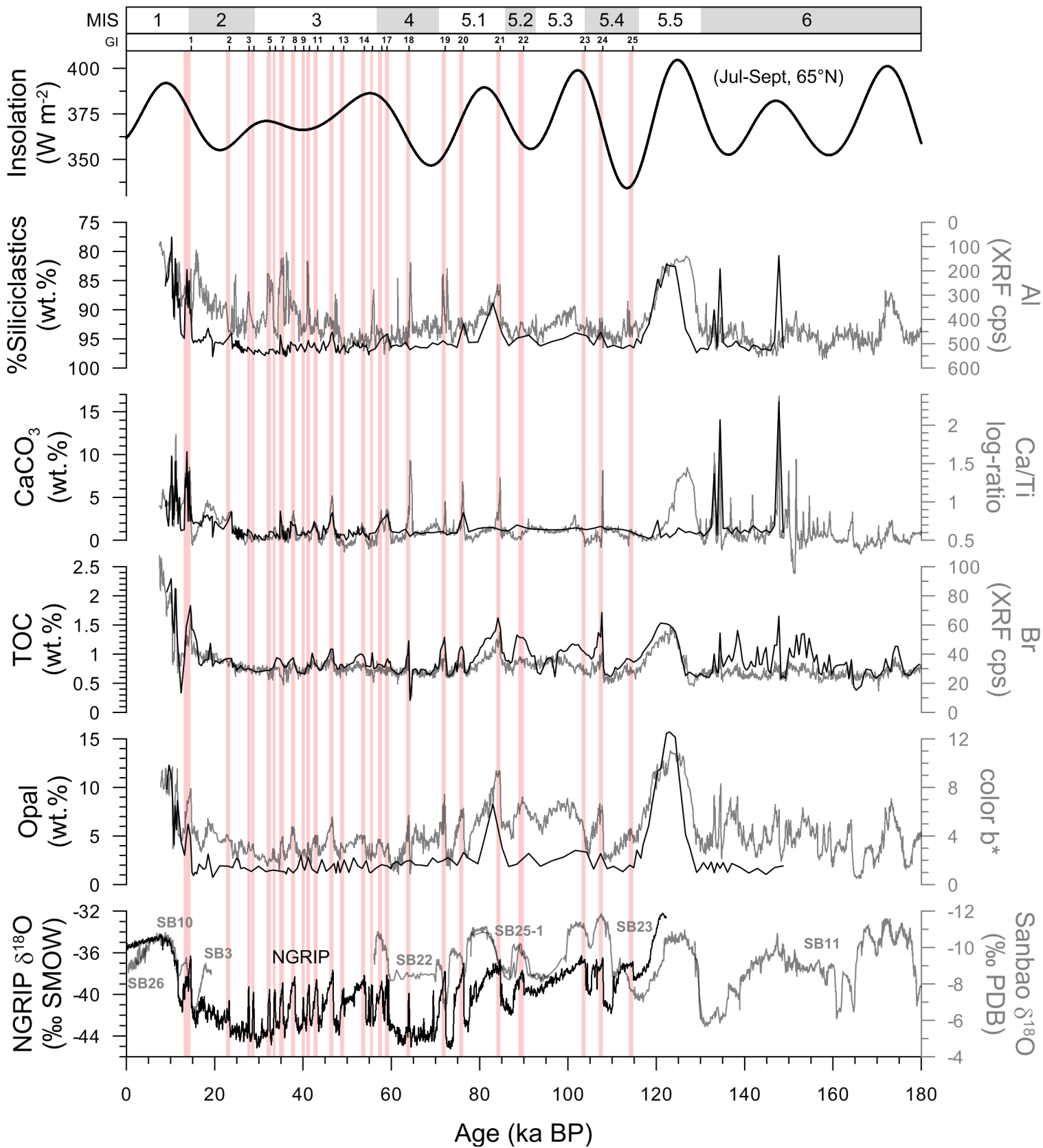
1253 Figure 7: Comparison of  $\delta^{15}\text{N}_{\text{bulk}}$  from SO201-2-85KL (black line) with other  
1254 sedimentary (solid lines) and diatom-bound (dotted lines)  $\delta^{15}\text{N}$  records from the  
1255 subarctic North Pacific and its marginal seas (cf. Figure 1). The timing of Greenland  
1256 interstadials and MIS boundaries are indicated.

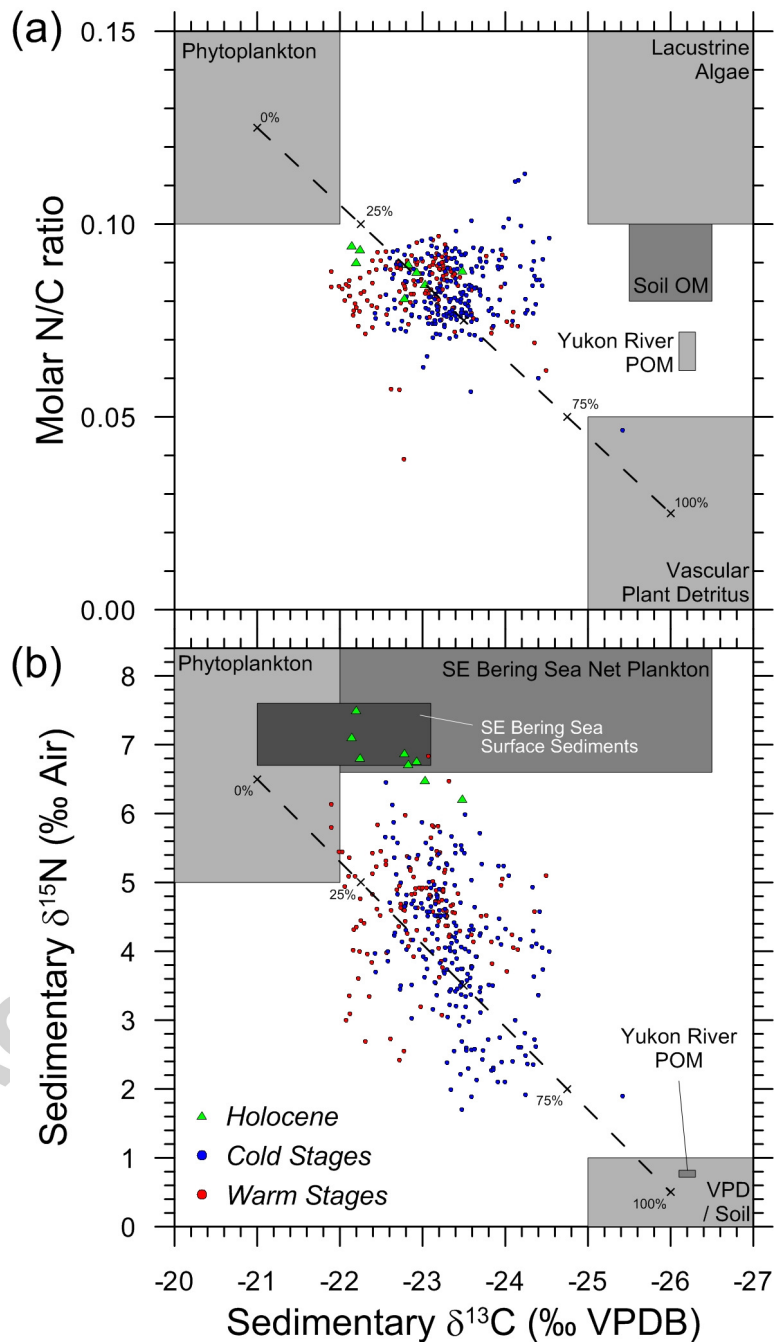
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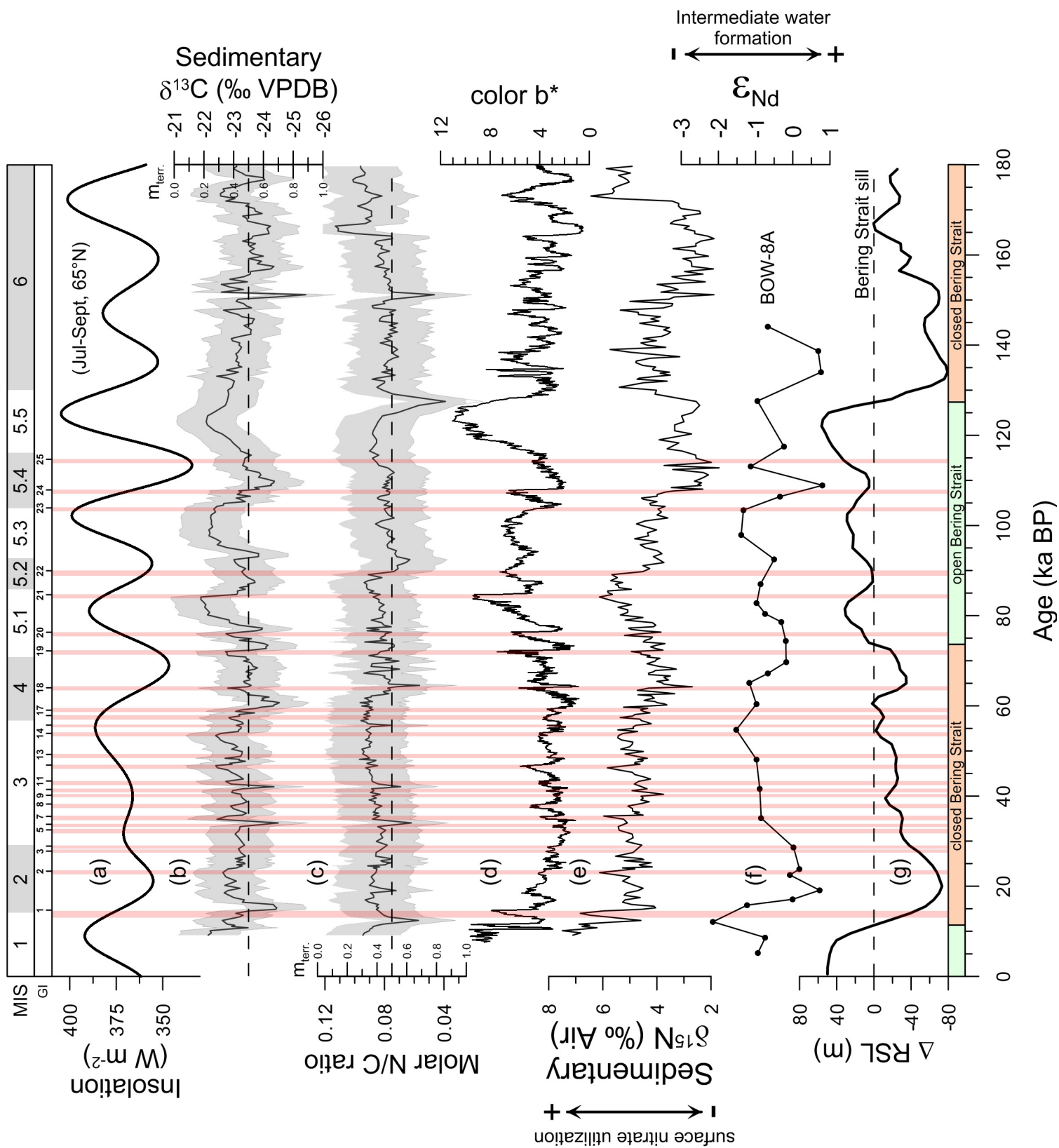




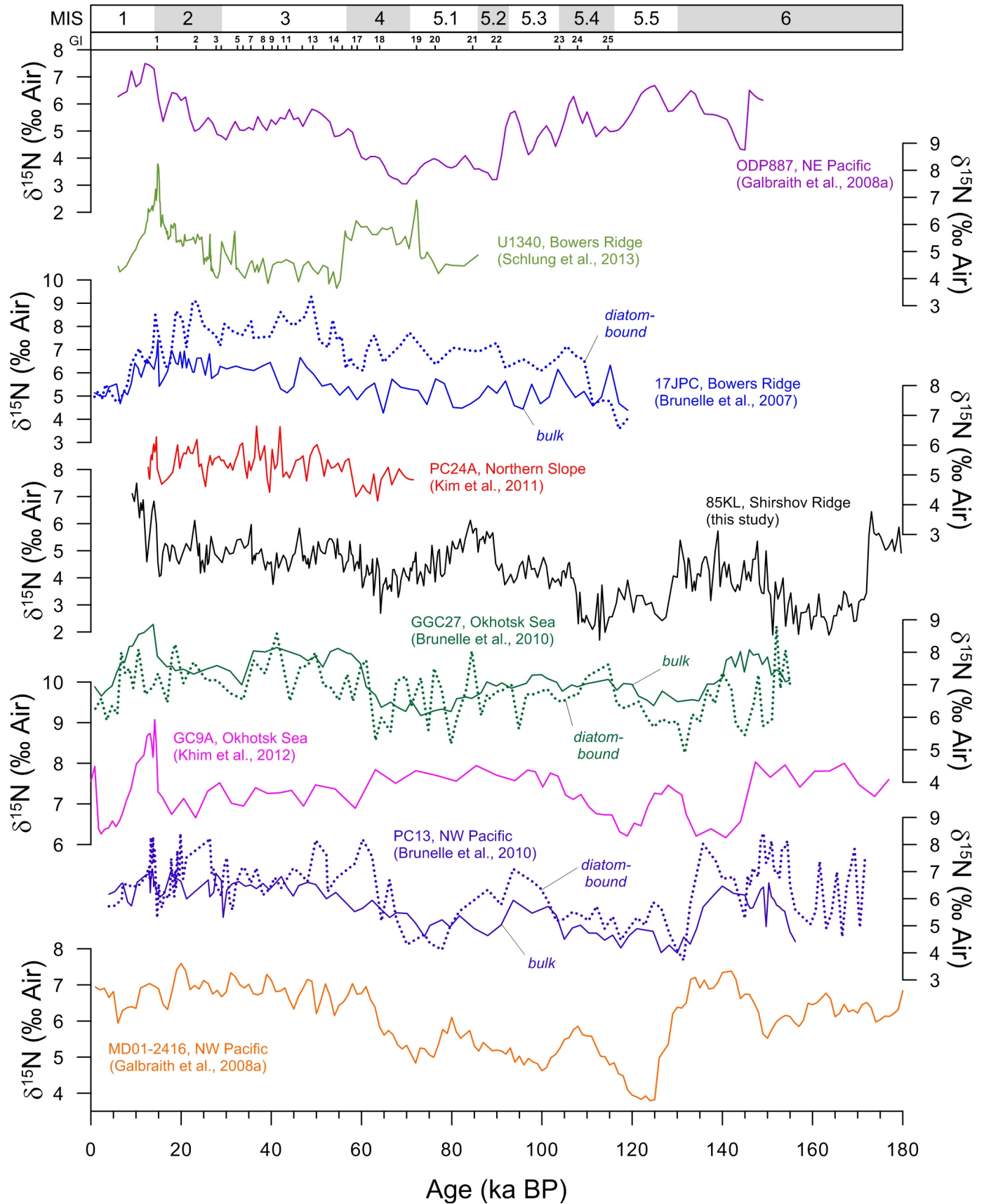












**Table 1** Ranges, averages, and variability of  $\delta^{13}\text{C}$ , molar N/C ratio, and  $m_{\text{terr}}$  in Core SO201-2-85KL.

Variable	Minimum	Maximum	Average	StDev.
$\delta^{13}\text{C}$ (‰ VPDB)	-25.4	-21.9	-23.2	0.6
molar N/C ratio	0.04	0.11	0.08	0.01
$m_{\text{terr}}$ via $\delta^{13}\text{C}$	18%	88%	45%	11%
$m_{\text{terr}}$ via N/C	12%	86%	42%	8%

**Table 2** Parameters for Calculation of  $m_{\text{terr}}$  Using Eq. (1).

Variable	Minimum	Maximum	Average
<i>Marine Endmember Composition</i>			
$d^{13}\text{C}_{\text{mar}}$	-20 ‰	-22 ‰	-21‰
$d^{15}\text{N}_{\text{mar}}$	5.0 ‰	/	6.5 ‰
N/C <sub>mar</sub>	0.100	0.150	0.125
<i>Terrestrial Endmember Composition</i>			
$d^{13}\text{C}_{\text{terr}}$	-25 ‰	-27 ‰	-26 ‰
$d^{15}\text{N}_{\text{terr}}$	0 ‰	1.0 ‰	0.5 ‰
N/C <sub>terr</sub>	0	0.050	0.025