

1 **Millennial-scale variations of sedimentary oxygenation in the western**
2 **subtropical North Pacific and its links to North Atlantic climate**

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20 **Key Points**

21 1. This study reconstructs the history of sedimentary oxygenation processes at
22 mid-depths in the western subtropical North Pacific since the last glacial period.

23 2. Sediment-bound redox-sensitive proxies reveal millennial-scale variations in
24 sedimentary oxygenation that correlated closely to changes in the North Pacific
25 Intermediate Water.

26 3. A millennial-scale out-of-phase relationship between deglacial ventilation in the
27 western subtropical North Pacific and the formation of North Atlantic Deep Water is
28 suggested.

29 4. A larger CO₂ storage at mid-depths of the North Pacific corresponds to the
30 termination of atmospheric CO₂ rise during the Bölling-Alleröd interval.

31 **Abstract**

32 The deep ocean carbon cycle, especially carbon sequestration and outgassing, is
33 one of the mechanisms to explain variations in atmospheric CO₂ concentrations on
34 millennial and orbital timescales. However, the potential role of subtropical North
35 Pacific subsurface waters in modulating atmospheric CO₂ levels on millennial
36 timescales is poorly constrained. An increase in respired CO₂ concentration in the
37 glacial deep ocean due to biological pump generally corresponds to deoxygenation in
38 the subsurface layer. This link thus offers a chance to study oceanic ventilation and
39 coeval export productivity based on redox-controlled, sedimentary geochemical
40 parameters. Here, we investigate a suite of geochemical proxies in a sediment core
41 from the Okinawa Trough to understand sedimentary oxygenation variations in the
42 subtropical North Pacific over the last 50,000 years (50 ka). Our results suggest that
43 enhanced mid-depth western subtropical North Pacific (WSTNP) sedimentary
44 oxygenation occurred during cold intervals and after 8.5 ka, while oxygenation
45 decreased during the Bölling-Alleröd (B/A) and Preboreal. The enhanced sedimentary
46 oxygenation in the WSTNP is aligned with intensified formation of North Pacific
47 Intermediate Water (NPIW) during cold spells, while better sedimentary oxygenation
48 seems to be linked to an intensified Kuroshio Current after 8.5 ka. The enhanced
49 formation of NPIW during Heinrich Stadial 1 (HS1) was likely driven by the
50 perturbation of sea ice formation and sea surface salinity oscillations in high-latitude
51 North Pacific. The diminished sedimentary oxygenation during the B/A due to
52 decreased NPIW formation and enhanced export production, indicates an expansion
53 of oxygen minimum zone in the North Pacific and enhanced CO₂ sequestration at
54 mid-depth waters, along with termination of atmospheric CO₂ concentration increase.
55 We attribute the millennial-scale changes to intensified NPIW and enhanced abyss
56 flushing during deglacial cold and warm intervals, respectively, closely related to
57 variations in North Atlantic Deep Water formation.

58 **Keywords:** sedimentary oxygenation; millennial timescale; North Pacific
59 Intermediate Water; North Atlantic Deep Water; subtropical North Pacific

60 **1. Introduction**

61 A more sluggish deep ocean ventilation combined with a more efficient
62 biological pump widely thought to facilitate enhanced carbon sequestration in the
63 ocean interior, leading to atmospheric CO₂ drawdown during glacial cold periods
64 (Sigman and Boyle, 2000). These changes are tightly coupled to bottom water
65 oxygenation and sedimentary redox changes on both millennial and orbital timescales
66 (Hoogakker et al., 2015; Jaccard and Galbraith, 2012; Sigman and Boyle, 2000).
67 Reconstruction of past sedimentary oxygenation is therefore crucial for understanding
68 changes in export productivity and renewal of deep ocean circulation (Nameroff et al.,
69 2004). Previous studies from North Pacific margins as well as open subarctic Pacific
70 have identified drastic variations in export productivity and ocean oxygen levels at
71 millennial and orbital timescales using diverse proxies such as trace elements
72 (Cartapanis et al., 2011; Chang et al., 2014; Jaccard et al., 2009; Zou et al., 2012),
73 benthic foraminiferal assemblages (Ohkushi et al., 2016; Ohkushi et al., 2013;
74 Shibahara et al., 2007) and nitrogen isotopic composition ($\delta^{15}\text{N}$) of organic matter
75 (Addison et al., 2012; Chang et al., 2014; Galbraith et al., 2004; Riethdorf et al., 2016)
76 in marine sediment cores. These studies suggested that both North Pacific
77 Intermediate Water (NPIW) and export of organic matter regulate the sedimentary
78 oxygenation variation during the last glaciation and Holocene in the subarctic Pacific.
79 By contrast, little information exists on millennial-scale oxygenation changes to date
80 in the western subtropical North Pacific (WSTNP).

81 The modern NPIW precursor waters are mainly sourced from the NW Pacific
82 marginal seas (Shcherbina et al., 2003; Talley, 1993; You et al., 2000), spreading into
83 the subtropical North Pacific at intermediate depths of 300 to 800 m (Talley, 1993).
84 The pathway and circulation of NPIW have been identified by You (2003), who
85 suggested that cabbeling, a mixing process to form a new water mass with increased
86 density than that of parent water masses, is the principle mechanism responsible for
87 transforming subpolar source waters into subtropical NPIW along the
88 subarctic-tropical frontal zone. More specifically, a lower subpolar input of about 2 Sv
89 (1 Sv = $10^6 \text{ m}^3/\text{s}$) is sufficient for subtropical ventilation (You et al., 2003). Benthic

90 foraminiferal $\delta^{13}\text{C}$, a quasi-conservative tracer for water mass, from the North Pacific
91 indicates an enhanced ventilation (higher $\delta^{13}\text{C}$) at water depths of < 2000 m during
92 the last glacial period (Keigwin, 1998; Matsumoto et al., 2002). Furthermore, on the
93 basis of both radiocarbon data and modeling results, Okazaki et al. (2010) suggested
94 the formation of deep water in the North Pacific during the early deglaciation in
95 Heinrich Stadial 1 (HS1). Enhanced NPIW penetration was further explored using
96 numerical model simulations (Chikamoto et al., 2012; Gong et al., 2019; Okazaki et
97 al., 2010). In contrast, substantial effects of intensified NPIW formation during
98 Marine Isotope Stage (MIS) 2 and 6 on the ventilation and nutrient characteristics of
99 lower latitude mid-depth Eastern Equatorial Pacific have been suggested by recent
100 studies (Max et al., 2017; Rippert et al., 2017). The downstream effects of intensified
101 NPIW are also reflected in the record of $\delta^{13}\text{C}$ of *Cibicides wuellerstorfi* in core PN-3
102 from the middle Okinawa Trough (OT), where lower deglacial $\delta^{13}\text{C}$ values were
103 attributed to enhanced OC accumulation rates due to higher surface productivity by
104 (Wahyudi and Minagawa, 1997).

105 The Okinawa Trough is separated from the Philippine Sea by the Ryukyu Islands
106 and is an important channel of the northern extension of the Kuroshio in the WSTNP
107 (Figure 1). Initially the OT opened at the middle Miocene (Sibuet et al., 1987) and
108 since then, it has been a depositional center in the East China Sea (ECS), receiving
109 large sediment supplies from nearby rivers (Chang et al., 2009). Surface
110 oceanographic characteristics of the OT over glacial-interglacial cycles are largely
111 influenced by the Kuroshio and ECS Coastal Water (Shi et al., 2014); the latter is
112 related to the strength of summer East Asian monsoon (EAM) sourced from the
113 western tropical Pacific. Modern physical oceanographic investigations showed that
114 intermediate waters in the OT are mainly derived from horizontal advection and
115 mixing of NPIW and South China Sea Intermediate Water (Nakamura et al., 2013).
116 These waters intrude into the OT through two ways: (i) deeper part of the Kuroshio
117 enters the OT through the channel east of Taiwan (sill depth 775 m) and (ii) through
118 the Kerama Gap (sill depth 1100 m). In the northern OT, the subsurface water mainly
119 flows through horizontal advection through the Kerama Gap from the Philippine Sea

120 (Nakamura et al., 2013). Recently, Nishina et al. (2016) found that an overflow
121 through the Kerama Gap controls the modern deep-water ventilation in the southern
122 OT.

123 Both surface characteristics and deep ventilation in the OT varied significantly
124 since the last glaciation. During the last glacial period, the mainstream of the
125 Kuroshio likely migrated to the east of the Ryukyu Islands or also became weaker due
126 to lower sea levels (Shi et al., 2014; Ujiié and Ujiié, 1999; Ujiié et al., 2003) and the
127 hypothetical emergence of a Ryukyu-Taiwan land bridge (Ujiié and Ujiié, 1999). In a
128 recent study, based on the Mg/Ca-derived temperatures in surface and thermocline
129 waters, and planktic foraminiferal indicators of water masses from two sediment cores
130 located in the northern and southern OT, Ujiié et al. (2016) argued that the
131 hydrological conditions of the North Pacific Subtropical Gyre since MIS 7 is
132 modulated by the interaction between the Kuroshio and the NPIW. Besides the
133 Kuroshio, the flux of East Asian rivers to the ECS, which is related to the summer
134 EAM and the sea level oscillations coupled with topography have also been regulating
135 the surface hydrography in the OT (Chang et al., 2009; Kubota et al., 2010; Sun et al.,
136 2005; Yu et al., 2009).

137 Based on benthic foraminiferal assemblages, previous studies have implied a
138 reduced oxygenation in deep waters of the middle and southern OT during the last
139 deglacial period (Jian et al., 1996; Li et al., 2005), but a strong ventilation during the
140 Last Glacial Maximum (LGM) and the Holocene (Jian et al., 1996; Kao et al., 2005).
141 High sedimentary $\delta^{15}\text{N}$ values, an indicator of increased denitrification in the
142 subsurface water column, also occurred during the late deglaciation in the middle OT
143 (Kao et al., 2008). Inconsistent with these results, Dou et al. (2015) suggested an oxic
144 depositional environment during the last deglaciation in the southern OT based on
145 weak positive cerium anomalies. Furthermore, Kao et al. (2006) hypothesized a
146 reduced ventilation of deepwater in the OT during the LGM due to the reduction of
147 KC inflow using a 3-D ocean model. Thus, the patterns and reasons that caused
148 sedimentary oxygenation in the OT remain controversial.

149 **2. Paleo-redox proxies**

150 The sedimentary redox conditions are governed by the rate of oxygen supply from
151 the overlying bottom water and the rate of oxygen removal from pore water (Jaccard
152 et al., 2016), processes that are related to the supply of oxygen by ocean circulation
153 and organic matter respiration, respectively. Contrasting geochemical behaviors of
154 redox-sensitive trace metals (Mn, Mo, U, etc.) have been used to reconstruct bottom
155 water and sedimentary oxygen changes (Algeo, 2004; Algeo and Lyons, 2006;
156 Crusius et al., 1996; Dean et al., 1997; Tribovillard et al., 2006; Zou et al., 2012), as
157 their concentrations readily respond to redox condition of the depositional
158 environment (Morford and Emerson, 1999).

159 In general, enrichment of Mn with higher speciation states (Mn (III) and Mn (IV))
160 in the form of Mn-oxide coatings is observed in marine sediments, when oxic
161 conditions prevail into greater sediment depths as a result of low organic matter
162 degradation rates and well-ventilated bottom water (Burdige, 1993). Under reducing
163 conditions, the authigenic fraction of Mn (as opposed to its detrital background) is
164 released as dissolved Mn (II) species into the pore water and thus its concentration is
165 usually low in suboxic (O_2 and HS^- absent) and anoxic (HS^- present) sediments. In
166 addition, when Mn enrichment occurs in oxic sediments as solid phase Mn
167 oxyhydroxides, it may lead to co-precipitation of other elements, such as Mo
168 (Nameroff et al., 2002).

169 The elements Mo and U behave conservatively in oxygenated seawater, but are
170 preferentially enriched in oxygen-depleted water (Morford and Emerson, 1999).
171 However, these two trace metals behave differently in several ways. Molybdenum can
172 be enriched in both oxic sediments, such as the near surface manganese-rich horizons
173 in continental margin environments (Shimmield and Price, 1986) and in anoxic
174 sediments (Nameroff et al., 2002). Under anoxic conditions, Mo can be reduced either
175 from the +6 oxidation state to insoluble MoS_2 , though this process is known to occur
176 only under extremely reducing conditions, such as hydrothermal and/or diagenesis
177 (Dahl et al., 2010; Helz et al., 1996) or be converted to particle-reactive
178 thiomolybdates (Vorlichek and Helz, 2002). Zheng et al. (2000) suggested two critical
179 thresholds for Mo scavenging from seawater: 0.1 μM hydrogen sulfide (H_2S) for

180 Fe-S-Mo co-precipitation and 100 μM H_2S for Mo scavenging as Mo-S or as
181 particle-bound Mo without Fe. Although Crusius et al. (1996) noted insignificant
182 enrichment of sedimentary Mo under suboxic conditions, Scott et al. (2008) argued
183 that burial flux of Mo is not so low in suboxic environments. Excess concentration of
184 Mo ($\text{Mo}_{\text{excess}}$) in sediments thus suggests the accumulation of sediments either in
185 anoxic (H_2S occurrence) or well oxygenated conditions (if $\text{Mo}_{\text{excess}}$ is in association
186 with Mn-oxides).

187 In general, U is enriched in anoxic sediments ($>1 \mu\text{M} \text{ H}_2\text{S}$), but not in oxic
188 sediments ($>10 \mu\text{M} \text{ O}_2$) (Nameroff et al., 2002). Accumulation of U depends on the
189 content of reactive organic matter (Sundby et al., 2004) and U precipitates as uraninite
190 (UO_2) during the conversion of Fe (III) to Fe (II) in suboxic conditions (Morford and
191 Emerson, 1999; Zheng et al., 2002). One of the primary removal mechanisms for U
192 from the ocean is via diffusion across the sediment-water interface of reducing
193 sediments (Klinkhammer and Palmer, 1991). Under suboxic conditions, soluble U (VI)
194 is reduced to insoluble U (IV), but free sulfide is not required for U precipitation
195 (McManus et al., 2005). Jaccard et al. (2009) suggested that the presence of excess
196 concentration of U (U_{excess}) in the absence of Mo enrichment is indicative of a suboxic,
197 but not sulfidic condition, within the diffusional range of the sediment-water interface.
198 The felsic volcanism is also a primary source of uranium (Maithani and Srinivasan,
199 2011). Therefore, the potential input of uranium from active volcanic sources around
200 the northwestern Pacific to the adjacent sediments should not be neglected.

201 In this study, we investigate a suite of redox-sensitive elements and the ratio of
202 Mo/Mn along with productivity proxies from a sediment core retrieved from the
203 northern OT to reconstruct the sedimentary oxygenation in the WSTNP over the last
204 50ka. Based on that, we propose that multiple factors, such as NPIW ventilation, the
205 strength of the Kuroshio Current and export productivity, control the bottom
206 sedimentary oxygenation in the OT on millennial timescales since the last glacial.

207 **3. Oceanographic setting**

208 Surface hydrographic characteristics of the OT are mainly controlled by the
209 warmer, more saline, oligotrophic Kuroshio water and cooler, less saline, nutrient-rich

210 Changjiang Diluted Water, and the modern flow-path of the former is influenced by
211 the bathymetry of the OT (Figure 1a). The Kuroshio Current originates from the
212 North Equatorial Current and flows into the ECS from the Philippine Sea through the
213 Suao-Yonaguni Depression. In the northern OT, Tsushima Warm Current (TWC), a
214 branch of the Kuroshio, flows into the Japan Sea through the shallow Tsushima Strait.
215 Volume transport of the Kuroshio varies seasonally due to the influence of the EAM
216 with a maximum of 24 Sv in summer and a minimum of 20 Sv in autumn across the
217 east of Taiwan (Qu and Lukas, 2003).

218 A lower sea surface salinity (SSS) zone in summer relative to the one in winter in
219 the ECS migrates toward the east of OT, indicating enhanced impact of the
220 Changjiang discharge associated with summer EAM (Figures 2a and b). An estimated
221 ~80% of the mean annual discharge of the river Changjiang is supplied to the ECS
222 (Ichikawa and Beardsley, 2002) and in situ observational data show a pronounced
223 negative correlation between the Changjiang discharge and SSS in July (Delcroix and
224 Murtugudde, 2002). Consistently, previous studies from the OT reported such close
225 relationship between summer EAM and SSS back to the late Pleistocene (Chang et al.,
226 2009; Clemens et al., 2018; Kubota et al., 2010; Sun et al., 2005).

227 Despite the effects of EAM and the Kuroshio, evidence of geochemical tracers
228 (temperature, salinity, oxygen, nutrients and radiocarbon) collected during the World
229 Ocean Circulation Experiment (WOCE) in the Pacific (transects P24 and P03) favors
230 the presence of low salinity, nutrient-enriched intermediate and deep waters (Talley,
231 2007). Dissolved oxygen content is <100 $\mu\text{mol/kg}$ at water depths below 600 m in the
232 OT, along WOCE transects PC03 and PC24 (Talley, 2007). Modern oceanographic
233 observations at the Kerama Gap reveal that upwelling in the OT is associated with the
234 inflow of NPIW and studies using a box model predicted that overflow through the
235 Kerama Gap is responsible for upwelling ($3.8\text{--}7.6 \times 10^{-6} \text{m s}^{-1}$) (Nakamura et al.,
236 2013; Nishina et al., 2016).

237 **4. Materials and methods**

238 **4.1. Chronostratigraphy of core CSH1**

239 A 17.3 m long sediment core CSH1 ($31^\circ 13.7' \text{N}$, $128^\circ 43.4' \text{E}$; water depth: 703

240 m) was collected from the northern OT, close to the main stream of Tsushima Warm
241 Current (TWC) (Figure 1b) and within the depth of NPIW (Figure 1c) using a piston
242 corer during *Xiangyanghong09* Cruise in 1998, carried out by the First Institute of
243 Oceanography, Ministry of Natural Resources of China. This location is enabling us
244 to reconstruct millennial-scale changes in the properties of TWC and NPIW. Core
245 CSH1 mainly consists of clayey silt and silt with occurrence of plant debris at some
246 depth intervals (Ge et al., 2007) (Figure 3a). In addition, three layers of volcanic ash
247 were observed at depths of 74–106 cm, 782–794 cm, 1570–1602 cm. These three
248 intervals can be correlated with well-known ash layers, Kikai-Akahoya (K-Ah; 7.3
249 ka), Aira-Tanzawa (AT; 29.24 ka) and Aso-4 (roughly around MIS 5a) (Machida,
250 1999), respectively. The core was split and sub-sampled at 4 cm interval and then
251 stored in the China Ocean Sample Repository at 4 °C until analysis.

252 Previously, paleoceanographic studies have been conducted and a set of data has
253 been investigated for core CSH1, including the contents of planktic foraminifers as
254 well as their carbon ($\delta^{13}\text{C}$) and oxygen isotope ($\delta^{18}\text{O}$) compositions (Shi et al., 2014),
255 pollen (Chen et al., 2006), paleomagnetism (Ge et al., 2007) and CaCO_3 (Wu et al.,
256 2004). An age model for this core has been constructed by using ten Accelerator Mass
257 Spectrometry (AMS) ^{14}C dates and six oxygen isotope ($\delta^{18}\text{O}$) age control points. The
258 whole 17.3 m core contains *ca.* 88 ka-long record of continuous sedimentation (Shi et
259 al., 2014).

260 Notably, the original age model, which used constant radiocarbon reservoir ages
261 throughout core CSH1 are suitable to reveal orbital-scale Kuroshio variations (Shi et
262 al., 2014), but insufficient to investigate millennial-scale climatic events. A higher
263 abundance of *Neogloboquadrina pachyderma (dextral)*, e. g. that occurred during
264 warmer intervals, such as the Bölling-Alleröd (B/A), has been challenging to explain.
265 On the other hand, paired measurements of $^{14}\text{C}/^{12}\text{C}$ and ^{230}Th ages from Hulu Cave
266 stalagmites suggest magnetic field changes have greatly contributed to high
267 atmospheric $^{14}\text{C}/^{12}\text{C}$ values at HS4 and the Younger Dryas (YD) (Cheng et al., 2018).
268 Thus a constant reservoir age ($\Delta\text{R}=0$) assumed when calibrating foraminiferal
269 radiocarbon dates using CALIB 6 software and the Marine 13 calibration dataset

270 (Reimer et al., 2013) for core CSH1 may cause large chronological uncertainties.

271 Here, we recalibrated the radiocarbon dates using updated CALIB 7.04 software
272 with Marine 13 calibration dataset (Reimer et al., 2013). Moreover, on the basis of
273 significant correlation between planktic foraminifera species *Globigerinoides ruber*
274 $\delta^{18}\text{O}$ and Chinese stalagmite $\delta^{18}\text{O}$ (Cheng et al., 2016), a proxy of summer EAM
275 related to SSS of the ECS, we improve the age model for core CSH1 (Figures 3b-d).
276 Overall, the new chronological framework is similar to the one previously reported by
277 Shi et al. (2014), but with more dates. In order to compare with published results
278 associated with ventilation changes in the North Pacific, here we mainly report the
279 history of sedimentary oxygenation in the northern OT since the last glacial period.
280 Linear sedimentation rate varied between ~10 and 40 cm/ka with higher
281 sedimentation rates (around 30-40 cm/ka) between ~24 ka and 32.5 ka. The new age
282 control points are shown in Table 2.

283 **4.2. Chemical analyses**

284 Sediment subsamples for geochemical analyses were freeze-dried and ground to
285 a fine powder with an agate mortar and pestle. Based on the age model, 85
286 subsamples from core CSH1, representing a temporal resolution of about 600 years
287 (every 4 cm interval) were selected for detailed geochemical analyses of major and
288 minor elements, and total carbon (TC), organic carbon (TOC) and nitrogen (TN)
289 contents. The pretreatment of sediment and other analytical methods have been
290 reported elsewhere (Zou et al., 2012).

291 TC and TN were determined with an elemental analyzer (EA; Vario EL III,
292 Elementar Analysen systeme GmbH) in the Key Laboratory of Marine Sediment and
293 Environment Geology, First Institute of Oceanography, Ministry of Natural Resources
294 of China, Qingdao. Carbonate was removed from sediments by adding 1M HCl to the
295 homogenized sediments for total organic carbon (TOC) analysis using the same
296 equipment. The content of calcium carbonate (CaCO_3) was calculated using the
297 equation:

298
$$\text{CaCO}_3 = (\text{TC} - \text{TOC}) \times 8.33$$

299 where 8.33 is the ratio between the molecular weight of carbonate and the atomic

300 weight of carbon. National reference material (GSD-9), blank sample and replicated
301 samples were used to control the analytical process. The relative standard deviation of
302 the GSD-9 for TC, TN and TOC is $\leq 3.4\%$.

303 About 0.5 g of sediment powder was digested in double distilled HF:HNO₃ (3:1),
304 followed by concentrated HClO₄, and then re-dissolved in 5% HNO₃. Selected major
305 and minor elements such as aluminum (Al) and manganese (Mn) were determined by
306 inductively coupled plasma optical emission spectroscopy (ICP-OES; Thermo
307 Scientific iCAP 6000, Thermo Fisher Scientific), as detailed elsewhere (Zou et al.,
308 2012). In addition, Mo and U were analyzed with inductively coupled plasma mass
309 spectrometry (ICP-MS; Thermo Scientific XSERIES 2, Thermo Fisher Scientific), as
310 described in Zou et al. (2012). Precision for most elements in the reference material
311 GSD-9 is $\leq 5\%$ relative standard deviation. The excess fractions of U and Mo were
312 estimated by normalization to Al:

313 Excess fraction = total_{element}— (element/Al_{average shale} \times Al), with U/Al_{average shale} =
314 0.307×10^{-6} and Mo/Al_{average shale} = 0.295×10^{-6} (Li and Schoonmaker, 2014).

315 In addition, given the different geochemical behaviors of Mn and Mo and
316 co-precipitation and adsorption processes associated with the redox cycling of Mn, we
317 calculated the ratio of Mo to Mn, assuming that higher Mo/Mn ratio indicates lower
318 oxygen content in the depositional environment and vice versa. In combination with
319 the concentration of excess uranium, we infer the history of sedimentary oxygenation
320 in the subtropical North Pacific since the last glaciation.

321 **5. Results**

322 **5.1. TOC, TN, and CaCO₃**

323 The content of CaCO₃ varies from 8.8 to 35% (Figure 4a) and it mostly shows
324 higher values with increasing trends during the last deglaciation. In contrast, the
325 content of CaCO₃ is low and exhibits decreasing trends during the late MIS 3 and the
326 LGM (Figure 4a). TN content shows a larger variation compared to TOC (Figure 4b),
327 but it still strongly correlates with TOC ($r = 0.74$, $p < 0.01$) throughout the entire core.
328 Concentration of TOC ranges from 0.5 to 2.1% and it shows higher values with stable
329 trends during the last glacial phase (MIS 3) (Figure 4c). Molar ratios of TOC/TN vary

330 around 10, with higher ratios at the transition into the LGM (Figure 4d),
331 corresponding to higher linear sedimentation rate (Figure 4e).

332 Both TOC and CaCO_3 have been used as proxies for the reconstruction of past
333 export productivity (Cartapanis et al., 2011; Lembke-Jene et al., 2017; Röhleman et
334 al., 1999). Molar C/N ratios of >10 (Figure 4c) suggest that terrigenous organic
335 sources significantly contribute to the TOC concentration in core CSH1. The TOC
336 content therefore may be not a reliable proxy for the reconstruction of surface water
337 export productivity during times of the LGM and late deglaciation, when maxima in
338 C/N ratios co-occur with decoupled trends between CaCO_3 and TOC concentrations.

339 Several lines of evidence support CaCO_3 as a reliable productivity proxy,
340 particularly during the last deglaciation. The strong negative correlation coefficient (r
341 = -0.85 , $p<0.01$) between Al and CaCO_3 in sediments throughout core CSH1
342 confirms the biogenic origin of CaCO_3 against terrigenous Al (Figure 4f). Generally,
343 terrigenous dilution decreases the concentrations of CaCO_3 . An inconsistent
344 relationship between CaCO_3 contents and sedimentation rates indicates a minor effect
345 of dilution on CaCO_3 . Furthermore, the increasing trend in CaCO_3 associated with
346 high sedimentation rate during the last deglacial interval indicates a substantial
347 increase in export productivity (Figures 4a and d). The high coherence between
348 CaCO_3 content and alkenone-derived sea surface water (SST) (Shi et al., 2014)
349 indicates a direct control on CaCO_3 by SST. Moreover, a detailed comparison between
350 CaCO_3 concentrations and the previously published foraminiferal fragmentation ratio
351 (Wu et al., 2004) shows, apart from a small portion within the LGM, no clear
352 co-variation between them. These evidence suggest that CaCO_3 changes are driven
353 primarily by variations in carbonate primary production, and not overprinted by
354 secondary processes, such as carbonate dissolution through changes in the lysocline
355 depth and dilution by terrigenous materials. Likewise, a similar deglacial trend in
356 CaCO_3 is also observed in core MD01-2404 (Chang et al., 2009), indicating a
357 ubiquitous, not local picture in the OT. All these lines of evidence thus support CaCO_3
358 of core CSH1 as a reliable productivity proxy to a first order approximation.

359 **5.2. Redox-sensitive Elements**

Figure 4 shows time series of selected redox-sensitive elements (RSEs) and proxies derived from them. Mn shows higher concentrations during the LGM and HS1 (16 ka–22.5 ka) and middle-late Holocene, but lower concentrations during the last deglacial and Preboreal periods (15.8 ka–9.5 ka, Figure 4g). Generally, concentrations of excess Mo and excess U (Figures 4j and l) show coherent patterns with those of Mo and U (Figures 4i and k), but both are out-of-phase with Mn over the last glacial period (Figure 4h). Pronounced variations in U concentration after 8.5 ka are related to the occurrence of discrete volcanic materials. A significant positive Eu anomaly (Zhu et al., 2015) confirms the occurrence of discrete volcanic materials and its dilution effects on terrigenous components since 7 ka. Occurrence of discrete volcanic material is likely related to intensified Kuroshio Current during the mid-late Holocene, as supported by higher hydrothermal Hg concentrations in sediments from the middle OT (Lim et al., 2017). A negative correlation between Mn and Mo_{excess} during the last glaciation and the Holocene, and the strong positive correlation between them during the LGM and HS1 (Figures 5a and b) further corroborate the complex geochemical behaviors of Mn and Mo. A strong positive correlation between Mo_{excess} and Mn (Figure 5b) may be attributed to co-precipitation of Mo by Mn-oxyhydroxide under oxygenated conditions. Here, we thus use the Mo/Mn ratio, instead of excess Mo concentration to reconstruct variations in sedimentary redox conditions in our study area. Overall, the Mo/Mn ratio shows similar downcore pattern to that of Mo_{excess} with higher ratios during the last deglaciation, but lower ratios during the LGM and HS1. A strong correlation ($r = 0.69$) between Mo/Mn ratio and excess U concentration (excluding the data of Holocene, due to the contamination with volcanic material, Figure 5c) further corroborates the integrity of Mo/Mn as an indicator of sedimentary oxygenation changes.

Rapidly decreasing Mo/Mn ratios indicate a well oxygenated sedimentary environment after ~8 ka (Figure 4h). Both higher Mo/Mn ratios and excess U concentrations, together with lower Mn concentrations suggest suboxic depositional conditions during the late deglacial period (15.8 ka–9.5 ka), whereas lower ratios during the LGM, HS1 and HS2 indicate relatively better oxygenated sedimentary

390 conditions. A decreasing trend in Mo/Mn ratio and excess U concentration from 50 ka
391 to 25 ka also suggest higher sedimentary oxygen levels.

392 **6.Discussion**

393 **6.1. Constraining paleoredox conditions in the Okinawa Trough**

394 In general, three different terms, hypoxia, suboxia and anoxia, are widely used to
395 describe the degree of oxygen depletion in the marine environment (Hofmann et al.,
396 2011). Here, we adopt the definition of oxygen thresholds by Bianchi et al. (2012) for
397 oxic ($>120 \text{ }\mu\text{mol/kg O}_2$), hypoxic ($<60\text{--}120 \text{ }\mu\text{mol/kg O}_2$) and suboxic ($<2\text{--}10 \text{ }\mu\text{mol/kg O}_2$)
398 conditions, whereas anoxia is the absence of measurable oxygen.

399 Proxies associated with RSEs, such as sedimentary Mo concentration (Lyons et
400 al., 2009; Scott et al., 2008) have been used to constrain the degree of oxygenation in
401 seawater. Algeo and Tribovillard (2009) proposed that open-ocean systems with
402 suboxic waters tend to yield U_{excess} enrichment relative to $\text{Mo}_{\text{excess}}$, resulting in
403 sediment $(\text{Mo}/\text{U})_{\text{excess}}$ ratio less than that of seawater (7.5-7.9). Under increasingly
404 reducing and occasionally sulfidic conditions, the accumulation of $\text{Mo}_{\text{excess}}$ increase
405 relative to that of U_{excess} leading the $(\text{Mo}/\text{U})_{\text{excess}}$ ratio either is equal to or exceeds
406 with that of seawater. Furthermore, Scott and Lyons (2012) suggested a non-euxinic
407 condition with the presence of sulfide in pore waters, when Mo concentrations range
408 from $> 2 \text{ }\mu\text{g/g}$, the crustal average to $< 25 \text{ }\mu\text{g/g}$, a threshold concentration for euxinic
409 condition. Given that the northern OT is located in an open oceanic setting, we use
410 these two proxies to evaluate the degree of oxygenation in sediments.

411 Both bulk Mo concentration (1.2-9.5 $\mu\text{g/g}$) and excess (Mo/U) ratio (0.2-5.7) in
412 core CSH1 suggest that oxygen-depleted conditions have prevailed in the deep water
413 of the northern OT over the last 50 ka (Figure 4m). However, increased excess Mo
414 concentrations with higher Mo/U ratios during the last termination (18 ka-9 ka)
415 indicate more reducing conditions compared to the Holocene and the last glacial
416 period, though Mo concentrations were less than 25 $\mu\text{g/g}$, a threshold for euxinic
417 deposition proposed by Scott and Lyons (2012).

418 The relative abundance of benthic foraminifera species that thrive in different
419 oxygen concentrations has also been widely used to reconstruct variations in bottom

420 water ventilation, such as the enhanced abundance of *Bulimina aculeata*, *Uvigerina*
421 *peregrina* and *Chilostomella oolina* found under oxygen-depleted conditions in the
422 central and southern OT from 18 ka to 9.2 ka (Jian et al., 1996; Li et al., 2005). An
423 oxygenated bottom water condition is also indicated by abundant benthic foraminifera
424 species *Cibicidoides hyalina* and *Globocassidulina subglobosa* after 9.2 ka (Jian et al.,
425 1996; Li et al., 2005) in cores E017 (1826 m water depth) and 255 (1575 m water
426 depth) and high benthic $\delta^{13}\text{C}$ values (Wahyudi and Minagawa, 1997) in core PN-3
427 (1058 m water depth) from the middle and southern OT during the postglacial period.
428 The poorly-ventilated deep water in the middle and southern OT inferred by benthic
429 foraminiferal assemblages during the last deglaciation correlates with the one in the
430 northern OT referring to our RSEs (Figure 4). A link thus can be hypothesized
431 between deep-water ventilation and sedimentary oxygenation in the OT. Overall, a
432 combination of our proxy records of RSEs in core CSH1 with other records shows
433 oxygen-rich conditions during the last glaciation and middle and late Holocene (since
434 8.5 ka) intervals, but oxygen-poor conditions during the last deglaciation.

435 **6.2. Causes for sedimentary oxygenation variations**

436 Our observed pattern of RSEs in core CSH1 suggests that drastic changes in
437 sedimentary oxygenation occurred on orbital and millennial timescales over the last
438 glaciation in the OT. In general, four factors can regulate the redox condition in the
439 deep water column: (i) O_2 solubility, (ii) export productivity and subsequent
440 degradation of organic matter, (iii) vertical mixing, and (iv) lateral supply of oxygen
441 through intermediate and deeper water masses (Ivanochko and Pedersen, 2004;
442 Jaccard and Galbraith, 2012). These processes have been invoked in previous studies
443 to explain the deglacial Pacific-wide variations in oxygenation by either one or a
444 combination of these factors (Galbraith and Jaccard, 2015; Moffitt et al., 2015;
445 Praetorius et al., 2015). Our data also suggest drastic variations in sedimentary
446 oxygenation over the last 50 ka. However, the mechanisms responsible for
447 sedimentary oxygenation variations in the basin-wide OT and its connection with
448 ventilation of the open North Pacific remain unclear. In order to place our core results
449 in a wider regional context, we compare our proxy records of sedimentary

450 oxygenation (U_{excess} concentration and Mo/Mn ratio) and export productivity (CaCO_3)
451 (Figures 6a, b, c) with abundance of *Pulleniatina obliquiloculata* (an indicator of
452 Kuroshio strength) and sea surface temperature (Shi et al., 2014), bulk sedimentary
453 nitrogen isotope (an indicator of denitrification) (Kao et al., 2008), benthic
454 foraminifera $\delta^{13}\text{C}$ (a proxy for ventilation) in cores PN-3 and PC23A (Rella et al.,
455 2012; Wahyudi and Minagawa, 1997), abundance of benthic foraminifera (an
456 indicator of hypoxia) in core E017 (Li et al., 2005) and ODP Site 1017 (Cannariato
457 and Kennett, 1999) (Figures 6d-k).

458 **6.2.1. Effects of regional ocean temperature on deglacial deoxygenation**

459 Warming ocean temperatures lead to lower oxygen solubility. In the geological
460 past, solubility effects connected to temperature changes of the water column thought
461 to enhance or even trigger hypoxia (Praetorius et al., 2015). Shi et al. (2014) reported
462 an increase in SST of around 4°C (from $\sim 21^\circ\text{C}$ to $\sim 24.6^\circ\text{C}$) during the last
463 deglaciation in core CSH1 (Figure 6d). Based on thermal solubility effects, a hypothetical
464 warming of 1°C would reduce oxygen concentrations by about $3.5\text{ }\mu\text{mol/kg}$ at water temperatures around 22°C (Brewer and Peltzer, 2016), therefore a \sim
465 4°C warming at core CSH1 (Shi et al., 2014) could drive a conservative estimate of a
466 drop of $<15\text{ }\mu\text{mol/kg}$ in oxygen concentration, assuming no large salinity changes.
467 However, given the semi-quantitative nature of our data about oxygenation changes,
468 which seemingly exceed an amplitude of $>15\text{ }\mu\text{mol/kg}$, we suggest that other factors,
469 e.g. local changes in export productivity, regional influences such as vertical mixing
470 due to changes of the Kuroshio Current, and far-field effects may have played
471 decisive roles in shaping the oxygenation history of the OT.

473 **6.2.2. Links between deglacial primary productivity and sedimentary**
474 **deoxygenation**

475 Previous studies have suggested the occurrence of high primary productivity in
476 the entire OT during the last deglacial period (Chang et al., 2009; Jian et al., 1996;
477 Kao et al., 2008; Li et al., 2017; Shao et al., 2016; Wahyudi and Minagawa, 1997).
478 Such an increase in export production was due to favorable conditions for
479 phytoplankton blooms, which were likely induced by warm temperatures and maxima

480 in nutrient availability, the latter being mainly sourced from increased discharge of
481 the Changjiang River, erosion of material from the ongoing flooding of the shallow
482 continental shelf in the ECS, and upwelling of Kuroshio Intermediate Water (Chang
483 et al., 2009; Li et al., 2017; Shao et al., 2016; Wahyudi and Minagawa, 1997). On the
484 basis of sedimentary reactive phosphorus concentration, Li et al. (2017) concluded
485 that export productivity increased during warm episodes but decreased during cold
486 spells on millennial timescales over the last 91 ka in the OT. Gradually increasing
487 concentrations of CaCO_3 in core CSH1 during the deglaciation (Figure 6a) and little
488 changes in foraminiferal fragmentation ratios (Wu et al., 2004), are indicative of high
489 export productivity in the northern OT. Accordingly, our data indicate that an increase
490 in export productivity during the last deglaciation, which was previously evidenced by
491 concentrations of reactive phosphorus (Li et al., 2017) and CaCO_3 (Chang et al., 2009)
492 from the middle OT, and thus was a pervasive, synchronous phenomenon in the entire
493 study region at the outermost extension of the ECS.

494 Similar events of high export productivity have been reported in the entire North
495 Pacific due to increased nutrient supply, high SST, reduced sea ice cover, etc.
496 (Crusius et al., 2004; Dean et al., 1997; Galbraith et al., 2007; Jaccard and Galbraith,
497 2012; Kohfeld and Chase, 2011). In most of these cases, increased productivity were
498 thought to be responsible for oxygen depletion in mid-depth waters, due to
499 exceptionally high oxygen consumption. However, the productivity changes during
500 the deglacial interval, very specifically CaCO_3 , are not fully consistent with the trends
501 of excess U and Mo/Mn ratio (Figures 6b and c). The sedimentary oxygenation thus
502 cannot be determined by export productivity alone.

503 **6.2.3 Effects of the Kuroshio dynamics on sedimentary oxygenation**

504 The Kuroshio Current, one of the main drivers of vertical mixing, has been
505 identified as the key factor in controlling modern deep ventilation in the OT (Kao et
506 al., 2006). However, the flow path of the Kuroshio in the OT during the glacial
507 interval remains a matter of debate. Planktic foraminiferal assemblages in sediment
508 cores from inside and outside the OT indicated that the Kuroshio migrated to the east
509 of the Ryukyu Islands during the LGM (Ujiié and Ujiié, 1999). Subsequently, Kao et

510 al. (2006) based on modeling results suggested that the Kuroshio still enters the OT,
511 but the volume transport was reduced by 43% compared to the present-day transport
512 and the outlet of Kuroshio switches from the Tokara Strait to the Kerama Gap at -80
513 and -135m lowered sea level. Combined with sea surface temperature (SST) records
514 and ocean model results, Lee et al. (2013) argued that there was little effect of
515 deglacial sea-level change on the path of the Kuroshio, which still exited the OT from
516 the Tokara Strait during the glacial period. Because the main stream of the Kuroshio
517 Current is at a water depth of ~150 m, the SST records are insufficient to decipher past
518 changes of the Kuroshio (Ujié et al., 2016). On the other hand, low abundances of *P.*
519 *obliquiloculata* in core CSH1 in the northern OT (Figure 6e) indicate that the main
520 flow path of the Kuroshio migrated to the east of the Ryukyu Island (Shi et al., 2014).
521 Such a flow change would have been caused by the proposed block of the
522 Ryukyu-Taiwan land bridge by low sea level (Ujié and Ujié, 1999) and an overall
523 reduced Kuroshio intensity (Kao et al., 2006), effectively suppressing the effect of the
524 Kuroshio on deep ventilation in the OT. Our RSEs data show that oxygenated
525 sedimentary conditions were dominant in the northern OT throughout the last glacial
526 period (Figures 6b, c). The Kuroshio thus likely had a weak or even no effect on the
527 renewal of oxygen to the sedimentary environment during the last glacial period.
528 More recently, lower hydrothermal total Hg concentration during 20 ka - 9.6 ka,
529 associated with reduced intensity and/or variation in flow path of KC, relative to that
530 of Holocene recorded in core KX12 - 3 (1423 water depth) (Lim et al., 2017), further
531 validates our inference.

532 On the other hand, the gradually increased alkenone-derived SST and abundance
533 of *P. obliquiloculata* (Figures 6d and e) from 15 ka onwards indicates an intensified
534 Kuroshio Current. At present, mooring and float observations revealed that the KC
535 penetrates to 1200 m isobath in the East China Sea (Andres et al., 2015). However,
536 the effect of Kuroshio on sedimentary oxygenation was likely very limited during the
537 glacial period and only gradually increasing throughout the last glacial termination.
538 Therefore, while its effect on our observed deglacial variation in oxygenation may
539 provide a slowly changing background condition in vertical mixing effects on the

540 sedimentary oxygenation in the OT, it cannot account for the first order, rapid
541 oxygenation changes, including indications for millennial-scale variations, that we
542 observe between 18 ka and 9 ka.

543 Better oxygenated sedimentary conditions since 8.5 ka coincided with intensified
544 Kuroshio (Li et al., 2005; Shi et al., 2014), as indicated by rapidly increased SST and
545 *P. obliquiloculata* abundance in core CSH1 (Figures 6d and e) and *C.hyalinea*
546 abundance in core E017 (Figure 6i). Re-entrance of the Kuroshio into the OT (Shi et
547 al., 2014) with rising eustatic sea level likely enhanced the vertical mixing and
548 exchange between bottom and surface waters, ventilating the deep water in the OT.
549 Previous comparative studies based on epibenthic $\delta^{13}\text{C}$ values indicated
550 well-ventilated deep water feeding both inside the OT and outside off the Ryukyu
551 Islands during the Holocene (Kubota et al., 2015; Wahyudi and Minagawa, 1997). In
552 summary, enhanced sedimentary oxygenation regime observed in the OT during the
553 Holocene is mainly related to the intensified Kuroshio, while the effect of the
554 Kuroshio on OT oxygenation was limited before 15 ka.

555 **6.2.4. Effects of GNPIW on sedimentary oxygenation**

556 Relatively stronger oxygenated Glacial North Pacific Intermediate Water
557 (GNPIW), coined by (Matsumoto et al., 2002), has been widely documented in the
558 Bering Sea (Itaki et al., 2012; Kim et al., 2011; Rella et al., 2012), the Okhotsk Sea
559 (Itaki et al., 2008; Okazaki et al., 2014; Okazaki et al., 2006; Wu et al., 2014), off east
560 Japan (Shibahara et al., 2007), the eastern North Pacific (Cartapanis et al., 2011;
561 Ohkushi et al., 2013) and western subarctic Pacific (Keigwin, 1998; Matsumoto et al.,
562 2002). The intensified formation of GNPIW due to additional source region in the
563 Bering Sea was proposed by Ohkushi et al. (2003) and Horikawa et al. (2010). Under
564 such conditions, the invasion of well-ventilated GNPIW into the OT through the
565 Kerama Gap would have replenished the water column oxygen in the OT, although
566 the penetration depth of GNPIW remains under debate (Jaccard and Galbraith, 2013;
567 Max et al., 2014; Okazaki et al., 2010; Rae et al., 2014). Both a gradual decrease in
568 excess U concentration and an increase in Mo/Mn ratio during the last glacial period
569 (25 ka-50 ka) validate such inference, suggesting pronounced effects of intensified

570 NPIW formation in the OT.

571 During HS1, a stronger formation of GNPIW was supported by proxy studies
572 and numerical simulations. For example, on the basis of paired benthic-planktic (B-P)
573 ^{14}C data, enhanced penetration of NPIW into a much deeper water depth during HS1
574 relative to the Holocene has been revealed in several studies (Max et al., 2014;
575 Okazaki et al., 2010; Sagawa and Ikehara, 2008), which was also simulated by several
576 models (Chikamoto et al., 2012; Gong et al., 2019; Okazaki et al., 2010). On the other
577 hand, increased intermediate water temperature in the subtropical Pacific recorded in
578 core GH08-2004 (1166 m water depth) (Kubota et al., 2015) and young deep water
579 observed in the northern South China Sea during HS1 (Wan and Jian, 2014) along
580 downstream region of NPIW are also related to intensified NPIW formation.
581 Furthermore, the pathway of GNPIW from numerical model simulations (Zheng et al.,
582 2016) was similar to modern observations (You, 2003). Thus, all these evidence imply
583 a persistent, cause and effect relation between GNPIW ventilation, the intermediate
584 and deep water oxygen concentration in the OT and sediment redox state during HS1.
585 In addition, our RSEs data also suggested a similarly enhanced ventilation in HS2
586 (Figures 6b and c) that is also attributed to intensified GNPIW formation.

587 Hypoxic conditions during the B/A have been also widely observed in the mid-
588 and high-latitude North Pacific (Jaccard and Galbraith, 2012; Praetorius et al., 2015).
589 Our data of excess U concentration and Mo/Mn ratio recorded in core CSH1 (Figures
590 6b and c), together with enhanced denitrification and *B.aculeata* abundance (Figures
591 6f and h), further reveal the expansion of oxygen-depletion at mid-depth waters down
592 to the subtropical NW Pacific during the late deglacial period. Based on high relative
593 abundances of radiolarian species, indicators of upper intermediate water ventilation
594 in core PC-23A, Itaki et al. (2012) suggested that a presence of well-ventilated waters
595 was limited to the upper intermediate layer (200 m–500 m) in the Bering Sea during
596 warm periods, such as the B/A and Preboreal. Higher B-P foraminiferal ^{14}C ages,
597 together with increased temperature and salinity at intermediate waters recorded in
598 core GH02-1030 (off East Japan) supported a weakened formation of NPIW during
599 the B/A (Sagawa and Ikehara, 2008). These lines of evidence indicate that the

boundary between GNPIW and North Pacific Deep Water shoaled during the B/A, in comparison to HS1. Based on a comparison of two benthic foraminiferal oxygen and carbon isotope records from off northern Japan and the southern Ryukyu Island, Kubota et al. (2015) found a stronger influence of Pacific Deep Water on intermediate-water temperature and ventilation at their southern than the northern locations, though both sites are located at similar water depths (1166 m and 1212 m for cores GH08-2004 and GH02-1030, respectively). Higher excess U concentration and low Mo/Mn ratio in our core CSH1 during the B/A and Preboreal suggest reduced sedimentary oxygenation, consistent with reduced ventilation of GNPIW, contributing to the subsurface water deoxygenation in the OT.

During the YD, both Mo/Mn ratio and excess U show a slightly decreased oxygen condition in the northern OT. By contrast, benthic foraminiferal $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values in a sediment core collected from the Oyashio region suggested a strengthened formation and ventilation of GNPIW during the YD (Ohkushi et al., 2016). This pattern possibly indicates a time-dependent, varying contribution of distal GNPIW to the deglacial OT oxygenation history, and we presume a more pronounced contribution of organic matter degradation due to high export productivity during this period, as suggested by increasing CaCO_3 content.

6.3. Subtropical North Pacific ventilation links to North Atlantic Climate

One of the characteristic climate features in the Northern Hemisphere, in particular the North Atlantic is millennial-scale oscillation during glacial and deglacial periods. These abrupt climatic events have been widely thought to be closely related to varying strength of Atlantic Meridional Overturning Circulation (AMOC) (Lynch-Stieglitz, 2017). One of dynamic proxies of ocean circulation, $^{231}\text{Pa}/^{230}\text{Th}$ reveals that severe weakening of AMOC only existed during Heinrich stadials due to increased freshwater discharges into the North Atlantic (Böhm et al., 2015; McManus et al., 2004). On the other hand, several mechanisms, such as sudden termination of freshwater input (Liu et al., 2009), atmospheric CO_2 concentration (Zhang et al., 2017), enhanced advection of salt (Barker et al., 2010) and changes in background climate (Knorr and Lohmann, 2007) were proposed to explain the reinvigoration of

630 AMOC during the B/A.

631 Our RSEs data in the Northern OT and endobenthic $\delta^{13}\text{C}$ in the Bering Sea
632 (Figures 7a-c) both show a substantial millennial variability in intermediate water
633 ventilation in the subtropical North Pacific. Notably, enhanced ventilation during HS1
634 and HS2 and oxygen-poor condition during the B/A respectively correspond to the
635 collapse and resumption of AMOC (Figure 7d). Such out-of-phase millennial-scale
636 pattern is consistent with the results of various modeling simulations (Chikamoto et
637 al., 2012; Menzel et al., 2014; Okazaki et al., 2010; Saenko et al., 2004), although
638 these models had different boundary conditions and causes for the observed effects in
639 GNPIW formation, and ventilation ages derived from B-P ^{14}C (Freeman et al., 2015;
640 Max et al., 2014; Okazaki et al., 2012). These lines of evidence confirm a persistent
641 link between the ventilation of North Pacific and the North Atlantic climate
642 (Lohmann et al., 2019). Such links have also been corroborated by proxy data and
643 modeling experiment between AMOC and East Asian monsoon during the 8.2 ka
644 event (Liu et al., 2013), the Holocene (Wang et al., 2005) and 34 ka–60 ka (Sun et al.,
645 2012). The mechanism linking East Asia with North Atlantic has been attributed to an
646 atmospheric teleconnection, such as the position and strength of Westerly Jet and
647 Mongolia-Siberian High (Porter and Zhisheng, 1995). However, the mechanism
648 behind such out-of-phase pattern between the ventilation in the subtropical North
649 Pacific and the North Atlantic deep water formation remains unclear.

650 Increased NPIW formation during HS1 may have been caused by enhanced
651 salinity-driven vertical mixing through higher meridional water mass transport from
652 the subtropical Pacific. Previous studies have proposed that intermediate water
653 formation in the North Pacific hinged on a basin-wide increase in sea surface salinity
654 driven by changes in strength of the summer EAM and the moisture transport from
655 the Atlantic to the Pacific (Emile-Geay et al., 2003). Several modeling studies found
656 that freshwater forcing in the North Atlantic could cause a widespread surface
657 salinification in the subtropical Pacific Ocean (Menzel et al., 2014; Okazaki et al.,
658 2010; Saenko et al., 2004). This idea has been tested by proxy data (Rodríguez-Sanz
659 et al., 2013; Sagawa and Ikehara, 2008), which indicated a weakened summer EAM

660 and reduced transport of moisture from Atlantic to Pacific through Panama Isthmus
661 owing to the southward displacement of Intertropical Convergence Zone caused by a
662 weakening of AMOC. Along with this process, as predicted through a general
663 circulation modeling, a strengthened Pacific Meridional Overturning Circulation
664 would have transported more warm and salty subtropical water into the high-latitude
665 North Pacific (Okazaki et al., 2010). In accordance with comprehensive Mg/Ca
666 ratio-based salinity reconstructions, however, Riethdorf et al. (2013) found no clear
667 evidence for such higher salinity patterns in the subarctic northwest Pacific during
668 HS1.

669 On the other hand, a weakened AMOC would deepen the wintertime Aleutian
670 Low based on modern observation (Okumura et al., 2009), which is closely related to
671 the sea ice formation in the marginal seas of the subarctic Pacific (Cavalieri and
672 Parkinson, 1987). Once stronger Aleutian Low, intense brine rejection due to sea ice
673 expansion, would have enhanced the NPIW formation. Recently modeling-derived
674 evidence confirmed that enhanced sea ice coverage occurred in the southern Okhotsk
675 Sea and off East Kamchatka Peninsula during HS1 (Gong et al., 2019). In addition,
676 stronger advection of low-salinity water via the Alaskan Stream to the subarctic NW
677 Pacific was probably enhanced during HS1, related to a shift of the Aleutian Low
678 pressure system over the North Pacific, which could also increase sea ice formation,
679 brine rejection and thereafter intermediate water ventilation (Riethdorf et al., 2013).

680 During the late deglaciation, ameliorating global climate conditions, such as
681 warming Northern Hemisphere, and a strengthened Asian summer monsoon, are a
682 result of changes in insolation forcing, greenhouse gases concentrations, and variable
683 strengths of the AMOC (Clark et al., 2012; Liu et al., 2009). During the B/A, a
684 decrease in sea ice extent and duration was indicated by combined reconstructions of
685 SST and mixed layer temperatures from the subarctic Pacific (Riethdorf et al., 2013).
686 At that time, the rising eustatic sea level (Spratt and Lisiecki, 2016) would have
687 supported the intrusion of Alaska Stream into the Bering Sea by deepening and
688 opening glacial closed straits of the Aleutian Islands chain, while reducing the
689 advection of the Alaska Stream to the subarctic Pacific gyre (Riethdorf et al., 2013).

690 In this scenario, saltier and more stratified surface water conditions would have
691 inhibited brine rejection and subsequent formation and ventilation of NPIW (Lam et
692 al., 2013), leading to a reorganization of the Pacific water mass, closely coupled to the
693 collapse and resumption modes of the AMOC during these two intervals.

694 **6.4 Increased storage of CO₂ at mid-depth water in the North Pacific at the B/A**

695 One of the striking features of RSEs data is higher Mo/Mn ratios and excess U
696 concentrations across the B/A, supporting an expansion of Oxygen Minimum Zone in
697 the North Pacific (Galbraith and Jaccard, 2015; Jaccard and Galbraith, 2012; Moffitt
698 et al., 2015) and coinciding with the termination of atmospheric CO₂ concentration
699 rise (Marcott et al., 2014) (Figure 7e). As described above, it can be related to the
700 upwelling of nutrient- and CO₂-rich Pacific Deep Water due to resumption of AMOC
701 and enhanced export production. Notably, boron isotope data measured on
702 surface-dwelling foraminifera in core MD01-2416 situated in the western subarctic
703 North Pacific did reveal a decrease in near-surface pH and an increase in pCO₂ at the
704 onset of B/A (Gray et al., 2018), indicating that the subarctic North Pacific is a source
705 of relatively high atmospheric CO₂ concentration at that time. Here we cannot
706 conclude that the same processes could have occurred in the subtropical North Pacific
707 due to the lack of well-known drivers to draw out of the old carbon in the deep sea
708 into the atmosphere. In combination with published records from the North Pacific
709 (Addison et al., 2012; Cartapanis et al., 2011; Crusius et al., 2004; Galbraith et al.,
710 2007; Lembke-Jene et al., 2017; Shibahara et al., 2007), an expansion of
711 oxygen-depletion zone during the B/A suggest an increase in respired carbon storage
712 at mid-depth waters of the North Pacific, which likely stalls the rise of atmospheric
713 CO₂. Our results support the findings by Galbraith et al. (2007). Given the sizeable
714 volume of the North Pacific, potentially, once the respired carbon could be emitted to
715 the atmosphere in stages, which would bring the planet out of the last ice age (Jaccard
716 and Galbraith, 2018).

717 **7. Conclusions**

718 Our geochemical results of sediment core CSH1 revealed substantial changes in
719 intermediate water redox conditions in the northern Okinawa Trough over the last 50

720 ka on orbital and millennial timescales. Enhanced sedimentary oxygenation mainly
721 occurred during cold intervals, such as the last glacial period, Heinrich stadials 1 and
722 2, and during the middle and late Holocene, while diminished sedimentary
723 oxygenation prevailed during the Bölling-Alleröd and Preboreal. The sedimentary
724 oxygenation variability presented here provides key evidence for the substantial
725 impact of ventilation of NPIW on the sedimentary oxygenation in the subtropical
726 North Pacific and shows out-of-phase pattern with North Atlantic Climate during the
727 last deglaciation. The linkage is attributable to the disruption of NPIW formation
728 caused by climate changes in the North Atlantic, which is transferred to the North
729 Pacific via atmospheric and oceanic teleconnections. We also suggest an expansion of
730 oxygen-depleted zone and accumulation of respired carbon at the mid-depth waters
731 from previously reported subarctic locations into the western subtropical the North
732 Pacific during the B/A, coinciding with the termination of atmospheric CO₂ rise. A
733 step-wise injection of such respired carbon into the atmosphere would be helpful to
734 maintain high atmospheric CO₂ levels during the deglaciation and bring the planet out
735 of the last ice age.

736

737 **Data availability.** All raw data are available to all interested researchers upon request.

738

739 **Author Contributions.** J.J.Z. and X.F.S. conceived the study. A.M.Z. performed
740 geochemical analyses of bulk sediments. J.J.Z., X.F.S. K.S. and X.G. led the write up
741 of the manuscript. All other authors provided comments on the manuscript and
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745

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1154

1155 **Captions**

1156 **Table 1.** Locations of different sediment core records and their source references
1157 discussed in the text.

1158

1159 **Table 2.** Age control points adopted between planktic foraminifera species
1160 *Globigerinoides ruber* $\delta^{18}\text{O}$ of Core CSH1 and Chinese stalagmite $\delta^{18}\text{O}$ (Cheng et al.,
1161 2016) for tuning the age model between 10 ka and 60 ka in this study. A linear
1162 interpolation was assumed between age control points.

1163

1164 **Figure 1.** (a) Spatial distribution of dissolved oxygen content at 700 m water depth in
1165 the North Pacific. Black arrows denote simplified Kuroshio and Oyashio circulations
1166 and North Pacific Intermediate Water (NPIW) in the North Pacific. The red thick
1167 dashed line indicates transformation of Okhotsk Sea Intermediate Water (OSIW) by
1168 cabbeling the subtropical NPIW along the subarctic-tropical frontal zone (You, 2003).
1169 The light brown solid line with arrow indicates the spreading path of subtropical
1170 NPIW from northeast North Pacific southward toward the low-latitude northwest
1171 North Pacific (You, 2003). Yellow solid lines with arrow represent two passages
1172 through which NPIW enter into the Okinawa Trough. This figure was created with
1173 Ocean Data View (odv.awi.de). (b) Location of sediment core CSH1 investigated in
1174 this study (red diamond). Also shown are locations of sediment cores PN-3, E017, 255
1175 and MD012404 investigated previously from the Okinawa Trough, GH08-2004 from
1176 the East of Ryukyu Island, GH02-1030 off the east of Japan, PC-23A from the Bering
1177 Sea and ODP Site 1017 from the northeastern Pacific. Letters A to E represent the
1178 sediment cores from and near the OT. The detailed information for these cores is
1179 shown in Table 1.

1180

1181 **Figure 2.** Spatial distribution of sea surface salinity in the East China Sea. (a) summer
1182 (July to September); (b) winter (January to March). Lower sea surface salinity in
1183 summer relative to that of winter indicates strong effects of summer East Asian
1184 Monsoon.

1185
1186 **Figure 3.** (a) Lithology and oxygen isotope ($\delta^{18}\text{O}$) profile of planktic foraminifera
1187 species *Globigerinoides ruber* (*G.ruber*) in core CSH1. (b) Plot of ages versus depth
1188 for core CSH1. Three known ash layers are indicated by solid red rectangles. (c) Time
1189 series of linear sedimentation rate (LSR) from core CSH1. (d) Comparison of age
1190 model of core CSH1 with Chinese Stalagmite composite $\delta^{18}\text{O}$ curve of (Cheng et al.,
1191 2016). Tie points for CSH1 core chronology (Table 2) in Figures 3c and 3d are
1192 designated by colored crosses.

1193
1194 **Figure 4.** Age versus (a) CaCO_3 concentration, (b) Total nitrogen (TN) concentration,
1195 (c) Total organic carbon (TOC) concentration, (d) C/N molar ratio, (e) linear
1196 sedimentation rate (LSR), (f) Al concentration, (g) Mn concentration, (h) Mo/Mn ratio,
1197 (i) Mo concentration, (j) excess Mo concentration, (k) U concentration and (l) excess
1198 U concentration and (m) $(\text{Mo}/\text{U})_{\text{excess}}$ ratio in core CSH1. Light gray and dark gray
1199 vertical bars indicate different sediment intervals in core CSH1. 8.2 ka, PB, YD, B/A,
1200 HS1, LGM and HS2 refer to 8,200 year cold event, Preboreal, Younger Dryas, Bölling
1201 - Alleröd, Heinrich Stadial 1, Last Glacial Maximum and Heinrich Stadial 2,
1202 respectively, which were identified in core CSH1. Blue solid diamonds in Figure 4m
1203 indicate the age control points.

1204
1205 **Figure 5.** Scatter plots of $\text{Mo}_{\text{excess}}$ vs Mn concentrations and U_{excess} concentration vs
1206 Mo/Mn ratio at different time intervals in core CSH1. A various correlation is present
1207 in core CSH1 at different time intervals, which shows their complicated geochemical
1208 behaviors (Figs.5a and b). Strong positive correlation between Mo/Mn ratio and
1209 U_{excess} concentration (Fig.5c) suggest that Mo/Mn ratio is a reliable proxy to track
1210 sedimentary redox conditions in the geological past.

1211
1212 **Figure 6.** Proxy-related reconstructions of mid-depth sedimentary oxygenation at site
1213 CSH1 (this study) compared with oxygenation records from other locations of the
1214 North Pacific and published climatic and environmental records from the Okinawa

1215 Trough. From top to bottom: (a) CaCO_3 concentration, (b) U_{excess} concentration, (c)
1216 Mo/Mn ratio, and (d) sea surface temperature (SST) (Shi et al., 2014), (e) abundance
1217 of *P.obliquiloculata* in core CSH1 (Shi et al., 2014), (f) bulk sedimentary organic
1218 matter $\delta^{15}\text{N}$ in core MD01-2404 (Kao et al., 2008), (g) $\delta^{13}\text{C}$ of epibenthic
1219 foraminiferal *C.wuellerstorfi* in core PN-3 (Wahyudi and Minagawa, 1997), (h)
1220 relative abundance of *B. aculeata* (hypoxia-indicating species) and (i) *C.hyalinea*
1221 (oxygen-rich indicating species) (Li et al., 2005), (j) dysoxic taxa (%) in core ODP
1222 167-1017 in the northeastern Pacific (Cannariato and Kennett, 1999) and (k) $\delta^{13}\text{C}$ of
1223 benthic foraminiferal *Uvigerina akitaensis* in core PC23A in the Bering Sea (Rella
1224 et al., 2012). Light gray and dark gray vertical bars are the same as those in Figure 4.

1225
1226 **Figure 7.** Proxy records favoring the existence of out-of-phase connections between
1227 the subtropical North Pacific and North Atlantic during the last deglaciation and
1228 enhanced carbon storage at mid-depth waters. (a) U_{excess} concentration in core CSH1;
1229 (b) Mo/Mn ratio in core CSH1; (c) benthic $\delta^{13}\text{C}$ record in core PC-23A in the Bering
1230 Sea (Rella et al., 2012); (d) Indicator of strength of Atlantic Meridional Ocean
1231 Circulation ($^{231}\text{Pa}/^{230}\text{Th}$) (Böhm et al., 2015; McManus et al., 2004); (e) Atmospheric
1232 CO_2 concentration (Marcott et al., 2014). Light gray and dark gray vertical bars are
1233 the same as those in Figure 4.

1234

Table 1

Label in Figure 1b	Station	Latitude (°N)	Longitude (°E)	Water depth (m)	Area	Reference
	CSH1	31.23	128.72	703	Okinawa Trough	this study
A	PN-3	28.10	127.34	1058	Okinawa Trough	Wahyudi and Minagawa, (1997)
B	MD012404	26.65	125.81	1397	Okinawa Trough	Kao et al., (2008)
C	E017	26.57	126.02	1826	Okinawa Trough	Li et al., (2005)
D	255	25.20	123.12	1575	Okinawa Trough	Jian et al., (1996)
E	GH08-2004	26.21	127.09	1166	East of Ryukyu Island	Kubota et al. (2015)
	GH02-1030	42.23	144.21	1212	Off Japan	Sagawa and Ikehara, (2008)
	PC-23A	60.16	179.46	1002	Bering Sea	Rella et al.,(2012)
	ODP Site1017	34.54	239.11	955	NE Pacific	Cannariato and Kennett, (1999)

1 **Table 2**

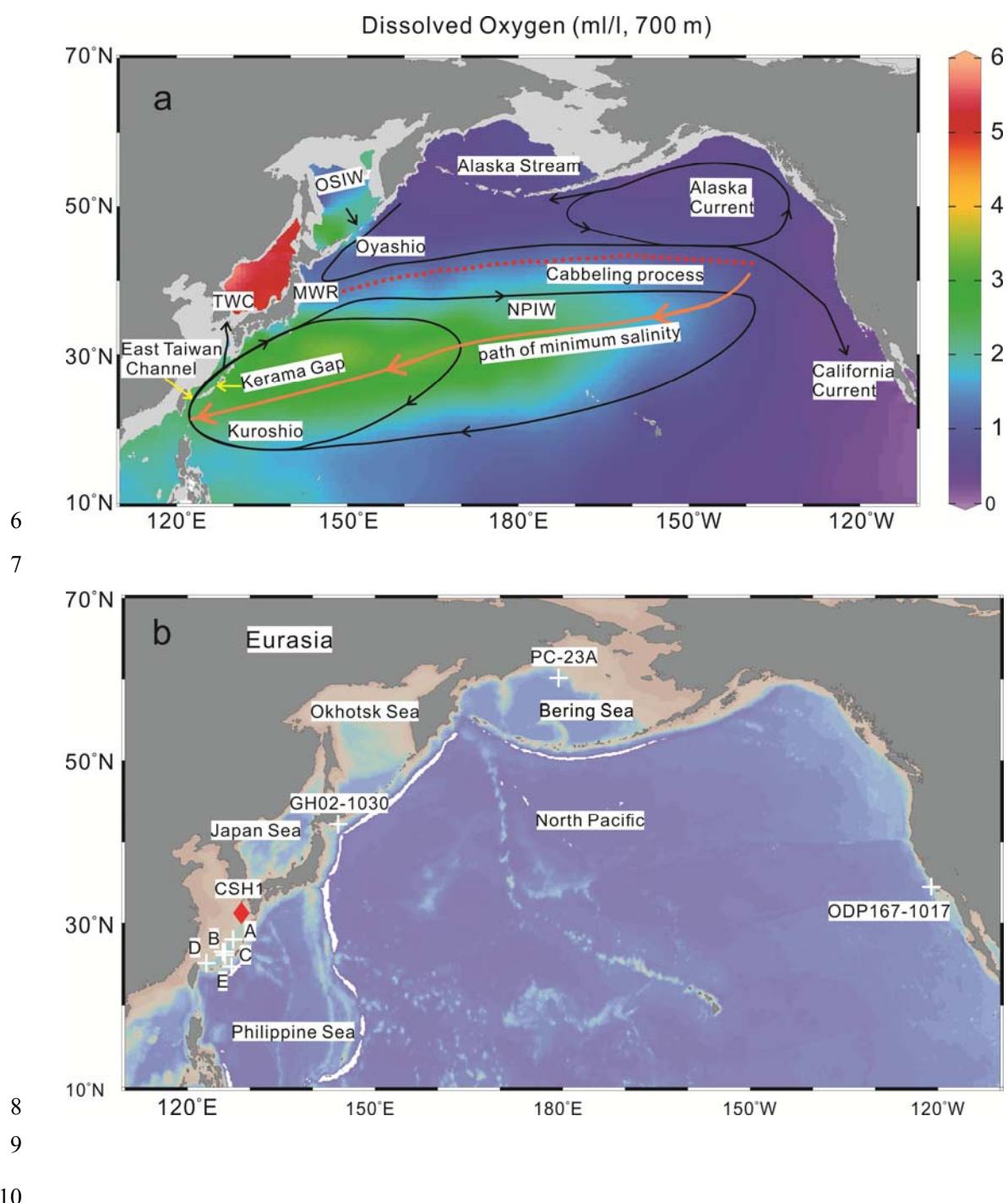
2

Depth(cm)	AMS ^{14}C (yr)	Error (yr)	Calibrated Age (yr)	Tie Point Type	LSR (cm/ka)	Source
10	3420	± 35	3296	^{14}C		Shi et al., (2014)
106	7060	± 40	7545	^{14}C	22.59	Shi et al., (2014)
218			12352	Stalagmite, YD	23.30	This study
322			16029	Stalagmite, H1	28.28	This study
362			19838	Stalagmite	10.50	This study
506			24163	Stalagmite, H2	33.29	This study
698			28963	Stalagmite, DO4	40.00	This study
834			32442	Stalagmite, DO5	39.09	This study
938			37526	Stalagmite, DO8	20.46	This study
978			39468	Stalagmite, H4	20.60	This study
1058			46151	Stalagmite, DO12	11.97	This study
1122			49432	Stalagmite, DO13	19.51	This study
1242			52831	Stalagmite, DO14	35.30	This study
1282			57241	Stalagmite, DO16	9.07	This study
1346			61007	Stalagmite, H6	16.99	This study
1530		± 2590	73910	MIS4/5	14.26	Shi et al., (2014)
1610		± 3580	79250	MIS 5.1	14.98	Shi et al., (2014)

3

4

5 Fig.1

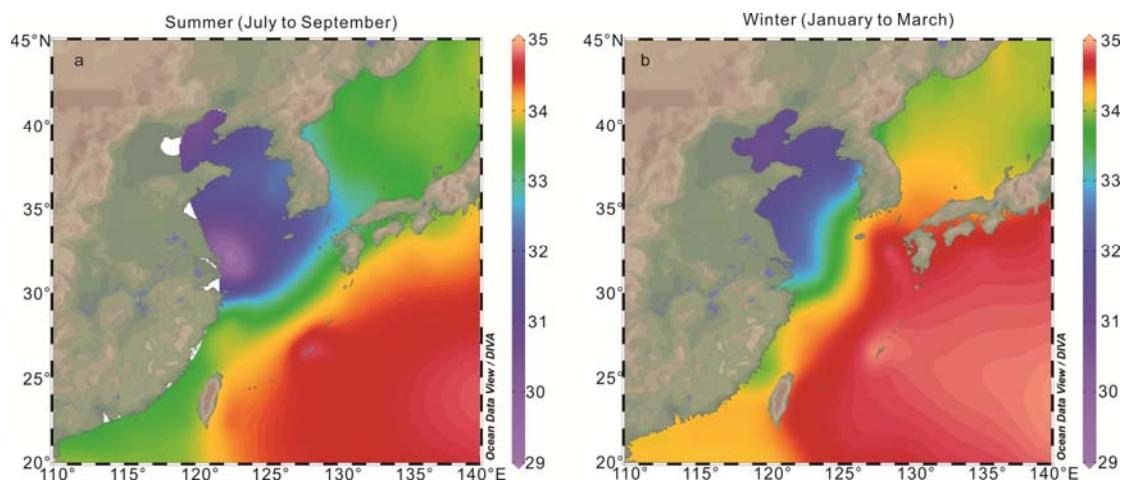


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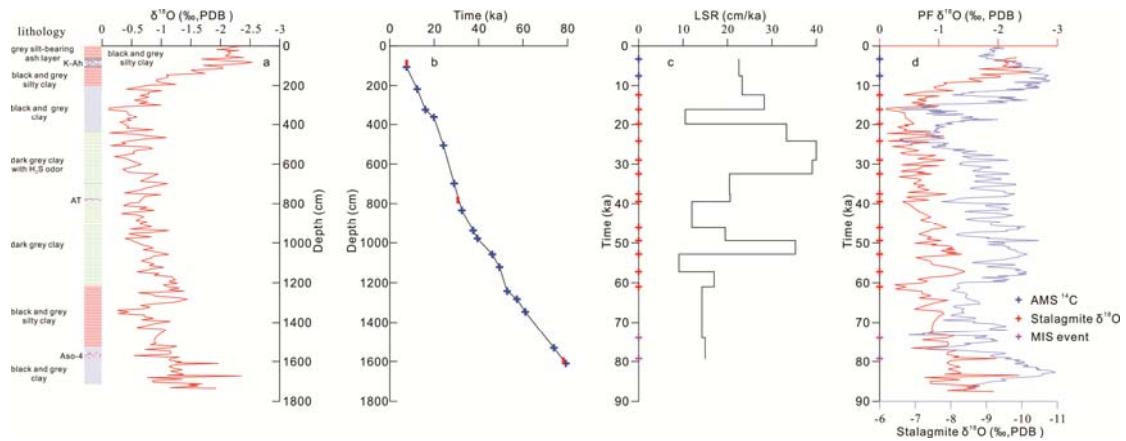
11 Fig.2



12

13

14 Fig.3

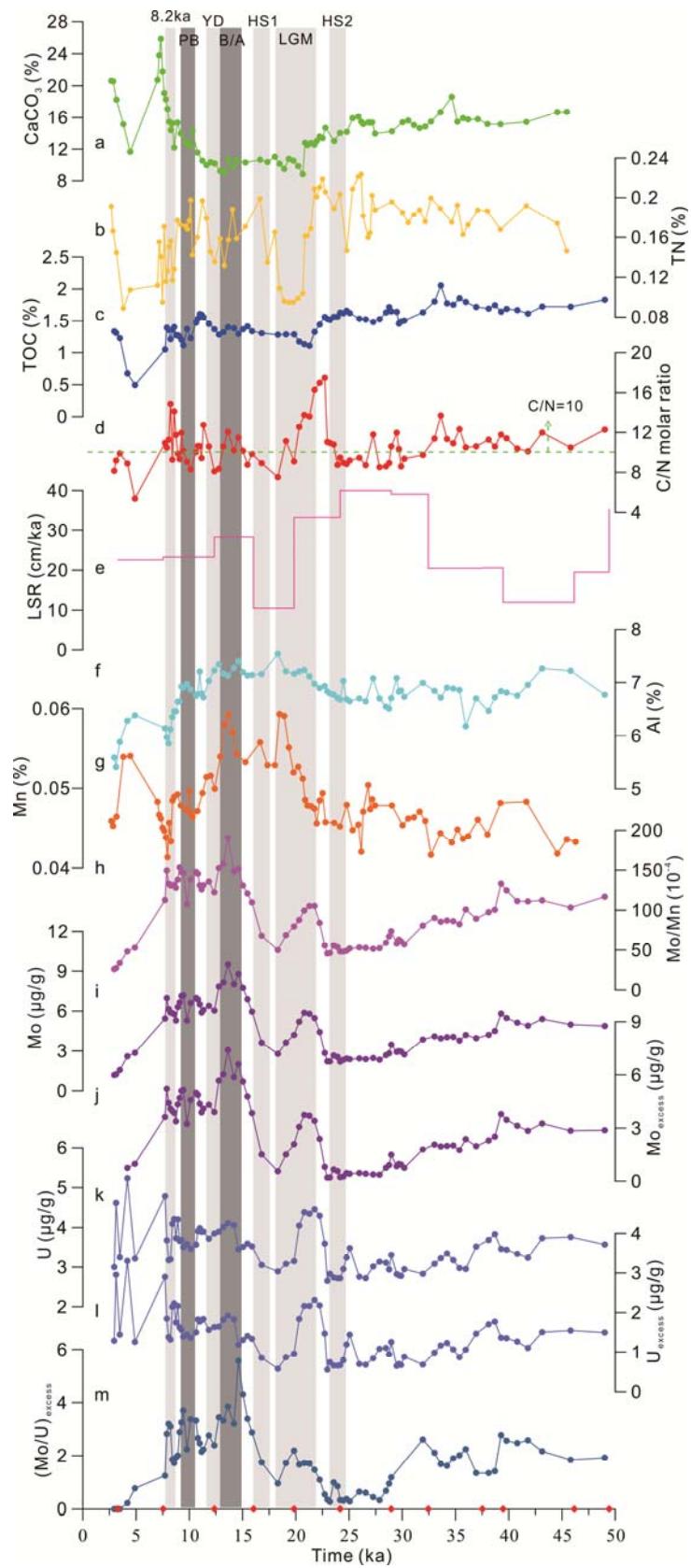


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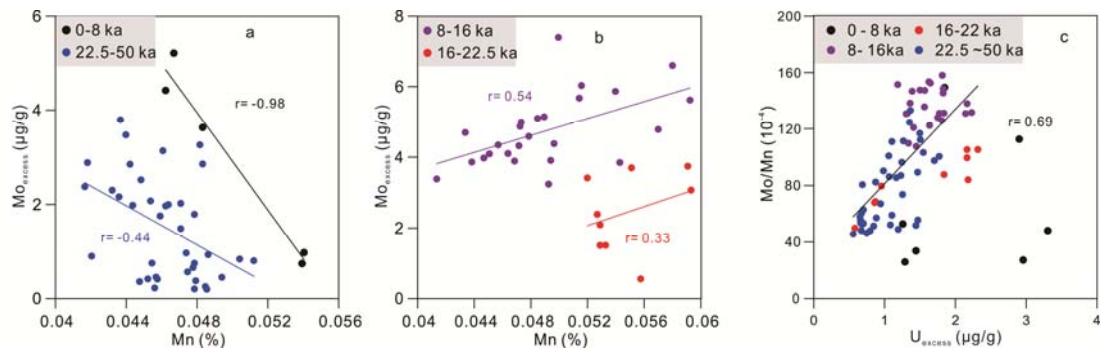
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18 Fig.4



21 Fig.5

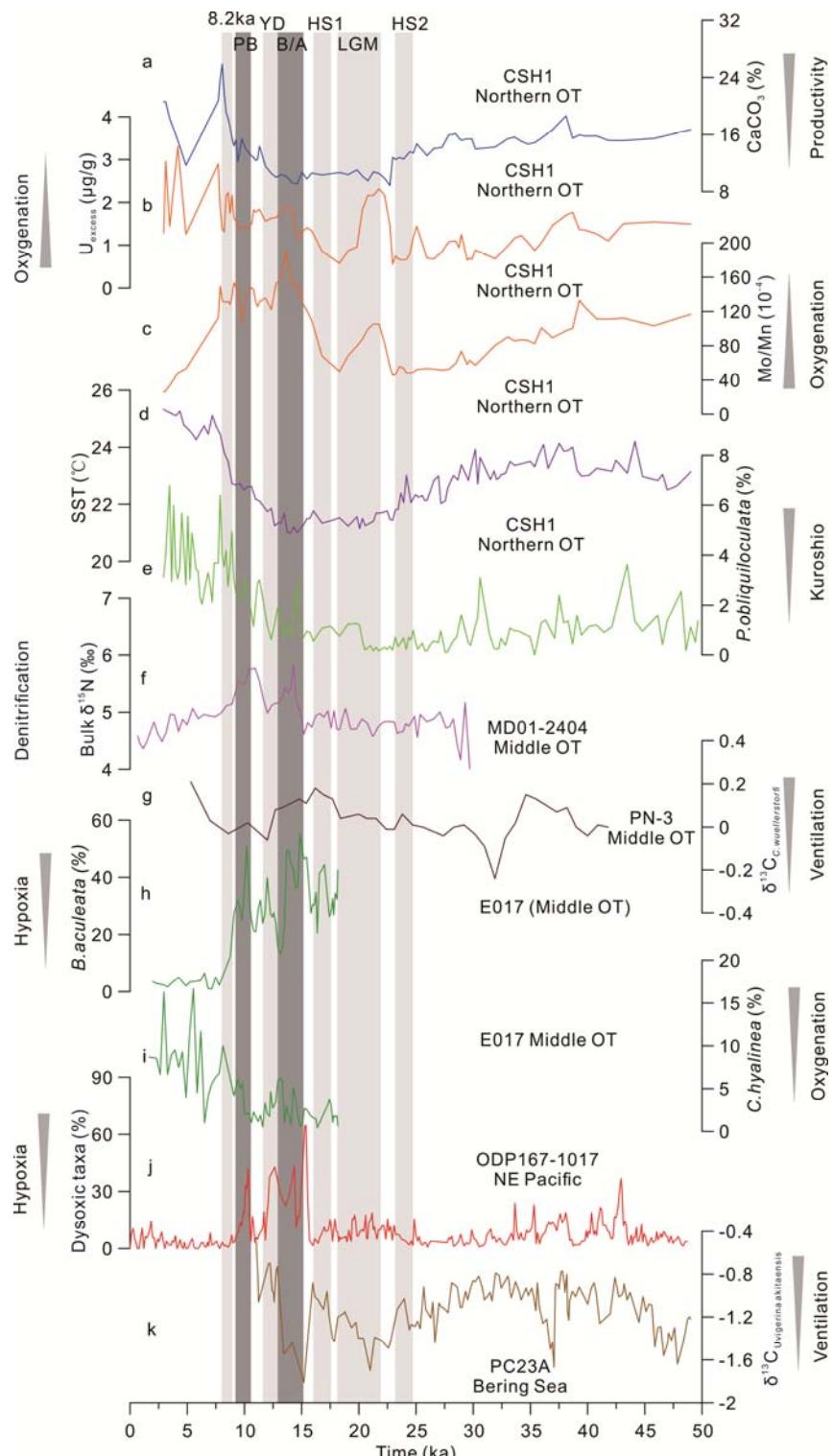
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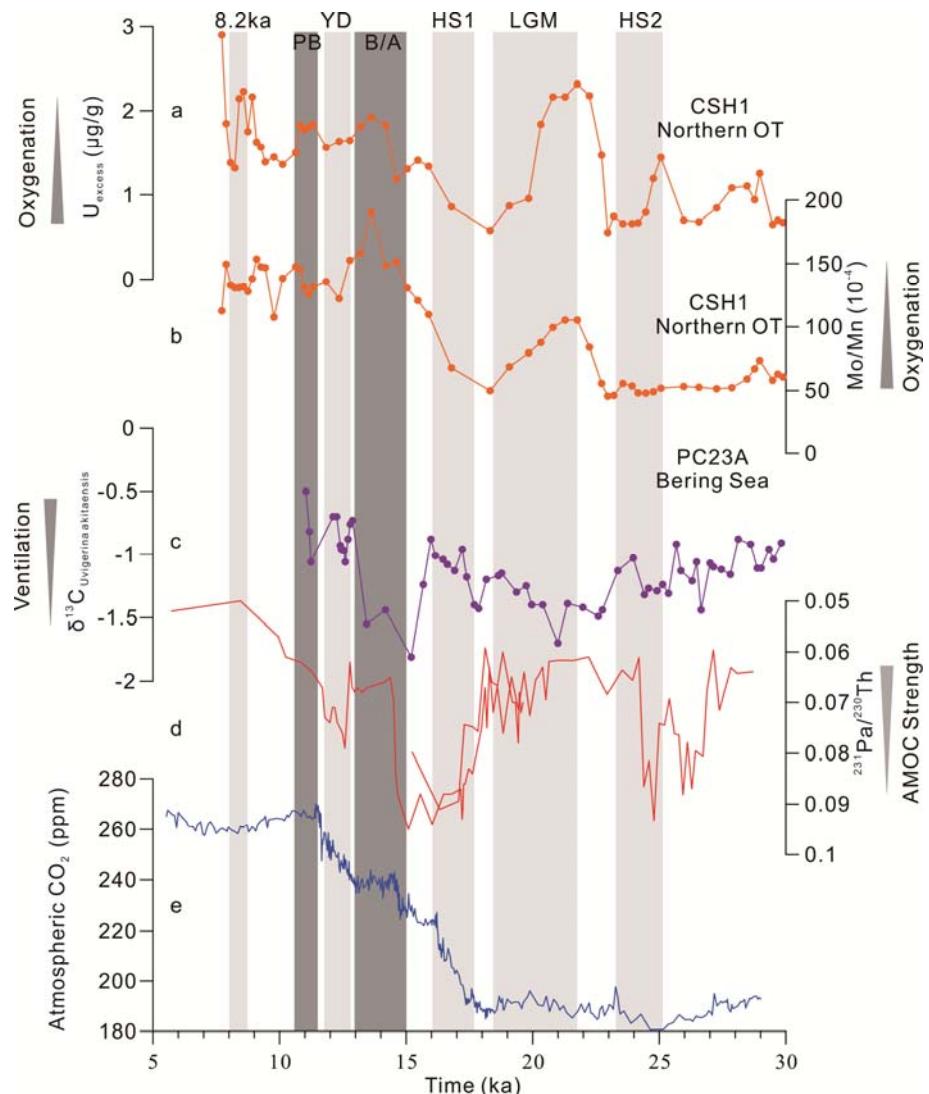
25 Fig.6



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27

28 Fig. 7



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