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

Jianjun Zou[1,2], Xuefa Shi[1,2], Aimei Zhu[1], Selvaraj Kandasamy[3], Xun Gong[4], Lester Lembke-Jene[4], Min-Te Chen[5], Yonghua Wu[1,2], Shulan Ge[1,2], Yanguang Liu[1,2], Xinru Xue[1], Gerrit Lohmann[4], Ralf Tiedemann[4]

[1]Key Laboratory of Marine Sedimentology and Environmental Geology, First Institute of Oceanography, MNR, Qingdao 266061, China

[2]Laboratory for Marine Geology, Qingdao National Laboratory for Marine Science and Technology, Qingdao, 266061, China

[3]Department of Geological Oceanography and State Key Laboratory of Marine Environmental Science, Xiamen University, Xiamen361102, China

[4]Alfred-Wegener-Institut Helmholtz-Zentrum für Polar- und Meeresforschung, Bussestr. 24, 27570 Bremerhaven, Germany

[5]Institute of Applied Geosciences, National Taiwan Ocean University, Keelung 20224, Taiwan

Corresponding authors:

Jianjun Zou (zoujianjun@fio.org.cn); Xuefa Shi (xfshi@fio.org.cn)

Key Points

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

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

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

4. A larger CO_2 storage at mid-depths of the North Pacific corresponds to the termination of atmospheric CO_2 rise during the Bölling-Alleröd interval.

Abstract

Deep ocean carbon cycle, especially carbon sequestration and outgassing, is one of the competitive mechanisms to explain variations in atmospheric CO_2 concentrations on orbital and millennial timescales. However, the potential role of subtropical North Pacific subsurface waters in modulating atmospheric CO_2 levels on millennial timescales is poorly constrained. Further, an increase in respired CO_2 concentration in the glacial deep ocean due to biological pump generally corresponds to deoxygenation in the subsurface layer. This link thus offers a chance to visit oceanic ventilation and the coeval export productivity based on redox-controlled, sedimentary geochemical parameters. Here we investigate a suite of geochemical proxies in a sediment core CSH1 to understand the sedimentary oxygenation variations in the subtropical North Pacific over the last 50 thousand years (ka). Our results suggest that enhanced sedimentary oxygenation at mid-depths of the subtropical North Pacific occurred during the cold intervals and after 8.5 ka, while decreased oxygenation during the Bölling-Alleröd (B/A) and Preboreal. The enhanced sedimentary oxygenation in the subtropical North Pacific is aligned with intensified formation of North Pacific Intermediate Water (NPIW) during cold spells, while the ameliorated sedimentary oxygenation seems to be linked to the intensified Kuroshio Current since 8.5 ka. The enhanced formation of NPIW during HS1 can be driven by the perturbation of sea ice formation and sea surface salinity oscillation in high-latitude North Pacific. The diminished sedimentary oxygenation during the B/A due to upwelling of aged, nutrient-rich deep water and enhanced export production, indicates an enhanced CO_2 sequestration at mid-depth waters, along with a slight increase in atmospheric CO_2 concentration. We attribute these millennial-scale changes to intensified NPIW and enhanced abyss flushing during deglacial cold and warm intervals, respectively, on the basis of background climate change due to shift in North Atlantic Deep Water formation.

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

1. Introduction

The sluggish ocean ventilation and efficient biological pump in the ocean facilitate carbon sequestration in the ocean interior, linking to atmospheric CO_2 drawdown, which in turn play a crucial role in regulating sedimentary oxygen on millennial and orbital timescales (Hoogakker et al., 2015; Jaccard and Galbraith, 2012; Sigman and Boyle, 2000). Reconstruction of past sedimentary oxygenation is therefore crucial for understanding changes in export productivity and the renewal rate of deep ocean circulation (Nameroff et al., 2004). Previous studies from eastern and western North Pacific margins and subarctic Pacific have identified drastic variations in export productivity and ocean oxygen levels at glacial-interglacial timescales using diverse proxies such as trace elements (Cartapanis et al., 2011; Chang et al., 2014; Jaccard et al., 2009; Zou et al., 2012), benthic foraminiferal assemblages (Ohkushi et al., 2016; Ohkushi et al., 2013; Shibahara et al., 2007) and nitrogen isotopic composition ($\delta^{15}N$) of organic matter (Addison et al., 2012; Chang et al., 2014; Galbraith et al., 2004; Riethdorf et al., 2016) in marine sediment cores. These studies further suggested that both North Pacific Intermediate Water (NPIW) and export of organic matter regulate the sedimentary oxygenation variation during the last glaciation and Holocene in the subarctic North Pacific. By contrast, little information exists on millennial-scale oxygenation changes to date in the western subtropical North Pacific.

The modern NPIW is mainly sourced from the NW Pacific marginal seas (Shcherbina et al., 2003; Talley, 1993; You et al., 2000), and then it spreads into subtropical North Pacific at intermediate depths of 300 to 800 m (Talley, 1993). The pathway and circulation of NPIW have been identified by You (2003), who suggested that cabbeling, a mixing process to form a new water mass with increased density than that of parent water masses, is the principle mechanism responsible for transforming subpolar source waters into subtropical NPIW along the subarctic-tropical frontal zone. More specifically, You et al. (2003) argued that a lower subpolar input of about 2 Sv (1 Sv = 10^6 m^3/s) is sufficient for subtropical ventilation. Benthic foraminiferal $\delta^{13}C$, a quasi-conservative tracer for water mass, from the North Pacific suggested an enhanced ventilation (enriched $\delta^{13}C$) at water depths of < 2000 m during the last glacial period (Keigwin, 1998; Matsumoto et al., 2002). Furthermore, on the basis of both radiocarbon data and modeling results, Okazaki et al. (2010) provided further insight into the formation of deep water in the North Pacific during early deglaciation. Enhanced NPIW penetration is further explored using numerical model simulations (Chikamoto et al., 2012; Gong et al., 2019; Okazaki et al., 2010). More recently, Max et al (2017) identified the substantial effects of intensified NPIW on $\delta^{13}C$ of deep-dwelling planktic foraminifera *Globorotaloides hexagonus* in the Eastern Equatorial Pacific during Marine Isotope Stage (MIS) 2. Subsequently, Rippert et al. (2017) confirmed that such enhanced effect of NPIW also occurred during MIS 6. The downstream effects of intensified NPIW also can be seen in the record of $\delta^{13}C$ of *Cibicides wuellerstorfi* in core PN-3 from the middle Okinawa Trough (OT), whereas lower deglacial $\delta^{13}C$ values were attributed to enhanced OC accumulation rates due to higher surface productivity by Wahyudi and Minagawa (1997).

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

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

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

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

2. Paleo-redox proxies

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

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

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

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

In general, U is enriched in anoxic sediments (>1 μM H_2S), but not in oxic sediments (>10 μM O_2) (Nameroff et al., 2002). Accumulation of U depends on the content of reactive organic matter (Sundby et al., 2004) and U precipitates as uraninite (UO_2) during the conversion of Fe (III) to Fe (II) in suboxic conditions (Morford and Emerson, 1999; Zheng et al., 2002). One of the primary removal mechanisms for U from the ocean is via diffusion across the sediment-water interface of reducing sediments (Klinkhammer and Palmer, 1991). Under suboxic conditions, soluble U (VI) is reduced to insoluble U (IV), but free sulfide is not required for U precipitation (McManus et al., 2005). Jaccard et al. (2009) suggested that the presence of excess concentration of U (U_{excess}) in the absence of Mo enrichment is indicative of a suboxic, but not sulfidic condition, within the diffusional range of the sediment-water interface. The felsic volcanism is also a primary source of uranium (Maithani and Srinivasan, 2011). Therefore, the potential input of uranium from active volcanic sources around the northwestern Pacific to the adjacent sediments should not be neglected.

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

3. Oceanographic setting

Surface hydrographic characteristics of the OT are mainly controlled by the warmer, more saline, oligotrophic Kuroshio water and cooler, less saline, nutrient-rich Changjiang Diluted Water, and the modern flow-path of the former is influenced by the bathymetry of the OT (Figure 1a). The Kuroshio Current originates from the North Equatorial Current and flows into the ECS from the Philippine Sea through the Suao-Yonaguni Depression. In the northern OT, Tsushima Warm Current (TWC), a branch of the Kuroshio, flows into the Japan Sea through the shallow Tsushima Strait. Volume transport of the Kuroshio varies seasonally due to the influence of the EAM with a maximum of 24 Sv in summer and a minimum of 20 Sv in autumn across the east of Taiwan (Qu and Lukas, 2003).

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

Despite the effects of EAM and the Kuroshio, evidence of geochemical tracers (temperature, salinity, oxygen, nutrients and radiocarbon-$\Delta^{14}C$) collected during the World Ocean Circulation Experiment (WOCE) Expeditions in the Pacific (transects P24 and P03) favors the presence of low saline, nutrient-enriched intermediate and deep waters (Talley, 2007). Dissolved oxygen content is <100 μmol/kg at water depths of below 600 m in the OT along WOCE transects PC03 and PC24 (Talley, 2007). Modern oceanographic observations at the Kerama Gap reveal that upwelling in the OT is associated with the inflow of NPIW and studies using box model predicted that overflow through the Kerama Gap is responsible for upwelling (3.8–7.6 $\times 10^{-6}$m s^{-1}) (Nakamura et al., 2013; Nishina et al., 2016).

4. Materials and methods

4.1. Chronostratigraphy of core CSH1

A 17.3 m long sediment core CSH1 (31° 13.7' N, 128° 43.4' E; water depth: 703 m) was collected from the northern OT, close to the main stream of Tsushima Warm Current (TWC) (Figure 1b) and within the depth of NPIW (Figure 1c) using a piston corer during *Xiangyanghong*09 Cruise in 1998. This location is thus enabling us to reconstruct millennial-scale changes in the properties of TWC and NPIW. The expedition was carried out by the First Institute of Oceanography, Ministry of Natural Resources of China. Core CSH1 mainly consists of clayey silt and silt with occurrence of plant debris at some depth intervals (Ge et al., 2007) (Figure 3a). In addition, three layers of volcanic ash were observed at depths of 74−106 cm, 782−794 cm, 1570−1602 cm and these three intervals can be correlated with well-known ash layers, Kikai-Akahoya (K-Ah; 7.3 ka), Aira-Tanzawa (AT; 29.24 ka) and Aso-4 (roughly around MIS 5a) (Machida, 1999), respectively. The core was split and sub-sampled at every 4 cm interval and then stored in China Ocean Sample Repository at 4 °C until analysis.

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

It is noteworthy that previous age control points with constant radiocarbon reservoir throughout core CSH1 are used to reveal orbital-scale Kuroshio variations (Shi et al., 2014), but insufficient to investigate millennial-scale climatic events. On the basis of original age model, a higher abundance of *Neogloboquadrina pachyderma* (dextral) that occurred during warmer intervals, such as the B/A, has been challenging to explain reasonably. On the other hand, paired measurements of $^{14}C/^{12}C$ and ^{230}Th ages from Hulu Cave stalagmites suggest magnetic field change has greatly contributed to high atmospheric $^{14}C/^{12}C$ values at HS4 and the YD (Cheng et al., 2018). Thus a constant reservoir age assumed when calibrating foraminiferal radiocarbon dates using CALIB 6 software and the Marine 13 calibration dataset (Reimer et al., 2013) for core CSH1 may cause large chronological uncertainties.

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

4.2. Chemical analyses

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

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

$CaCO_3 = (TC-TOC) \times 8.33$

where 8.33 is the ratio between the molecular weight of carbonate and the atomic weight of carbon. National reference material (GSD-9), blank sample and replicated samples were used to control the analytical process. The relative standard deviation of the GSD-9 for TC, TN and TOC is $\leqslant$ 3.4%.

About 0.5 g of sediment powder was digested in double distilled HF:HNO_3 (3:1), followed by concentrated $HClO_4$, and then re-dissolved in 5% HNO_3. Selected major and minor elements such as aluminum (Al) and manganese (Mn) were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES; Thermo Scientific iCAP 6000, Thermo Fisher Scientific), as detailed elsewhere (Zou et al., 2012). In addition, Mo and U were analyzed with inductively coupled plasma mass spectrometry (ICP-MS; Thermo Scientific XSERIES 2, Thermo Fisher Scientific), as described in Zou et al. (2012). Precision for most elements in the reference material GSD-9 is $\leqslant$ 5% relative standard deviation. The excess fractions of U and Mo were estimated by normalization to Al:

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

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

5. Results

5.1. TOC, TN, and $CaCO_3$

The content of $CaCO_3$ varies from 8.8 to 35% (Figure 4a) and it mostly shows higher values with increasing trends during the last deglaciation. In contrast, the content of $CaCO_3$ is low and exhibits decreasing trends during the late MIS 3 and the LGM (Figure 4a). TN content shows a larger variation compared to TOC (Figure 4b), but it still strongly correlates with TOC ($r = 0.74$, $p<0.01$) throughout the entire core.

Concentration of TOC ranges from 0.5 to 2.1% and it shows higher values with stable trends during the last glacial phase (MIS 3) (Figure 4c). Molar ratios of TOC/TN vary around 10, with higher ratios at the transition into the LGM (Figure 4d), corresponding to higher linear sedimentation rate (Figure 4e).

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

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

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 4l) show coherent patterns with those of Mo and U (Figures 4i and 4k), but both are out-of-phase with Mn over the last glacial period (Figure 4h). It should be noted that pronounced variations in U concentration since 8.5 ka is related to the occurrence of discrete volcanic materials. A significant positive Eu anomaly (Zhu et al., 2015) together with more radiogenic Nd values (unpublished data) from the same core 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 5b) further corroborate the complicated 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 use Mo/Mn ratio, instead of excess Mo concentration to reconstruct variations in sedimentary redox conditions in the 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 contamination of volcanic material, Figure 5c) further corroborates the integrity of Mo/Mn as an indicator of sedimentary oxygenation changes.

Rapidly decreasing Mo/Mn ratio indicates an oxygenated sedimentary environment since ~8 ka (Figure 4h). Both higher Mo/Mn ratios and excess U concentration, together with lower Mn concentrations suggest a deoxygenated depositional condition 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 condition. A decreasing trend in Mo/Mn ratio and excess U concentration from 50 ka to 25 ka also suggest higher sedimentary oxygen levels.

6.Discussion

6.1. Constraining paleoredox conditions in the Okinawa Trough

In general, three different terms, hypoxia, suboxia and anoxia, are widely used to describe the degree of oxygen depletion in the marine environment (Hofmann et al., 2011). Here, we adopt the definition of oxygen thresholds by Bianchi et al. (2012) for oxic (>120 μmol/kg O_2), hypoxic (<60–120 μmol/kg O_2) and suboxic (<2–10 μmol/kg O_2) conditions, whereas anoxia is the absence of measurable oxygen.

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

Both bulk Mo concentration (1.2-9.5 μg/g) and excess (Mo/U) ratio (0.2-5.7) in core CSH1 suggest that oxygen-depleted conditions may have prevailed in the deep water of the northern OT over the last 50 ka (Figure 4m). However, increased excess Mo concentration with enhanced Mo/U ratio during the last termination (18ka-9 ka) indicate a stronger reducing condition compared to the Holocene and the last glacial period, though Mo concentration is less than 25 μg/g, a threshold for euxinic deposition proposed by Scott and Lyons (2012).

The relative abundance of benthic foraminifera species that thrive in different oxygen concentrations also have been widely used to reconstruct the variations in bottom water ventilation, such as enhanced abundance of *Bulimina aculeata*, *Uvigerina peregrina and Chilostomella oolina* found under oxygen-depleted conditions in the central and southern OT during the last deglaciation (18 ka -9.2 ka) (Jian et al., 1996; Li et al., 2005). An oxygenated bottom water condition is also indicated by abundant benthic foraminifera species *Cibicidoides hyalina* and *Globocassidulina subglobosa* after 9.2 ka (Jian et al., 1996; Li et al., 2005) in cores E017 (1826 m water depth), 255 (1575 m water depth) and high benthic $\delta^{13}C$ values (Wahyudi and Minagawa, 1997) in core PN-3 (1058 m water depth) from the middle and southern OT during the postglacial period. The poorly-ventilated deep water in the middle and southern OT inferred by benthic foraminiferal assemblages during the last deglaciation is coeval with the one in the northern OT referring to our RSEs (Figure 4). A clear linkage thus can be established between deep-water ventilation and sedimentary oxygenation in the OT. Overall, a combination of our proxy records of RSEs in core CSH1 with other records shows oxygen-rich conditions during the last glaciation and middle and late Holocene (since 8.5 ka) intervals, but oxygen-poor conditions during the last deglaciation.

6.2. Causes for sedimentary oxygenation variations

As discussed above, the pattern of RSEs in core CSH1 suggests that drastic changes in sedimentary oxygenation occurred on orbital and millennial timescales over the last glaciation in the OT. In general, four factors can regulate the redox condition in the deep water column: (i) O_2 solubility, (ii) export productivity and subsequent degradation of organic matter, (iii) vertical mixing, and (iv) lateral supply of oxygen through intermediate and deeper water masses (Ivanochko and Pedersen, 2004; Jaccard and Galbraith, 2012). These processes have been invoked in previous studies to explain the deglacial Pacific-wide variations in oxygenation by either one or a combination of these factors (Galbraith and Jaccard, 2015; Moffitt et al., 2015;

Praetorius et al., 2015). Our data also suggest drastic variations in sedimentary oxygenation over the last 50 ka. However, the mechanisms responsible for sedimentary oxygenation variations in the basin-wide OT and its connection with ventilation of the open North Pacific remain unclear. In order to place our core results in a wider regional context, here, we compare our proxy records of sedimentary oxygenation (U_{excess} concentration and Mo/Mn ratio) and export productivity ($CaCO_3$) (Figures 6a, b, c) with abundance of *Pulleniatina obliquiloculata* (an indicator of Kuroshio strength) and sea surface temperature (Shi et al., 2014), bulk sedimentary nitrogen isotope (an indicator of denitrification) (Kao et al., 2008), benthic foraminifera $\delta^{13}C$ (a proxy for water mass) in cores PN-3 and PC23A (Rella et al., 2012; Wahyudi and Minagawa, 1997), abundance of benthic foraminifera (an indicator of hypoxia) in core E017 (Li et al., 2005) and ODP167 site 1017 (Cannariato and Kennett, 1999) (Figures 6d - k).

6.2.1. Effects of regional ocean temperature on deglacial deoxygenation

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

6.2.2. Links between deglacial primary productivity and sedimentary deoxygenation

Previous studies have suggested the occurrence of high primary productivity in the entire OT during the last deglacial period (Chang et al., 2009; Jian et al., 1996; Kao et al., 2008; Li et al., 2017; Shao et al., 2016; Wahyudi and Minagawa, 1997). Such an increase in export production was due to favorable conditions for bloom development, which were likely induced by warm temperatures and maxima in nutrient availability, the latter being mainly sourced from increased discharge of the Changjiang River, erosion of material from the ongoing flooding of the shallow continental shelf in the ECS, and upwelling of Kuroshio Intermediate Water (Chang et al., 2009; Li et al., 2017; Shao et al., 2016; Wahyudi and Minagawa, 1997). On the basis of sedimentary reactive phosphorus concentration, Li et al. (2017) concluded that export productivity increased during warm episodes but decreased during cold spells on millennial timescales over the last 91 ka in the OT. Gradually increasing concentrations of $CaCO_3$ in core CSH1 during the deglaciation (Figure 6a) and little changes in foraminiferal fragmentation ratios (Wu et al., 2004), are indicative of high export productivity in the northern OT. Accordingly, our data indicate that an increase in export productivity during the last deglaciation, which was previously evidenced by concentrations of reactive phosphorus (Li et al., 2017) and $CaCO_3$ (Chang et al., 2009) from the middle OT, and thus was a pervasive, synchronous phenomenon of entire study region at the outermost extension of the ECS.

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

6.2.3 Effects of the Kuroshio dynamics on sedimentary oxygenation

The Kuroshio Current, one of the main drivers of vertical mixing, has been identified as the key factor in controlling modern deep ventilation in the OT (Kao et al., 2006). However, the flow path of the Kuroshio in the OT during the glacial interval remains a matter of debate. Planktic foraminiferal assemblages in sediment cores from inside and outside the OT indicated that the Kuroshio have migrated to the east of the Ryukyu Islands during the LGM (Ujiié and Ujiié, 1999). Subsequently, Kao et al. (2006) based on modeling results suggested that the Kuroshio still enters into the OT, but the volume transport was reduced by 43% compared to the present-day transport and the outlet of Kuroshio switches from the Tokara Strait to the Kerama Gap at -80 and -135m lowered sea level. Combined with sea surface temperature (SST) records and ocean model results, Lee et al. (2013) argued that there was little effect of deglacial sea-level change on the path of the Kuroshio, which still exited the OT from the Tokara Strait during the glacial period. Because the main stream of the Kuroshio Current is at a water depth of ~150 m, the SST records are insufficient to decipher past changes of the Kuroshio (Ujiié et al., 2016). On the other hand, low abundances of *P. obliquiloculata* in core CSH1 in the northern OT (Figure 6e) indicate that the main flow path of the Kuroshio may have migrated to the east of the Ryukyu Island (Shi et al., 2014). Such a flow change would have been caused by the proposed block of the Ryukyu-Taiwan land bridge by low sea level (Ujiié and Ujiié, 1999) and an overall reduced Kuroshio intensity (Kao et al., 2006), effectively suppressing the effect of the Kuroshio on deep ventilation in the OT. Our RSEs data show that oxygenated sedimentary conditions were dominant in the northern OT throughout the last glacial period (Figures 6b, c). The Kuroshio thus likely had a weak or even no effect on the renewal of oxygen to the sedimentary environment during the last glacial period. More recently, lower hydrothermal total Hg concentration during 20 ka - 9.6 ka, associated with reduced intensity and/or variation in flow path of KC, relative to that of Holocene recorded in core KX12－3 (1423 water depth) (Lim et al., 2017), further validates our inference.

On the other hand, the gradually increased alkenone-derived SST and abundance of *P.obliquiloculata* (Figures 6d and 6e) from 15 ka onwards indicates an intensified Kuroshio Current. At present, mooring and float observations revealed that the KC penetrates to 1200 m isobath in the East China Sea (Andres et al., 2015). However, as mentioned above, the effect of Kuroshio on the sedimentary oxygenation was likely very limited during the glacial period and only gradually increasing throughout the last glacial termination. Therefore, while its effect on our observed deglacial variation in oxygenation may provide a slowly changing background condition in vertical mixing effects on the sedimentary oxygenation in the OT, it cannot account for the first order, rapid oxygenation changes, including indications for millennial-scale variations, that we observe between 18 ka and 9 ka.

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

6.2.4. Effects of GNPIW on sedimentary oxygenation

Relatively stronger oxygenated Glacial North Pacific Intermediate Water (GNPIW), coined by (Matsumoto et al., 2002), has been widely documented in the Bering Sea (Itaki et al., 2012; Kim et al., 2011; Rella et al., 2012), the Okhotsk Sea (Itaki et al., 2008; Okazaki et al., 2014; Okazaki et al., 2006; Wu et al., 2014), off east Japan (Shibahara et al., 2007), the eastern North Pacific (Cartapanis et al., 2011; Ohkushi et al., 2013) and western subarctic Pacific (Keigwin, 1998; Matsumoto et al., 2002). The intensified formation of GNPIW due to the displacement of source region to the Bering Sea was proposed by Ohkushi et al. (2003) and then is confirmed by Horikawa et al. (2010). Under such conditions, the invasion of well-ventilated

GNPIW into the OT through the Kerama Gap would have replenished the water column oxygen in the OT, although the penetration depth of GNPIW remains under debate (Jaccard and Galbraith, 2013; Okazaki et al., 2010; Rae et al., 2014). Both a gradual decrease in excess U concentration and an increase in Mo/Mn ratio during the last glacial period (25 ka-50 ka) validate such inference, suggesting pronounced effects of intensified NPIW formation in the OT.

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

Hypoxic conditions during the B/A have been also widely observed in the mid- and high-latitude North Pacific (Jaccard and Galbraith, 2012; Praetorius et al., 2015). Our data of excess U concentration and Mo/Mn ratio recorded in core CSH1 (Figures 6b and 6c), together with enhanced denitrification and *B.aculeata* abundance (Figures 6f and 6h), further reveal the expansion of oxygen-depletion at mid-depth waters down to the subtropical NW Pacific during the late deglacial period. Based on high relative abundances of radiolarian species, indicators of upper intermediate water ventilation in core PC-23A, Itaki et al. (2012) suggested that a presence of well-ventilated waters was limited to the upper intermediate layer (200 m–500 m) in the Bering Sea during warm periods, such as the B/A and Preboreal. Higher B-P foraminiferal ^{14}C ages, together with increased temperature and salinity at intermediate waters recorded in core GH02-1030 (off East Japan) supported a weakened formation of NPIW during 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, Mo/Mn ratio and excess U show a slightly decreased oxygen condition in the northern OT. By contrast, benthic foraminiferal $\delta^{18}O$ and $\delta^{13}C$ 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 characteristics climate features in the Northern Hemisphere, in particular the North Atlantic is millennial-scale oscillations during the glacial and deglacial periods. These abrupt climatic events have been widely thought to be related to varying strength of Atlantic Meridional Overturning Circulation (AMOC) (Lynch-Stieglitz, 2017). One of dynamic proxies of ocean circulation, $^{231}Pa/^{230}Th$ 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 AMOC during the B/A.

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

Increased NPIW formation during HS1 may have been caused by enhanced salinity-driven vertical mixing through higher meridional water mass transport from the subtropical Pacific. Previous studies have proposed that intermediate water formation in the North Pacific hinged on a basin-wide increase in sea surface salinity driven by changes in strength of the summer EAM and the moisture transport from the Atlantic to the Pacific (Emile-Geay et al., 2003). Several modeling studies found that freshwater forcing in the North Atlantic could cause a widespread surface salinification in the subtropical Pacific Ocean (Menviel et al., 2014; Okazaki et al., 2010; Saenko et al., 2004). This idea has been tested by proxy data (Rodríguez-Sanz et al., 2013; Sagawa and Ikehara, 2008), which indicated a weakened summer EAM and reduced transport of moisture from Atlantic to Pacific through Panama Isthmus owing to the southward displacement of Intertropical Convergence Zone caused by a weakening of AMOC. Along with this process, as predicted through a general circulation modeling, a strengthened Pacific Meridional Overturning Circulation would have transported more warm and salty subtropical water into the high-latitude North Pacific (Okazaki et al., 2010). In accordance with comprehensive Mg/Ca ratio-based salinity reconstructions, however, Riethdorf et al. (2013) found no clear evidence for such higher salinity patterns in the subarctic northwest Pacific during HS1.

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

During the late deglaciation, ameliorating global climate conditions, such as warming Northern Hemisphere, and a strengthened Asian summer monsoon, are a result of changes in insolation forcing, greenhouse gases concentrations, and variable strengths of the AMOC (Clark et al., 2012; Liu et al., 2009). During the B/A, a decrease in sea ice extent and duration, as well as reduced advection of Alaska Stream waters were indicated by combined reconstructions of SST and mixed layer temperatures from the subarctic Pacific (Riethdorf et al., 2013). At that time, the rising eustatic sea level (Spratt and Lisiecki, 2016) would have supported the intrusion of Alaska Stream into the Bering Sea by deepening and opening glacial closed straits of the Aleutian Islands chain, while reducing the advection of the Alaska Stream to the subarctic Pacific gyre (Riethdorf et al., 2013). In this scenario, saltier and more stratified surface water conditions would have inhibited brine rejection and subsequent formation and ventilation of NPIW (Lam et al., 2013), leading to a reorganization of the Pacific water mass, closely coupled to the collapse and resumption modes of the AMOC during these two intervals.

6.4 Increased storage of CO_2 at mid-depth water in the North Pacific at the B/A

One of the striking features of RSEs data is higher Mo/Mn ratio and excess U concentration at the B/A, supporting an expansion of Oxygen Minimum Zone in the North Pacific (Galbraith and Jaccard, 2015; Jaccard and Galbraith, 2012; Moffitt et al., 2015) and coinciding with the termination of atmospheric CO_2 concentration rise (Marcott et al., 2014) (Figure 7e). As described above, it can be related to the upwelling of nutrient- and CO_2-rich Pacific Deep Water due to resumption of AMOC and enhanced export production. Although here we are unable to distinguish these two reasons from each other, boron isotope data measured on surface-dwelling foraminifera in core MD01-2416 situated in the western subarctic North Pacific did reveal a decrease in near-surface pH and an increase in pCO_2 at this time (Gray et al., 2018). That is to say, subarctic North Pacific is a source of relatively high atmospheric CO_2 concentration at the B/A. Here we cannot conclude that the same processes could have occurred in the subtropical North Pacific due to the lack of well-known drivers to draw out of the old carbon in the deep sea into the atmosphere. However, an expansion of oxygen-depletion zone in the entire North Pacific suggest an increase in respired carbon storage at intermediate-depth in the subtropical North Pacific, which likely stalls the rise of atmospheric CO_2. Our results support the findings by Galbraith et al. (2007) and are consistent with the hypothesis of deglacial flushing of respired carbon dioxide from an isolated, deep ocean reservoir (Marchitto et al., 2007; Sigman and Boyle, 2000). Given the sizeable volume of the North Pacific, potentially, once the respired carbon could be emitted to the atmosphere in stages, which would play an important role in propelling the Earth out of the last ice age (Jaccard and Galbraith, 2018).

7. Conclusions

Our geochemical results of sediment core CSH1 revealed substantial changes in intermediate water redox conditions in the northern Okinawa Trough over the last 50 ka on orbital and millennial timescales. Enhanced sedimentary oxygenation mainly occurred during cold intervals, such as the last glacial period, Heinrich stadials 1 and 2, and during the middle and late Holocene, while diminished sedimentary oxygenation prevailed during the Bölling-Alleröd and Preboreal. The sedimentary oxygenation variability presented here provides key evidence for the substantial impact of ventilation of NPIW on the sedimentary oxygenation in the subtropical North Pacific and shows out-of-phase pattern with North Atlantic Climate during the last deglaciation. The linkage is attributable to the disruption of NPIW formation caused by climate changes in the North Atlantic, which is transferred to the North Pacific via atmospheric and oceanic teleconnections. We also suggest an expansion of oxygen-depleted zone and accumulation of respired carbon at the mid-depth waters of the North Pacific during the B/A, coinciding with the termination of atmospheric CO_2 rise. A step-wise injection of such respired carbon into the atmosphere, the mechanism likely to propel the Earth out of glacial climate, would be helpful to maintain high atmospheric CO_2 levels during the deglaciation.

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

Author Contributions. J.J.Z. and X.F.S. conceived the study. A.M.Z. performed geochemical analyses of bulk sediments. J.J.Z., X.F.S. K.S. and X.G. led the write up of the manuscript. All other authors provided comments on the manuscript and contributed to the final version of the manuscript.

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Captions

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

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

Figure 1. (a) Spatial distribution of dissolved oxygen content at 700 m water depth in the North Pacific. Black arrows denote simplified Kuroshio and Oyashio circulations and North Pacific Intermediate Water (NPIW) in the North Pacific. The red thick dashed line indicates transformation of Okhotsk Sea Intermediate Water (OSIW) by cabbeling the subtropical NPIW along the subarctic-tropical frontal zone (You, 2003). The light brown solid line with arrow indicates the spreading path of subtropical NPIW from northeast North Pacific southward toward the low-latitude northwest North Pacific (You, 2003). Yellow solid lines with arrow represent two passages through which NPIW enter into the Okinawa Trough. This figure was created with Ocean Data View (odv.awi.de). (b) Location of sediment core CSH1 investigated in this study (red diamond). Also shown are locations of sediment cores PN-3，E017，255 and MD012404 investigated previously from the Okinawa Trough, GH08-2004 from the East of Ryukyu Island, GH02-1030 off the east of Japan, PC-23A from the Bering Sea and ODP167-1017 from the northeastern Pacific. Letters A to E represent the sediment cores from and near the OT. The detailed information for these cores is shown in Table 1.

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

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

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

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

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

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

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

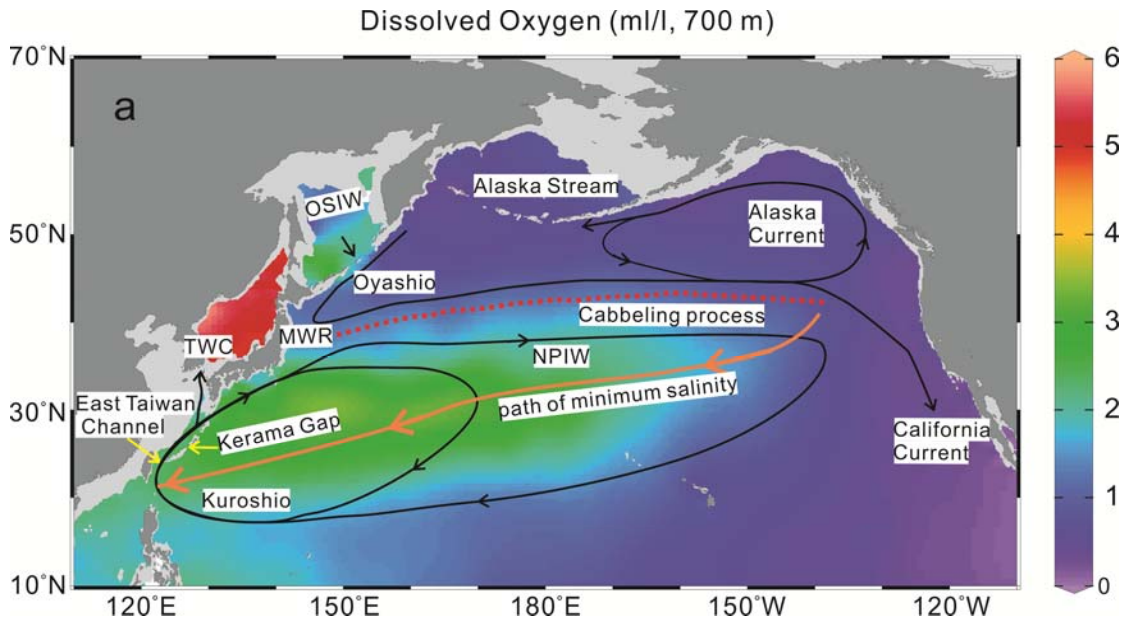
Table 1

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)
	ODP167-1017	34.54	239.11	955	NE Pacific	Cannariato and Kennett, (1999)

Table 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)

Fig.1


Dissolved Oxygen (ml/l, 700 m)
70°N
50°N
30°N
10°N
120°E
150°E
180°E
150°W
120°W
a
6
5
4
3
2
1
0
Alaska Stream
OSIW
Alaska Current
Oyashio
Cabbeling process
MWR
TWC
NPIW
East Taiwan Channel
path of minimum salinity
Kerama Gap
California Current
Kuroshio

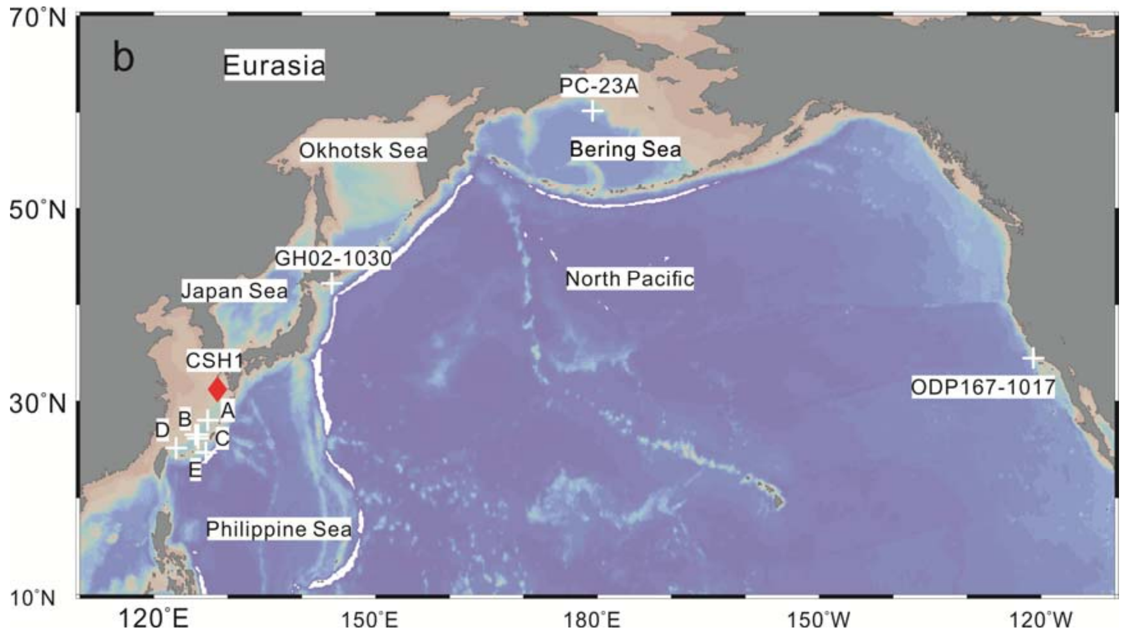

70°N
50°N
30°N
10°N
120°E
150°E
180°E
150°W
120°W
b
Eurasia
PC-23A
Okhotsk Sea
Bering Sea
GH02-1030
North Pacific
Japan Sea
CSH1
A
B
C
D
E
ODP167-1017
Philippine Sea

Fig.2

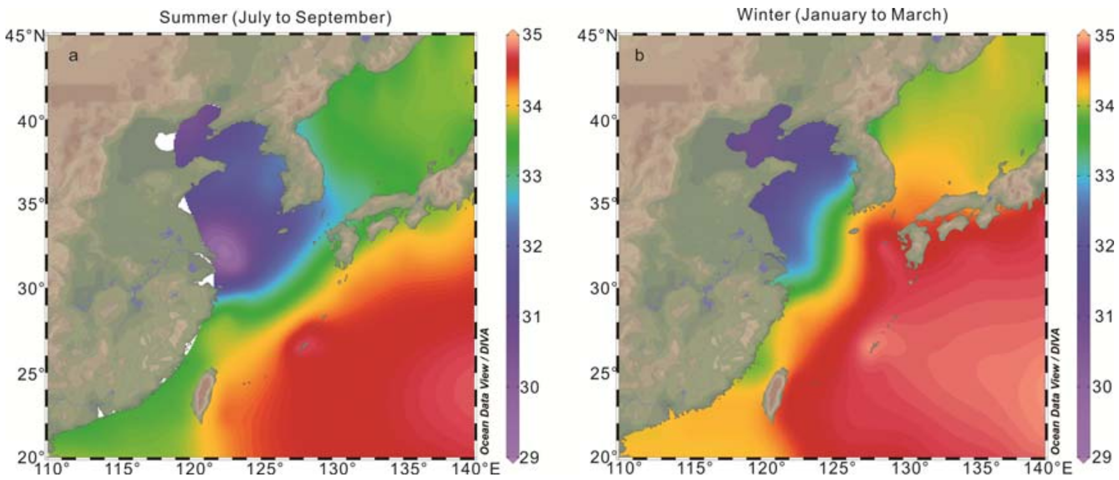

Summer (July to September)
Winter (January to March)
a
b
45°N
40°
35°
30°
25°
20°
110°
115°
120°
125°
130°
135°
140°E
Ocean Data View / DIVA
35
34
33
32
31
30
29

Fig.3

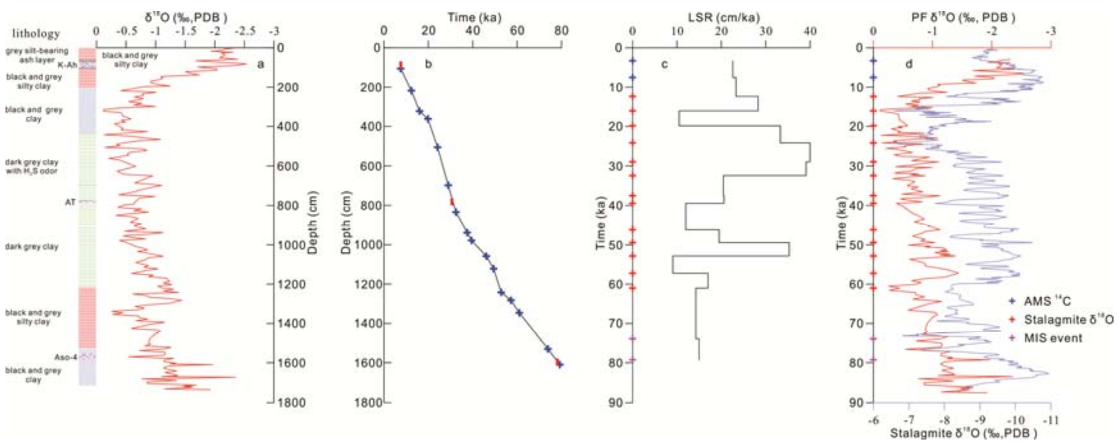
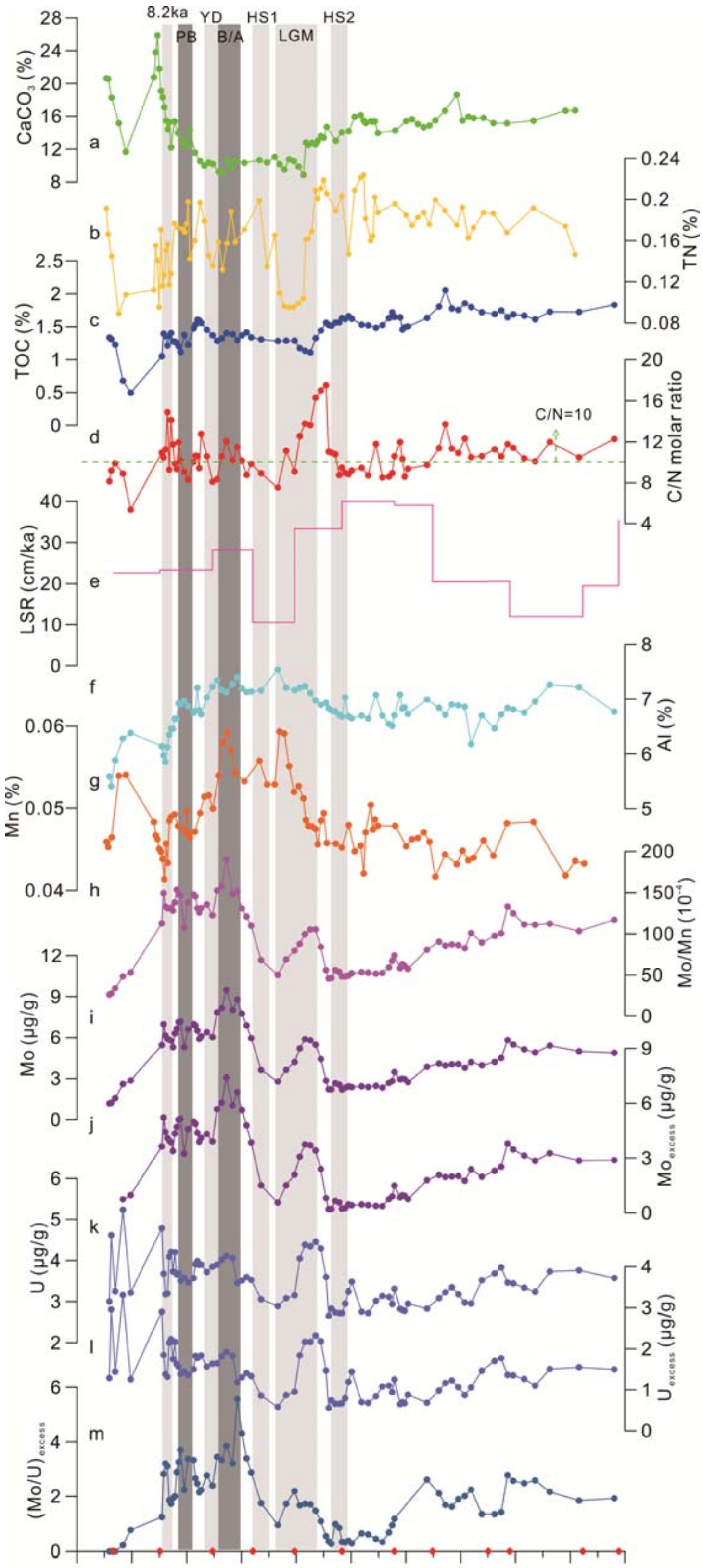
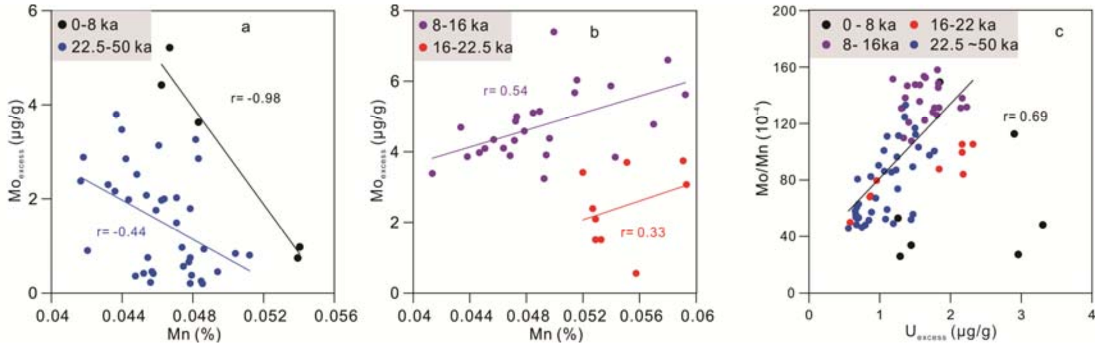

lithology
grey silt-bearing ash layer
K-Ah
black and grey silty clay
black and grey clay
dark grey clay with H2S odor
AT
dark grey clay
black and grey silty clay
Aso-4
black and grey clay
δ18O (‰,PDB)
Depth (cm)
Time (ka)
LSR (cm/ka)
PF δ18O (‰, PDB)
Stalagmite δ18O (‰,PDB)
AMS 14C
Stalagmite δ18O
MIS event
a
b
c
d

Fig.4



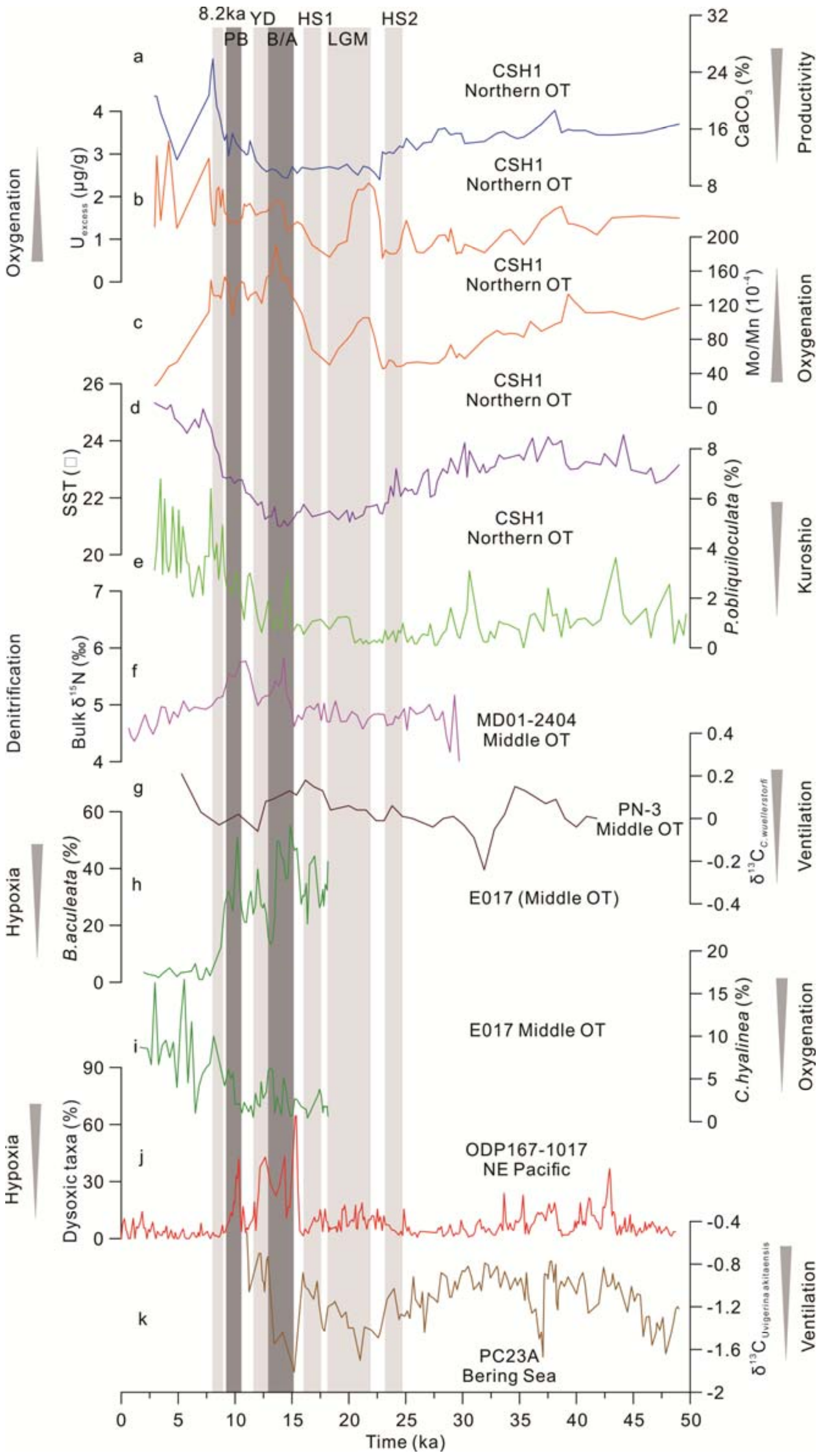
8.2ka
YD
HS1
HS2
PB
B/A
LGM
$CaCO_3$ (%)
TN (%)
TOC (%)
C/N molar ratio
C/N=10
LSR (cm/ka)
Al (%)
Mn (%)
Mo/Mn (10^{-4})
Mo (μg/g)
Mo_{excess} (μg/g)
U (μg/g)
U_{excess} (μg/g)
$(Mo/U)_{excess}$
a
b
c
d
e
f
g
h
i
j
k
l
m
Time (ka)

Fig.5



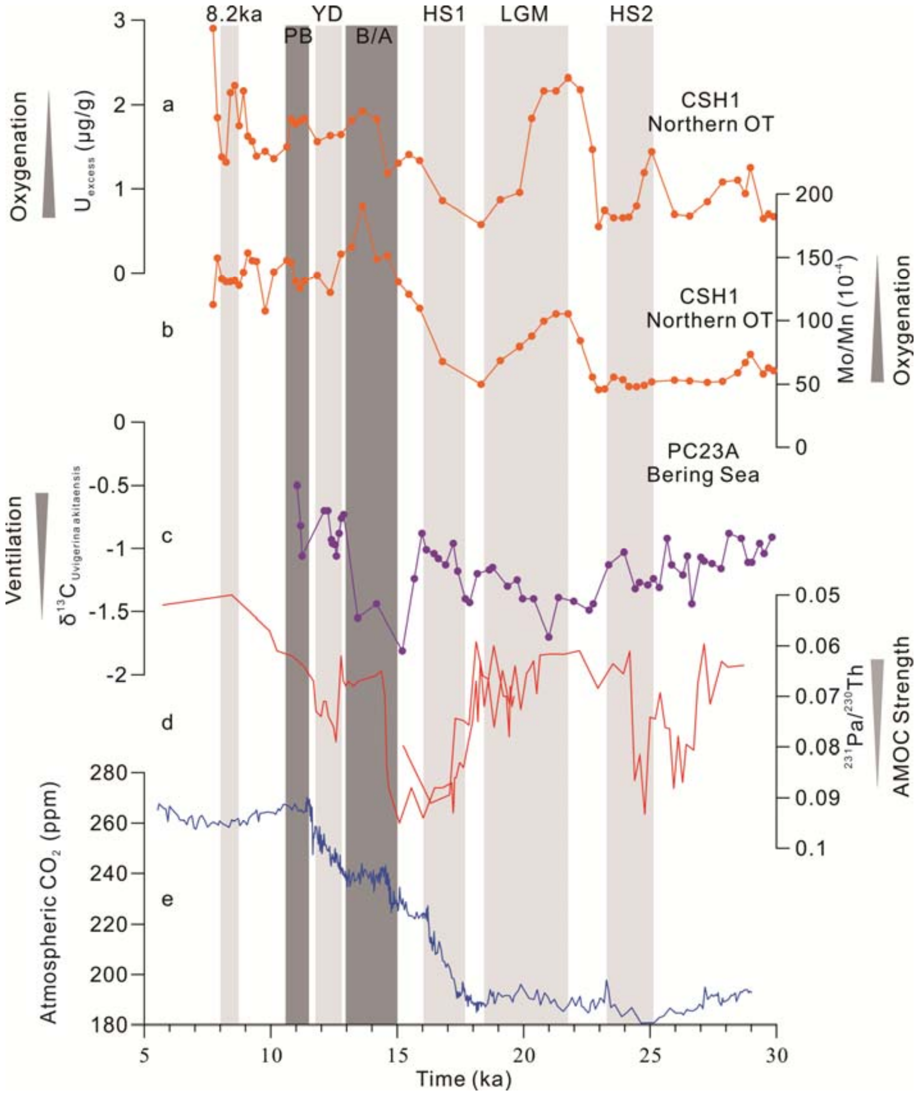
0-8 ka
22.5-50 ka
a
r= -0.98
r= -0.44
Mo_excess (μg/g)
Mn (%)
8-16 ka
16-22.5 ka
b
r= 0.54
r= 0.33
Mo_excess (μg/g)
Mn (%)
0 - 8 ka
16-22 ka
8- 16ka
22.5 ~50 ka
c
r= 0.69
Mo/Mn (10^-4)
U_excess (μg/g)

Fig.6



8.2ka
YD
HS1
HS2
PB
B/A
LGM
a
CSH1 Northern OT
CaCO3 (%)
Productivity
Oxygenation
Uexcess (μg/g)
b
CSH1 Northern OT
CSH1 Northern OT
c
Mo/Mn (10⁻⁴)
Oxygenation
CSH1 Northern OT
d
SST
CSH1 Northern OT
e
P.obliquiloculata (%)
Kuroshio
Denitrification
Bulk δ15N (‰)
f
MD01-2404 Middle OT
g
PN-3 Middle OT
δ13C C.wuellerstorfi
Ventilation
Hypoxia
B.aculeata (%)
h
E017 (Middle OT)
E017 Middle OT
i
C.hyalinea (%)
Oxygenation
Hypoxia
Dysoxic taxa (%)
j
ODP167-1017 NE Pacific
k
PC23A Bering Sea
δ13C Uvigerina akitaensis
Ventilation
Time (ka)

Fig.7



8.2ka
YD
HS1
LGM
HS2
PB
B/A
Oxygenation
U_{excess} (μg/g)
a
CSH1
Northern OT
b
CSH1
Northern OT
Mo/Mn (10^{-4})
Oxygenation
PC23A
Bering Sea
Ventilation
$\delta^{13}C_{Uvigerina\ akitaensis}$
c
d
$^{231}Pa/^{230}Th$
AMOC Strength
Atmospheric CO_2 (ppm)
e
Time (ka)