Point-by-point response to referee comments

Response to reviewer

Key:

- Reviewers' comments

- Author's response

- Modified text in the manuscript

Anonymous Referee #1

Received and published: 18 July 2019

Review Zou et al 'Millennial-scale variations of sedimentary oxygenation in the

westernsubtropical North Pacific and its link to North Atlantic climate.

Zou et al. study sedimentary redox conditions in the Okinawa Trough, and use

this as a proxy to infer bottom water oxygen concentrations (I think, it is not always

very clear from the manuscript). The title promises more than the paper delivers; the

link with North Atlantic climate is only mentioned briefly and explanation is

sometimes unclear.

The authors present interesting data, but the paper itself needs work. There is so

much information (several times incorrectly referenced), and some information seems

irrelevant. There is also a lot of internal discussion within the paper without reaching

firm conclusions. While the authors are critical about their own proxy, they are less so

about others and this needs to be improved.

Reply#1: We thank Reviewer#1 for taking the time to review our manuscript and

for the constructive comments, which contribute improving our manuscript. Our work

adds an important element to understand the process in the subtropical North Pacific

during the last deglaciation. Our data suggest a substantial impact of NPIW on

sedimentary oxygenation in the western subtropical North Pacific and also an

expansion of oxygen minimum zone in the North Pacific during the B/A.

According to the comments of Reviewers #1 and #2, we have carried out a

thorough revision of our entire manuscript, including Figures 3-7, and use the "track

changes" to display our revisions the text. In the revised manuscript, we re-phrased

the abstract, amended the age model (see new Figure 3), changed sentences and paragraphs as outlined in the revised text, added new evidence of benthic foraminifera abundance of core E017 (see new Figure 6) and added text to the discussion on the connection of our time series to North Atlantic climate. We hope that we have addressed Reviewer #1' concerns appropriately.

In the following we provide point-by-point responses to all the Reviewer's comments in blue as well as excerpts from the manuscript in green.

Comments: Authors should check that their references are appropriate. Several references are not put in the right context.

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Reply#2: Thanks. Checked and Revised.
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The following references were included in the revised MS:
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in Line 66 for reference Hoogakker et al., 2015

in Line 75 for reference Addison et al., 2012

in Lines 100 for reference Max et al 2017; Rippert et al. 2017

in Line 107 for reference Sibuet et al., 1987

in Line 170 for reference Morford and Emerson, 1999

in Line 333 for references Cartapanis et al., 2011; Lembke-Jene et al., 2017

in Line 348 for reference Shi et al., 2014

in Line 356 for reference Chang et al., 2009

in Line 368 for reference Zhu et al., 2015

in Line 372 for reference Lim et al., 2017

in line 396 for reference Bianchi et al. 2012

in lines 444-445 for references Galbraith and Jaccard, 2015; Moffitt et al., 2015;

Praetorius et al., 2015

in line 465 for reference Brewer and Peltzer, 2016

in line 496 for reference Galbraith et al., 2007

in line 530 for reference Lim et al., 2017

in line 535 for reference Andres et al., 2015

in line 551 for reference Kubota et al., 2015

in line 578 for reference Kubota et al., 2015

in line 623 for reference Lynch-Stieglitz, 2017

in lines 625-630 for references Böhm et al., 2015; McManus et al., 2004; Liu et al., 2009; Zhang et al., 2017; Barker et al., 2010; Knorr and Lohmann, 2007;

in lines 697-698 for references Galbraith and Jaccard, 2015; Jaccard and Galbraith, 2012; Moffitt et al., 2015

in lines 709-710 for references Addison et al., 2012; Cartapanis et al., 2011; Crusius et al., 2004; Galbraith et al., 2007; Lembke-Jene et al., 2017; Shibahara et al., 2007

The following original references were removed in the revised MS:

in line 65 Lu et al., 2016;

in line 400 Savrda and Bottjer, 1991

in line 471 Benson and Krause, 1984

in line 541 Matsumotoet al. (2002)

in line 557 Kubota et al., 2010

in line 565 Wang and Wang, 2008

in line 584 Kubota et al., 2010

There are several other studies that deal with the North Pacific and NPIW, which are not referenced here; this includes work by Rippert et al. (2017), Max et al. (2017).

Reply#3: Thanks. These two references have been included in the revised manuscript and we briefly introduced their main findings.

In particular, both these papers highlight the substantial effects of NPIW on subsurface water composition of Eastern Tropical North Pacific and potent roles in regulating global climate. Likewise, the effect of NPIW's ventilation on the western subtropical North Pacific is observed in our study. Our study further validates the role of NPIW in its downstream oceanic environment.

We have added Lines 97-100

"In contrast, substantial effects of intensified NPIW formation during Marine Isotope Stage (MIS) 2 and 6 on the ventilation and nutrient characteristics of lower latitude mid-depth Eastern Equatorial Pacific have been suggested by recent studies (Max et al., 2017; Rippert et al., 2017)."

Abstract: Lines 44-50: these sentences go around the bushes. Really what you want to say is that sedimentary oxygenation conditions at mid-depth in the subtropical western North Pacific were more or less similar over the last 50,000 years, apart from the Bolling-Allerod and Pre-boreal. However, it may not be possible to compare with Holocene data, as this may be compromised by ash. This is not made very clear in the manuscript.

Reply#4: Thanks for your suggestion. We have rephrased these sentences in the revised manuscript. The sentence was amended as follows. The text was modified as follows (Lines 42-45):

"Our results suggest that enhanced mid-depth western subtropical North Pacific (WSTNP) sedimentary oxygenation occurred during cold intervals and after 8.5 ka, while oxygenation decreased during the Bölling-Alleröd (B/A) and Preboreal."

As suggested by the Reviewer, our data suggest well-oxygenated water during the cold intervals apart from the B/A and Preboreal. For the Holocene, increased sedimentary oxygenation is attributed to an intensified Kuroshio Current, although discrete volcanic materials, indicating by positive Eu anomaly (Zhu et al., 2015), would dilute the Holocene data. Modern observation suggest the Kuroshio can reach to the seafloor at 1200 m in isobath in the East China Sea (Andres et al., 2015). In the geological past, both various proxy data and modeling simulation suggest intensified Kuroshio re-entered the OT at early Holocene 9-9.6 ka (Chang et al., 2015; Diekmann et al., 2008; Dou et al., 2016; Lim et al., 2017; Zheng et al., 2016). In our previous study based on the same core CSH1 (Zhu et al., 2015), we have suggested that the occurrence of discrete volcanic materials during the Holocene is closely related to enhanced Kuroshio intensity. More recently, increased total Hg concentration in the sediments from the middle Okinawa since 9.3 ka (Lim et al., 2017) was also suggested, which was explained to be hydrothermal Hg source (due to much higher concentrations than potential terrigenous end-members) and to be brought to the site location by intensified Kuroshio Current. Although the focus of this manuscript is not the ventilation changes during the Holocene, the Kuroshio Current does play a crucial role in controlling the ventilation of the OT during the Holocene.

We have now added more information into the section 5.2 Redox-sensitive Elements. Lines 366-372:

"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)."

Lines 59-61: how does it seem to be driven? Is this not something you are proposing? Then it is not seem.

Reply#5: Thanks. Revised. "seem to be driven" is replaced by "was likely driven". Lines 48-51 in the revised MS. The sentence now reads:

"The enhanced formation of NPIW during Heinrich Stadial 1 (HS1) was likely driven by the perturbation of sea ice formation and sea surface salinity oscillations in high-latitude North Pacific."

The authors mix up NADW and the Atlantic Meridional Overturning Circulation. For a good description of AMOC see recent paper by Frajke-Williams et al. (2019).

Reply#6: Thanks. We checked the paper by Frajke-Williams et al. (2019). In the revised manuscript, we replaced AMOC by North Atlantic Deep Water and reworded these sentences. Lines 51-57 in the revised MS. The sentences now read:

"The diminished sedimentary oxygenation during the B/A due to decreased NPIW formation and enhanced export production, indicates an expansion of oxygen minimum zone in the North Pacific and enhanced CO₂ sequestration at mid-depth waters, along with termination of atmospheric CO₂ concentration increase. We attribute the millennial-scale changes to intensified NPIW and enhanced abyss flushing during deglacial cold and warm intervals, respectively, closely related to variations in North Atlantic Deep Water formation."

Introduction: Lines 70-73: not sure how to interpret this. Where is the respired carbon stored? At the sediment-seawater interface, in sedimentary pore-waters, or in seawater? The study of Lu et al. (2016) deals with I/Ca in planktic foraminifer in the Pacific Sector of the Southern Ocean, to reconstruct upper ocean oxygenation, the part of respired carbon in their paper refers to a different study (Hoogakker et al., 2015).

Reply#7: We have reworded the first sentence for a clearer statement. Additionally, the reference (Lu et al., 2016) was replaced by reference (Hoogakker et al., 2015).

The sentence was amended as follows, Lines 61-66:

"A more sluggish deep ocean ventilation combined with a more efficient biological pump widely thought to facilitate enhanced carbon sequestration in the ocean interior, leading to atmospheric CO₂ drawdown during glacial cold periods (Sigman and Boyle, 2000). These changes are tightly coupled to bottom water oxygenation and sedimentary redox changes on both millennial and orbital timescales (Hoogakker et al., 2015; Jaccard and Galbraith, 2012; Sigman and Boyle, 2000)."

Lines 76-83: the study of Cartapanis is from the northeastern Pacific, but not high latitude or subarctic.

Reply#8: The study by Cartapanis et al. (2011) presents high-resolution redox-sensitive trace metals in a sediment core from the Eastern Tropical North Pacific and proposed improved intermediate water oxygenation during Heinrich events. The enhanced ventilation during Heinrich intervals is consistent with our inference. Therefore, we would like to continue to keep this reference in the revised manuscript and rephrased the expression of the context in Lines 69-72. Now the sentence reads:

"Previous studies from North Pacific margins as well as open subarctic Pacific have identified drastic variations in export productivity and ocean oxygen levels at millennial and orbital timescales using diverse proxies such as trace elements (Cartapanis et al., 2011; Chang et al., 2014; Jaccard et al., 2009; Zou et al., 2012),"

Line 92: explain what cabbeling is, not everyone will have heard of the term.

Reply#9: Done. Cabbeling is a mixing process to form a new water mass with increased density than that of parent water masses. Lines 85-86 in the revised MS. The sentence was changed to:

"..... 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"

Lines 95- 97: do the data really show this? The one core at 1km is about 0.04 per mil lighter (and within error), but crucially there is no Holocene equivalent for the 0.7 km core.

Reply#10: These two studies show enhanced formation of North Pacific Intermediate Water during the last glaciation. On the basis of >30 sediment cores on the northern Emperor Seamounts and in the Okhotsk Sea with a water depth of 1000 to 4000 m, Keigwin (1998) found that there was a better ventilated and relatively fresher water mass above 2000 m in the far northwestern Pacific compared to deep waters. Matsumoto et al. (2002) compiled and compared available nutrient proxies (δ^{13} C) in sediment cores with water depth of 740 m to 3320 m in the North Pacific and found a presence of distinctive water masses below and above 2000 m water depth in the glacial Pacific with higher benthic δ^{13} C in the upper 2000 m water.

Line2 149-152: do you mean 'is governed by' instead of 'is the balance between'.

Reply#11: Revised. Lines 150-153 in the revised MS. Now the sentence reads:

"The sedimentary redox conditions are 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."

Figure 1. O2 map, and locations of cores. Are all the cores discussed in the paper? Is it worrying that the main core CSH1 is from just south of Japan and perhaps should not be considered an open ocean core? What do the letters A to E stand for?

Reply#12: All cores shown in Figure 1b have been discussed in this manuscript. For example, benthic δ^{13} C in core PN-3 was used to indicate ventilation change in the OT. Concentrations of CaCO₃ and reactive phosphorus recorded in core MD01-2404 were used to correlate with our productivity proxy. Benthic foraminiferal assemblages in cores E017 and 225 retrieved from the middle and southern OT were used to indicate the ventilation of deep water mass in the OT. Deep-water temperatures in cores GH08-2004 and GH02-1030, benthic δ^{13} C in core PC23A and benthic foraminiferal assemblages in ODP Site 1017 have implicated the ventilation of North Pacific Intermediate Water.

Core CSH1 is situated in the northern Okinawa Trough at a water depth of 703 m. In this area, both surface and deep water can be continuously replenished by water masses from open ocean.

Letters A to E stand for sediment cores previously reported in and near the Okinawa Trough and are shown in Table 1.

Setting: Do details of discharge and SSS add anything to this study?

Reply#13: Sea surface salinity (SSS) in the East China Sea is closely related to the intensity of the summer East Asian Monsoon (EAM). On the basis of this relationship, a recent study from the northern Okinawa Trough (U1429) has extended this relationship back to 400 ka (Clemens et al., 2018). In order to discern millennial-scale variability, we correlate planktic δ^{18} O of core CSH1 with Chinese stalagmite δ^{18} O, an indicator of summer EAM to establish the age model for core CSH1 in our study. We would like to add the details of discharge and SSS in the manuscript to let the readers know the details.

Material and methods: What causes the high accumulation rates in this core? As the accumulation rates vary significantly, how do these influence the patterns in redox elements etc?

Reply#14: In the northern Okinawa Trough, previous sediment provenance studies suggested the terrigenous sediments are mainly sourced from the Yellow and Changjiang Rivers, from China and short mountainous rivers (Japanese or even

Taiwanese rivers) from the surrounding islands, as well as eolian dust, volcanic and hydrothermal materials from Yellow River, Changjiang and part of Korea and Japan Rivers (Beny et al., 2018; Li et al., 2015; Zhao et al., 2018; Zhao et al., 2017). Variation in sedimentation rate has been attributed to changes in eustatic sea level, EAM intensity, path and/or intensity of Kuroshio Current. Generally, sea level is thought to be the first-order factor for controlling linear sedimentation rate changes (Beny et al., 2018; Li et al., 2015; Zhao et al., 2017).

Our data show that there is no coherent relationship between linear sedimentation rate and concentrations of redox sensitive elements. For example, high sedimentation rate between 24.2 ka and 32.4 ka (around 40 cm/ka) corresponds to decreasing concentrations of redox sensitive elements. On the other hand, lower sedimentation between 16 ka and 19.8 ka also corresponds to lower concentrations of redox sensitive elements. Therefore, linear sedimentation rate is not deemed to be a crucial factor in controlling concentrations of redox sensitive elements in core CSH1.

Lines 339-340: preservation of TOC and CaCO3 are influenced by many factors and not a widely used paleo-export proxy.

Reply#15: We fully agree that preservation of TOC and CaCO₃ are influenced by many factors, including supply, dissolution, organic matter degradation, terrigenous dilution, etc. Some factors can be ruled out and at times these two proxies have been used to reconstruct export productivity. In this study, C/N molar shows substantial contribution of terrigenous organic matter to total organic carbon, therefore it is not a suitable proxy for productivity reconstruction.

In the revised manuscript, we showed multiple lines of evidence to support the utility of CaCO₃ as a reliable productivity proxy, including (1) a strong negative correlation with terrigenous Al of core CSH1; (2) weak dilution effects of terrigenous material on CaCO₃; (3) similar pattern to sea surface temperature of core CSH1 (Shi et al., 2014) (the data have been included in Figure 6); (4) similar deglacial trends in CaCO₃ and reactive phosphorus reported in core MD012404 retrieved from the middle OT (Chang et al., 2009; Li et al., 2017). All these lines of evidence support CaCO₃ as a proxy for productivity in the study area. We have added these information

in lines 339-358 in the revised MS:

"Several lines of evidence support CaCO₃ as a reliable productivity proxy, particularly during the last deglaciation. The strong negative correlation coefficient (r = -0.85, p<0.01) between Al and CaCO₃ in sediments throughout core CSH1 confirms the biogenic origin of CaCO₃ against terrigenous Al (Figure 4f). Generally, terrigenous dilution decreases the concentrations of CaCO₃. An inconsistent relationship between CaCO₃ contents and sedimentation rates indicates a minor effect of dilution on CaCO₃. Furthermore, the increasing trend in CaCO₃ associated with high sedimentation rate during the last deglacial interval indicates a substantial increase in export productivity (Figures 4a and d). The high coherence between CaCO₃ content and alkenone-derived sea surface water (SST) (Shi et al., 2014) indicates a direct control on CaCO₃ by SST. Moreover, a detailed comparison between CaCO₃ 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 CaCO₃ 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, a similar deglacial trend in CaCO₃ 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 CaCO₃ of core CSH1 as a reliable productivity proxy to a first order approximation."

Line 354-357: have you checked that it is an extant biological component that makes up the high CaCO3 going from B/A to ~ 8000 years? Can you explain the differences in the LSR figures between Figures 3 and 4? For examples, in Figure 3 highest rates occur centred around 22 kyrs as part of a large interval of high LSR (from 30 to 20), whereas in Figure 4 this occurs earlier (33 to 24 kyrs).

Reply#16: Seven samples at 8.23ka (120-124cm), 9.26ka (144-148cm), 10.98ka (184-188cm), 11.66ka (200-204cm), 12.92ka (232-236cm), 14.05ka (264-268cm), and 15.18ka (296-300cm) in core CSH1 were analyzed by Modular Stereo

Microscope (Zeiss SteREO Discovery V12) to look into the sediment components. It

is clear that abundant biogenic tests, especially foraminiferal tests, are observed

during these sediment intervals (Figure A1). On the other hand, increased

concentration of CaCO3 is highly coherent with the abundance of planktic

foraminifera species G.ruber and SST (Shi et al., 2014), indicating a substantial effect

of SST on CaCO₃.

In the original manuscript, we made a mistake for LSR in Figure 3. In the revised

manuscript, this issue has been corrected (Figure 3).

In addition, high sedimentation (>40-60 cm/ka) in the original manuscript mainly

occurred during HS2 and HS3. This can be caused by uncertainties of age control

points at 23.476 ka (DO2) and 29.995 ka (H3). In the revised manuscript, these two

age control points have been eliminated from the Chinese Stalagmite tuned age model.

Even with this more conservative tuning approach, the conclusions on sedimentary

oxygenation variations remain the same as before and robust.

Lines 365-266: if U concentrations are affected by volcanic material over the last

8.5 yrs, then surely so are other sedimentary properties? I would like to see an

argument in the main text discussing why certain proxy methods are deemed not to be

influenced by this volcanic material, whilst others are. If it turns out that interval

should not be used than this creates the complication of not being able to compare the

down core data with more modern.

Reply#17: The occurrence of volcanic material has been confirmed by positive

Eu anomalies and it has substantial effects on concentration of terrigenous materials

(Zhu et al., 2015). This argument has been included in the revised text. Although the

focus of this manuscript is not the ventilation changes during the Holocene, the

Kuroshio Current does play a crucial role in controlling the ventilation of the OT

during the Holocene. Please see **Reply#4**.

Line 370: change 'seems' to 'may'.

Reply#18: Revised. "seem to" was replaced by 'may". Line 376.

Lines 372-377: are there any other studies that use Mo/Mn ratios as a sedimentary oxygenation proxy, to support your interpretation?

Reply#19: To our knowledge, Mo/Mn ratios are not used as sedimentary oxygenation proxy in previous studies. In this study, we use both Mo/U ratio and excess U concentration to reconstruct sedimentary oxygenation changes. Among these, excess U concentration has been widely used for past sedimentary oxygenation changes (Jaccard and Galbraith, 2013; Jaccard et al., 2016; Jaccard et al., 2009). The strong positive correlation coefficient between excess U and Mo/Mn ratio in core CSH1 indicates its reliability and supports our interpretation. In contrast the ratio of Mo/Mn is easier to qualitatively assess indication of low vs. high oxygen environments, supported by the individual contents of Mn and Mo, respectively.

Line 380: define oxygen deficient.

Reply#20: In the revised manuscript, "oxygen-deficient" was replaced by "suboxic". The sentence now reads as follows, Lines 387-388:

".....together with lower Mn concentrations suggest suboxic depositional conditions during the late deglacial period (15.8 ka-9.5 ka)....."

The oxygen thresholds was given in lines 396-398.

Discussion: Lines 387-392: I would recommend the authors to use more appropriate scheme that is used for sea-water that includes hypoxic, for example as defined by Bianchi et al. (2012). You will also found that suboxic is classified as $< 2-10 \mu mol/l$.

Reply#21:Thanks. The oxygen content scheme for seawater developed by Bianchi et al.(2012) has been adopted in the revised manuscript. Lines 396-398 in the revised MS. Following the Reviewer's suggestion we changed the sentence to:

"Here, we adopt the definition of oxygen thresholds by Bianchi et al. (2012) for oxic (>120 μ mol/kg O₂), hypoxic (<60–120 μ mol/kg O₂) and suboxic (<2–10 μ mol/kg O₂) conditions, whereas anoxia is the absence of measurable oxygen."

Lines 403-405: I do not understand this reasoning. You have not linked weakly restricted basin settings with euxinia?

Reply#22: We thank the reviewer for this suggestion. The previous manuscript had inaccurate statement. Now, we changed this sentence to make it more explicit. The sentence was amended as follows. Lines 409-410 in the revised MS.

"Given that the northern OT is located in an open oceanic setting, we use these two proxies to evaluate the degree of oxygenation in sediments."

It is confusing talking about ppm in the main text, whilst Figure 4 gives concentrations in $\mu g/g$.

Reply#23: Corrected. In the revised manuscript, the uniform concentration unit, μ g/g has been used for all trace elemental concentrations in the main text. Lines 411 and 417 in the revised MS.

Lines 406-412: Mo/U ratios are not shown in the manuscript. This is out of the blue.

Reply#24: Revised. Mo/U ratio has been included in new Figure 4m.

Lines 413-425: two studies. More importantly though, Figure 4 shows no benthic foraminifera data, and it is therefore impossible to confirm this claim of ventilation pattern from benthic foraminiferal assemblages to be similar to that of the RSEs. It would also be good to see a more critical discussion about this proxy.

Reply#25: We understand the point raised by the Reviewer. For this study, the benthic foraminiferal species were not counted in core CSH1. The benthic foraminiferal census data in cores E017 (1826 m water depth), 255 (1575 m water depth) are used to indicate the variations in ventilation of the middle and southern OT. The age model for core 255 (core length 655cm) was determined by two AMS ¹⁴C dates of *N. dutertrei* at depth 370 cm (9.17 ka) and 590 cm (18.8 ka) (Jian et al., 1996), whereas the age model for core E017 was established by six age control points (Li et al., 2005). Although the down-core abundance of hypoxia-affine species in both studies are similar to each other (Li et al., 2005), we here focus on the benthic foraminiferal census data from core E017.

We recalibrated the AMS ¹⁴C dates using the CALIB 7.04 software with the Marine 13 calibration dataset (Reimer et al., 2013) (ΔR=0) and compare the profiles of oxygen-like species and hypoxia-like species with our Mo/Mn and excess U. For the sake of simplicity, the abundance of *Bulimina aculeata* (hypoxia-indicating species) and *Cibicidoides hyalina* (oxygen-rich indicating species) have been included in new Figure 6 in the revised manuscript. High relative abundance of *B.aculeata* and low *C. hyalina* suggest the dominance of a hypoxic environment, whereas oxic conditions prevailed after ~7 ka. This is consistent with our RSE data, suggesting a widespread occurrence of oxygen-depleted water in the Okinawa Trough during the last deglaciation.

Lines 425-428: No. There is at least 800 meter water depth difference between your core and the others. The core of the current study is situated just above the low oxygen zone, whereas those of the other two studies are in /below the low oxygen zone.

Reply#26: Thanks for your comments. The seafloor bathymetry is much deeper in the southern OT, and shoals gradually toward the northern OT. Although our core is above the sill depth (1100 m), while others below (1100 m), previous investigations show the hydrographic characteristics in the mid-depth and deep OT are mainly regulated by the NPIW in the western boundary region of the Philippine Sea that flows into the OT through the Kerama Gap (1100 m) and the channel east of Taiwan (775 m) (Nakamura et al., 2013). Thus, ventilation signals recorded in these cores are mainly controlled by the same physical processes, though export productivity in different areas also exerts some impacts on deepwater and sedimentary oxygenation.

Lines 439-448: why are you looking at one NE Pacific to find out what is happening at your core site? There are several studies from across the Pacific that show something happening around the same time period (for example see Moffit et al., 2015), and Galbriath and Jaccard (2015), so rather than repeating the same discussion for a very small area, it would be easier to build up on those results.

Reply#27: At present, a contrasting distribution of dissolved oxygen

concentration of the subsurface water can be observed in the eastern and western North Pacific margins (Figure 1), which is characterized by strong Oxygen Minimum Zone in the eastern margin and oxic condition in the western margin. The benthic foraminiferal assemblages from ODP site 1017 exhibit a strengthening of the OMZ during warm periods and weakening during cold periods (Cannariato and Kennett, 1999). The question whether an expanded OMZ can extend toward the western NW Pacific remains elusive in the geological past during warming intervals. In fact, the key question involved is how to explain the cause of oxygenation variation on basin-wide scale. Comparison of our results from the western North Pacific with those of the eastern North Pacific aids to understand the mechanism behind sedimentary oxygen changes.

We have now added more information into the section 6.2 and all these references have been included in Lines 442-457.

"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, 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 δ^{13} C (a proxy for ventilation) 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 ODP Site 1017 (Cannariato and Kennett, 1999) (Figures 6d-k)."

Lines 454-457: no, at those high temperatures you would only get a reduction in O2 of \sim 3 for one degree warming, and 15 for a four degree warming (assuming no

large salinity changes). Higher glacial salinity would cause less reduction in O2.

Reply#28: Corrected. The reference by Brewer and Peltzer (2016) has been included in the revised manuscript. Lines 463-465. The sentence was changed to:

"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),"

Lines 457-458: sentence does not make sense.

Reply#29: We have removed the sentence in the revised manuscript.

Lines **848-846** (484-486?) : does not make sense. How does subsurface water oxygen consumption lead to lower oxygen concentrations in deeper waters?

Reply#30: Thanks for your suggestion. We have removed the sentence in the revised manuscript. The replenishment of oxygen in deep water is controlled by both lateral advection and vertical supply. The oxygen consumption in subsurface water would reduce the oxygen supply, thus lead to a lower oxygen concentration in deeper waters.

Lines 491-494: again not taking into account other factors that influence CaCO3 accumulation and preservation in sediments. For discussion on Kuroshio Current: see Lim et al. (2017).

Reply#31: Please see **Reply #16** for CaCO₃.

Total Hg concentrations (Lim et al., 2017) has been invoked to explain variations in the intensity of Kuroshio. Interestingly, they concluded that the intensity of Kuroshio strengthened rapidly since 9.3 ka, whereas weakening and/or changing path of Kuroshio occurred during the last glaciation (20 ka - 9.3 ka). This conclusion further confirms weakening effect of Kuroshio on ventilation during the glacial period but an increase since 8.5 ka, consist with our inference. We have added Lines 528-531.

"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."

Line 544-550: coined? Matsumoto et al. (2002) discuss one radiocarbon age from the Santa Barbara basin in relation to oxygen content, but at no point do they propose that GNPIW was stronger oxygenated. Cartapanis et al. (2011) and Ohkushi et al. (2013) discuss that the NE OMZ Pacific strengthened and weakened at millennial time scales, not glacial interglacial timescales. Also around the equatorial Pacific it is suggested that there was no difference in intermediate water oxygenation between the last glacial and Holocene (Hoogakker et al., 2018). Further down in the South Pacific Lu et al. (2016) suggest that upper waters were depleted in oxygen during the last glacial period.

Reply#32:In Matsumoto et al. (2002), published in Quaternary Science Reviews (2002, 21, 1693-1704), they used a compilation of benthic foraminiferal δ^{13} C to reveal the deep hydrography of the North Pacific. On page 1700 of this paper, they stated that "Although a water mass that reaches 2000 m should not be called intermediate water in the sense of modern physical oceanography, here we will refer to it as glacial NPIW (GNPIW) for the lack of a better name."

According to our understanding, GNPIW refers to the nature of NPIW during cold intervals in the geological past, which is used to describe the state of NPIW at a variety of timescales, such as glacials, stadials, and Heinrich cold events. It should be noted that, since then, this term have been widely used in the literature related to NPIW (Cartapanis et al., 2011; Max et al., 2017; Worne et al., 2019).

Lines 550-556: generalised comment, what about brine (aka Kim et al. 2011).

Reply#33: Thanks. In the revised Manuscript, we shorten these sentences. Lines 562-563 in the revised MS. The sentence now was changed to:

"The intensified formation of GNPIW due to additional source region in the Bering Sea was proposed by Ohkushi et al. (2003) and Horikawa et al. (2010)."

Lines 556-559: what is intensified GNPIW?

Reply#34:"intensified GNPIW" means improved formation of NPIW. Here, we

also use enhanced NPIW formation to replace intensified GNPIW. Lines 569-570 in the revised MS. The sentence was amended as follows.

".....validate such inference, suggesting pronounced effects of intensified NPIW formation in the OT."

Discussion lines 560-571: needs tightening, it is unclear where this goes and how it relates to this study?

Reply#35: We have rewrote these sentences. In the revised manuscript, the aim of this paragraph is to clarify the process of intensified GNPIW during HS1 and its substantial control on sedimentary oxygenation of the northern OT. Lines 571-586 in the revised MS. We have changed the sentences as follows:

"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) ¹⁴C 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 c) that is also attributed to intensified GNPIW formation."

Line 629: what is ocean ventilation seesaw? There is hardly any explanation for this in the main text, and Figure 7 shows strength of AMOC in the Atlantic and compares this with the current study.

Reply#36: In the revised manuscript, the "ocean ventilation seesaw" has been replaced by "the mechanism behind such out-of-phase pattern between the ventilation in the subtropical North Pacific and the North Atlantic deep water formation" Lines 647-649 in the revised MS.

On the other hand, we also have added some discussion about the North Atlantic Climate in the 1st paragraph of section 6.3 and modified the text of the following sections to make it more logical.

Lines 619-630

"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, ²³¹Pa/²³⁰Th 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 CO₂ 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."

Lines 631-632

"Our RSEs data in the Northern OT and endobenthic $\delta^{13}C$ in the Bering Sea (Figures 7a-c) both....."

Lines 647-649

"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."

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Captions

Figure A1 Photomicrographs with Modular Stereo Microscope (Zeiss SteREO Discovery V12) show that both detrital and biogenic components of sediment coarse fraction (>63 μm) for 8.23 ka (120-124 cm), 9.26 ka (144-148 cm), 10.98 ka (184-188 cm), 11.66 ka (200-204 cm), 12.92 ka (232-236 cm), 14.05 ka (264-268 cm), and 15.18 ka (296-300 cm) in core CSH1 at 200 X magnification.

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 d are designated by colored crosses.

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.

Figure A1



Figure 3

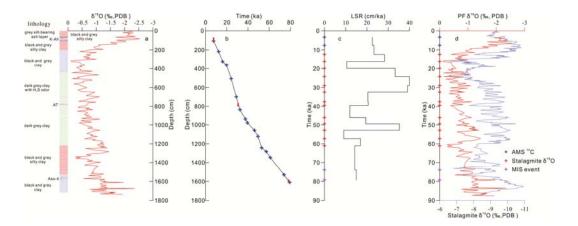
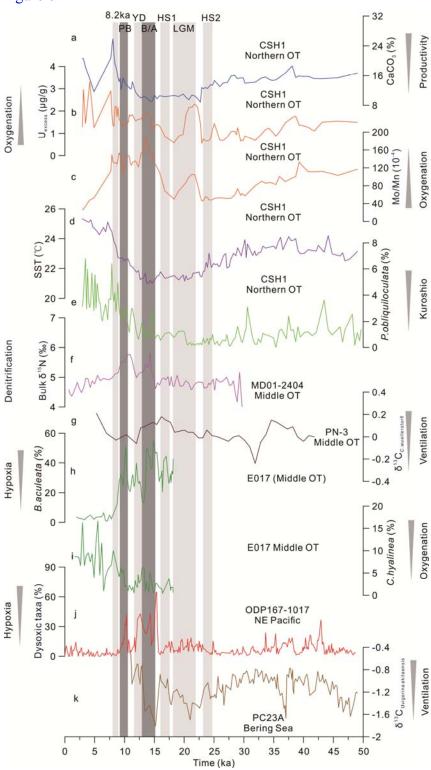


Figure 6



Point-by-point response to referee comments

Response to reviewer

Key:

- Reviewers' comments

- Author's response

- Modified text in the manuscript

Anonymous Referee #2

Received and published: 24 July 2019

Zou et al., present a rather interesting study focusing on reconstructing the

oxygenation history in the Okinawa Trough covering the last 50 kyrs. Specifically, the

authors attempt to disentangle the typically confounding influence of export

production (and by inference the oxygen consumption related to organic matter

degradation) and bottom water ventilation on the sedimentary redox condition. Their

geochemical records largely corroborate previous findings in that oxygenation

patterns at intermediate depths in the North Pacific were primarily controlled by the

production and ventilation of North Pacific Intermediate Water. Specifically,

conditions were generally better oxygenated during stadials, when NPIW was

generally better ventilated and vertically expanded. Furthermore, their data support a

general expansion of oxygen-depleted waters at intermediate depths during the B/A

occupying large swaths of the North Pacific.

The manuscript is well documented and quite detailed in places. The

argumentation could be somewhat streamlined (see comments below) and would

certainly benefit from editorial support. I would also recommend the argumentation to

focus on the aspects outlined in the title and abstract.

Reply#1: We thank the reviewer for the recognition of significance of this study.

Based on the suggestions of the reviewer, we revised our manuscript thoroughly with

a focus on sedimentary oxygenation changes in the subtropical North Pacific and its

linkages to North Atlantic Climate. In the following, we address each specific point

raised by the reviewer.

1. 35 – deep ocean carbon sequestration is certainly one the reasons potentially explaining lower glacial pCO2 concentrations, but certainly not the only one. Please rephrase to avoid unnecessary confusion

Reply#2: Thanks for your suggestion. We rephrased this sentence as the following. Lines 32-34 in the revised MS.

"The deep ocean carbon cycle, especially carbon sequestration and outgassing, is one of the mechanisms to explain variations in atmospheric CO2 concentrations on millennial and orbital timescales."

1. 36 – I would suggest rephrasing as follows – However, the potential role of subtropical North Pacific subsurface waters in modulating.

Reply#3: Agreed and we have done so. Thanks. Lines 34-36. The sentence was amended as follows.

"However, the potential role of subtropical North Pacific subsurface waters in modulating atmospheric CO₂ levels on millennial timescales is poorly constrained."

1. 48 and throughout – why is HS1 so much different from HS2 when considering their respective oxygenation history?

Reply#4: During HS1 and HS2, our RSEs suggest enhanced sedimentary oxygenation. As suggested by the Reviewer, a slight difference in the structure of ventilation mode can be observed (Figure 7). We think this slight discrepancy could be related to different climatic background of HS1 and HS2. Records from paleoclimate archives, such as ice core and Chinese cave stalagmites, show some differences in structure, duration, amplitude between HS1 and HS2. Such differences are thought to be related to the climate background state, such as, CO₂ concentration, ice sheet volume, AMOC intensity, sea ice extent and source of freshwater, etc. (Flückiger et al., 2006; Hemming, 2004; Kaspi et al., 2004; Lynch-Stieglitz et al., 2014). The response of NPIW to HS1 and HS2 events could be different, and thus cause a slight difference in sedimentary oxygenation. On the other hand, the discrepancy in export productivity (CaCO₃) during these two cold intervals in core

CSH1 could also play a role in controlling sedimentary oxygenation.

1. 62 – agreed. But I would add that in addition of the flushing of a poorly ventilated deep water mass upon the resumption of NADW, many export production records show a drastic increase during the B/A (e.g. Kohfeld& Chase, 2011), which could account for enhance oxygen removal associated with organic matter respiration upstream of the core site location.

Reply#5: Agreed with your suggestion and revised. Lines 51-52. The sentence was changed to

".....due to decreased NPIW formation and enhanced export production....."

1. 70 – AT the sediment-water interface

Reply#6: We reworded the sentence. Lines 61-66 in revised Manuscript. It was amended as follows.

"A more sluggish deep ocean ventilation combined with a more efficient biological pump widely thought to facilitate enhanced carbon sequestration in the ocean interior, leading to atmospheric CO₂ drawdown during glacial cold periods (Sigman and Boyle, 2000). These changes are tightly coupled to bottom water oxygenation and sedimentary redox changes on both millennial and orbital timescales (Hoogakker et al., 2015; Jaccard and Galbraith, 2012; Sigman and Boyle, 2000)."

1. 83 . . . in marine sediment cores

Reply#7: Corrected. Line 76. The sentence was amended as follows.

".....in marine sediment cores."

1. 86 . . . in the subarctic North Pacific.

Reply#8: Corrected. Line 78. The sentence was amended as follows.

"..... last glaciation and Holocene in the subarctic North Pacific."

1. 92 – I would suggest to briefly explain what cabbelingmeans

Reply#9: Done. Cabbeling is a mixing process to form a new water mass with increased density than that of parent water masses. We have added in Lines 85-86.

".....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....."

1. 96 and throughout (incl. Fig. 6) – this may well be a semantic issue, but benthic d13C cannot be considered as a ventilation proxy per se, as the isotopic value can be obfuscated by air-sea gas exchange in locations where subsurface water masses form.

Reply#10: We agree with the comment by the Reviewer that benthic δ^{13} C changes are influenced by a variety of factors, such as air-sea equilibration, ocean-circulation changes, and productivity changes, etc. Previous studies also revealed benthic δ^{13} C patterns at basin-wide scales can reflect ocean-circulation changes (Charles and Fairbanks, 1992; Charles et al., 1996; Ninnemann and Charles, 2002). In this study we also noticed similar trends in benthic δ^{13} C during ~22 ka and ~14 ka between cores PN-3 (Okinawa Trough) and PC23A (Bering Sea) (Figure 4), despite their great distance. In the revised manuscript, we add additional information for benthic δ^{13} C. The sentence was amended Lines 89-92.

"Benthic foraminiferal δ^{13} C, a quasi-conservative tracer for water mass, from the North Pacific suggested an enhanced ventilation (higher δ^{13} C) at water depths of < 2000 m during the last glacial period (Keigwin, 1998; Matsumoto et al., 2002)."

1. 151 – processes that are related to the supply of oxygen by ocean circulation and. . .

Reply#11: Revised. Lines 150-153. The sentence was changed to:

"The sedimentary redox conditions are 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."

1. 163 – technically one should specify that under reducing conditions it is the authigenic fraction of Mn (as opposed to its detrital background) that remains low.

Reply#12: Revised. Lines 162-163. The sentence was amended as follows:

"Under reducing conditions, the authigenic fraction of Mn (as opposed to its detrital background)"

1. 168 – please add adequate reference

Reply#13: The reference (Morford and Emerson 1999) has been included in the revised manuscript. Line 170.

1. 195 – volcanism

Reply#14: Revised. Line 198. "volcanic" has been changed to "volcanism".

§3 – I would suggest to substantially shorten this paragraph as the general oceanographic setting is already outlined in the introduction. I would recommend focusing on the aspects directly relevant to the argumentation (nutrient, dissolved O2).

Reply#15: Thanks for your suggestion. In the revised manuscript, we removed the 1st paragraph of section 3 and then we reworded some sentences of previous 3rd paragraph of section 3. Lines 218-227. The paragraph was amended as follows.

"A 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 (Figures 2a and b). An estimated ~80% of the mean annual discharge of the river 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)."

1. 287 – why is the sedimentation rate so high during HS2 when both export production and detrital input (based on Al) are low? I would suggest verifying the age pointers for that interval.

Reply#16: Thanks for your suggestion and we have verified the age control points (Figure 3 and Table 2). In the original manuscript, higher sedimentation rate mainly occurred during HS2 and HS3 (> 40-60 cm/ka) and is mainly caused by uncertainties of age control points at 23.476 ka (DO2) and 29.995 ka (H3). In the revised manuscript, these two age control points have been eliminated. Even with this

more conservative tuning approach, the conclusions remain the same as before and robust.

1.291 - 85 samples covering 50 kyrs cannot provide an average time resolution of 200 yrs.

Reply#17: We thank the reviewer to point out this mistake. Now we have corrected it to ~ 600 years. Lines 286-287 in revised manuscript. The sentence was changed to:

"..... representing a temporal resolution of about 600 years (every 4 cm interval)....."

1. 345-346 – Maybe. But it may also suggest that the sedimentary CaCO3 content could be directly controlled by dilution. I would interpret the export productivity records with caution.

Reply#18: We thank the reviewer for the very helpful comments and suggestions. In the revised manuscript, we have ruled out the effects of some factors, such as dilution and dissolution, on CaCO₃. On the other hand, we have reworded this section to cautiously interpret CaCO₃ as a reliable proxy for export production at core CSH1. We have added the following information to Lines 339-358.

"Several lines of evidence support CaCO₃ as a reliable productivity proxy, particularly during the last deglaciation. The strong negative correlation coefficient (r = -0.85, p<0.01) between Al and CaCO₃ in sediments throughout core CSH1 confirms the biogenic origin of CaCO₃ against terrigenous Al (Figure 4f). Generally, terrigenous dilution decreases the concentrations of CaCO₃. An inconsistent relationship between CaCO₃ contents and sedimentation rates indicates a minor effect of dilution on CaCO₃. Furthermore, the increasing trend in CaCO₃ associated with high sedimentation rate during the last deglacial interval indicates a substantial increase in export productivity (Figures 4a and d). The high coherence between CaCO₃ content and alkenone-derived sea surface water (SST) (Shi et al., 2014) indicates a direct control on CaCO₃ by SST. Moreover, a detailed comparison between CaCO₃ 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 CaCO₃ 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, a similar deglacial trend in CaCO₃ 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 CaCO₃ of core CSH1 as a reliable productivity proxy to a first order approximation."

1.371-372 – interesting idea!

Reply#19: Thanks.

1. 427 (438?) – supply seems more adequate than provision.

Reply#20: Agreed and revised. Line 440. "provision" was replaced by "supply".

1. 487-488 – please also consider citing Galbraith et al., 2007.

Reply#21: The paper has been included in the revised manuscript. Line 496.

1. 489-490 – please keep in mind that O2 can be consumed upstream of the core site location as the removal of O2 in relation to organic matter degradation is integrated over the flow path of a give subsurface water mass.

Reply#22: Thanks for your suggestion. We agree with the comment.

Fig. 6C – shouldn't the grey triangle right of the vertical axis be flipped upside down (i.e. high Mo/Mn coincident with low oxygenation)?

Reply#23: Thanks. High Mo/Mn ratio indicates low oxygen condition. The sign in Fig. 6c has been corrected.

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Millennial-scale variations of sedimentary oxygenation in the western 1 2 subtropical North Pacific and its links to the North Atlantic climate 3 Jianjun Zou^{1,2}, Xuefa Shi^{1,2}, Aimei Zhu¹, Selvaraj Kandasamy³, Xun Gong⁴, Lester 4 Lembke-Jene⁴, Min-Te Chen⁵, Yonghua Wu^{1,2}, Shulan Ge^{1,2}, Yanguang Liu^{1,2}, Xinru 5 Xue¹, Gerrit Lohmann⁴, Ralf Tiedemann⁴ 6 ¹Key Laboratory of Marine Sedimentology and Environmental Geology, First Institute 7 of Oceanography, MNR, Qingdao 266061, China 8 9 ²Laboratory for Marine Geology, Qingdao National Laboratory for Marine Science and Technology, Qingdao, 266061, China 10 11 ³Department of Geological Oceanography and State Key Laboratory of Marine Environmental Science, Xiamen University, Xiamen361102, China 12 ⁴Alfred-Wegener-Institut Helmholtz-Zentrum für Polar- und Meeresforschung, Am 13 14 Handelshafen 12Bussestr. 24, 27570 Bremerhaven, Germany ⁵Institute of Applied Geosciences, National Taiwan Ocean University, Keelung 20224, 15 Taiwan 16 17 Corresponding authors: 18 带格式的:无下划线,字体颜色:自 Jianjun Zou (zoujianjun@fio.org.cn); Xuefa Shi (xfshi@fio.org.cn) 19 带格式的:无下划线,字体颜色:自 20 带格式的:无下划线,字体颜色:自 **Key Points** 21 带格式的: 无下划线, 字体颜色: 自 1. This study reconstructs the History history of sedimentary oxygenation processes at 22 带格式的: 无下划线, mid-depths in the western subtropical North Pacific since the Last last gGlacial period 23 **带格式的**:无下划线,字体颜色:自动设置 is reconstructed using sediment-bound geochemical proxies. 24 带格式的:无下划线,字体颜色:自 2. Sediment-bound rRedox-sensitive proxies reveal millennial-scale variations in 25 **带格式的:** 无下划线, 动设置 字体颜色: 自 sedimentary oxygenation that correlated closely to changes in the North Pacific 26 Intermediate Water. 27 **带格式的**:无下划线,字体颜色:自动设置 3. A millennial-scale out-of-phase relationship between deglacial ventilation in the 28 western subtropical North Pacific and the formation of North Atlantic Deep Water is 29

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suggested.

A. A larger CO₂ storage at mid-depths of the North Pacific corresponds to the

termination of atmospheric CO_2 rise during the Bölling-Alleröd interval.

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Abstract

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The deep ocean carbon cycle, especially carbon sequestration and outgassing, is one of the mechanisms to explain variations in Lower glacial atmospheric CO₂ concentrations on millennial and orbital timescales have been attributed to carbon sequestration in deep oceans. However, the potential roles of voluminous subtropical North Pacific subsurface waters in modulating atmospheric CO₂ levels on millennial timescales are is poorly constrained. Further, aAn increase in respired CO₂ concentration in the glacial deep ocean due to biological pump generally is coeval withcorresponds to less deoxygenation in the subsurface layer. This link thus offers a chance to visit study oceanic ventilation and the coeval export productivity based on redox-controlled, sedimentary geochemical parameters. Here, we investigate a suite of sediment geochemical proxies in a sediment core from the Okinawa Trough to understand the sedimentary oxygenation variations in the subtropical North Pacific (core CSH1) over the last 50,000, thousand years (50 ka). Our results Our results / suggest that enhanced mid-depth western subtropical North Pacific (WSTNP) sedimentary oxygenation suggest that enhanced occurred occurred at mid depths of the subtropical Northwest Pacific duringduring cold the past glacial periodintervals and after 8.5 ka, while oxygenation decreased during the Bölling-Alleröd (B/A) and Preboreal. The especially pronounced for the North Atlantic millennial scale abrupt cold events of the Younger Dryas, Heinrich Stadial (HS) 1 and 2. On the other hand, oxygen depleted seawater is found during the Bölling Alleröd (B/A) and Preboreal. Our findings of enhanced sedimentary oxygenation in the subtropical North Pacific WSTNP, is aligned with intensified formation of North Pacific Intermediate Water (NPIW) during cold spells, while the amelioratedbetter, sedimentary oxygenation since 8.5 kg seems to be linked with to the an intensified Kuroshio Current after 8.5 kg. The enhanced formation of NPIW during Heinrich Stadial 1 (HS1) was likely driven by the perturbation of sea ice formation and sea surface salinity oscillations in the high-latitude North Pacific, The In our results, ddiminished sedimentary oxygenation during the B/A due to decreased NPIW formationdecreased NPIW formation and enhanced export production, indicates an expansion of oxygen

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minimum zone in the North Pacific and enhanced CO₂ sequestration at mid-depth waters, along with slight increase termination of in atmospheric CO₂ concentration increase. Mechanistically, we We speculate that attribute -these millennial-scale changes were linked to the intensified NPIW and enhanced abyss flushing also a drastic increase in export productivity) during deglacial cold and warm intervals, respectively, closely related to variations in strength of North Atlantic Deep Water formation, leading to intensification of NPIW formation and enhanced abyss flushing during deglacial cold and warm intervals, respectively. Enhanced formation of NPIW seem to be driven by the perturbation of sea ice formation and sea surface salinity oscillation in high latitude North Pacific through atmospheric and oceanic teleconnection. During the B/A, decreased sedimentary oxygenation likely resulted from an upward penetration of aged deep water into the intermediate-depth in the corresponding to a resumption of Atlantic Meridional Overturning Circulation.

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

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1. Introduction

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The A more sluggish deep ocean ventilation combined with a more efficient biological pump widely thought to depletes dissolved oxygen in the sediment water interface and facilitates enhanced the carbon storage sequestration of respired in the ocean interior-carbon, leading to atmospheric CO₂ drawdown during glacial cold periods (Sigman and Boyle, 2000). These changes are tightly coupled towhich in turn playss a role in regulating bottom water oxygenation and sedimentary oxygenredox changes, potentially linking to atmospheric CO2 changes on both millennial and orbital and millennial timescales (Hoogakker et al., 2015; Jaccard and Galbraith, 2012; Sigman and Boyle, 2000). The 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 both high-latitude North Pacific marginss as well as open subarctic Pacific have identified drastic variations in export productivity and marine ocean oxygen levels at millennial and orbital millennialglacial-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 (δ^{15} N) of organic matter (Addison et al., 2012; Chang et al., 2014; Galbraith et al., 2004; Riethdorf et al., 2016) in marine sediment cored sediments. These studies further suggested that both North Pacific Intermediate Water (NPIW) and export of organic carbon-matter regulate the sedimentary oxygenation variation during the last glaciation and Holocene in the northeast subarctic North Pacific. By contrast, little information exists on millennial-scale oxygenation changes to date in the western subtropical North Pacific (WSTNP).

The modern NPIW precursor waters are mainly sourced from the NW Pacific of marginal seas (Shcherbina et al., 2003; Talley, 1993; You et al., 2000), and then it spreadings into the 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.

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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 m³/s) is sufficient for subtropical ventilation (You et al., 2003). Benthic foraminiferal $\delta^{13}C$ -data, a quasi-conservative tracer for water mass, from the North Pacific suggested indicates an enhanced ventilation (enriched higher, δ^{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 insightsuggested into the formation of deep water in the North Pacific during the early deglaciation in Heinrich Stadial 1 (HS1). Enhanced NPIW penetration is was further explored using numerical model simulations (Chikamoto et al., 2012; Gong et al., 2019; Okazaki et al., 2010). The downstream effects of intensified GNPIW can be seen in the record of δ^{13} C of *Cibicides wuellerstorfi* in core PN-3 from the middle Okinawa Trough (OT), whereas lower deglacial 8¹³C values were attributed to enhanced OC accumulation rates due to higher surface productivity by Wahyudi and Minagawa (1997), In contrast, substantial effects of intensified NPIW formation during Marine Isotope Stage (MIS) 2 and 6 on the ventilation and nutrient characteristics of lower latitude mid-depth Eastern Equatorial Pacific have been suggested by recent studies (Max et al., 2017; Rippert et al., 2017). Rippert et al. (2017) The downstream effects of intensified NPIW are also reflected in the record of δ^{13} C of Cibicides wuellerstorfi in core PN-3 from the middle Okinawa Trough (OT), where lower deglacial δ^{13} C values were attributed to enhanced OC accumulation rates due to higher surface productivity by (Wahyudi and Minagawa, 1997).

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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 WSTNP (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 oceanographic characteristics of the OT over

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glacial-interglacial cycles are largely influenced by the Kuroshio and East China SeaECS 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-mixing of NPIW and South China Sea Intermediate Water (Nakamura et al., 2013). These waters intrude into the OT through two ways (Nakamura et al., 2013); (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 horizontal advection 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.

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Both surface characterisitics hydrography and deep ventilation in the OT varied greatly significantly 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), further argued that the hydrological conditions of the 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 East China Sea (ECS), which is related to the summer EAM and the sea level oscillations coupled with topography are also have also been regulating the surface hydrography in the Okinawa TroughOT (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 带格式的:无下划线,字体颜色:自

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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) hypothesized 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 Thus, the patterns and that caused sedimentary oxygenation in the OT reasons unclear controversial.

2. Paleo-redox proxies

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The sSedimentary redox conditionss is are the balance between 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 elosely related to the supply of oxygen by ocean circulationadvection of submarine ocean circulation and organic matter respiration, respectively. Contrasting geochemical behaviors of redox-sensitive trace metals (Mn, Mo, U, etc.) have been extensively 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 conditions prevails into greater sediment depths as a result of low organic matter degradation rates and well-ventilated bottom water (Burdige, 1993). In—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 (O₂ 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

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(Nameroff et al., 2002)

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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 MoS₂, 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_uM hydrogen sulfide (H₂S) for Fe-S-Mo co-precipitation and 100 μM H₂S 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 (H₂S occurrence) or well oxygenated conditions (if Mo_{excess} is in association with Mn-oxides).

In general, U is enriched in anoxic sediments (>1 μM H₂S), but not in oxic sediments (>10 μM O₂) (Nameroff et al., 2002). Accumulation of U depends on the content of reactive organic matter (Sundby et al., 2004) and U precipitates as uraninite (UO₂) 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.

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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 WSTNP 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 time-scales since the last glacial.

3. Oceanographic setting

The OT resulted from the collision of the Philippine and Eurasian plates and initially opened at the middle Miocene (Sibuet et al., 1987). Since that time, the OT has been a depositional center in the ECS and receives large sediment supplies from nearby rivers (Chang et al., 2009). At present, water depth in the axial part of the OT deepens from 500 m in the north to ~2700 m in the south.

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 (1 Sv = 10⁶ m³/s) in summer and a minimum of 20 Sv in autumn across the east of Taiwan (Qu and Lukas, 2003).

Figures 2a and b show the A 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 (Figures

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2a and b). An estimated ~80% of The the mean annual discharge of the river Changiang is 0.028 Sy and ~80% of its total discharge is supplied to the ECS 域代码已更改 (Ichikawa and Beardsley, 2002). In and in situ observational data of surface hydrography along the ship track from Taiwan Strait to Korea Strait and around the entrance of the Tsushima Strait in the northern part of the ECS show a lower SSS in summer and a pronounced negative correlation between the Changjiang discharge and 域代码已更改 SSS in July (Delcroix and Murtugudde, 2002). Lower SSS in summer than that in winter suggests stronger effects of summer EAM on surface hydrography over the 域代码已更改 Kuroshio Current (Sun et al., 2005). Consistently, previous studies from the Okinawa 域代码已更改 TroughOT reported such close relationship between summer EAM and SSS back to 域代码已更改 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- Δ^{14} C) collected during the World Ocean Circulation Experiment (WOCE) Expeditions in the Pacific (transects P24 and P03) favors the presence of low salinesalinity, nutrient-enriched intermediate 域代码已更改 and deep waters (Talley, 2007). Dissolved oxygen content is <100 μM-mol/kg at water 域代码已更改 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

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4. Materials and methods

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4.1. Chronostratigraphy of core CSH1

 $\times 10^{-6}$ m s⁻¹) (Nakamura et al., 2013; Nishina et al., 2016).

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 <u>Tsushima Warm Current (TWC)</u> (Figure 1b) and within the depth of NPIW (Figure 1c) using a piston corer during *Xiangyanghong*09 Cruise in 1998, <u>carried out by the First Institute of Oceanography, Ministry of Natural Resources of China</u>. This location is <u>thus</u> enabling us to reconstruct millennial-<u>scale</u> changes in the properties of TWC and NPIW. <u>The</u>

in the OT is associated with the inflow of NPIW and studies using a box model

predicted that overflow through the Kerama Gap is responsible for upwelling (3.8–7.6

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 t. 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 the China Ocean Sample Repository at 4 °C until analysis.

Previously, some paleoceanographic studies have been conducted and a set of data have has been investigated for core CSH1, including the contents of planktic foraminifers as well as their carbon (δ^{13} C) and oxygen isotope (δ^{18} O) compositions (Shi et al., 2014), pollen (Chen et al., 2006), paleomagnetism (Ge et al., 2007) and CaCO₃ (Wu et al., 2004). An age model for this core has been constructed by using ten Accelerator Mass Spectrometry (AMS) ¹⁴C dates and six oxygen isotope (δ^{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 Notably, previous the original age control points model, which used with constant radiocarbon reservoir ages throughout core CSH1 are used suitable 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, Δa higher abundance of *Neogloboquadrina pachyderma* (*dextral*), e. g. that occurred during warmer intervals, such as the Bölling-Alleröd (B/A), has been challenging to explain-reasonably. On the other hand, paired measurements of ¹⁴C/¹²C and ²³⁰Th ages from Hulu Cave stalagmites suggest magnetic field changes has have greatly contributed to high atmospheric ¹⁴C/¹²C values at HS4 and the YD (Cheng et al., 2018). Thus a constant reservoir age (ΔR=0) assumed when calibrating foraminiferal radiocarbon dates using CALIB 6 software and the Marine_13 calibration dataset (Reimer et al., 2013) for Core—core CSH1 may cause large chronological uncertainties.

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Here, we therefore recalibrated the radiocarbon dates using updated CALIB 7.04 software with Marine 13 calibration dataset (Reimer et al., 2013). Moreover, on the basis of significant correlation between planktic foraminiferal species *Globigerinoides* ruber δ¹⁸O and Chinese stalagmite δ¹⁸O (Cheng et al., 2016), a proxy of summer EAM related to SSS of the ECS, we re established improve 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 60 40 cm/ka with higher sedimentation rates (around 30-40 cm/ka) between ~24 ka and 32.5 ka. The new age control points were are 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, representing a with a temporal resolution of about 600 years (every 4 cm sampling interval) were selected for detailed geochemical analyses of major and minor elements, and total contents of carbon (TC), organic carbon (TOC) and nitrogen (TN) contents. 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 (CaCO₃) was calculated using the equation:

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

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

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samples were used to control the analytical process. The relative standard deviation of the GSD-9 for TC, TN and TOC is \leq 3.4%.

About 0.5 g of sediment powder was digested in double distilled HF:HNO₃ (3:1), followed by concentrated HClO₄, and then re-dissolved in 5% HNO₃. 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 \leq 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} × Al), with U/Al_{average shale} = 0.307×10^{-6} and Mo/Al_{average shale} = 0.295×10^{-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, given-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 CaCO₃

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),

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corresponding to higher linear sedimentation rate (Figure 4a4e). The content of CaCO₃ varies from 8.8 to 35% (Figure 4e) and it mostly shows higher values with increasing trends during the last deglaciation. Conversely, the content of CaCO₃ is low and exhibits decreasing trends during late MIS 3 and the LGM (Figure 4e).

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Both TOC and CaCO₃ have been are widely—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 CaCO₃ and TOC concentrations.

Several lines of evidence support CaCO₃ as a reliable productivity proxy, particularly during the last deglaciation. (Shi et al., 2014)In addition, tThe strong negative correlation coefficient (r = -0.85, p<0.01) between Al and CaCO₃ in sediments throughout core CSH1 confirms the biogenic origin of CaCO₃ against terrigenous Al (Figure 4f). Generally, terrigenous dilution decreases the concentrations of CaCO₃. An inconsistent relationship between CaCO₃ contents and sedimentation rates indicates a minor effect of dilution on CaCO₃. Furthermore, the increasing trend in CaCO₃ associated with high sedimentation rate during the last deglacial interval indicates a substantial increase in export productivity (Figures 4a and d). The high coherence between CaCO₃ content and alkenone-derived sea surface water (SST) (Shi et al., 2014) indicates a direct control on CaCO₃ by SST. Moreover, a detailed comparison between CaCO₃ concentrations and the previously published foraminiferal fragmentation ratio (Wu et al., 2004) elearly shows, apart from a small portion within the LGM, no clear co-variation between them. This These evidence suggests that CaCO₃ changes are primarily 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

materialsthrough changes in the lysocline depth. Likewise, a similar deglacial trend in

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CaCO₃ is also observed in core MD01-2404 (Chang et al., 2009), indicating a ubiquitous, not local picture in the OT. On the other hand, terrigenous dilution generally decreases the content of CaCO₃. All these lines of evidence thus support CaCO₃ of core CSH1 as a reliable productivity proxy to a first order approximation. The increasing trend of CaCO₃ associated with high sedimentation rate (Figures 4a, e) indicates a substantial increase in export productivity during the last deglacial interval. Thus, we can confidently use CaCO₃ content as productivity proxy to a first order approximation.

5.2. Redox-sensitive Elements

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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-4, Figure 4g). Generally, concentrations of excess Mo and excess U (Figures 4j and -1) 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). It should be noted that pPronounced variations in U concentration since after 8.5 ka isare related to the occurrence of discrete volcanic materials. -A significant positive Eu anomaly (Zhu et al., 2015) (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 Moexcess 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 complex geochemical behaviors of Mn and Mo. A strong positive correlation between Mo_{excess} and Mn (Figure 5b) seems tomay 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 stateconditions in our study area. Overall, the Mo/Mn ratio shows similar downcore pattern to that of

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Mo_{excess} with higher values ratios during the last deglaciation, but lower values 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 indicates an well oxygenated sedimentary environment since after ~8 ka (Figure 4h). Both higher Mo/Mn ratios and excess U concentrations, together with lower Mn concentrations suggest an oxygen-deficient suboxic sedimentary depositional conditions environment during the late deglacial period (15.8 ka-9.5 ka), whereas lower values ratios during the LGM, HS1 and HS2 indicate relatively better oxygenated sedimentary conditions. 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

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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 O₂), hypoxic (<60–120 μmol/kg O₂) and suboxic (<2–10 μmol/kg O₂) conditions, whereas anoxia is the absence of measurable oxygenGenerally, redox states in waters can be classified as oxic (>89 μmol/L O₂), suboxic (-8.9 89 μmol/LO₂), anoxic nonsulfidic (<89 μmol/L O₂, 0 μmol/L H₂S), and anoxic sulfidic or euxinic (0 ml O₂/L, >0 μmol/L H₂S) (Savrda and Bottjer, 1991).

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} a, nd to resultresulting 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

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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 ppmµg/g, the crustal average to < 25 ppmµg/g, a threshold concentration for euxinic condition. Given that the northernOkinawa Trough OT is located in an open oceanic settingweakly restricted basin settings, we use these two above mentioned proxies to evaluate the degree of oxygenation in sediments.

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Both bulk Mo concentration (1.2-9.5 ppmµg/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 concentrations with enhanced-higher Mo/U ratios during the last termination (18 ka-9 ka) indicate a strongermore reducing conditions compared to the Holocene and the last glacial period, though Mo concentrations is were less than 25 ppmµg/g, a threshold for euxinic deposition proposed by Scott and Lyons (2012).

The relative abundance of benthic foraminiferal species that thrive in different oxygen concentrations also haves also been widely used to reconstruct the variations in bottom water ventilation, such as the enhanced abundance of Bulimina aculeata, Uvigerina peregrina and Chilostomella oolina found under oxygen-depleted conditions in the central and southern OT during from the last deglaciation 18 ka to 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) and 255 (1575 m water depth) and high benthic δ^{13} C values (Wahyudi and Minagawa, 1997) in cores E017 (water depth 1826 m), 225 (water depth 1575 m) in core and PN-3 (water depth 1058 m water depth) from the middle and southern OT (Figures 1 and 3) during the postglacial period. The inferred ventilation pattern poorly-ventilated deep water in the middle and southern OT inferred from by benthic foraminiferal assemblages during the last deglaciation is correlateseonsistent with the one inferred from RSEs in theis northern OTstudy referring to our RSEs (Figure 4). A link thus can be hypothesized between deep-water 域代码已更改

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ventilation and sedimentary oxygenation in the OT. Although we did not carry out benthic foraminiferal species analyses for our core CSH1, it is reasonable to infer based on RSEs that the deepwater in the northern OT was also in a prominent oxygen poor condition during the late deglacial interval. A clear link thus can be built between the ventilation of deep water and the sedimentary oxygenation in the OT. In briefOverall, 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 late-last deglaciation deglacial period.

6.2. Causes for sedimentary oxygenation variations

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As discussed above, the Our observed 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 Okinawa TroughOT. In general, four factors can regulate the redox condition in the deep water column: (i) O₂ solubility, (ii) export productivity and subsequent degradation of organic matter, (iii) vertical mixing, and (iv) lateral provision 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). In the OT, the oxygen deficiency during the late deglacial period can be caused either by one and/or a combination of more than one of these factors. Our data also suggest drastic variations in sedimentary oxygenation over the last 50 ka. However, In order to uncover the mechanisms responsible for sedimentary oxygenation variations in the basin wide OTin 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, we compare our proxy records of sedimentary oxygenation (Uexcess concentration and Mo/Mn ratio) and export productivity (CaCO₃) (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

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al., 2008), benthic foraminifera δ¹³C (a proxy for ventilation) in cores PN-3 and PC23A (Rella et al., 2012; Wahyudi and Minagawa, 1997), a proxy for water mass, in core PC23A (Rella et al., 2012), abundance of benthic foraminifera (an indicator of hypoxia) in core E017 (Li et al., 2005) and ODP Site 1017 (Cannariato and Kennett, 1999) the NE Pacific, an indicator of anoxic condition (Cannariato and Kennett, 1999), abundance of *Pulleniatina obliquiloculata*, an indicator of the Kuroshio strength (Shi et al., 2014) (Figures 6d-k).

6.2.1. Effects of regional ocean temperature on deglacial deoxygenation

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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). For instance, Shi et al. (2014) reported an increase in SST of around 4°C (from ~21°C to ~24.6°C) during the last deglaciation in core CSH1 (Figure 6d). Based on thermal solubility effects, a hypothetical warming of $\pm 1^{\circ}$ C at our site—would reduce oxygen concentrations by about 8-3.5 μMmol/kg at water temperatures around 22°C (Brewer and Peltzer, 2016) (Shi et al., 2014) could drive a conservative estimate of a drop of <15 μmol/kg drastic in drop of oxygen concentration-by, assuming no large salinity changes <30 μM in subsurface water of the OT. Therefore, we assume that the late deglacial hypoxia in the OT underwent a similar increase in ocean temperatures. However, given the semi-quantitative nature of our data about oxygenation changes, which seemingly exceed an amplitude of $> \frac{30-15}{\mu mol/kgM}$, we suggest that other factors, in particular processes likec.g. local changes in export productivity, regional influences such as vertical mixing due to changes of the Kuroshio Current, as well as and far-field effects all may have played some 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).

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Such an increase in export production was due to favorable conditions for phytoplankton blooms—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 CaCO₃ 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 reported evidenced by concentrations of reactive phosphorus (Li et al., 2017) and CaCO₃ (Chang et al., 2009) from the middle and southern OT during the last deglaciation, and thus was a pervasive, synchronous phenomenon of in the entire study region at the outermost extension of the ECS.

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As a consequence, high export productivity lowers oxygen concentrations in deeper waters, due to subsurface consumption of oxygen by remineralization of organic matter. Similar events of high export productivity have been extensively 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, the increaseds in productivity were thought to be likely also responsible for oxygen depletion in mid-depth waters, due to exceptionally high oxygen consumption. However, the productivity changes during the deglacial interval, very specifically CaCO₃, are not fully consistent with the trends of excess U and Mo/Mn ratio (Figures 46b and c). The sedimentary oxygenation thus cannot be determined by export productivity alone.

6.2.3 Effects of the Kuroshio dynamics on sedimentary oxygenation

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593 The Kuroshio Current, one of the main drivers of vertical mixing, has been 594 identified as the key factor in controlling modern deep ventilation in the Okinawa 域代码已更改 TroughOT (Kao et al., 2006). However, the flow path of the Kuroshio in the Okinawa 595 TroughOT during the glacial interval remains a matter of debate. Planktic 596 597 foraminiferal assemblages in sediment cores from inside and outside the Okinawa 598 TroughOT indicated that the Kuroshio have migrated to the east of the Ryukyu 域代码已更改 Islands during the LGM (Ujiié and Ujiié, 1999). Subsequently, Kao et al. (2006) 599 域代码已更改 based on modeling results suggested that the Kuroshio still enters-into the Okinawa 600 TroughOT, 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 602 603 Kerama Gap at -80 and -135m lowered sea level. Combined with sea surface 域代码已更改 604 temperature (SST) records and ocean model results, Lee et al. (2013) argued that there 605 was little effect of deglacial sea-level change on the path of the Kuroshio, which still 606 exited the Okinawa TroughOT 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 607 域代码已更改 SST records are insufficient to decipher past changes of the Kuroshio (Ujiié et al., 608 609 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 610 域代码已更改 611 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 612 域代码已更改 sea level (Ujiié and Ujiié, 1999) and an overall reduced Kuroshio intensity (Kao et al., 613 域代码已更改 2006), effectively suppressing the effect of the Kuroshio on deep ventilation in the OT. 614 Our RSEs data show that oxygenated sedimentary conditions were dominant in the 615 616 northern OT throughout the last glacial period (Figures 6a, b, c, d). The Kuroshio thus 617 likely had a weak or even no effect on the renewal of oxygen to the sedimentary 618 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 619 variation in flow path of KC, relative to that of Holocene recorded in core KX12 - 3 620 域代码已更改 (1423 water depth) (Lim et al., 2017), further validates our inference(Lim et 621 域代码已更改 域代码已更改 622 2017).

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On the other hand, the gradually increasing increased alkenone-derived SST and abundance of *P.obliquiloculata* (Figures 6d and e) 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). Matsumoto et al. (2002) suggested that the influence of the present Kuroshio can reach to the bottom depth of the permanent thermocline, which is approximately at 1000 m water depth. 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; including indications for millennial-scale variations (Figure 6).

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 e) and *C.hyalinea* abundance in core E017 (Figure 6i). The rRe-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 δ^{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, during the Holocene our observed 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

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域代码已更改 653 Bering Sea (Itaki et al., 2012; Kim et al., 2011; Rella et al., 2012), the Okhotsk Sea 域代码已更改 (Itaki et al., 2008; Okazaki et al., 2014; Okazaki et al., 2006; Wu et al., 2014), off east 654 域代码已更改 Japan (Shibahara et al., 2007), the eastern North Pacific (Cartapanis et al., 2011; 655 域代码已更改 域代码已更改 Ohkushi et al., 2013) and western subarctic Pacific (Keigwin, 1998; Matsumoto et al., 656 带格式的:字体颜色:自动设置 657 2002). The intensified ventilation formation of GNPIW due to additional source region in the Bering Sea is firstly attributable to was proposed by the displacement of 658 域代码已更改 659 formation source region to the Bering Sea Ohkushi et al. (2003) and and then is 域代码已更改 further confirmed by Horikawa et al. (2010). Under such conditions, the invasion of 660 well-ventilated GNPIW into the OT through the Kerama Gap would have replenished 661 the water column oxygen in the OT, although the penetration depth of GNPIW 662 域代码已更改 663 remains under debate (Jaccard and Galbraith, 2013; Max et al., 2014; Okazaki et al., 664 2010; Rae et al., 2014). Both a gradual decrease in excess U concentration and an 665 increase in Mo/Mn ratio during the last glacial period (25 ka-50 ka) validate such 666 inference, suggesting pronounced effects of intensified GNPIW formation in the OT. During HS1, a stronger formation of GNPIW was supported by proxy studies 667 域代码已更改 and numerical simulations (Chikamoto et al., 2012; Gong et al., 2019; Jaccard and 668 域代码已更改 域代码已更改 Galbraith, 2013; Max et al., 2014; Okazaki et al., 2010), recorded in the North Pacific 669 域代码已更改 670 by a variety of studies. For example, -oOn the basis of paired benthic-planktic (B-P) 域代码已更改 域代码已更改 ¹⁴C data, eand model simulations, nhanced penetration of Okazaki et al. (2010) 671 域代码已更改 suggested that NPIW penetrated into a much deeper water depth of ~2500 to 3000 m 672 域代码已更改 during HS1 relative to the Holocene has been revealed in several studies (Max et al., 673 2014; Okazaki et al., 2010; Sagawa and Ikehara, 2008), which was also simulated by 674 域代码已更改 several models (Chikamoto et al., 2012; Gong et al., 2019; Okazaki et al., 2010). On 675 676 the other hand, increased intermediate water temperature in the subtropical Pacific 域代码已更改 677 recorded in core GH08-2004 (1166 m water depth) (Kubota et al., 2015) and young 域代码已更改 deep water observed in the northern South China Sea during HS1 (Wan and Jian, 678 679 2014) along downstream region of NPIW are also related to intensified NPIW 域代码已更改 formation. Furthermore, In contrast, Max et al. (2014) argued against deep water 680 formation in the North Pacific and showed that GNPIW was well-ventilated only to 681

intermediate water depths (< 1400 m). Various mid- and high-latitude North Pacific

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records of B P ¹⁴C age offsets at the intermediate water depth (<600 2000 m) showed an active production of GNPIW during HS1 (Max et al., 2014; Sagawa and Ikehara, 2008). (Kubota et al., 2015)Moreover, Kubota et al. (2010) reported increased subsurface water temperatures related to enhanced GNPIW contributions during HS1 at a water depth of 1166m (GH08, and young deep water was observed in the northern South China Sea during HS1 (Wan and Jian, 2014).

All these multiple lines of evidence imply the presence of well ventilated intermediate water in the upper 2000 m of the North Pacific during HS1. At this point, the effect of a strong GNPIW likely reached the South China Sea (Wan and Jian, 2014; Zheng et al., 2016), further to the south the Okinawa Trough. 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 has been established between GNPIW ventilation, the intermediate and deep water oxygen concentration in theof OT deepwater and sediment redox state during HS1. In addition, our RSEs data—also suggested a similarly enhanced ventilation in HS2 (Figures 6b and c) (Figure 6) that must is also also be attributed to intensified GNPIW formation.

Hypoxic conditions during the Bölling Alleröd (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, both of excess U concentrations and Mo/Mn ratio recorded in core CSH1 (Figures 6b- and cd), together with enhanced denitrification and B.aculeata abundance (Figures 6f and h), 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 ¹⁴C ages, together with increased temperature and salinity at intermediate waters temperature and salinity recorded in core GH02-1030 (off East Japan) supported a weakened formation of NPIW during

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the B/A (Sagawa and Ikehara, 2008). These lines of evidence indicate that the 713 boundary between GNPIW and North Pacific Deep Water shoaled during the B/A, in 714 comparison to HS1. Based on a comparison of two benthic foraminiferal oxygen and 715 carbon isotope records from off northern Japan and the southern Ryukyu Island, 716 717 Kubota et al. (2015), found a stronger influence of Pacific Deep Water on 718 intermediate-water temperature and ventilation at their southern than the northern 719 locations, although both sites are located at similar water depths (1166 m and -1212) m for cores GH08-2004 and GH02-1030, 1212 m, respectively). Higher excess U 720 721 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 722

GNPIW, contributing to the subsurface water suboxia deoxygenation in the OT.

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During the YD, <u>both</u> Mo/Mn ratio and excess U show a slightly decreased oxygen condition in the northern OT. <u>In-By</u> contrast, benthic foraminiferal δ^{18} O and δ^{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 CaCO₃ 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, ²³¹Pa/²³⁰Th 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 CO₂ concentration (Zhang et al., 2017), enhanced advection of salt (Barker et al., 2010) (Liu et al., 2009) and changes

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in background climate (Knorr and Lohmann, 2007) were proposed to explain the reinvigoration of AMOC during the B/A.

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Our RSEs data in the Northern OT and endobenthic δ ¹³C in the Bering Sea (Figures 7a-c) both show a substantial millennial variability in intermediate water ventilation in the subtropical North Pacific. Notably, both enhanced ventilation during HS1 and HS2 and oxygen-poor condition during the B/A respectively correspond to the collapse and resumption of Atlantic meridional overturning circulation (AMOC) (Bohm et al., 2015; McManus et al., 2004) (Figure 7-d). Such out-of-phase millennial-scale pattern This 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 seenarios boundary conditions and causes for the observed effects in GNPIW formation, and ventilation ages derived from B-P₁¹⁴C (Freeman et al., 2015; Max et al., 2014; Okazaki et al., 2012). These lines of evidence reveal 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 using 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 oceanic ventilation seesaw out-of-phase pattern between the ventilation in the subtropical North Pacific and the North Atlantic deep water formation and North Pacific is still remains unclear.

Increased NPIW formation of 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

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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 TCZ 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 observation (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-intense brine rejection due togaccompanied by expanded sea ice expansion formation, would have enhanced the NPIW formation. Recently our modeling-derived evidence suggests confirmed that enhanced sea ice coverage occurred in the southern Okhotsk Sea and off East Kamchatka Peninsula during HS1 (Gong et al., 2019). In addition, higher 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 was, as well as reduced advection of Alaska

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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.

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

One of the striking features of RSEs data is higher Mo/Mn ratios and excess U concentrations at across the B/A, indicating supporting an expansion of Oxygen Minimum Zonea substantial oxygen-poor condition in the subtropical North Pacific (Galbraith and Jaccard, 2015; Jaccard and Galbraith, 2012; Moffitt et al., 2015) and coinciding with the- termination of atmospheric CO₂ concentration rise (Marcott et al., 2014) (Figure 747e). As described above, it can be related to the upwelling of nutrient- and CO₂-rich Pacific Deep Water due to resumption of AMOC and enhanced export production. Notably, bAlthough 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 pCO2 at the onset of B/A at this time (Gray et al., 2018). That is to say, indicating that the subarctic North Pacific is a source of relatively high atmospheric CO₂ concentration at that time 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. In combination with published records from the North Pacific (Addison et al., 2012; Cartapanis et al., 2011; Crusius et al., 2004; Galbraith et al., 2007; Lembke-Jene et al., 2017; Shibahara et al., 2007), However, and expansion of oxygen-depletion zone during the BA in the entire North

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Pacific suggest an increase in respired carbon storage at intermediatemid-depth waters of in-the-subtropical North Pacific, which likely stalls the rise of atmospheric CO₂. 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 bring the planet out of the last ice age (Jaccard and Galbraith, 2018).

7. Conclusions

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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 in the past. 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 highlights shows out-of-phase pattern with North Atlantic Climate during the last deglaciationthe major role of Atlantic Meridional Overturning Circulation in regulating the variations in sedimentary oxygenation in the Okinawa Trough through ventilation of NPIW. 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. Combined with other published records. Wwe also suggest an expansion of oxygen-depleted zone and accumulation of respired carbon at the mid-depth waters from previously reported subarctic locations into the western subtropical of the North Pacific at during the B/A, coinciding with the termination of atmospheric CO₂ rise. A step-wise injection of such respired carbon into the atmosphere would be helpful to maintain high atmospheric CO₂ levels during the deglaciation and bring the planet out of the last ice age.

Once the release of the sequestered carbon into the atmosphere in stages, it would

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be helpful to maintain high atmospheric CO2 levels during the deglaciation and to 863 propel the earth out of the glacial climate. 864 865 866 Data availability. All raw data are available to all interested researchers upon request. 867 Author Contributions. J.J.Z. and X.F.S. conceived the study. A.M.Z. performed 868 geochemical analyses of bulk sediments. J.J.Z., X.F.S. K.S. and X.G. led the write up 869 of the manuscript. All other authors provided comments on the manuscript and 870 871 contributed to the final version of the manuscript. 872 873 **Competing interests:** The authors declare no competing interests. 874 875 Acknowledgements 876 Financial support was provided by the National Program on Global Change and Air-Sea Interaction (GASI-GEOGE-04), by the National Natural Science Foundation 877 of China (Grant Nos.: 41476056, 41876065, 41420104005, 41206059, and U1606401) 878 879 and by the Basic Scientific Fund for National Public Research Institutes of China 880 (No.2016Q09) and International Cooperative Projects in Polar Study (201613) and Taishan Scholars Program of Shandong. This study is a contribution to the bilateral 881 882 Sino-German collaboration project (funding through BMBF grant 03F0704A -883 SIGEPAX). XG, LLJ, GL, RT thank the bilateral Sino-German collaboration 884 NOPAWAC project (BMBF grant No. 03F0785A).LLJ and RT acknowledge financial support through the national Helmholtz REKLIM Initiative. We would like to thank 885 886 the anonymous reviewers, who helped to improve the quality of this manuscript. The 887 data used in this study are available from the authors upon request

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1284 **Captions** 带格式的: 两端对齐 Table 1. Locations of different sediment core records and their source references 1285 discussed in the text. 1286 1287 1288 Table 2. Age control points adopted between planktic foraminifera species 带格式的:字体:倾斜 Globigerinoides ruber δ^{18} O of Core CSH1 and Chinese stalagmite δ^{18} O (Cheng et al., 1289 2016) for tuning the age model between 10 ka and 60 ka in this study. A linear 1290 1291 interpolation was assumed between age control points. 1292 Figure 1. (a) Spatial distribution of dissolved oxygen content at 700 m water depth in 1293 1294 the North Pacific. Black arrows denote simplified Kuroshio and Oyashio circulations 1295 and North Pacific Intermediate Water (NPIW) in the North Pacific. The red thick 1296 dashed line indicates transformation of Okhotsk Sea Intermediate Water (OSIW) by 域代码已更改 1297 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 1298 NPIW from northeast North Pacific southward toward the low-latitude northwest 1299 域代码已更改 1300 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 1301 1302 Ocean Data View (odv.awi.de). (b) Location of sediment core CSH1 investigated in 1303 this study (red diamond). Also shown are locations of sediment cores PN-3, E017, 255 1304 and MD012404 investigated previously from the Okinawa Trough, GH08-2004 from 1305 the East of Ryukyu Island, GH02-1030 off (PN-3, E017, 255 and MD012404; white cross line), the east of Japannorthern and southern Japan (GH08-2004 and 1306 1307 GH02-1030), PC-23A from the Bering Sea (PC-23A) and ODP Site 1017 from the northeastern Pacific (ODP167-1017). Letters A to E represent the sediment cores from 1308 and near the OT. The detailed information for these cores can be is shown seen in 1309 Table 1. 1310 1311 Figure 2. Spatial distribution of sea surface salinity in the East China Sea. (a) summer 1312 1313 (July to September); (b) winter (January to March). Lower sea surface salinity in

1314 summer relative to that of winter indicates strong effects of summer East Asian 1315 Monsoon. 1316 **Figure 3.** (a) Lithology and oxygen isotope (δ^{18} O) profile of planktic foraminiferal 1317 1318 species Globigerinoides ruber (G.ruber) in core CSH1. (b) Plot of ages versus depth 1319 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 1320 域代码已更改 model of core CSH1 with Chinese Stalagmite composite δ¹⁸O curve of (Cheng et al., 1321 带格式的:字体:五号 2016). Tie points for CSH1 core chronology (Table 2) in Figures 2b 3c and 2e 3d are 1322 designated by blue and red solidcolored dots crosses. 1323 1324 Figure 4. Age versus (a) CaCO₃ concentration, linear sedimentation rate (LSR), (b) 1325 Total nitrogen (TN) concentration, C/N molar ratio, (c) Total organic carbon (TOC) 1326 concentration, (d) C/N molar ratio, Total nitrogen (TN) concentration, (e) linear 1327 sedimentation rate (LSR), CaCO₃ concentration, (f) Al concentration, (g) Mn 1328 1329 concentration, (h) Mo/Mn ratio, (i) Mo concentration, (j) excess Mo concentration, (k) 带格式的:下标 1330 U concentration and (l) excess U concentration and (m) (Mo/U) excess ratio in core CSH1. Light gray and dark gray Gray and black vertical bars indicate different 1331 sediment intervals in core CSH1. MIS indicates Marine Isotope Stage. 8.2 ka, PB, YD, 1332 B/A, HS1, LGM and HS2 refer to 8,200 year cold event, Preboreal, Younger Dryas, 1333 1334 Bölling - Alleröd, Heinrich Stadial 1, Last Glacial Maximum and Heinrich Stadial 2, respectively, which were identified in core CSH1. Blue solid diamonds in Figure 31 1335 4m indicate the age control points. 1336 1337 Figure 5. Scatter plots of Mo_{excess} vs Mn concentrations and U_{excess} concentration vs 1338 1339 Mo/Mn ratio at different time intervals in core CSH1. A various correlation is present 1340 in core CSH1 at different time intervals, which shows their complicated geochemical 1341 behaviors (Figs.5a and b). Strong positive correlation between Uexees concentration and Mo/Mn ratio and Uexcess concentration (Fig.5c) suggest that Mo/Mn ratio they are 1342 is a reliable proxy suitable to track sedimentary redox conditions in the geological 1343

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Figure 6. Proxy-related reconstructions of intermediate watermid-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) CaCO₃ concentration, (b) U_{excess} concentration, (c) Mo/Mn ratio, and (d) Mn concentrationsea surface temperature (SST-) (Shi et al., 2014), and (e) abundance of *P.obliquiloculata* in core CSH1 (Shi et al., 2014) and (f) bulk sedimentary organic matter δ¹⁵N of TOC in core MD01-2404 (Kao et al., 2008), (g) δ¹³C of epibenthic foraminiferal *C.wuellerstorfi* in core PN-3 (Wahyudi and Minagawa, 1997), (h) relative abundance of *B. aculeata* (hypoxia-indicating species) and (i) *C.hyalinea* (oxygen-rich indicating species) (Li et al., 2005) (hj) Dysoxie-dysoxic taxa (%) in core ODP 167-1017 in the northeastern Pacific (Cannariato and Kennett, 1999) and (ik) δ¹³C of benthic foraminiferal Uvigerina akitaensisthe in core PC23A in the Bering Sea (Rella et al., 2012). Light gray Gray and dark grayblack vertical bars are the same as those in Figure 4.

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Figure 7. Proxy records favoring the existence of <u>out-of-phase connections oceanic</u> ventilation seesaw between the subtropical North Pacific and North Atlantic during the last deglaciation and enhanced carbon storage at mid-depth waters. (a) <u>U</u> excess concentration in core CSH1; Atmospheric CO₂ concentration (Marcott et al., 2014) (b) Mo/Mn ratio in core CSH1; Indicator of strength of Atlantic Meridional Ocean Circulation (²³¹Pa/²³⁰Th) (Böhm et al., 2015; McManus et al., 2004); (c) benthic δ¹³C record in core PC-23A in the Bering Sea (Rella et al., 2012); (d) Indicator of strength of Atlantic Meridional Ocean Circulation (²³¹Pa/²³⁰Th) (Böhm et al., 2015; McManus et al., 2004); Mo/Mn ratio in core CSH1; (e) Atmospheric CO₂ concentration (Marcott et al., 2014) U excess concentration in core CH1. Light gray and dark gray vertical bars are the same as those in Figure 4. Blue diamonds are the same as those in Figure 3.

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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)
В	MD012404	26.65	125.81	1397	Okinawa Trough	Kao et al., (2008)
С	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. (20102015)*
	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 <u></u>	34.54	239.11	955	NE Pacific	Cannariato and Kennett, (1999)

◆--- 带格式的: 两端对齐

--- 带格式表格

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带格式的: 到齐到网格,制表位: 不在 19, 78 字符 + 39, 55 字符 **带格式的**: 段落间距段前: 0 磅, 段后: 0 磅, 行距: 单倍行距, 与下段不同页, 段中分页

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
466 –			23476	Stalagmite, DO2	28.59	This study
506			24163	Stalagmite, H2	<u>58.2233.29</u>	This study
698			28963	Stalagmite, DO4	40.00	This study
746 –			29995	Stalagmite, H3	46.51	This study
834			32442	Stalagmite, DO5	35 <u>39.09</u> 96	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)

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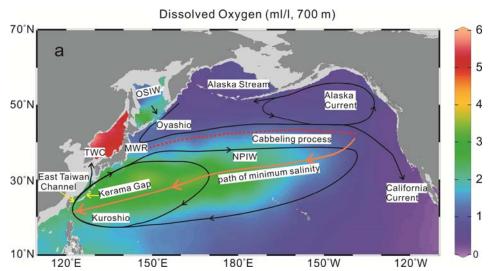
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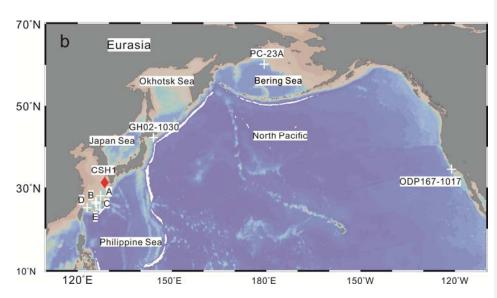
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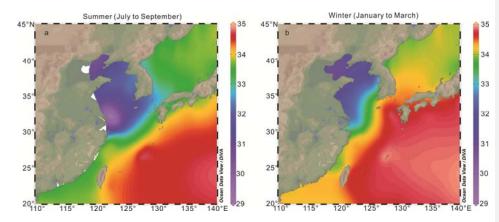
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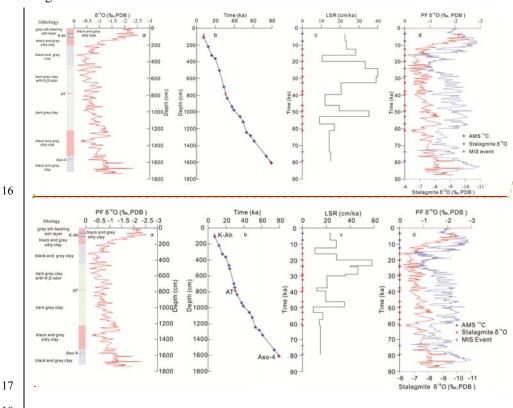


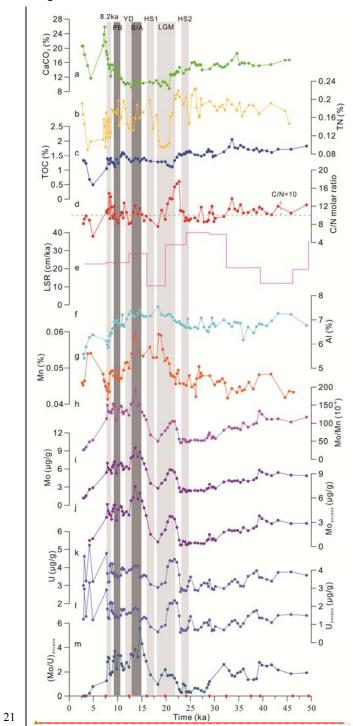




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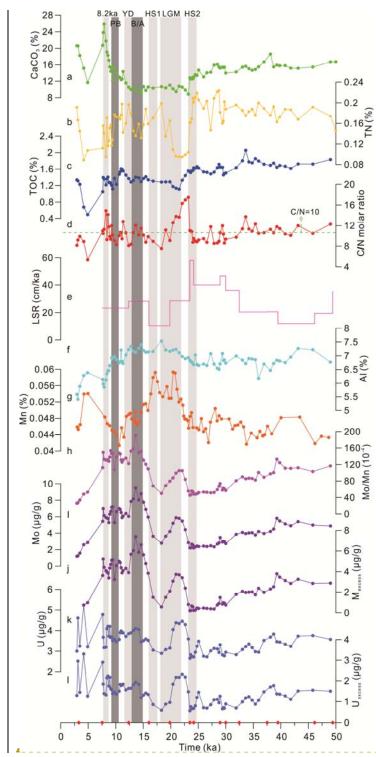
15 Fig.3

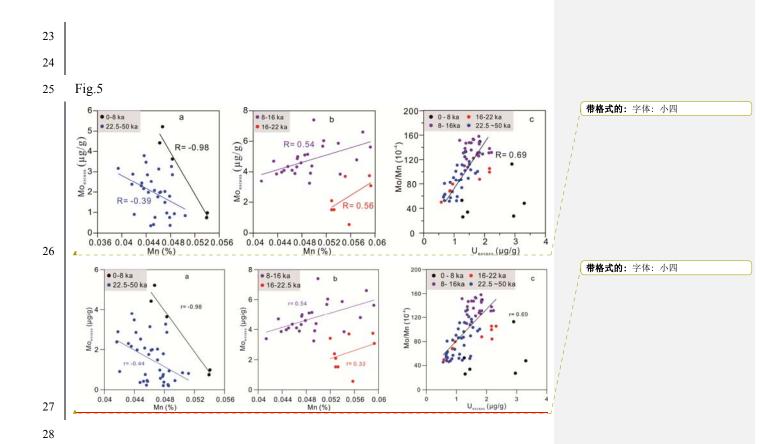




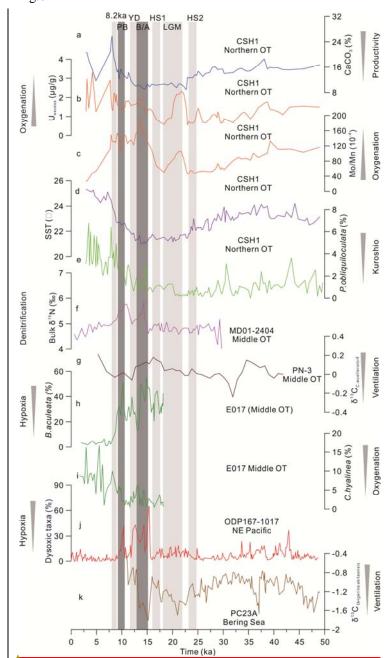
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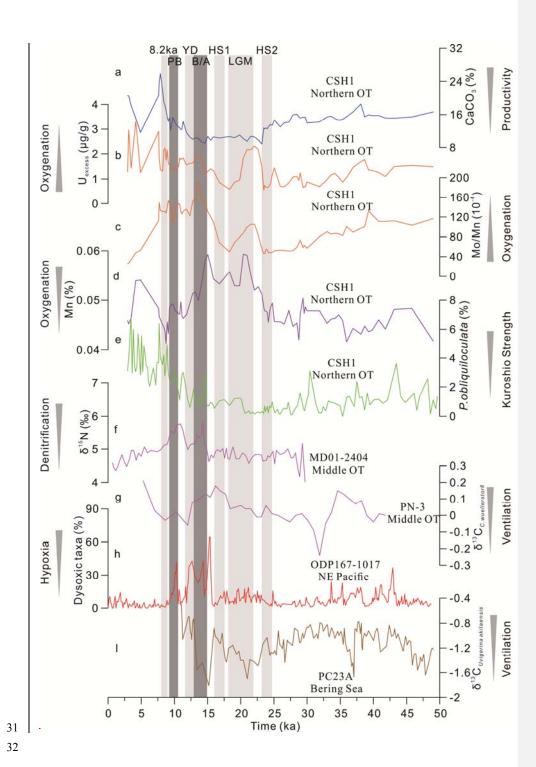




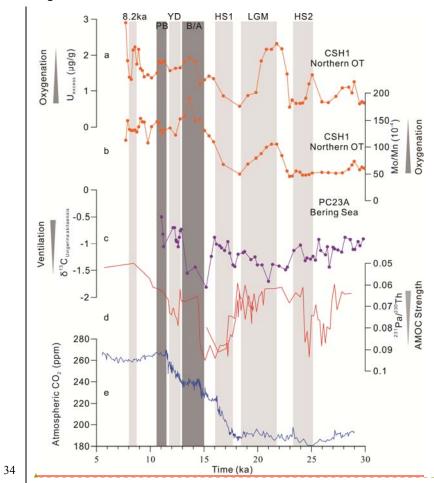
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