



Changes of deep Pacific overturning circulation and carbonate chemistry during middle Miocene East Antarctic ice sheet expansion

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ARTICLE INFO

Article history:

Received 18 June 2017

Received in revised form 25 November 2017

Accepted 1 December 2017

Available online xxxx

Editor: M. Frank

Keywords:

deep Pacific Ocean circulation

middle Miocene

carbonate chemistry

East Antarctic ice sheet expansion

ABSTRACT

East Antarctic ice sheet expansion (EAIE) at ~ 13.9 Ma in the middle Miocene represents a major climatic event during the long-term Cenozoic cooling, but ocean circulation and carbon cycle changes during this event remain unclear. Here, we present new fish teeth isotope (ϵNd) and benthic foraminiferal B/Ca records from the South China Sea (SCS), newly integrated meridional Pacific benthic foraminiferal $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ records and simulated results from a biogeochemical box model to explore the responses of deep Pacific Ocean circulation and carbon cycle across EAIE. The ϵNd and meridional benthic $\delta^{13}\text{C}$ records reveal a more isolated Pacific Deep Water (PDW) and a sluggish Pacific meridional overturning circulation during the post-EAIE with respect to the pre-EAIE owing to weakened southern-sourced deep water formation. The deep-water $[\text{CO}_3^{2-}]$ and calcium carbonate mass accumulation rate in the SCS display markedly similar increases followed by recoveries to the pre-EAIE level during EAIE, which were probably caused by a shelf–basin shift of CaCO_3 deposition and strengthened weathering due to a sea level fall within EAIE. The model results show that the $\sim 1\text{‰}$ positive $\delta^{13}\text{C}$ excursion during EAIE could be attributed to increased weathering of high- $\delta^{13}\text{C}$ shelf carbonates and a terrestrial carbon reservoir expansion. The drawdown of atmospheric CO_2 over the middle Miocene were probably caused by combined effects of increased shelf carbonate weathering, expanded land biosphere carbon storage and a sluggish deep Pacific meridional overturning circulation.

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1. Introduction

Earth's climate underwent dramatic cooling during the middle Miocene climate transition (MMCT) around 14.2–13.8 Ma, manifested by a 6–7 °C sea surface temperatures (SST) drop in the high-latitude southwest Pacific Ocean (Shevenell et al., 2004). At ~ 13.9 Ma, a $\sim 1.0\text{‰}$ increase in benthic foraminiferal $\delta^{18}\text{O}$ suggests a large ice sheet expansion in East Antarctic (e.g., Tian et al., 2013), equivalent to ~ 40 – 60 m sea level drop (John et al., 2004). During this so-called East Antarctic Ice-sheet Expansion (EAIE), the ocean carbon reservoir also showed a prominent perturbation, characterized by large positive excursions in benthic and planktonic foraminiferal $\delta^{13}\text{C}$ in the Pacific Ocean (e.g., Woodruff and Savin, 1991; Holbourn et al., 2005; Tian et al., 2014). These positive

excursions are known as Carbon-isotope Maxima (CM) (Vincent and Berger, 1985; Woodruff and Savin, 1991). In total, six CM events, numbered from CM1 to CM6 and paced by 400-kyr eccentricity cycles, have been identified (e.g., Holbourn et al., 2013; Shevenell et al., 2004; Shevenell and Kennett, 2004; Tian et al., 2013, 2014; Woodruff and Savin, 1991). CM6 event is the largest carbon isotope shift, showing a prominent $\sim 1.0\text{‰}$ increase in benthic foraminiferal $\delta^{13}\text{C}$ during ~ 13.9 – 13.8 Ma.

Explanations for the causal relationship between deep ocean circulation, EAIE and CM6 mainly fall into two categories. One hypothesis (so-called “CO₂ hypothesis”) suggests that EAIE was triggered by an atmospheric $p\text{CO}_2$ drop which was driven by oceanic carbon reservoir changes including variations in carbonate chemistry, organic carbon production, and remineralization (Diester-Haass et al., 2009; Kürschner et al., 2008; Raymo and Ruddiman, 1992; Vincent and Berger, 1985). Existing atmospheric $p\text{CO}_2$ reconstructions have much lower time resolutions than benthic $\delta^{18}\text{O}$ records, and different proxies show contradictive results during the middle Miocene (Beerling and Royer, 2011; Foster et al., 2012). It is yet difficult to verify the “CO₂ hypothesis”,

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given the poor timing control of atmospheric $p\text{CO}_2$ relative to the ice sheet growth in Antarctic. The other hypothesis (“ocean circulation hypothesis”) states that poleward heat/moisture flux changes, driven by ocean circulation reorganization, were responsible for the ice sheet buildup and global cooling during the middle Miocene (e.g., Holbourn et al., 2005, 2013; Shevenell et al., 2004; Tian et al., 2009; Woodruff and Savin, 1991). Holbourn et al. (2013) suggested that following EAIE intermediate and deep water productions in the Southern Ocean strengthened the meridional overturning circulation and ventilation of the deep Pacific. However, this deep circulation pattern failed to explain the absence of CM6 event registered in the deep western Pacific Ocean (Tian et al., 2009). At present, deep Pacific Ocean circulation regime, carbonate chemistry changes and atmospheric $p\text{CO}_2$ are poorly constrained and their climatic roles remain unclear during MMCT.

To better understand the link between deep Pacific Ocean circulation, carbon cycle and EAIE during the middle Miocene, the middle Miocene requires high-quality paleoceanographic records based on well-calibrated proxies from multiple sites in the Pacific Ocean. In this study, we present new neodymium isotopic (ϵNd) records using fossil fish teeth, carbonate ion concentrations ($[\text{CO}_3^{2-}]$) derived from benthic foraminiferal boron-to-calcium (B/Ca) ratios, and carbonate accumulation rates from the western Pacific Ocean during MMCT. Combined with published benthic foraminiferal $\delta^{13}\text{C}$ records, we investigate the deep Pacific Ocean circulation changes and their links to Antarctic ice sheet expansion and carbon cycle during this transition. Finally, we use a biogeochemical box model to diagnose mechanisms causing the carbon cycle perturbation associated with EAIE.

2. Modern deep Pacific Ocean circulation

Today, the intermediate Pacific Ocean is occupied by two fresh, low-salinity water masses, the Antarctic Intermediate Water (AAIW) and the North Pacific Intermediate Water (NPIW) (Figs. 1b and c). AAIW is ventilated in the Southern Ocean and traceable throughout the South Pacific. By contrast, NPIW is confined to the subtropical North Pacific and ventilated in the Okhotsk Sea (Talley et al., 2011), where Dense Shelf Water is produced in coastal polynyas by brine rejection during winter sea-ice production. No deep water is formed in the modern subarctic Pacific, as surface waters are isolated from subsurface waters by a steep halocline, which leads to a robust surface ocean stratification in this region (Fig. 1c). AAIW spreads northward at $\sim 500\text{--}1500$ m water depth to $\sim 10^\circ\text{S}$ where it mixes with NPIW from the north.

In the South Pacific Ocean, water masses underlying AAIW include high-salinity Circumpolar Deep Water (CDW) and Antarctic Bottom Water (AABW) (Figs. 1b and c). Densest AABW is usually confined south of 50°S in the abyssal ocean. Depending on their densities, CDW can be further divided into Upper Circumpolar Deep Water (UCDW) and Lower Circumpolar Deep Water (LCDW), which flow northward into the central Pacific Ocean Basin (Figs. 1a and b). The southern-sourced deep waters are diffusively transformed into Pacific Deep Water (PDW), with little contribution of surface waters from the North Pacific (Figs. 1a and b). Hence, PDW represents an aged water mass characterized by low dissolved oxygen, high nutrients and negative $\delta^{13}\text{C}$ compared to relatively younger water masses formed in the Southern Ocean (Fig. 1b). The southward PDW return flow contributes to UCDW at $2000\text{--}3000$ m depth in the South Pacific (Figs. 1a and b).

3. Materials and methods

3.1. ODP Site 1148

Ocean Drilling Program (ODP) Site 1148 ($18^\circ 50.169'\text{N}$, $116^\circ 33.939'\text{E}$; 3294 m water depth) is located at the lower con-

tinental slope of the northern South China Sea (SCS) (Fig. 1a). During Quaternary, the deep SCS is connected to the open Pacific Ocean via Luzon Strait with the deepest sill depth of 2650 m, below which deep SCS waters are well mixed showing similar physicochemical characteristics as deep waters in the open Pacific Ocean (Qu et al., 2006). The tectonic history of the SCS was closely related to India-Eurasia continental collision and the subduction of the Pacific Plate. The previous researches suggested the age of the SCS is from ~ 32 Ma to 17 or 16 Ma (Taylor and Hayes, 1980, 1983; Briais et al., 1993; Seton et al., 2012). This result is further supported by the recently acquired deep tow magnetic anomalies and International Ocean Discovery Program (IODP) Expedition 349 cores, which indicated the onset of seafloor of SCS occurred between 32–31.5 Ma and seafloor spreading was terminated at ~ 15 Ma (Li et al., 2014). Then, the SCS oceanic crust was subducted eastward beneath the West Philippine Sea Plate along the Manila Trench, which produced an accretion prism and a volcanic arc between Taiwan and Luzon Islands (Huang et al., 1997; Queano et al., 2007). At around 10 Ma, the Palawan and the Borneo/Kalimantan were subducted beneath the double subduction zone between the SCS oceanic crust and the Philippine plate, which established the semi-closed SCS formation and likely caused significant uplift of the central sill of between the Taiwan and Luzon in the Late Miocene (Queano et al., 2007; Seton et al., 2012). At least before 10 Ma, the deep SCS was entirely open to the deep west Pacific Ocean, allowing the use of ODP Site 1148 to infer deep Pacific Ocean circulation changes. ODP Site 1148 unfolds an 853-m-thick sediment succession covering the time since the Late Oligocene (Wang et al., 2000). The Middle Miocene lithology mainly contains olive-gray and reddish-brown clays with nanofossils, and show a sedimentation rate of ~ 15 mm/kyr and carbonate contents of $\sim 40\%$ (Wang et al., 2000). The age model for ODP Site 1148 is readjusted to that of IODP Site U1337 (Tian et al., 2013) by correlating their benthic $\delta^{18}\text{O}$ records (Tian et al., 2008).

3.2. Fish teeth ϵNd

In the ocean, seawater Nd is mainly derived from continental weathering. The interocean contrast of deep water ϵNd is dominated by the distribution of ancient and juvenile crusts surrounding the ocean. The modern Southern Ocean sourced waters have ϵNd values ranging from -8 to -9 , while ϵNd of deep North Pacific waters span from -4 to -6 (e.g., Frank, 2002). Neodymium has a short residence time of $300\text{--}1000$ yrs in the ocean, shorter than the mean oceanic mixing time (e.g., Scher et al., 2015). This renders seawater ϵNd to be sensitive to ocean circulation and water mass mixing. Deep-water ϵNd can be recorded by fish teeth apatite, not biased by post-deposition processes (Martin and Scher, 2004). This allows the use of fish teeth ϵNd to reconstruct ocean circulation changes in the past.

We measured ϵNd in fossil fish teeth from ODP Site 1148 to investigate Pacific overturning circulation changes during EAIE. For each sample, 2–5 fish teeth debris were picked from the $>150\text{-}\mu\text{m}$ size fraction, and checked by scanning electron microscopy. Fish teeth samples were then cleaned by quartz-distilled water and methanol, followed by reductive, oxidative, and weak acid cleaning steps. The cleaned fish teeth were finally dissolved in quartz-distilled 0.25 M HCl. Nd was then purified using the single column method (Scher et al., 2015), with the only change being a doubling column length to improve separation of samarium. ϵNd were measured on a Neptune multiple collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at the Center for Elemental Mass Spectrometry (University of South Carolina). The mass fractionation was corrected by normalizing $^{146}\text{Nd}/^{144}\text{Nd}$ to 0.7219 and using exponential-fractionation correction. During analysis, each sample

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