



Invariance of the carbonate chemistry of the South China Sea from the glacial period to the Holocene and its implications to the Pacific Ocean carbonate system

Yiming Luo^{a,b,c,*}, Markus Kienast^a, Bernard P. Boudreau^a

^a Department of Oceanography, Dalhousie University, 1355 Oxford Street, Halifax, NS B3H 4R2, Canada

^b Geophysical Institute, University of Bergen and Bjerknes Centre for Climate Research, Allegaten 70, Bergen 5007, Norway

^c School of Marine Sciences, Sun Yat-Sen University, A405 Main Building, Zhuhai, Guangdong 519082, China

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ABSTRACT

Substantial and correlated changes in marine carbonate (CaCO_3) content of oceanic sediments commonly accompany the transitions from cold glacial periods to warm interglacial periods. The South China Sea (SCS) is said to be ocean-dominated at depth, and its CaCO_3 records should reflect and preserve the effects of changes in the carbonate chemistry of the (western) Pacific Ocean. Using published and newly acquired CaCO_3 data and a model for carbonate compensation dynamics, we show that a significant change with respect to carbonate saturation is unlikely to have occurred in the SCS during the last glacial–interglacial transition. Instead, the results from a carbonate deposition model argue that the saturation state of the SCS was largely invariant; a separate diagenetic model argues that changes in sediment CaCO_3 content can be explained by alterations in lithogenic input. In turn, this could indicate that the carbonate ion concentration of the (western) Pacific at depths shallower than the sill to the SCS (ca. 2,400 m) has not changed appreciably between the last glacial period and the present interglacial.

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1. Introduction

Global climate change on glacial–interglacial timescales is linked causatively to profound differences in the fluxes and accumulations of terrestrial and marine sedimentary materials in the ocean (e.g. Francois et al., 1990, 1993; Winckler et al., 2008). Accumulation of components, such as carbonate (e.g. Lyle et al., 2002; Francois et al., 1990; Farrell and Prell, 1989) or dust (e.g. Winckler et al., 2008; Kienast et al., 2016), are consequently a reflection of both cause and effect of dramatic changes in ocean biogeochemistry and continental climate on multi-centennial timescales.

The CaCO_3 content observed with depth (time) in sediment cores from broad areas of the Pacific Ocean has long been established to fluctuate generally with Pleistocene glacial–interglacial climate cycles (e.g., Berger, 1973; Farrell and Prell, 1989), but the ultimate cause for this “Pacific-style” cyclicity with higher glacial CaCO_3 -content compared to interglacials remains moot. While carbonate production has been shown to be largely invariant on glacial–interglacial time scales, based on ^{230}Th -normalized

CaCO_3 flux reconstructions (e.g., Anderson et al., 2008), microfossil records have been interpreted to reflect changes in production at some sites (Bordiga et al., 2013). Proxy evidence for changes in carbonate-ion concentrations (CO_3^{2-}) has been ambiguous as well. Foraminiferal fragmentation indices have been interpreted to reflect better glacial CaCO_3 preservation and thus higher carbonate ion concentrations (Sun et al., 2017; Mekik et al., 2012). Foraminiferal shell weights, on the other hand, suggest lower carbonate ion concentrations for the entire Pacific during the glacial time than today (Broecker and Clark, 2003), whereas foraminiferal B/Ca implies modest changes in CO_3^{2-} , on the order of -5 to $3 \mu\text{mol/kg}$ between the LGM and the late Holocene (Yu et al., 2013; Kerr et al., 2017).

These ambiguous or equivocal results mean that despite the long-established and significant cyclicity in sedimentary CaCO_3 content, paleoceanographers and marine geologists have no clear idea of the behavior the Pacific carbonate chemistry over the past ~ 20 ka, i.e. the LGM to Holocene transition, which constitutes a serious gap in our knowledge. As a result, we have been considering other sources of data and information that might shed light onto the problem. In particular, we considered the possibility that marginal, ocean-dominated, seas might better archive changes in Pacific carbonate chemistry through their sediment CaCO_3 records. Here we consider the changes recorded in the

* Corresponding author at: School of Marine Sciences, Sun Yat-Sen University, A405 Main Building, Zhuhai, Guangdong 519082, China.

E-mail address: luoyiming@mail.sysu.edu.cn (Y. Luo).

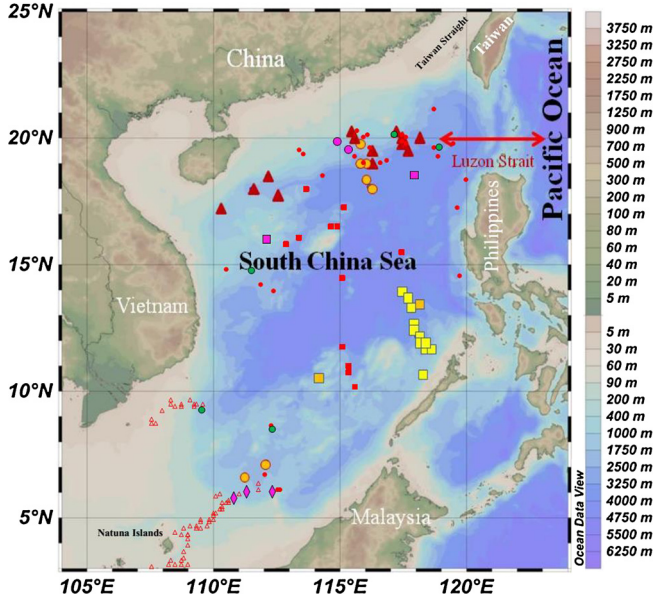


Fig. 1. Location of the South China Sea (SCS), primary geographic features mentioned in the text, and the location of sediment cores discussed in this study. Large, dark red, triangles are cores that contain the early-Holocene (8.5–11 ka) low $\text{CaCO}_3\%$ event in north SCS (Huang et al., 2015); pink diamonds are sediment cores north of Natuna island in the Southern SCS (Steinke et al., 2003); green circles are unpublished data from this study; orange circles are the cores compiled by Wang et al. (1995), which are on or close to the continental slope; orange squares are those far from the continental slope, also compiled by Wang et al. (1995); small red circles and red squares are new data reported in this study for locations close to or far from the continental slope, respectively; pink circles and squares are the cores close to or far from the continental slope, respectively reported by Li (1993); light yellow squares are sediment cores in east SCS (Thunell et al., 1992); red, unfilled triangles in southern SCS are data measured by Paulsen (1998). (Details regarding all the sediment cores can be found in Table S1 and S2 in supplementary materials.) (For interpretation of the colors in the figure(s), the reader is referred to the web version of this article.)

CaCO_3 content of sediment cores from the South China Sea (SCS), the largest, semi-enclosed marginal sea connected to the Pacific Ocean (Fig. 1). Currently, the deep water in the SCS is thought to be “ocean-dominated” (Dai et al., 2013), influenced by ~ 3 Sv of incoming Pacific intermediate/deep water through the Luzon straight from the Philippines Sea (deepest sill at about 2400 m in the Bashi Channel, according to Wang et al., 2011). Consequently, it is widely believed that changes in seawater chemistry of the SCS closely mirror those of the Pacific Ocean today and did so during the late Quaternary (Miao et al., 1994; Chen et al., 1997; Wei et al., 1998). In addition, extensive paleoceanographic studies of the SCS have generated a large database of past geochemical conditions (Wang and Li, 2009).

Earlier data from Thunell et al. (1992) and Wang et al. (1995) show that, like Pacific sediments, the CaCO_3 content of SCS sediments below a water depth of 3,500 m was higher during the LGP (Last Glacial Period) than during the Holocene. This might suggest that the deep SCS simply mirrors the carbonate chemistry of the deep western Pacific, as expected. Here, we present a new, more complete, data set of CaCO_3 records for SCS sediments and then introduce a relatively simple model that predicts carbonate dynamics within the SCS basin, i.e., saturation and compensation depths. With this model we test the consistency and viability of the competing proposed causes: changing ocean chemistry versus changing inputs. We show that the ocean depth where the CaCO_3 falls to zero (compensation depth) seems not to have changed in the SCS, suggesting that the carbonate chemistry of the SCS and thus the Pacific Ocean are invariant on glacial–interglacial timescales, or that the SCS is not (Pacific-)ocean dominated and

was invariant in its own right. Finally, using a separate diagenetic model, we can account for the full CaCO_3 depth profiles, including the recorded swift and strong decrease of CaCO_3 content during the early Holocene (9–11 ka) in northern SCS sediment cores as a result of changing rates of non-carbonate sedimentation, rather than changes in carbonate chemistry.

2. Methods

2.1. Data

Sampling locations are displayed in Fig. 1, and the resulting data are displayed in Figs. 2–4 and tabulated in Table S1, S2 and S4 of the SI. The data consist of a combination of new measurements of carbonate content (as weight percent carbonate, $\text{wt}\text{CaCO}_3\%$) and previously published data (Huang et al., 2015; Steinke et al., 2003; Wang et al., 1995; Li, 1993; Thunell et al., 1992; Paulsen, 1998; Zhang et al., 2015).

2.2. Model equations for carbonate chemistry

Oceanic carbonate dynamics are characterized by three critical depths in the ocean. The first is the carbonate (calcite) saturation horizon, where the water is exactly saturated with respect to CaCO_3 (calcite); its position can be expressed as (Boudreau et al., 2010a):

$$Z_{\text{sat}} = \alpha^{-1} \ln\left(\frac{[\text{Ca}^{2+}]_{\text{D}}[\text{CO}_3^{2-}]_{\text{D}}}{K_{\text{sp}}^1}\right) \quad (1)$$

where α is an attenuation constant derived from empirical fits to the calcite solubility data, $[\text{Ca}^{2+}]_{\text{D}}$ and $[\text{CO}_3^{2-}]_{\text{D}}$ (mM) are the concentrations of calcium and carbonate ions in the deep ocean, respectively, K_{sp}^1 is the solubility product at 1 atm pressure and at the temperature and salinity of the deep ocean, which is assumed homogeneous (Boudreau et al., 2010a).

The carbonate (calcite) compensation depth, Z_{cc} , is the depth where the CaCO_3 (calcite) dissolution rate at the sediment–water interface equals the rate of biogenic CaCO_3 deposition; this balance can be expressed mathematically to obtain an equation for Z_{cc} (Boudreau et al., 2010a):

$$Z_{\text{cc}} = \alpha^{-1} \ln\left(\frac{[\text{Ca}^{2+}]_{\text{D}}[\text{CO}_3^{2-}]_{\text{D}}}{K_{\text{sp}}^1} + F_{\text{B}}[\text{Ca}^{2+}]_{\text{D}} / \{K_{\text{sp}}^1 A_{\text{D}} k_{\text{c}}\}\right) \quad (2)$$

where F_{B} ($\mu\text{mol cm}^{-2} \text{yr}^{-1}$) is the export flux of CaCO_3 (calcite) onto deep-ocean sediments, and excluding CaCO_3 deposition to sediments on shelves, A_{D} (m^2) is the area of the deep seafloor, and k_{c} is the mass-transfer coefficient for dissolution (Boudreau et al., 2010b; Boudreau, 2013). The position of Z_{cc} is independent of the sedimentation rate of non-carbonate components, as can be discerned from Equation (2).

Finally, the snowline (Z_{SL}) is the critical depth where calcite first disappears entirely from the sediment (Zeebe and Westbrock, 2003), which is identical to Z_{cc} at (quasi-)steady state. The snowline will be different from Z_{cc} if the deep water is undergoing fast acidification (non-steady state).

3. Results and discussion

3.1. Late Holocene sediments

The CaCO_3 content of Late-Holocene SCS sediments displays, on average, well defined trends, despite appreciable scatter (Fig. 2). This scatter is not surprising, as the CaCO_3 of sediments can be influenced by variable in-situ conditions, such as local CaCO_3 productivity, intensity of CaCO_3 dissolution, and delivery of lithogenic

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