Quaternary International 349 (2014) 257–269

Contents lists available at ScienceDirect

Quaternary International

journal homepage: www.elsevier.com/locate/quaint

Carbon isotopic records of middle Holocene corals and environmental causes in northern South China Sea

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ARTICLE INFO

Article history:
Available online 7 September 2014

Keywords:
Coral $\delta^{13}C$
Solar radiation
Atmospheric CO_2 concentration
Salinity
Holocene
South China Sea

ABSTRACT

Two high-resolution $\delta^{13}C$ records of ~5.4 and ~4.4 ka coral colonies *Porites lutea* from east coast of Hainan Island in the northern South China Sea (SCS) provide insights into the complex links between coral carbon isotopes and environment changes. The $\delta^{13}C$ of the ~5.4 ka coral offered a 40-year growth history with an average of −2.62‰ and amplitude of 2.96‰, the ~4.4 ka coral's revealed a 55-year growth history with an average of −3.12‰ and amplitude of 4.21‰, which revealed higher coral $\delta^{13}C$ and increased seasonality in the middle Holocene. Our analysis suggests that the higher coral $\delta^{13}C$ was the combined result of higher radiation, salinity and nutrient level and lower atmosphere CO_2 concentration, which might increase coral $\delta^{13}C$ through strengthening photosynthesis of symbiont zooxanthella in the context of kinetic and metabolic fractionations. The increased seasonality was mostly produced by strengthened radiation.

The decreasing standard deviation of the ~5.4 ka coral annual $\delta^{13}C$ revealed weakened interannual changes of atmospheric CO_2, salinity, and nutrient. By contrast, the increasing deviation of the ~4.4 ka coral $\delta^{13}C$ suggests strengthened interannual changes of these variables. The ~5.4 ka coral $\delta^{13}C$ showed a long-term increasing trend at a rate of 0.33% increase y^{-1}, which was attributed to natural decrease in atmospheric CO_2, and increasing salinity and nutrient level. However, the ~4.4 ka coral $\delta^{13}C$ showed a long term decreasing trend at a rate of 0.25% decrease y^{-1}, which was ascribed to unusual increase in atmospheric CO_2, and decreasing salinity and nutrient level.

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1. Introduction

Keith and Weber (1965) first found that unlike other organisms, corals frequently form their skeletons out of "isotopic equilibrium" with their environment. The isotopic disequilibrium is attributed to the combined effect of kinetic and metabolic fractionation (McConnaughey, 1989a, b). Kinetic fractionation is associated with CO_2 hydration and hydroxylation, and it results in depletion of ^{13}C in carbonate. In this model, the coral pumps protons from the region in the skeleton forming raising the pH and lowering pCO_2. Molecular CO_2 then diffuses across the coral basal epithelium and reacts with H_2O and OH^- to produce HCO_3^- and CO_3^{2-}. This raises the $CaCO_3$ saturation state, and allows for rapid calcification. The starting CO_2 has relatively low ^{13}C content. Kinetic discrimination against the heavy isotopes during CO_2 reactions further lowers the ^{13}C content of product HCO_3^-. During fast calcification, this isotopically depleted HCO_3^- precipitates before it can isotopically equilibrate with seawater dissolved inorganic carbon (DIC). In contrast, the effect of the metabolic fractionation on coral $\delta^{13}C$ is more complicated (Swart, 1983; McConnaughey, 1989a, b). According to the common carbon pool assumption, photosynthesis preferentially uses ^{12}C, leaves the DIC pool enriched in ^{13}C (Goreau, 1977;

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http://dx.doi.org/10.1016/j.quaint.2014.08.030
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Swart, 1983). As the metabolic demand for carbon increases with coral growth, the pool of available $^{12}CO_2$ in the coral becomes relatively depleted causing increase to the $\delta^{13}C$ signature of coral (Erez, 1978; Swart, 1983; McConnaughey, 1989b). As photosynthesis is the faster reaction in light, the reservoir enriched in ^{13}C caused a $\delta^{13}C$ increase with light (Goreau, 1977). On the contrary, during respiration of organic matter, addition of ^{12}C-enriched CO_2 decreases the $\delta^{13}C$ of the skeletal carbonate (Smith and Kroopnick, 1981) due to reduction in photosynthesis and decrease in metabolic processes (Grottoli et al., 2004).

Factors such as cloud cover can play a key role in carbon assimilation of symbiotic corals (Fairbanks and Dodge, 1979; Quinn et al., 1993; Sun et al., 2008) where $\delta^{13}C$ signature has been shown to be a recorder of light level or solar radiation due to radiation induced changes in metabolic rate (Coles and Jokiel, 1978; Swart, 1983; McConnaughey, 1989a; Grottoli and Wellington, 1999), such as, moderate increase in the rate of photosynthesis, related to increase in light intensity, appears to increase the coral skeleton $\delta^{13}C$, while decrease in light results in low $\delta^{13}C$ in the skeleton (Swart et al., 2005). Obviously, metabolic effects are responsible for large shifts of skeletal $\delta^{13}C$, which results primarily from the seasonally variant ambient light incident on the coral surface (Coles and Jokiel, 1978; Swart et al., 2005, 2010).

However, characteristics of interannual variations in coral $\delta^{13}C$ are difficultly explained by solar radiation, but easily interpreted by other factors, such as, sea surface salinity (SSS) indicating seawater components related to terrestrial runoff and rainfall (Swart et al., 1996; Su et al., 2007; Craig et al., 2010), nutrient (Grottoli and Wellington, 1999), spawning (Gagan et al., 1996), El Niño-Southern Oscillation (ENSO) event and blenching (Grottoli et al., 2004). Coral $\delta^{13}C$ shows a significant positive response to salinity (Pretet et al., 2014), which is related to precipitation and river discharge related DIC in the coastal ocean (Grove et al., 2012). Moreover, seawater zooplankton, composition, and DIC are used by coral as food and calcification (Gupta et al., 2006), because on the one hand, coral is autotrophic and depend on its symbiont for nutrition, on the other it is heterotrophic and depend on plankton predation (Levy et al., 2006). Coral $\delta^{13}C$ increases as abundant nutrient and rich food can strengthen the both algal photosynthesis and coral calcification (Grottoli and Wellington, 1999). Thus, $\delta^{13}C$ reflects the different DIC proportions of calcified CO_2 sources from seawater and coral respiration (Rollion-Bard et al., 2003), which may be independent of illuminations (Furla et al., 2000). In non-zooxanthellate coral, the fractionation of the stable isotopes is controlled by the pH of the calcification fluid, as which decreases the skeletal $\delta^{13}C$ decreases (Adkins et al., 2003). Significantly more negative $\delta^{13}C$ explains that coral may respire a significant proportion of isotopically negative substances, such as lipids (Swart et al., 2005). Obviously, $\delta^{13}C$ apparent disequilibrium in coral is the result of a mixing among metabolic CO_2 related with solar radiation, DIC, salinity, nutrient and more other factors coming directly from seawater.

In addition to the seasonal and interannual variation in coral $\delta^{13}C$, several workers have remarked upon long term trend towards lower $\delta^{13}C$ value within modern coral skeleton, which is attributed to the ^{13}C Suess effect due to the addition of anthropogenically derived CO_2 to the atmosphere (Nozaki et al., 1978; Swart et al., 2010). Coral $\delta^{13}C$ has a positive correlation with pH as CO_2 increases in atmosphere and seawater (Sabine et al., 2004), which is closely linked to the anthropogenic CO_2 emissions from burning of fossil fuels (Hemming et al., 1998). Coral $\delta^{13}C$ may also identify long term processes of coral growth under high nutrient loads and potential disturbances, suggesting that mariculture and other human related stressors influence on the carbon fractionation of coral skeleton (Levy et al., 2010). Therefore, distinguishing and evaluating various processes and environmental variables are necessary if coral $\delta^{13}C$ values are used to reconstruct marine environments.

In this work, the two fossil coral colonies *Porites lutea* OC1 and OC2, and modern coral *P. lutea* QG5 were collected at east coast of Hainan Island in the northern South China Sea (SCS), a region well positioned to monitor the Asian monsoon. Previous works have discussed the evolution of Asian monsoon variability in the middle Holocene revealed by the $\delta^{18}O$ records of the two fossil corals (Sun et al., 2005; Su et al., 2010), based on the researches of relationships between the modern coral skeletal $\delta^{18}O$ and monsoonal sea surface temperature (SST), SSS and fresh-water input (Su et al., 2006). It has been studied for associations between the modern coral calcification and $\delta^{13}C$ and solar radiation and cloud cover, climate warming due to increasing anthropogenic CO_2 emissions and human activities (Sun et al., 2008), and carbon isotope compositions of SSS indicating seawater inorganic and organic carbon components related to rainfall and freshwater inputs, and increasing atmospheric CO_2 concentration (Su et al., 2007). In contrast, the associations between the Holocene coral $\delta^{13}C$ and environment changes are poorly understood. In this study, the paper offers 40 and 55 year high resolution records of the two fossil coral $\delta^{13}C$, which are compared with a 17 year record of the modern coral QG5 $\delta^{13}C$. Consequently, Holocene environment and climate causes for coral $\delta^{13}C$ variability are examined and evaluated at seasonal, interannual and interdecadal scales.

2. Methods

2.1. Study site

The corals are located on eastern Hainan Island of the northern SCS (Fig. 1A), strongly affected by the Asian monsoon and northwestern Pacific Ocean. During the summer monsoonal seasons, wetter climate prevails in the area, with higher temperature (25–29 °C), lower salinity (4.1–33.7‰), and more plentiful rainfall (2000–2400 mm). During winter seasons, drier climate prevails with lower temperature (16–24 °C), higher salinity (20.5–35.5‰), and lower rainfall (1000–1200 mm). Sunshine is from 1750 to 2650 h annually, and the total annual radiation is about 4500–5900 MJ m^{-2}. Detailed millennial climate changes in geomorphology of the Holocene are appropriate for investigation, because periodic uplifts elevated a succession of fossil reefs above present sea level at Hainan Island. Abundant Holocene coral reefs have developed along the coastal shallow sea area around the northern SCS (Sun et al., 2005; Su et al., 2010).

2.2. Coral sampling

In February 2002, the two well-preserved fossil corals *P. lutea* OC1 (Su et al., 2010) and OC2 (Sun et al., 2005) were collected from an emergent Holocene coastal terrace at Oucun, eastern Hainan Island, ~10 km northeastern of Qionghai (Fig. 1A and B). A modern coral *P. lutea* QG5, growing in a similar reef environmental setting, was collected from the same platform on 5 May, 2002, ~5 km southwest of the paleo-reefs (Fig. 1B; Su et al., 2006). Tetragonal prism samples of the two fossil corals and one modern coral with cross-sections as squares with sides 30 cm were cut in parallel to their main growth axes.

2.3. Chronology and isotopic analysis

In the laboratory, the corals were cut into 5-mm-thick slabs in parallel with their main growth axes using an automatic stonecutter, as with the modern coral (Su et al., 2006). After air-drying,

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