EXPRESS LETTER

Mekong River discharge and the East Asian monsoon recorded by a coral geochemical record from Con Dao Island, Vietnam

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The offshore flow of the Mekong River is strongly governed by the East Asian monsoon (EAM). Monthly-resolved skeletal carbonate Sr/Ca, Ba/Ca and oxygen isotope ($\delta^{18}\text{O}_c$) data from 1980 to 2005 of a *Porites* coral from Con Dao Island, ~90 km from the Mekong River mouth, were analyzed for capturing the past flood events and the signal of East Asian winter monsoon. Ba/Ca time series is characterized with intra-annual double peaks, the first large one in March during the dry season and relative small one in August during the wet season. The low values of seawater $\delta^{18}\text{O}$ ($\delta^{18}\text{O}_{sw}$), deduced from both the Sr/Ca ratio and $\delta^{18}\text{O}_c$, in the wet season resulted from precipitation and/or freshwater input from the Mekong River. The difference in the seasonal characteristics of Ba/Ca and $\delta^{18}\text{O}_{sw}$ between flood and no-flood years could be attributed to the seasonal reversal of the regional ocean current derived from the EAM.

Keywords: coral skeleton, Ba/Ca, oxygen isotope, Con Dao Island, Mekong River

INTRODUCTION

The Mekong River is one of the largest river systems in the world. The climate in the lower Mekong region is dominated by the East Asian monsoon. Precipitation occurs mainly during the warm/wet season from May to November. River discharge reaches a maximum in September or October and a minimum in April (based on averaged water discharge data from the Tan Chau station from 1979 to 2005). The Mekong River plume is mostly geostrophic and exhibits a strong seasonal variability related to the monsoon wind and water current system, which is northward in winter and southward in summer (Tang *et al.*, 2006; Hein *et al.*, 2013; Unverricht *et al.*, 2014). The Mekong River in Vietnam represents a great potential for agriculture and aquaculture production. However, the knowledge about Mekong River discharge and past flood events have been very limited because of the

lack of observations and historical documents (Tran, 2009; Xue *et al.*, 2012). The paleoclimatic history of Mekong River discharge will contribute to prevent the disaster due to floods and climate changes, and to promote economic development in the Mekong Delta.

The geochemical compositions of *Porites* coral skeletons are affected by the prevailing environment at the time of skeletal deposition and high-resolution geochemical analyses of the skeletons provide useful information about seasonal variations in past sea surface climates and environments (Watanabe *et al.*, 2003). Coral skeletal Sr/Ca ratios have been used as a proxy for sea surface temperature (SST) (e.g., Mitsuguchi *et al.*, 2008; Bolton *et al.*, 2014). The coral stable oxygen isotopic record ($\delta^{18}\text{O}_c$) has also been used for the reconstruction of SST (Watanabe *et al.*, 2003; Cahyarini *et al.*, 2014). Using both the Sr/Ca ratio and $\delta^{18}\text{O}$, we can reconstruct the oxygen isotopic composition of sea water ($\delta^{18}\text{O}_{sw}$) as an indicator of sea surface salinity (SSS) related to freshwater runoff into the coral reefs. McCulloch *et al.* (1994) calculated $\delta^{18}\text{O}_{sw}$ and residual variation in coral $\delta^{18}\text{O}$ for the Great Barrier Reef of Australia. Their results were

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shown to correlate with the freshwater runoff from a nearby major river. This was used as proxy for the reconstruction of past flood events related to ENSO cycles in the western tropical Pacific corals.

Ba/Ca ratios in coral skeletons have been well calibrated with Ba concentration in seawater (La Vigne *et al.*, 2016; Gonneea *et al.*, 2017). Coral Ba/Ca ratios reflect changes in the seawater Ba/Ca composition due to oceanic upwelling, local rainfall, and suspended sediment flux associated with river discharge, land-use changes and coastal development (e.g., McCulloch *et al.*, 2003; Montaggioni *et al.*, 2006; Carriquiry and Horta-Puga, 2010; Horta-Puga and Carriquiry, 2012). McCulloch *et al.* (2003) indicated that the Ba/Ca ratios in coral skeletons from the Great Barrier Reef increased five to ten-fold after the arrival of Europeans in Australia. They argued that the sediment load from the Burdekin River increased with soil erosion rate due to increasing cattle grazing and agricultural activity around its catchment area and influenced on the coral reef environment. Ba/Ca ratios in coral skeletons have been used as powerful recorder of sediment loading by river discharge in the various coral reefs such as Great Barrier Reef (Jupiter *et al.*, 2008), the Gulf of Mexico (Carriquiry and Horta-Puga, 2010), and Madagascar (Maina *et al.*, 2012). In this paper, we attempted to establish the relationship between Mekong River discharge, flood events and coral Ba/Ca ratios, reconstructed $\delta^{18}\text{O}_{\text{sw}}$ based on a 26 year-old *Porites* coral core from Con Dao Island, Southern Vietnam.

MATERIALS AND METHODS

Coral sampling

From 2 to 8 June 2006, we drilled cores from two *Porites* coral colonies living at 20 m depth on the southwest side of Con Dao Island, Vietnam (N 8°33.43' E 106°33'), which is located 90 km away from the Mekong Delta coast (Fig. 1). The total length of the study core, CD-4, was 150 cm and 5 cm in diameter. The coral cores were cut into 5-mm-thick slabs using a diamond saw blade along the axis of major growth. Each coral slab was ultrasonically cleaned 3 times with deionized water for 10 minutes to remove surface contaminants. Then, the coral slabs were dried in an oven at 40°C for 48 h. The slabs were photographed under UV light and X-ray. The UV and X-ray photographs revealed luminescent bands and density bands on the coral skeletons, respectively (Supplementary Fig. S1). There were 82 bands in total, and all bands were clear. These bands were assumed to be annual growth bands covering the period AD 1924–2005. On the basis of the banding patterns in the UV-luminescence photograph and X-radiograph, measurement lines were determined on the coral slabs for sampling along the skeletal growth axis. A total of 1150 sub-samples were

Fig. 1. (a) Location of the East Sea and Mekong River of Vietnam. The climate of this area is strongly dominated by the East Asian monsoon: summer monsoon and winter monsoon correspond to southwesterly winds and northeasterly winds, respectively. Con Dao Island located in the ESVN, ~90 km from the mouth of the Mekong River. (b) Map of Con Dao Island showing the location of the sampling site at the southwest side of the island (red circle). Meteorological observation station (blue circle) located 15 km distance away from the sampling site.

taken from 0.8 mm diameter drill spots at 1 mm intervals with a drilling depth of 1 mm. In this paper, we have already analyzed 398 sub-samples corresponding to the period from 1980 to 2005.

Oxygen isotope analysis

Coral oxygen isotope ratios were measured with an isotope ratio mass spectrometer (IRMS; Finnigan MAT 253) connected with a carbonate device (Kiel IV) at Hokkaido University. The 20–40 μg carbonate powder

Fig. 2. Seasonal characteristics of (a) Sr/Ca ratios and $\delta^{18}\text{O}_{\text{coral}}$; (b) Calculated $\delta^{18}\text{O}_{\text{seawater}}$ and Ba/Ca ratio and climatological data calculated from monthly data; (c) SST and SSS and (d) Precipitation in the period from 1980 to 2005. Error bars indicate standard error.

was reacted with 100% H_3PO_4 at 70°C , and the emitted CO_2 gas was purified and introduced from the carbonate device to the IRMS. Oxygen isotope ratios are expressed as delta notation relative to Vienna Pee Dee Belemnite. The internal precision for $\delta^{18}\text{O}$ was 0.04‰ using replicate measurements of the NBS-19 standard (1σ , $n = 80$).

Fig. 3. The comparison between monthly averages of calculated (a) $\delta^{18}\text{O}_{\text{seawater}}$ and (b) Ba/Ca ratios during the period from 1980 to 2005, separated into flood years and no-flood years. Error bars indicate standard error.

Trace element analysis

Trace element concentrations in the coral skeletons were measured with inductively coupled plasma-atomic emission spectrometry (ICP-AES; iCAP6200) at Hokkaido University. The sample solution for coral Sr/Ca ratio analysis was prepared using 80–90 μg sample powder dissolved in 0.5 ml HNO_3 25% and milli-Q water, adjusting the Ca concentration to 7 ppm.

For Ba/Ca analysis, the 115–128 μg powder sample was rinsed with 2 ml milli-Q water in an ultrasonic bath for 10 minutes for removing the possible organic contamination. The solution was then centrifuged for 15 minutes and 1.8 ml milli-Q water was removed. The residual solution was mixed with 0.5 ml HNO_3 25% and milli-Q water to adjust the Ca concentration to 9 ppm. The ultrasonic nebulizer was coupled to ICP-AES to detect trace amounts of barium. Measurement accuracy was monitored by analyzing the *Porites* coral standard JCP-1. Analytical precision of the Sr/Ca and Ba/Ca determinations was 0.07% and 1.2%, respectively.

Climate data

The monthly SST, SSS and precipitation (Supplementary Fig. S2) have been recorded since January 1980 at the Con Dao meteorological observation station (Institute of Hydrology and Meteorology of Vietnam), which is located approximately 15 km away from the coral sampling site.

Age model

We used the coral Sr/Ca ratios to create an age model for all proxies. Minima and maxima of the Sr/Ca ratios were tied to the maxima and minima of observed SST from Con Dao Island. The Sr/Ca data and instrumental SST were re-sampled to a monthly resolution with 12 samples per year using AnalySeries software (Paillard *et al.*, 1996).

Data calculation

The seasonal variation of all proxies in period from 1980 to 2005 were calculated by using averaged data of each months during 26 years (Fig. 2). And the standard error was showed as standard deviation values (σ) of the means of each months (Figs. 2 and 3).

RESULTS AND DISCUSSION

Coral proxy calibrations

The coral Sr/Ca ratios and $\delta^{18}\text{O}$ showed 26 distinct annual cycles (Fig. S2). The average of the Sr/Ca ratios was 8.90 (mmol/mol), with values ranging from 8.71 to 9.09 (mmol/mol). The $\delta^{18}\text{O}$ averaged -5.68 (‰VPDB) and ranged from -5.04 to -6.51 (‰VPDB). The calibration of the coral Sr/Ca thermometer was obtained from the linear least squares regression between the coral Sr/Ca maxima (minima) and the SST maxima (minima) at Con Dao station from 1980 to 2005 as below:

$$\begin{aligned} \text{Sr/Ca (mmol/mol)} \\ = (-0.0504 \pm 0.004) * \text{SST (}^{\circ}\text{C)} + (10.305 \pm 0.17) \\ (r^2 = 0.8108, P < 0.01). \end{aligned}$$

The slope of the Sr/Ca - SST regression was -0.0504 ± 0.004 mmol/mol/ $^{\circ}\text{C}$, which lies within the range for slope values, from -0.0424 to -0.0608 mmol/mol/ $^{\circ}\text{C}$, recorded in other *Porites* corals in the East China Sea, South China Sea and East Sea of Vietnam (Supplementary Fig. S3).

We established a regression line between the SST data from Con Dao Island and $\delta^{18}\text{O}$, assuming that $\delta^{18}\text{O}$ reflected only SST variations, as follows:

$$\begin{aligned} \delta^{18}\text{O}_c (\text{‰VPDB}) = -0.2207 * \text{SST (}^{\circ}\text{C)} + 0.4568 \\ (r^2 = 0.8495; P < 0.01). \end{aligned}$$

Coral $\delta^{18}\text{O}$ is influenced by both SST and the $\delta^{18}\text{O}$ in seawater. The mean seasonal variation in SSS was approximately 6‰, with a maximum of 34‰ in February–April and a minimum of 28‰ in October. $\delta^{18}\text{O}_{\text{sw}}$ might be a significant contribution to coral $\delta^{18}\text{O}$. $\delta^{18}\text{O}_{\text{sw}}$ values was calculated from both the coral Sr/Ca ratio and $\delta^{18}\text{O}_c$. To isolate $\delta^{18}\text{O}_{\text{sw}}$, the SST component was removed from the $\delta^{18}\text{O}_c$ values using Sr/Ca-SST estimates (Cahyarini *et al.*, 2008). The average calculated $\delta^{18}\text{O}_{\text{sw}}$ was approximately 0.11 (‰VPDB), and the values ranged between -1.01 to 0.97 ‰. The error of calculated $\delta^{18}\text{O}_{\text{sw}}$ was ± 0.11 ‰ ($\pm \sigma$) (Following Nurhati *et al.*, 2011).

Factors controlling the Ba/Ca ratio

The coral Ba/Ca ratios varied from 2.51 to 8.45 ($\mu\text{mol/mol}$), with an average value of 3.7 ($\mu\text{mol/mol}$). The Ba/Ca ratios also showed seasonal variation with peaks typically occurring between January and March (Fig. S2).

There is still much debate about the possible influence factors on coral Ba/Ca. The main influence factors that can affect the coral Ba/Ca ratio include: (1) SST, (2) seasonal upwelling, (3) “vital effects” including extension rates, and phytoplankton blooms and (4) precipitation associated with river discharge and flood events.

SST has been known to be a thermodynamic driver regulating the incorporation of cations in corals and other marine carbonates. Previous studies reported the correlation between SST and the coral Ba/Ca ratio. Gaetani and Cohen (2006) suggested that barium incorporation into coral skeletons is inversely correlated with SST between 15 and 75 $^{\circ}\text{C}$. Moreover, Chen *et al.* (2011) showed that the maxima of coral Ba/Ca from Daya Bay, northern South China Sea, coincided with the winter minimum SSTs. However, in our study, we compared coral Ba/Ca values with SST from Con Dao Island and found that there was no association with SST ($r = 0.003$, $n = 398$, $P < 0.001$) (Fig. S2). Therefore, our results suggest that SST did not significantly affect coral Ba/Ca ratios.

Previous studies were successful in using coral Ba/Ca ratios as a proxy for seasonal upwelling (Montaggioni *et al.*, 2006). Upwelling is a regular phenomenon during summer (June to September) in the continental shelf of the East Sea in Vietnam (ESVN). Upwelling events along the coast of Vietnam are driven by the southwest monsoon, which causes strong seasonal winds parallel to the coast. The upwelling appears along the coast, ~ 270 km distance from Con Dao Island (Hu and Wang, 2016) (Supplementary Fig. S4). Moreover, our Ba/Ca peaks mostly occurred in cold winters (March) and not in the upwelling season (June to September). Furthermore, there is no evidence showing upwelling can affect the coral in Con Dao Island.

The underlying principle for inferring palaeoenvironmental conditions from coral geochemistry

is the assumption that coral physiology does not pose random influence upon the chemical composition of the skeleton. However, coral geochemistry might be modulated by biological influences (termed as ‘vital effects’) that affect the utility of coral skeleton for environment monitoring purposes. Gaetani and Cohen (2006) indicated “vital effects” accompanying coral growth influences on Ba incorporation in corals. In our core, the coral extension rates show quite stable through the 26 years, on average 12 mm/year with a range between 11 to 14.2 mm. In this study, we compared between annual growth rate and averaged Ba/Ca ratios in each years during period from 1980 to 2005. The result showed the weak positive correlation between them ($r = 0.242$, $n = 26$). Therefore, we assumed that coral growth rates did not significantly affect this coral record. Tang *et al.* (2006) demonstrated the major offshore phytoplankton bloom appear in the Gulf of Thailand during the winter northeast monsoon season and in ESVN during the summer southwest monsoon season. Phytoplankton blooms will combine with Mekong River discharge and influence on coral around our study area. Although complicated factors may influenced on coral Ba/Ca proxy, average seasonal profile of Ba/Ca ratios from 26 year records would eliminate or minimize noise due to other controlling factors.

In the coastal area, barium in coral skeletons mainly has been used as a record of terrestrial runoff (Prouty *et al.*, 2010). Sediments derived from Mekong Delta soils with a high rate of weathering and biological activities are usually rich in Ba. When these sediments are transported by river into coastal environments, Ba is desorbed from the sedimentary particles and released into the dissolved phase of seawater. Thus, the Ba/Ca ratios from coral in coastal areas can be used as a proxy for river discharge. In addition, the volume of freshwater flow from the mainland to coastal areas is highly dependent on rainfall, and the levels of coastal Ba increase with increases in river outflow, meaning that the coral Ba/Ca record could be used as a tracer of precipitation. Sinclair and McCulloch (2004) suggested that peaks in Ba/Ca ratios from coral in the Great Barrier Reef coincided with observed river flood plumes and local rainfall. Since Con Dao Island is close to the mouth of the Mekong River and Unverricht *et al.* (2014) show that Con Dao Island is strongly influenced by the Mekong River discharge, which causes high suspended sediment concentrations and phytoplankton blooms around the island (Tang *et al.*, 2006). Our Ba/Ca profiles showed the variation corresponding the seasonal discharge from Mekong River therefore Ba/Ca variation in the coral skeletons is also predominately controlled by terrestrial inputs from the Mekong River Delta and reflects the suspended sediment associated with Mekong River discharge.

Seasonal characteristics of coral geochemical records

We calculated the seasonal variation of all proxies in the period 1980–2005 and compared it with climatological data from Con Dao Island (Fig. 2). Sr/Ca ratios show maxima and minima in May and January corresponding to observed SST. The maximum of $\delta^{18}\text{O}_{\text{sw}}$ was determined to occur in May when SST and SSS are at a maximum, and the minimum of $\delta^{18}\text{O}_{\text{sw}}$ occurred in the rainy season because of the high amount of precipitation or freshwater input from the Mekong River. We can see the appearance of two peaks in the annual variation of the coral Ba/Ca ratio. The higher peak of $4.24 \mu\text{mol/mol}$ (± 1.25) occurred in March (winter), and the lower one of $3.72 \mu\text{mol/mol}$ (± 0.67) occurred in August (summer) when the precipitation was the highest.

Both of the peaks in the Ba/Ca profile could reflect the river sediment discharge corresponding to the cool/dry season and warm/wet season, respectively (McCulloch *et al.*, 2003). However, the peak in August was lower than the one in March, which means that although the river discharge reached a maximum in summer, the effect of the suspended sediment on Con Dao Island’s corals was not significant. The Ba/Ca ratios calculated in this research were compared with the annual river discharge recorded at the Tan Chau station (N $10^{\circ}48'$, E $105^{\circ}13'$) on the Mekong River and precipitation recorded at Con Dao Island meteorological station during the period from 1980 to 2005. There was no correlation between them with $r^2 = 0.0252$ and $r^2 = 0.0214$, respectively. This suggested that river discharge and local rainfall did not have a direct influence on Con Dao Island corals. To explain the difference between the effect of suspended sediment associated with river discharge, we considered the seasonal sediment transport model of the Mekong River related to the East Asian monsoon as follows (Supplementary Fig. S5): during the high flow season (May–October), a considerable part of riverine sediment was delivered to the Mekong River mouth and temporally deposited there; then during the low flow season (November–April), the previously deposited sediment was resuspended by strong mixings associated with the strong northeast monsoon and subsequently transported southwest along-shelf by coastal circulation (Hein *et al.*, 2013). The monsoon wind had an important role in changing the surface water current and sediment transportation in this area. The amount of suspended sediment effect on Con Dao Island in winter is higher than in summer. This is consistent with the results of our determined coral Ba/Ca ratio from Con Dao Island.

Influence of flood events related to Mekong River discharge and East Asian monsoon on coral records

Ba/Ca and $\delta^{18}\text{O}_{\text{sw}}$ could also be used to reconstruct past flood events. We investigated historical documents

about flood history in the Mekong Delta. During 1980–2005, seven large flood events were found in 1984, 1991, 1994, 1996, 2000, 2001 and 2002 (Tran, 2009). We calculated the seasonal variation of Ba/Ca and $\delta^{18}\text{O}_{\text{sw}}$ separately in flood years and no-flood years (Fig. 3). We found a significant increase of the Ba/Ca ratios and a decrease of $\delta^{18}\text{O}_{\text{sw}}$ in the warm/wet season in flood years. These seasonal characteristics correspond to larger amounts of freshwater discharge from the Mekong River when a flood occurred or a high amount of precipitation in the warm/wet season related to the East Asian summer monsoon. Our coral Ba/Ca and $\delta^{18}\text{O}_{\text{sw}}$ from Con Dao Island could be used as a possible proxy for capturing the past flood events in the Mekong Delta.

CONCLUSIONS

Our results suggested that the Mekong River discharge was influenced by the seasonal migration of the Asian monsoon and ocean currents. The maximum discharge of freshwater from the river reached our coral site in summer (from May to November), however the sediment discharge model of the Mekong River indicated the amount of suspended sediment influencing on Con Dao Island was high in winter (from December to April). Coral Ba/Ca record in Con Dao Island reflected the sediment discharge from the Mekong River. On the other hand, Ba/Ca and $\delta^{18}\text{O}_{\text{sw}}$ could be used as indicators of flooding from the Mekong Delta. During the period from 1980 to 2005, the difference in seasonal characteristics of geochemical signals in flood years and no-flood years was detected. During the flood years, in the warm/wet season, the Ba/Ca ratio and $\delta^{18}\text{O}_{\text{sw}}$ data significantly increased and decreased, respectively. These results reflect the increase in the Mekong River freshwater discharge and the sudden increase of precipitation when floods occurred in warm/wet season.

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SUPPLEMENTARY MATERIALS

URL (<http://www.terrapub.co.jp/journals/GJ/archives/data/53/MS552.pdf>)
 Figures S1 to S5
 Table S1