Quaternary International 392 (2016) 3–12

Contents lists available at ScienceDirect

Quaternary International

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

High resolution geochemical investigation of the bivalve shells (*Glycymeris* sp.) from shell mounds in Jeju Island, Korea: Late Holocene paleoclimatic implications related to East Asian Monsoon climate

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A R T I C L E I N F O

Article history:
Available online 15 August 2015

Keywords:
East Asian Monsoon
Holocene
Bivalve
Stable isotope
Trace element
Jeju Island

A B S T R A C T

Recent and shell mound bivalves (*Glycymeris* sp.) were collected from two sites (Sangmori and Udo, Jeju Island) to investigate paleoceanographic conditions using high resolution stable isotope and trace element data. The radiocarbon ages are about 3210 ± 50 and 4210 ± 60 BP for the Sangmori and Udo shell mounds, respectively. Both oxygen and carbon isotopic compositions of the shell mound bivalve shells show clear seasonal cycles with depleted values during summer and the coeval trend, which indicates that they were controlled by freshwater input. Similar ranges of both isotopes indicate that shallow seawater conditions near the Sangmori and Udo shell mounds are more or less similar. However, more negative oxygen and carbon isotope values of the shell mound bivalves than the recent counterpart suggest that surface seawater salinity near Jeju Island was lower due to the increase in Changjiang River Discharge. Mg contents of the recent bivalve reflect surface seawater temperature changes whereas Sr and Ba contents were controlled by nutrient concentrations directly influenced by Changjiang River Discharge. Geochemical data of the recent and shell mound bivalves may imply higher precipitation (more intense East Asian Summer Monsoon activity) at ~3.2 and 4.2 ka under the influence of the terminal stage of Holocene climatic optimum in south China or may reflect smaller fluctuating peaks during its decline.

1. Introduction

Recent temperature due to global warming is believed to induce severe weather changes such as heat sweltering and heavy snowfall, all of which resulted in significant social and economic damages past a few decades. Because of unpredictable weather changes, future climate change related to global warming has been a main global science subject. Paleoclimate results have been regarded as one of the most effective information to demonstrate the credibility of climate modeling. Thus centennial to millennial climate variations during the Holocene have been intensively investigated using various paleoclimate proxies such as ice cores, pelagic and lacustrine sediments, loess, tree rings, corals and speleothems (e.g., Beck et al., 1992; Jo et al., 2014). Cyclic climate changes during the Holocene have been reported and it was suggested that there were 7–8 intervals which were warmer or colder than present during the Holocene (Jian et al., 2000; Zhang et al., 2000; Wang et al., 2005). One of the most common methods to understand paleoceanographic changes is to use the geochemical data of planktonic and benthic foraminifera from pelagic sediments (e.g., Duplessy et al., 1970). Ice cores also have contributed significantly to understand atmospheric conditions (Siegenthaler et al., 2005). However, recently speleothems have been widely used (Lachniet, 2009; Jo et al., 2014) because they can also provide high resolution data such as annual changes (Baker et al., 1993). For this purpose, calcareous bivalves were investigated using growth laminae, oxygen and carbon isotopic compositions and trace elemental compositions to understand the seasonality as well as physico-chemical conditions of shallow marine environments (Andrus and Crowe, 2000; Davis, 2001; Watanabe et al., 2004; Stephens et al., 2008; Ford et al., 2010).

Shell mounds is known to be produced during the Neolithic age (since about 10,000 years ago) (Suzuki, 1989), and numerous

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http://dx.doi.org/10.1016/j.quaint.2015.07.050
1040-6182/

bivalve shells are commonly discovered at the sites along ancient shorelines. Due to relatively young ages, these shells have undergone little diagenetic alteration, hence original mineralogy, texture and their original geochemical compositions tend to be preserved, reflecting paleo-seawater conditions during growth (e.g., Schöne et al., 2011). Numerous shell mounds are distributed along the western and southern coasts of the Korean Peninsula and around Jeju Island (Korean Cultural Properties Investigation & Research Institute Association, 2010). This study has special advantages because very near-surface seawater conditions can be investigated due to well known habitats of shell mound bivalves and the living age of the shells can be well constrained. However, paleoclimatic research using shell mound bivalves near the Korean Peninsula has not been carried out yet. Therefore, the objective of this study is to delineate paleoclimatic conditions near Jeju Island from two late Holocene shell mound sites (Sangmori and Udo shell mounds). High resolution oxygen and carbon isotopic and trace elemental compositions of the shell mound as well as recent bivalves of a single taxon were analyzed for this purpose.

2. Climatic and oceanographic setting

Located at the middle latitudes between about 33 and 43° N in the northeast margin of Asia, the Korean peninsula and Jeju Island are strongly affected by the East Asian Monsoon system with pronounced seasonal fluctuations in humidity, temperature, and atmospheric circulation. A low-pressure cell above the central Eurasian continent during summer is associated with southwesterly winds that carry warm and moist air masses from the northwestern Pacific Ocean towards Japan and the Korean peninsula. During winter seasons, reversed pressure gradients with prevailing northwesterly winds carry dry and cool air masses from the Eurasian continent towards the Pacific Ocean. These prevailing tropospheric westerly winds of winter also contribute to cool to cold climate conditions in Korea (Jo et al., 2010).

Shallow marine seawater near Jeju Island is mostly influenced by the Taiwan Warm Current which is a branch of the Kuroshio Current and Changjiang River discharge (Beardsley et al., 1985; Chen et al., 1994). About 80% of freshwater input into East China Sea and Yellow Sea is contributed by the freshwater discharge from Changjiang River and their annual and seasonal contributions are quite variable. Their seasonal pathways can be changed, i.e., showing a narrow pathway southward along the coast from the river mouth and flowing and dispersing northeast toward Jeju Island (Beardsley et al., 1985). The Changjiang Diluted Water (CDW) which is a mixture between the Changjiang River discharge and the East China Sea seawater moves and dispersed east from June when southwesterly wind begins to prevail, forming the isolated watermass of 10–20 m in water depth with less than 30 psu in salinity (Lie et al., 2003). Sometimes extremely low saline water with less than 20 psu can be observed west of Jeju Island (Kim et al., 1998). The CDW with high temperature and low salinity usually expands northeast toward Jeju Island by seasonal winds. Due to clear density difference of the CDW from underlying the East China Sea watermass, it flows on the surface without vertical mixing. As a result, the separate watermass forms with the increase in sea surface temperature (SST), which is quite distinct from the relatively colder underlying watermass (Park et al., 2011).

3. Methods

The bivalve shells sampled for this study are the same genus (*Glycymeris* sp.) with round shape and thick shells which dwell in sands at the water depth of 5–20 m (Gwon et al., 1993). All the shells were collected from Jeju Island (Fig. 1a). The Sangmori Shell Mound is located about 300 m toward inland from the southwestern coast of Jeju Island (Fig. 1b). The bivalve shell (JSM-B01, *the Sangmori bivalve* herein) was excavated from sediments after removing vegetation on the surface and unearthing the 50 cm-thick soft sediments. The Udo Shell Mound is located about 100 m toward inland from the western coast of Udo Island, which is about 3.5 km east of Jeju Island (Fig. 1c). The bivalve shell (JUD-B01, *the Udo bivalve herein*) was sampled from a widespread sea shells in the Udo shell mound site and the best preserved shell as a whole skeleton was chosen. The recent bivalve (JUD-R-B01, *the Udo recent bivalve* herein) was collected from the Hagosudong Beach near the Udo Shell Mound by scuba diving (Fig. 1c). However, the bivalve was not alive, thus the exact age of the shell is not known. Collected shells were brushed and cleaned with water and organic matter was removed using 15% H_2O_2 for 24 h. The samples were additionally cleaned with ultrasonic water and dried in air. Before microdrilling the outermost organic layer (periostracum) of the recent shell was removed by polishing with sandpaper. After that, the shell was reacted with 15% H_2O_2 for 48 h to move residual organic matter. The sample was then washed and air-dried.

Seawater monitoring data of the Korea Hydrographic and Oceanographic Administration were used (KHOA, http://info.khoa.go.kr), and the amount of precipitation was used from the data by the Korea Meteorological Administration (KMA, http://www.kma.go.kr/weather/observation). SST and sea surface salinity (SSS) at two sites were monitored from January of 2003 to August of 2011 (Fig. 1). The monitoring site by KHOA is located in Hamo-ri, Daejeong-eup, Seoguipo City (33°13′N, 126°14′E) near the Sangmori Shell Mound. It was reported that the averaged SSTs in winter and summer are 14.4 °C and 23.8 °C, respectively, showing the seasonal variation of 9.4 C° (Fig. 2a). There is a distinctive seasonal salinity variation from ~32.9 psu during winter to ~28.2 psu during summer. The monitoring station near the Udo Shell Mound is near the coast in Ojori, Seongsan-eup, Seoguipo City (33°28′N, 126°55′E). The averaged SSTs in winter and summer are 11.8 °C and 24.1 °C, showing the seasonal range of 12.3 C° (Fig. 2b). There are distinctive seasonal salinity variations from ~34.0 psu during winter to ~31.3 psu during summer. The salinity values near Udo Island are higher than those near the Sangmori coast. The amount of precipitation near two studied sites show the typical rainfall trend of East Asian Monsoon (EAM) system, showing the concentrated amount of rainfall during summer. The daily amount of precipitation near the Udo Shell Mound monitored by the Seongsan Meteorological Station of KMA is about twice higher than that of the Sangmori Shell Mound by the Gosan Meteorological Station of KMA.

The mineralogy of the shells were determined by X-ray diffractometer and the ages of two shells collected from shell mounds were determined by radiocarbon dating at the Geochemical Analysis Center, Korea Institute of Geosciences and Mineral Resources. Microsamples were drilled out along the longest axis of the bivalves using a micromill for stable isotope analyses of carbon and oxygen. About 300–400 μg of microsamples was analyzed for stable isotopes at Leibniz Laboratory using the stable isotope mass spectrometer (Finningan MAT 253). Analytical error range for both isotopes is ±0.01‰. All the values for carbonate samples reported here are relative to VPDB (Vienna Pee Dee Belemnite).

High resolution analysis for trace element compositions of the Udo recent bivalve were carried out using the UV Laser Ablation System (213 nm Nd-YAG laser, New Wave UP213) connected to Laser Ablation Inductively Coupled Plasma Mass Spectrometer (LA-ICP-MS, Thermo Scientific Company X7). For the analysis, the bivalve shell was impregnated with epoxy resin for 24 h and cut into half. The impregnated slab was polished and trace elements were measured at the spacing of every 165 μm NIST612 was used