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Past 140-year environmental record in the northern South China Sea: Evidence from coral skeletal trace metal variations



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1. Introduction

Coral reefs are important parts of the ocean ecosystems. Because of biological mineralization, corals exhibit seasonal characteristics of carbonate deposits as speleothems (Cantillana et al., 1986; Couchoud et al., 2009). This is the reason why coral can become a chronometer (Al-Horani et al., 2003; Knutson et al., 1972). Corals can record environmental changes in the ocean, especially near the coast. Research on geochemical behavior of trace metals in coral skeletons, has been carried out since the 1970s (Banner, 1974; Johannes, 1975; St. John, 1974), and it was proposed that human activities have great impacts on the coral ecosystems. For example, in the case of heavy metals in the coral reefs of Hawaii (Banner, 1974), it was found that except for erosion of the coast, agriculture and sewage discharged carries high concentrations of nutrient elements into the ocean, therefore seawater pH was reduced and heavy metals were mobilized. Ocean environmental changes were always recorded in growth bands of coral skeletons through geochemical behavior of elements between seawater and corals. So

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ABSTRACT

About 140-year changes in the trace metals in *Porites* coral samples from two locations in the northern South China Sea were investigated. Results of PCA analyses suggest that near the coast, terrestrial input impacted behavior of trace metals by 28.4%, impact of Sea Surface Temperature (SST) was 19.0%, contribution of war and infrastructure were 14.4% and 15.6% respectively. But for a location in the open sea, contribution of War and SST reached 33.2% and 16.5%, while activities of infrastructure and guano exploration reached 13.2% and 14.7%. While the spatiotemporal change model of Cu, Cd and Pb in seawater of the north area of South China Sea during 1986–1997 were reconstructed. It was found that in the sea area Cu and Cd contaminations were distributed near the coast while areas around Sanya, Hainan had high Pb levels because of the well-developed tourism related activities.

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it is possible that ocean environment is reconstructed by measuring elemental changes in coral skeletons (Delaney et al., 1993; Linn et al., 1990; Shen and Boyle, 1987, 1988; Shen et al., 1987).

With research going on around the world (Ali et al., 2011; Dodge and Gilbert, 1984; Guzman and Jarvis, 1996; Linn et al., 1990; Mitchelmore et al., 2007; Reichelt-Brushett and McOrist, 2003; Rosales-Hoz et al., 2009; Shen and Boyle, 1988), various trace metals in corals were comprehensively studied. For example, besides its use as a signal for indicating upwelling (Lea et al., 1989), Ba could be an indicator of contamination by land use (Prouty et al., 2010) and flood events (Sinclair, 2005). Likewise, Cu, Zn and Pb are indicators of industrial and mining contamination in the ocean (David, 2003; Fallon et al., 2002; Kelly et al., 2009; Shen and Boyle, 1987). On the other hand, quantitative research on geochemical distribution behavior of trace metals between seawater and corals has been carried out but there are limited reports of qualitative studies. The quantitative research on distribution coefficients is mainly based on the chemical equilibrium of Ca²⁺ and other trace metals in the ocean during CaCO₃ deposit in coral skeleton (Shen and Boyle, 1987). The distribution coefficients could be evaluated using the ratio of trace metals to Ca both in coral and seawater. According to experiments, a homogeneous geochemical behavior with distribution coefficient K_D of ~ 1.0 for most elements in most



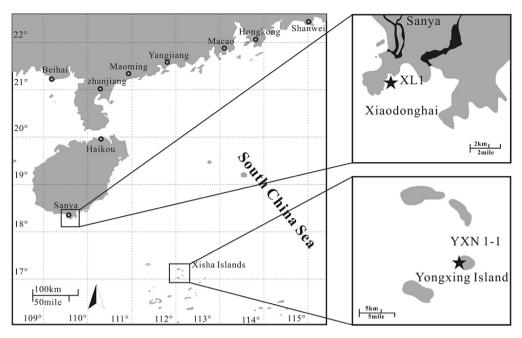


Fig. 1. North area of South China Sea and distribution of coral samples locations.

corals were found (Livingston and Thompson, 1971; Reuer et al., 2003). But in some cases, it is found that the distribution coefficient of Pb could reach ~2.3 (Linn et al., 1990; Shen and Boyle, 1987) while Cu reached ~0.3 (Linn et al., 1990; Livingston and Thompson, 1971). The results of these experiments supply a feasible and possible method to help reconstruct, understand and evaluate changes in trace metal concentrations in the seawater environment.

South China Sea is the biggest marginal sea of China covering tropics and subtropics, in which most corals are distributed. The research on the trace metals in coral of South China Sea which focused on climate change and paleoclimate reconstruction started in the early 2000s (Wei et al., 2000; Yu et al., 2002b, 2004, 2005). However, it is noteworthy that some studies on trace metals contamination in coral were also reported (Chen et al., 2010; Cheng et al., 2005; Huang et al., 2003; Peng et al., 2006; Yu et al., 2002a). These studies focus on short-term change in trace metals in coral (<50 years), and can potentially supply database for ocean environmental studies. By contrast, the current research focuses on the coral reef close to the mainland where anthropogenic activities are frequent and have great impacts on the ocean environment. Data on coral reefs, which are far away from the mainland, is rarely reported. But in a recent survey, it was found that the sea area far away from mainland appeared to be polluted (Gao et al., 2008). So the contamination of trace metals in coral needs more attention and requires further studies.

To study the long-term geochemical behavioral characteristics of trace metals in corals from the northern area of the South China Sea over the past century, we investigated the temporal changes of concentrations of some potentially harmful trace metals. Through PCA analysis with Varimax rotation, the trace metals in the coral of South China Sea were grouped according to their potential sources. Projection of principal components was used to plot out the events or activities impacting geochemical behaviors of trace metals during the past century. The contribution of these events or activities was calculated to evaluate the characteristics of geochemical behaviors of trace metals in different locations of the northern area of the South China Sea. Using data from other locations, a spatiotemporal change of trace metals in seawater in different years was reconstructed. This provides valuable data for the understanding of the potential sources of trace metals and the formulation of strategies for historical reconstruction of regional ocean environments of the South China Sea, and other tropics and subtropics ocean worldwide.

2. Materials and methods

2.1. Sample collection

Living *Porites* coral samples was collected from Xiaodonghai, Sanya city, southern Hainan Island and Yongxing Island of Xisha Islands during May 2006 and June 2008, and labeled as XL1 and YXN 1-1, respectively (Fig. 1). The coral samples were washed with freshwater; then sectioned using a water-lubricated diamond-bit masonry saw in order to obtain a set of parallel slabs that are ~8 mm thick. Dry coral slab was X-radiographed to show the annual growth bands which displayed growth procedure of coral aragonite (Fig. 2). The coral slabs were soaked and sterilized by 10% H₂O₂ for 48 h, and cleaned 3 times in an ultrasonic bath using Milli-Q water, and then air-dried in the oven at 60 °C for 48 h (Chen et al., 2010).

Annual-resolution sub-samples were sliced continuously using a ceramic knife along the growth bands of coral slabs and then ground to powder for measurement. A clear pattern of alternating bands of high and low density is visible in Fig. 2. Dating was accomplished by counting these annual bands (Knutson et al., 1972). As shown in the X-radiography of the coral slabs (Fig. 2), the length of each sub-sample depended on the thickness of each annual growth band sampled. Sample XL1 and YXN 1-1 contained coral skeletons growing between 1870–2006 and 1871–2008, covering 137 and 138 years record, respectively. Thus, a total of 137 sub-samples from sample XL1 and 138 sub-samples from YXN 1-1 were collected for analyses.

2.2. Geochemical analysis

Trace and major elements of samples, including Cr, Mn, Ni, Cu, Zn, Cd, Ba, Pb, Sr, U and Ca, were measured using a Thermo X-series II Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) at the Radiogenic Isotope Facility, the University of Queensland. In this work, only acid cleaned bottles and vials were used and ultra high purity water and nitric acid (70%, w/w) were prepared by sub-boiling distillation. Then, 2% HNO₃ was prepared for both preparation of standard solutions and sample digestions. Spiked 2% HNO₃ stock solution of 60 ppb was prepared with internal standard isotopes ⁶Li, ⁶¹Ni, ¹⁰³Rh, ¹¹⁵In ¹⁸⁷Re, ²⁰⁹Bi and ²³⁵U to correct for matrix effects of Ca and instrumental drift. Certified geochemical reference materials W-2, JCp-1 and BIR-1 were prepared as external standards, adding 60 ppb spiked solution and diluting to 6 ppb using 2% HNO₃. Analyzed data were assessed for accuracy and precision using quality assurance and quality control (QA/QC) program, which included reagent blanks, duplicat test, and certified geochemical reference materials (W-2, JCp-1 and BIR-1) with deviation <5%.

Small quantities (\sim 50 mg) of dried sub-samples were ground and mixed completely to make each sub-sample homogenous. Then \sim 2.5 $-\sim$ 3.0 mg of sub-samples were weighed into LDPE tubes and dissolved using 10 mL 6 ppb spiked

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