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Coral trace metal of natural and anthropogenic influences in the northern South China Sea



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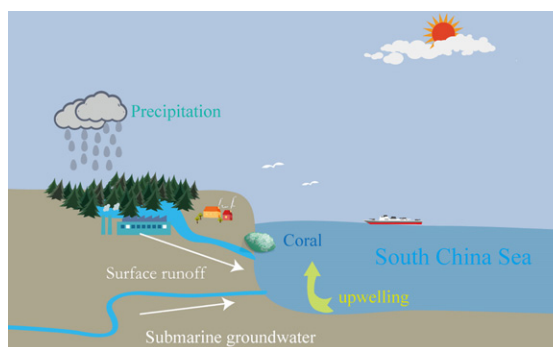
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HIGHLIGHTS

- Changes of trace metals in coral near Hainan Island are impacted by human activities.
- All coral records of trace metals are impacted by terrestrial inputs except Sr and U.
- Ba and REE in coastal seawater are dominated by the precipitation.

GRAPHICAL ABSTRACT



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ABSTRACT

The composition and concentrations of trace metals in coastal seawater have changed in parallel with variations in geochemical processes, climate and anthropogenic activities. To evaluate the response of trace metals in coastal seawater to climatic changes and human disturbances, we report annual-resolution trace element data for a Porites coral core covering ~100 years of continuous growth from a fringing reef in Xiaodonghai Bay in the northern South China Sea. The results suggested that the trace metal contents in the coral skeleton demonstrated decadal to interdecadal fluctuations with several large or small peaks in certain years with remarkable environmental significances. All of the trace metals in coastal surface seawater, especially Cr and Pb (related to industrial or traffic emissions), were impacted by terrestrial inputs, except for Sr and U, which were impacted by the surface seawater temperature (SST). Moreover, Mn, Ni, Fe and Co were also contributed by weapons and military supplies during wars, and Cu, Cd and Zn were further impacted by upwelling associated with their biogeochemical cycles. Ba and rare earth element (REE) in coastal surface seawater were dominated by runoff and groundwater discharge associated with precipitation. This study provided the potential for some trace metals (e.g., REE, Ba, Cu, Cd, and Zn) in coral skeletons to be used as proxies of natural (e.g., upwelling and precipitation) and anthropogenic (e.g., war and coastal construction) variability of seawater chemistry to enable the reconstruction of environmental and climatic changes through time.

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1. Introduction

Numerous archives consider that the trace metal composition and abundance of coastal seawater have changed in parallel with variations in geochemical processes, climate and anthropogenic activities (Chen et al., 2015; Fallon et al., 2002; Gothmann et al., 2015; Nguyen et al., 2013; Saha et al., 2016; Song et al., 2014; Sun et al., 2016). In these studies, measurements of trace metals in seawater or their proxies have provided valuable information on the environmental conditions of oceans and anthropogenic inputs of contaminants. Therefore, a thorough knowledge of the trace metal variations in coastal seawater can provide valuable insight into the interaction between major domains in the atmosphere-lithosphere-ocean system (Della Porta et al., 2015). However, understanding the natural variability of trace metals in seawater and its environmental significances require long term and geographically distributed records. A number of studies on the spatial distributions of trace metals in seawater have revealed that biological, physical, geochemical, and/or oceanic processes play critical roles in the composition and abundance of trace metals in seawater (Akagi et al., 2004; Chen et al., 2015). Though recent years have seen significant improvement in our understanding of the trace metal temporal variations in seawater (Chen et al., 2015; Liu et al., 2011; Nguyen et al., 2013; Song et al., 2014; Sun et al., 2016), the correlative work is still restricted to the trace element type (mainly trace metals, rarely rare earth element (REE)) or the length of time (mainly <20 years). Comparison studies between corals (same species) from similar locations remain sparse.

Trace metals can be incorporated into coral's aragonite lattice in proportion to their abundances in ambient seawater, and annually banded coral skeletons have been confirmed as a unique archive of trace metal temporal variations in surface seawater (Howard and Brown, 1984; Shen, 1986; Shen and Boyle, 1988). For their sensitivity to environmental changes, longevity and wide distribution in the tropical oceans (Gagan et al., 2000; Yu et al., 2005), corals can provide an excellent source of high-resolution geochemical records (Sadler et al., 2014; Yu, 2012). Corals, especially *Porites lutea*, have been widely applied as the recording materials for variability of trace metals at seasonal, inter-annual, decadal, and even secular timescales (Chen et al., 2010, 2015; Fallon et al., 2002; Nguyen et al., 2013; Song et al., 2014; Sun et al., 2016). Fallon et al. (2002), Chen et al. (2010), and Nguyen et al. (2013) discussed the impact on coral trace metal signatures from anthropogenic activities in Misima Island, Daya Bay, and Nha Trang Bay within 19, 14, and 14 years, respectively. Song et al. (2014) investigated changes over the past 140 years in 10 trace metals in coastal seawater, and quantified the contribution from different sources. Chen et al. (2015) investigated the interannual variation of Cu, Mn and V abundances in coral over 159 years and its relationship with the Pacific Decadal Oscillation (PDO) and East Asian Summer Monsoon (EASM). Sun et al. (2016) was the first to report that Hg coral records reveal the primary Hg emission sources (modern wars) during the past two centuries. Therefore, the long-term high-resolution records of trace metals provided by corals offer the potential to reveal the evolutionary history of seawater chemistry, which could benefit our knowledge of source identifications, geochemical behaviors, and probable environmental significances of trace metals in seawater.

In the present study, a long-lived *Porites* coral (1910–2006) in the northern South China Sea (SCS) was collected to investigate the temporal variations of lattice-bound trace metals in coral skeletons over the past century. We measured the annual contents of 28 trace metals in the coral skeletons instead of seawater samples to present the time series characteristics of trace metals in seawater. Through principal component analysis (PCA), trace metals in the coral were grouped according to their potential sources and geochemical behavioral characteristics in seawater. The environmental significance of trace metals in seawater could therefore be identified by the comparison between responses of trace metal composition and abundance and well-established instrumental data or reliable proxies. The main objectives of this study are

to (1) examine the reliability of coral geochemical proxies through comparison between two corals from similar locations, (2) evaluate the responses of trace metals in coastal seawater to natural processes and anthropogenic perturbations, and (3) reveal the possible environmental significance of trace metals in coastal seawater based on their time series characteristics.

2. Materials and methods

2.1. Sample collection and chronology

Xiaodonghai Bay (18° 37'N, 109° 30'E) is located on the east side of Sanya, Hainan Island, in the northern South China Sea (Fig. 1), characterized by a tropical oceanic monsoon climate. The PDO and EASM are dominant in the climatic changes of this area, and the average annual precipitation reaches ~1337 mm, with 90% occurring in the rainy season. Instrumental records from Yinggehai Ocean Observatory at Sanya indicate that the annual mean of monthly sea surface temperature (SST) is 27 °C. As part of the Sanya National Nature Reserve, Xiaodonghai Bay has suffered little disruption from local anthropogenic activities since 1990. Sanya City, adjacent to Xiaodonghai Bay, has undergone a significant boom period over recent decades. The reef coral community near Sanya has shifted from a flourishing state to a comparatively barren state during the past half century (Zhao et al., 2012). In Hainan Island, karst landforms are mainly distributed in the west and south, including Sanya, which is famous for the Luobi Cave as a tourist resort. Large submarine groundwater discharge has been observed in this coastal region (Wang et al., 2014). There is no record of volcanic eruptions around Hainan Island in the past century.

The core of a living and healthy colony of the hermatypic scleractinian coral *Porites lutea* labeled as XL2 was drilled (18° 12'N, 109° 30'E) at a water depth of 5 m in Xiaodonghai Bay in May 2006. After washed with fresh water, slabs of 8-mm thickness were cut from the core with a water-lubricated diamond-bit masonry saw, followed by X-ray radiography. To remove surface contaminants and organic matter, the coral slabs were soaked in and sterilized by 10% H₂O₂ for 48 h, cleaned 3 times in an ultrasonic bath using Milli-Q water, and then air-dried in the oven at 40 °C for 48 h. Knutson et al. (1972) confirmed that each high- and low-density band constitutes an annual couplet, generally representing 1 year of growth. An annual time series for the coral core was established along the major growth axis based on the annual density bands from X-ray images (Fig. S1 in the Supplementary materials). The sample contained coral skeletons growing from 1910 to 2006 covering ~100 years record, with the outermost band identified as the ongoing growth in 2006. Before collecting the coral samples, preliminary milling along the designated sample track should be undertaken to remove approximately 1 mm from the upper surface. Fine powder should be milled from 2-mm square-section grooves continuously along the maximum growth axis, homogenized to avoid any seasonal signatures and to obtain the annual subsamples for element analysis.

2.2. Geochemical analysis

All the lab works were accomplished at the Radiogenic Isotope Facility, the University of Queensland, Australia. Randomly chosen 2.5–3.0-mg samples from the completely ground and mixed samples (~50 mg) were weighed into LDPE tubes and dissolved using 10 mL 6-ppb spiked 2% HNO₃ solution. A spiked 2% HNO₃ stock solution of 60 ppb was prepared with internal standard isotopes ⁶Li, ⁶¹Ni, ¹⁰³Rh, ¹¹⁵In and ¹⁸⁷Re to correct for matrix effects of calcium (Ca) and instrumental drift. The certified geochemical reference materials W-2 and BIR-1 from the United States Geological Survey (USGS) and JCP-1 from the Geological Survey of Japan (GSJ) were prepared (W-2 was a calibration standard, and JCP-1 and BIR-1 standards were used for cross-checking the reproducibility), adding 60 ppb solution and

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