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Sedimentary records of recent anthropogenic eutrophication and metal contamination in Zhelin Bay, an important mariculture area in Southern China

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ABSTRACT

Dinoflagellate cysts (dinocysts), biogenic elements and metals were analyzed from sediment cores collected from Zhelin Bay of the South China Sea in December 2008 to understand the environmental changes over the past 50 years. Dinocyst concentrations ranged from 0 cysts/g to 770 cysts/g, and they were dominated by heterotrophic taxa. There was a clear increase trend upcore for biogenic elements, except for biogenic silica. Metals originated from both the lithogenic source and human activities, and significantly increased after 1985–1995. Environmental changes in the past 50 years can be divided into three stages: (1) before 1985, during which biogenic elements, cyst flux and metals were low; (2) from 1985 to the early 2000s, characterized by an obvious increase of dinocysts, TOC, TN and most metals, while TOC/TN and BSi decreased; and (3) after 2000, the period of rapid increase of dinocysts, TOC and TN but decrease of certain contamination metals.

Eutrophication and metal contamination have become a serious threat in Chinese coastal regions, which are undergoing rapid economic growth and population growth (Dai et al., 2007; Ip et al., 2007; Hu et al., 2008; Shi et al., 2010; Guo and Yang, 2016). Guangdong Province is located in the southeast area of China, and it is one of the fastest developing areas in China. Zhelin Bay is an important mariculture sea area in Guangdong Province. More than half of the water area in Zhelin Bay is now occupied by mariculture farms (Wang et al., 2008). Dense mariculture and intensive human activities resulted in high nutrient levels and metal contamination (Huang et al., 2004; Wang et al., 2013; Gu et al., 2014). However, there were few studies regarding the sedimentary records in core samples in the bay (Gu and Lin, 2016), and none of these studies examined dinoflagellate cysts (dinocysts).

In this study, dinocysts, metals and biogenic elements, including total organic carbon (TOC), total nitrogen (TN), total phosphorus (TP) and biogenic silica (BSi), were measured in sediment cores from Zhelin Bay. A correlation analysis (CA) and a principal component analysis (PCA) were used to define the potential sources of metals, and a cluster analysis was used to group different time periods based on these sedimentary parameters. The purposes of this study were to (1) reconstruct the eutrophication and metal contamination history in Zhelin Bay and (2) understand the relationships between environmental changes and anthropogenic activities.

In December 2008, triplicate sediment cores with lengths of 167 to 186 cm were collected at station Z2 (Fig. 1) by a diver using a steel pipe. After collection, cores were extruded and immediately sectioned into 2-cm intervals in situ. Samples with the same depth were mixed together and separated into two subsamples. One subsample was stored at 4 °C in the dark for dinocyst observation, and the other subsample was stored at −20 °C for the analyses of biogenic elements and metals.

Sediments (5 cm^3) were weighed and sieved through 125- and 20-μm meshes successively after sonication. The slurry remaining on the 20-μm mesh was used for cyst observation. Another 5 g of samples were used to measure the water content of the sediment. Cysts were counted with an inverted light microscope (Leica DMIRB, Leica Microsystems, Germany) at 400× magnification. At least 100 cysts were counted for each sample. In samples with few cysts, all of the treated samples were observed. Cysts in per gram of dry sediment (cysts/g) and per cube centimeter (cysts/cm^3) were calculated. The cyst fluxes were calculated by dividing the cyst concentration (cysts/cm^3) by the accumulation rate (cm/year).

Sediments for the Hg analysis were freeze-dried, and others were dried at 40 °C until reaching a constant weight. The dried sediments were ground gently and sieved through a 150-μm mesh for homogenization. TOC and TN were measured by a Perkin-Elmer 2400 Series II CHNS/O Analyzer (Perkin Elmer Inc., USA). BSi was measured by the molybdate blue spectrophotometric method after removing the carbonates and organics by 1 mol/L HCl and 10% H_2O_2 and digested using 0.5 mol/L Na_2CO_3 solution (Mortlock and Froelich, 1989). The US EPA Method 200.2 protocols (USEPA, 1994) were adapted to analyze As, Cd, Cu, Fe, Pb, Zn, and TP by an ICP/AES instrument (SPECTRO Spectro

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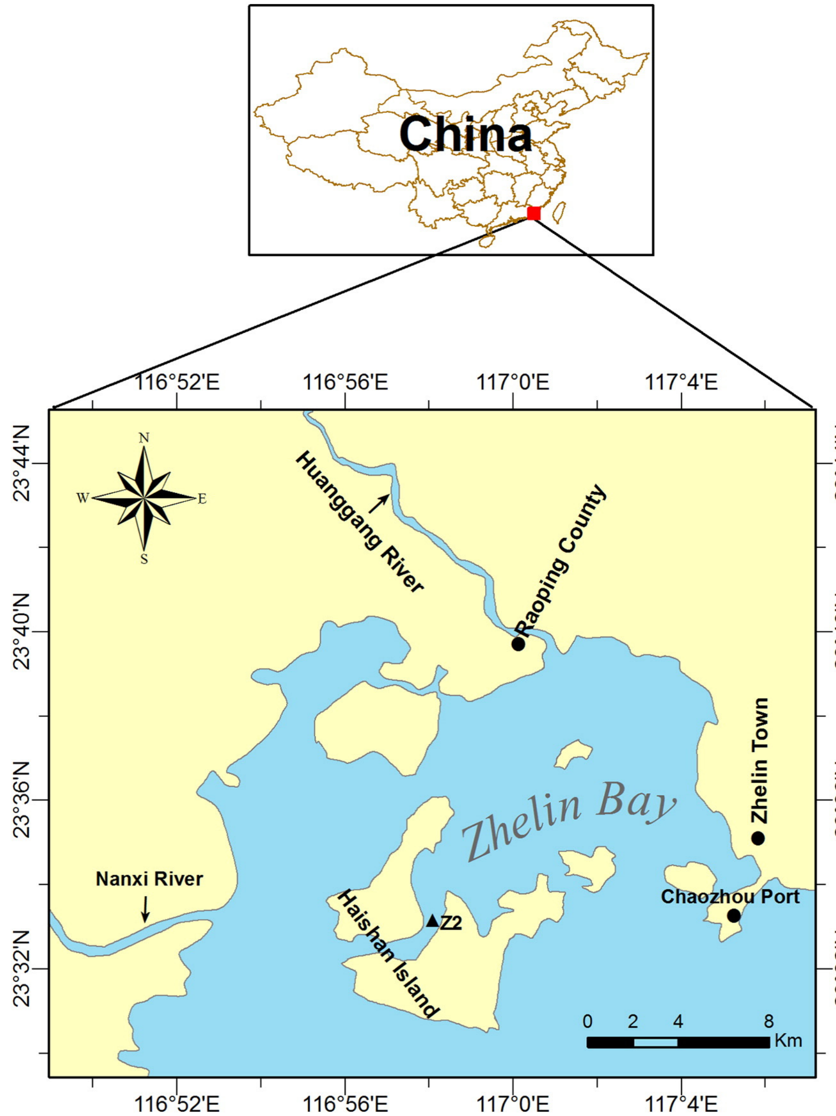

Fig. 1. Map and sampling stations in Zhelin Bay.

Ciros CCD inductively coupled plasma atomic emission spectrometry, Spectro Analytical Instruments, Germany). Hg was determined by combustion on a Direct Mercury Analyzer (Lumex Zeeman mercury RA-915+ Analyzer, Lumex-marketing JSC, Russia). The QA/QC was assessed by the analyses of blank reagents and 10 replicates of the certified reference material (Offshore Marine Sediment, GBW 07314, China). All of the reagents used in the analyses were ultrapure. The analytical results were in agreement with the certified values for all of the studied elements. The analytical precision was controlled within 5% for biogenic elements and within 10% for metals.

The sediment core was dated using the ^{210}Pb radiometric technique. The activities of ^{210}Pb and ^{226}Ra in the core were determined by a HP Ge λ Spectroscopy System (ORTEC GMX60P4, AMETEK, Inc., USA). The contribution of unsupported ^{210}Pb was calculated by subtracting the supported ^{210}Pb from the total ^{210}Pb. The supported ^{210}Pb was determined using the ^{226}Ra activity, assuming equilibrium between the two isotopes. According to the relatively high correlation between the core depths vs. natural logarithm (Ln) of the unsupported ^{210}Pb activity ($y = -111.3\ln(x) + 660.5$, $r^2 = 0.45$, n = 45), the average constant sedimentation rate of 3.5 cm/year was calculated using the constant initial concentration model. Therefore, the 186 cm core represented approximately 53 years of sediment deposition.

The *Z*-scoring standardized data were applied to eliminate the influence of different units of variance and make each determined variable have an equal weight. The two-tailed Pearson correlation analysis (CA) was used to evaluate the coefficients between all metallic and biogenic elements. Varimax rotation, with Kalser normalization, was chosen in PCA to maximize factor variance and to simplify the columns of the factor matrix. The cluster analysis, based on Euclidian distances, was used to compare and establish relationships between depths. All of the analyses were performed using the program SPSS 19.0 for Windows (SPSS Inc., Chicago, Illinois).

Cyst concentrations varied from 0 cysts/g to 770 cysts/g (Fig. 2a) with an average of 89.3 cysts/g. Cyst concentrations were low in sediments over 80 cm depth (before 1985), peaked between 40 cm and 70 cm (1988–1996), decreased between 30 cm and 40 cm (1997–2000), and increased thereafter and reached the maximum value in the topmost sediment. Cyst concentrations in this study were far lower than those previously reported (e.g., Harland et al., 2004; Wang et al., 2004; Pospelova et al., 2004, 2005), which were commonly higher than 1000 cysts/g (Radi et al., 2007). The low cyst assemblages in Zhelin Bay were due to the high sedimentary rate (3.5 cm/year). Furthermore, sediments from Zhelin Bay were predominantly composed of coarse sediments (>63 μm, sand and gravel), and the contribution of clay

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