



Historical record of lead accumulation and source in the tidal flat of Haizhou Bay, Yellow Sea: Insights from lead isotopes



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ABSTRACT

In order to investigate the historical records of lead contamination and source in coastal region of Haizhou Bay, Yellow Sea, a sediment core was collected from tidal flat, dated by ²¹⁰Pb and ¹³⁷Cs. Lead and its stable isotopic ratios were determined. The profiles of enrichment factor (EF) and Pb isotope ratios showed increasing trend upward throughout the core, correlating closely with the experience of a rapid economic and industrial development of the catchment. According to Pb isotopic ratios, coal combustion emission mainly contributed to the Pb burden in sediments. Based on end-member model, coal combustion emission dominated anthropogenic Pb sources in recent decades contributing from 48% to 67% in sediment. And the contribution of leaded gasoline was lower than 20%. A stable increase of coal combustion source was found in sediment core, while the contribution of leaded gasoline had declined recently, with the phase-out of leaded gasoline in China.

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Lead was released into the environment by human activities mainly through mining, and smelting of lead, which may pose potential adverse health effects to human, especially children (Nriagu, 1998). The sources of Pb such as coal combustion leaded gasoline and industrial activities also may be important contributors to environmental Pb contamination (Mukai et al., 1993; Zheng et al., 2004; Choi et al., 2007; Cheng and Hu, 2010). In aquatic environments, heavy metals such as Pb tend to associate with particulate matter due to their hydrophobic nature and are finally deposited to the underlying sediments (Gerritse et al., 1998). Therefore, concentrations of Pb in sediment cores from tidal flat provide a proxy for elucidating trends in deposition, associated with the socio-economical development of coastal cities. Chemostratigraphy of aquatic sediments has been conducted in many studies as a tool to reconstruct natural environmental changes, and to assess the extent of human activities in trace metal deposition (Hao et al., 2008; De Oliveira et al., 2009; Leorri et al., 2014). Traditional approaches, for example, statistical analysis method, chemical composition and the ratios of Pb and conservative/reference elements, are difficult to identify accurately the

potential sources. Given their fingerprinting properties, stable Pb isotopes can be used to discriminate among several sources and to identify environmental pathways of this metal (Ault et al., 1970; Chow and Earl, 1972; Sturges and Barrie, 1987; Carignan and Garipey, 1995; Gallon et al., 2005; Cheng and Hu, 2010; Louchouart et al., 2012; Allan et al., 2015; Gutiérrez-Caminero et al., 2015; Félix et al., 2015; Lee and Yu, 2016). In China, the majority of studies on coastal regions involve mainly reconstructions of anthropogenic activities based on trace metals (Li et al., 2000; Ip et al., 2004; Zhang et al., 2007; Chen et al., 2012; Yang et al., 2012; Zhang et al., 2013), but the few studies have examined ratios of stable Pb isotopes in sediments to identify the anthropogenic sources that contributed combustion-derived contaminants to the coastal regions (Hao et al., 2008; Zhang et al., 2008; Wang et al., 2014).

In this study, we report on the distribution of lead and stable Pb isotope ratios in a dated sediment core from tidal flat of Haizhou Bay, Yellow Sea. Haizhou Bay lies on the western margin of the South Yellow Sea, near the city of Lianyungang, and receives water inflow from the Linhong River (Fig. 1). Haizhou Bay is a major fishery base. Aquaculture industries are the main drivers of economic growth in Lianyungang, and its areas has grown by 4.3 times from 1995 to 2005 (OFBL, 2011). In addition, sediment quality around fish farming zones has been deteriorated in recent years (JOBST, 2001). Lianyungang Harbor, a natural deep-water harbor, is located on the southeast side of Haizhou Bay and is one of the most important harbors in China. In the past, the regions near Haizhou Bay experienced significant economic development. According to the recent reports, the Linhong River carried 2.26×10^8 t of domestic

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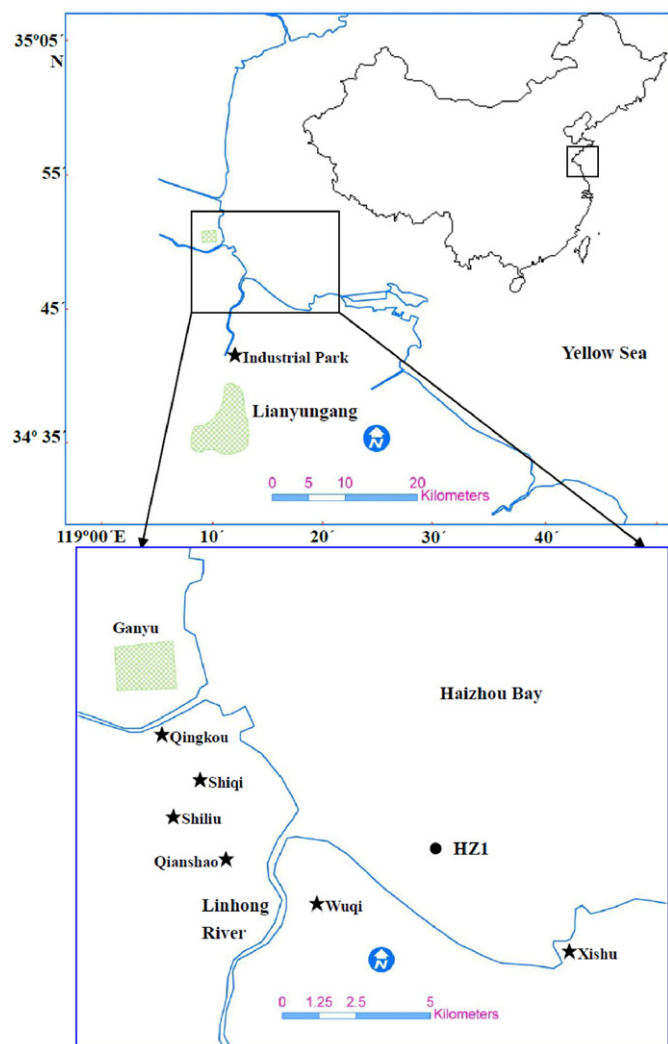


Fig. 1. Sampling site map.

sewage and industrial wastewater in 2010 (EPAL, 2011; OFBL, 2011), and a considerable portion of this wastewater was deposited and accumulated in the tidal flat. Here, we use the Pb concentrations and its isotopic ratios in a sediment core, to: 1) delineate the temporal trends of Pb concentrations and its isotopic ratios in tidal flat of Haizhou Bay recorded by a sedimentary core based on ^{210}Pb and ^{137}Cs dating; 2) elucidate anthropogenic impacts on the aquatic environment of Haizhou Bay in the recent decades; and 3) determine anthropogenic sources of Pb contamination.

Sediment core HZ1 with length 70 cm was manually collected from the south tidal flat of Haizhou Bay in April 2010 (Fig. 1). In the laboratory, the core was subsampled at 1 cm intervals. All the samples were immediately transferred to the laboratory and kept at $-20\text{ }^{\circ}\text{C}$.

About 0.25 g sample was digested with a mixture of concentrated $\text{HF-HClO}_4\text{-HNO}_3$ for the determination of total metal concentrations. The Al and Pb concentrations were analyzed using an inductively coupled plasma-optical emission spectrometer (ICP-OES) (Thermo Electro Co., ICAP 6300, USA). The accuracy of the analytical method was established with the standard reference material aquatic sediments GBW07312 (GSD-12). The recoveries for Al and Pb were 91%–101% and 83%–102%. Three replicates of each sample were run, and the coefficients of variation were Al, 0.62% and Pb, 2.85%. The analysis method of stable Pb isotopes was described by Jiang et al. (2006). Briefly, samples were digested using $\text{HF} + \text{HNO}_3$ to obtain the supernatant. The remaining dried residue was re-dissolved in $\text{HBr} + \text{HNO}_3$ to obtain the

supernatant, and then all supernatants were loaded into a column with 50 mm of AG 1-X8 anion exchange resin. The extracted Pb was purified in a second column. Approximately 100 ng of the Pb sample was loaded onto the single rhenium filament using the silica-gel technique. Then, the stable Pb isotopic compositions of samples were measured by the Finnigan Triton Thermal Ionization Mass Spectrometer (TIMS). Accuracy was checked by analyzing reference material SRM-981 from the National Institute of Standards and Technology (NIST). The measured values for NIST SRM-981 were $^{206}\text{Pb}/^{204}\text{Pb} = 16.8872 \pm 0.0043$ (certified value = 16.9371), $^{207}\text{Pb}/^{204}\text{Pb} = 15.4463 \pm 0.0071$ (certified value = 15.49135), $^{208}\text{Pb}/^{204}\text{Pb} = 36.4589 \pm 0.0185$ (certified value = 36.72132), $^{207}\text{Pb}/^{206}\text{Pb} = 0.913925 \pm 0.0173$ (certified value = 0.91464), and $^{208}\text{Pb}/^{206}\text{Pb} = 2.16211 \pm 0.0102$ (certified value = 2.1681) (2σ , external standard deviation, $n = 10$). The procedural blank for the bulk samples was less than 6 pg Pb.

^{210}Pb was determined by alpha counting of the ^{210}Po deposited onto Ag disks (Flynn, 1968) by using ^{209}Po as a yield tracer. The sediment was dissolved by adding a mixture of $\text{HNO}_3\text{:HCl:HF}$ (1:1:0.5 v/v/v) to 2 g of sediment and heating to $200\text{ }^{\circ}\text{C}$ overnight in closed Teflon containers. Counting was conducted by computerized multichannel α -spectrometry with gold-silicon surface barrier detectors (Model Otetc PLUS, ORTEC Co., USA). The 30 ~ 50 g of the sub-sample thus obtained was subjected to ^{137}Cs analysis. In brief ^{137}Cs was measured by γ -spectrometry using the 661.62 keV photo peak that results from the decay. The counting system is equipped with a 40% efficiency HPGe detector (GMX30P-A, ORTEC) interfaced to a digital γ -ray spectrometer (ORTEC DSPEC PlusTM). IAEA-327 and a sediment standard supplied by Bedford Institute of Oceanography were used to calibrate the detector.

The profiles of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs are shown in Fig. 2. The down-core profile of ^{137}Cs shows a clear maximum in activity at 66.5 cm depth (Fig. 2). Distinctly, the maximum peaks for ^{137}Cs in core at the depth of 66.5 cm were attributed to the maximum deposition in 1963 (UNSCEAR, 2000). Sedimentation rates (1.45 cm/yr) were derived from the CFCS method based on ^{210}Pb , which were calibrated based on ^{137}Cs time markers.

The profiles of Al and Pb for HZ1 core are shown in Fig. 3. The concentrations of Al fluctuated in the range of 4.03%–6.58%. Differing from

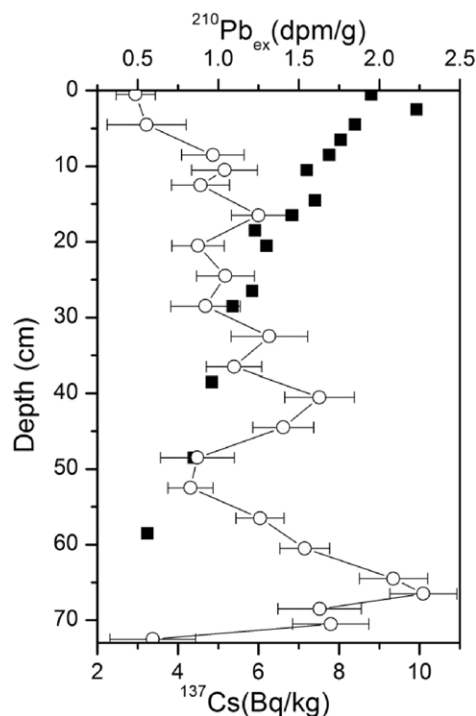


Fig. 2. Depth profiles of excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$) and ^{137}Cs in core HZ1.

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