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Tracking historical lead pollution in the coastal area adjacent to the Yangtze River Estuary using lead isotopic compositions

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The combination of Pb concentration, sedimentary flux, Pb isotopic composition and ^{210}Pb dating in the coastal ECS sediments revealed the historical Pb pollution in China.

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ABSTRACT

The rapid economic development in the Yangtze River Delta (YRD), China in the last three decades has had a significant impact on the environment of the East China Sea (ECS). Lead isotopic compositions of a ^{210}Pb dated sediment core collected from the coastal ECS adjacent to the Yangtze River Estuary were analyzed to track the Pb pollution in the region. The baseline Pb concentration in the coastal ECS sediments before the industrialization in China was 32 $\mu g\,g^{-1}$, and the corresponding $^{206}Pb/^{207}Pb$ ratio was 1.195. The high-resolution profiles of Pb flux and $^{206}Pb/^{207}Pb$ ratios had close relationships with the economic development and the history of the use of leaded gasoline in China, and they were clearly different from those of most European countries and United States.

1. Introduction

Lead in the aquatic environment has both natural and anthropogenic origins. Anthropogenic Pb is mostly from mining activities, industrial uses, coal burning and leaded gasoline because of the increasing number of automobiles on the road (Nriagu and Pacyna, 1988; Nriagu, 1998). Atmospheric fallout is the main anthropogenic input of Pb to the aquatic ecosystem (Nriagu and Pacyna, 1988) and leaded gasoline may be the main emission source until its phasing out in many countries (Soto-Jimenez et al., 2006). Stable Pb isotopic ratios ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$) have been used as a powerful tool to differentiate between the natural and anthropogenic Pb in the aquatic and atmospheric environments (Farmer et al., 1996; Moor et al., 1996; Monna et al., 1997; Shotyk et al., 1998; Lima et al., 2005).

The Yangtze River Delta (YRD) region, with a population of about 82.1 million in 2004 (Tuan and Ng, 2007), has become the largest and most developed economic region in China since 1978. The Yangtze River has the world's fifth largest discharge in water (9200 Mt yr^{-1}) and the fourth in sediment (480 Mt yr^{-1}) (Yang et al., 2006). Its sediments directly discharged into the East China Sea (ECS) are deposited mostly in the coastal mud due to the circulation systems and the Coriolis Force (Fig. 1) (Liu et al., 2007).

Anthropogenic Pb emitted into the atmosphere in the Yangtze River drainage basin enters the soil and aquatic environments by wet and dry deposition, and some of the Pb is eventually transported to the ECS by the river discharge. Atmospheric deposition directly over the ECS can be another source for Pb in the study area, since the ECS is located in the down wind area of the continental outflow of air pollutants to the northwest Pacific Ocean in winter and spring when northwestern winds prevail, owing to the East Asian Monsoon effect (Guo et al., 2006). The coastal mud of the ECS is a major sink for the fine-grained sediment and associated pollutants (Guo et al., 2007), making it an ideal place to study the regional environmental contamination record.

The rapid economic development in China in the past three decades has, unfortunately, had great pressures on the coastal environment (Guo et al., 2007; Ip et al., 2004, 2007). Historical pollution data would be very useful in understanding the impact of economic development on the environment.

There were several studies on the pollution and source of Pb in the ECS and Yellow Sea (YS). Pb concentration increased

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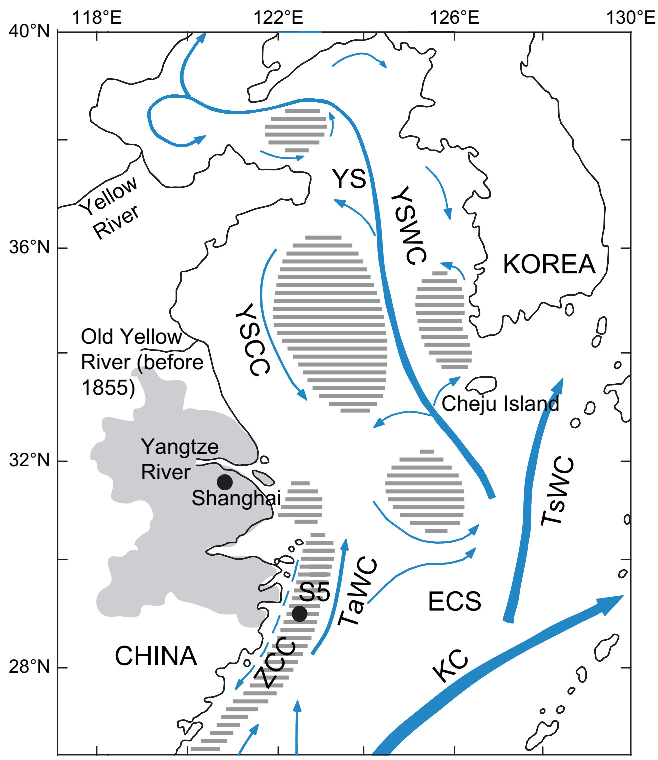

Fig. 1. Sampling site and general circulation system in the Yellow Sea and East China Sea. ECS: East China Sea; YS: Yellow Sea; KC: Kuroshio Current; TaWC: Taiwan Warm Current; TsWC: Tsushima Warm Current; YSCC: Yellow Sea Coastal Current; YSWC: Yellow Sea Warm Current; ZCC: Zhejiang Coastal Current. Circulation system and mud areas (dark patched areas) are after Guo et al. (2007). The dark area of the land in China is the economic region of the Yangtze River Delta.

remarkably from 1980 to 1997 in the coastal ECS sediments (Huh and Chen, 1999). Pb sources in the YS sediments were identified using stable Pb ratios (Choi et al., 2007). However, Pb source profiles determined by using stable Pb isotopes and Pb fluxes in the sediments of the ECS have not been reported, and more importantly, the impact of the recent phasing out of leaded gasoline in China since 1999 on the sediments has not been studied. In this work, high-resolution depositional records of Pb concentration, flux and stable Pb isotopic compositions in a dated sediment core collected from the coastal ECS mud adjacent to the YRE are used to track the Pb pollution in China for the past century.

2. Materials and methods

2.1. Sample collection

The sediment core (S5) was collected using a gravity corer deployed from the R/V Dong Fang Hong 2 of the Ocean University of China (OUC) in June 2003. The sampling site (122°30.01′E, 29°00.25′N) was located in the north coastal ECS (Fig. 1), and the water depth was 50.0 m. The length of the core was 148 cm. There was no significant loss of surface sediments or distortion of top sediment layer during the gravity coring. The core was stored at 4 °C in the vessel and was cut into 1–2 cm thick slices along the length using a stainless steel cutter after it was brought back to the laboratory at the university. Sub-samples were stored in pre-cleaned plastic bags and preserved at 4 °C until analysis.

2.2. Analysis of the grain size of the sediments

The grain size of the core sediments was determined by a laser particle size analyzer (Mastersizer 2000, Malvern Instruments, Ltd., UK) in the sedimentary dynamic laboratory of the College of Marine Geosciences, OUC. About 1 g of pre-homogenized sediment sample was pretreated using 10 ml of 30% H_2O_2 solution to decompose the organic matter, and then dispersed and homogenized using ultrasonic agitation for 30 s before being analyzed for grain size. The particle sizes of sediments were <4 μm for clay, 4–63 μm for silt and >63 μm for sand. The relative error of paired duplicate samples was less than 3% ($n = 8$).

2.3. Dating of the sediment core

The dominant fallout of ^{210}Pb in the ECS is from the wet and dry atmospheric depositions because riverine particles have low ^{210}Pb activities (DeMaster et al., 1985), and the water column (30–70 m) is so shallow that ^{226}Ra does not have sufficient time to decay to ^{210}Pb in the seawater (DeMaster et al., 1985).

S5 was dated at the Guangzhou Institute of Geochemistry, Chinese Academy of Science, and the detailed method of ^{210}Pb dating was described in Zhang et al. (2002). The ^{210}Pb activities in sediment sub-samples were determined by the analysis of the α radioactivity of its decay product ^{210}Po, on the assumption that the two are in equilibrium. The Po was extracted, purified, and self-plated onto silver disks at 75–80 °C in 0.5 M HCl, with ^{209}Po used as a yield monitor and tracer in quantification. Counting was conducted by computerized multi-channel α spectrometry with gold–silicon surface barrier detectors. Supported ^{210}Po was obtained by indirectly determining the α activity of the supporting parent ^{226}Ra, which was carried by co-precipitated $BaSO_4$. The fine-grained sediments of core S5 were clayey silt, and its down-core grain size variations were small (Fig. 2). These indicated that there was a stable sedimentary environment in this area, which could be attributed to the effect of the circulation system of the ECS and a relatively deeper water depth

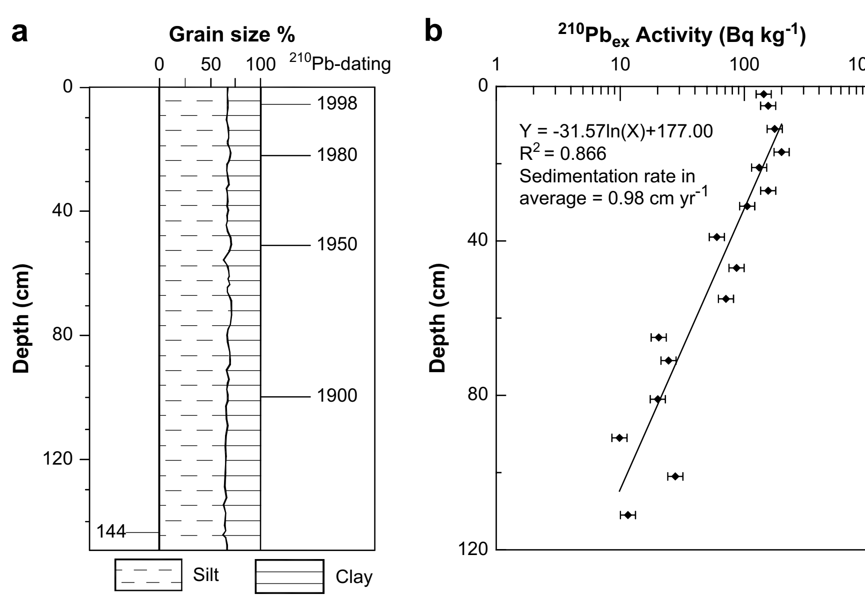

Fig. 2. Down-core grain size distributions and ^{210}Pb age dating of the core S5.