



## 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 <sup>210</sup>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 <sup>210</sup>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 μg g<sup>-1</sup>, and the corresponding <sup>206</sup>Pb/<sup>207</sup>Pb ratio was 1.195. The high-resolution profiles of Pb flux and <sup>206</sup>Pb/<sup>207</sup>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.

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### 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 (<sup>206</sup>Pb/<sup>207</sup>Pb and <sup>208</sup>Pb/<sup>207</sup>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; Shoty 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<sup>-1</sup>) and the fourth in sediment (480 Mt yr<sup>-1</sup>) (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).

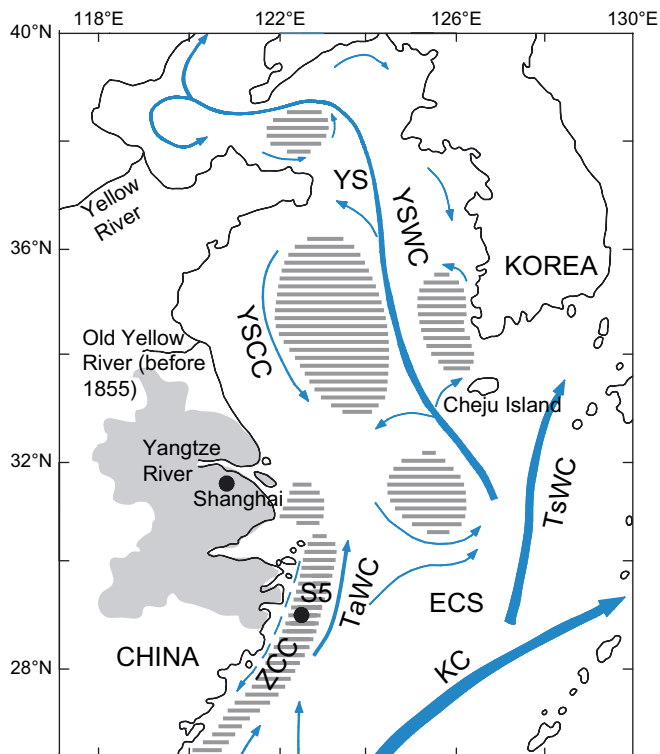
Anthropogenic Pb emitted into the atmosphere in the Yangtze River drainage basin enters the soil and aquatic environments by wet and dry deposition, and some of the Pb is eventually transported to the ECS by the river discharge. Atmospheric deposition directly over the ECS can be another source for Pb 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.

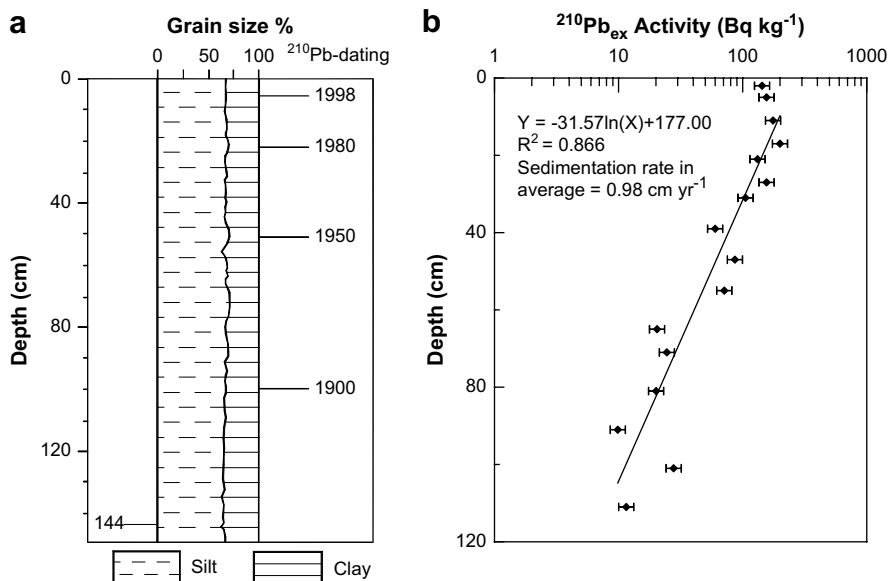
**2.2. Analysis of the grain size of the sediments**

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<sub>2</sub>O<sub>2</sub> 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 <sup>210</sup>Pb in the ECS is from the wet and dry atmospheric depositions because riverine particles have low <sup>210</sup>Pb activities (DeMaster et al., 1985), and the water column (30–70 m) is so shallow that <sup>226</sup>Ra does not have sufficient time to decay to <sup>210</sup>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 <sup>210</sup>Pb dating was described in Zhang et al. (2002). The <sup>210</sup>Pb activities in sediment sub-samples were determined by the analysis of the α radioactivity of its decay product <sup>210</sup>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 <sup>209</sup>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 <sup>210</sup>Po was obtained by indirectly determining the α activity of the supporting parent <sup>226</sup>Ra, which was carried by co-precipitated BaSO<sub>4</sub>. 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 <sup>210</sup>Pb age dating of the core S5.

(50 m) (Liu et al., 2007). Based on this, an average constant sedimentation rate of  $0.98 \text{ cm yr}^{-1}$  at site S5 could be calculated using a constant flux–constant sedimentation rate (cf:cs) model (Fig. 2) (Ugur and Yener, 2001). The results were in good agreement with those reported by Huh and Su (1999) using  $^{137}\text{Cs}$  at the same area. The data for S5 beyond 100 years were given only as a reference values.

#### 2.4. Elemental analysis

The elemental concentrations were measured using an Energy-dispersive Mini-probe Multi-element Analyzer (EMMA), a small desktop X-ray fluorescence (XRF) technique system, at the Institute of Environmental Geochemistry, University of Heidelberg, Germany. The analytical process and quality control used were according to Cheburkin and Shotykh (1996), and the sediment reference materials used were the NIST SRM 1646a (estuarine sediments), USGS MAG-1 (marine mud), and CCRMP LKSD-1 to LKSD-4 (set of lake sediments standards, Canada). The error was  $<10\%$  and the detection limit of Pb was  $2 \mu\text{g g}^{-1}$ .

#### 2.5. Stable Pb isotopic composition analysis

Stable Pb isotopic compositions were determined at Nanjing University, China, using a Finnigan Triton Thermal Ionization Mass Spectrometer (TIMS) according to the method by Jiang et al. (2006). Briefly, samples were digested using  $\text{HF} + \text{HNO}_3$ . The dried residue was re-dissolved in  $\text{HBr} + \text{HNO}_3$  and loaded into a column with  $50 \mu\text{m}$  of AG 1-X8 anion exchange resin. The extracted Pb was purified in a second column. Approximately  $100 \text{ ng}$  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 TIMS. The SRM 981 (common Pb) standard was used for mass fraction corrections (Todt et al., 1996) and repeated measurements of the SRM 981 standard yielded the following mass ratios:  $^{206}\text{Pb}/^{204}\text{Pb} = 16.8883 \pm 0.0042$ ,  $^{207}\text{Pb}/^{204}\text{Pb} = 15.4257 \pm 0.0062$ ,  $^{208}\text{Pb}/^{204}\text{Pb} = 36.4941 \pm 0.0197$ ,  $^{207}\text{Pb}/^{206}\text{Pb} = 0.913396 \pm 0.000143$ , and  $^{208}\text{Pb}/^{206}\text{Pb} = 2.16092 \pm 0.00064$  ( $2\sigma$ , external standard deviation,  $n=6$ ). The standard accepted values are:  $^{206}\text{Pb}/^{204}\text{Pb} = 16.9322$ ,  $^{207}\text{Pb}/^{204}\text{Pb} = 15.4855$ ,  $^{208}\text{Pb}/^{204}\text{Pb} = 36.6856$ ,  $^{207}\text{Pb}/^{206}\text{Pb} = 0.914559$ , and  $^{208}\text{Pb}/^{206}\text{Pb} = 2.166617$ . Pb isotopic ratios reported in this work were corrected according to the accepted value for the SRM 981.

### 3. Results and discussion

#### 3.1. Pb concentration and flux profiles

The sedimentary flux and concentration profiles of Pb in S5 are shown in Fig. 3. The Pb sedimentary flux was calculated from the corresponding concentration, the average sedimentation rate and

the dry density of the sediment (Guo et al., 2007). The sediment focusing for flux correction was not applied in this work since the sediment focusing factor was less than 1.2 in the northern coastal ECS (Huh and Su, 1999), meaning that the sedimentary inventories didn't greatly exceed the amounts from  $^{210}\text{Pb}$  atmospheric fallout (Lima et al., 2003).

The down-core Pb concentration profile followed that of the sedimentary fluxes very well (Fig. 3). The Pb concentrations (average,  $31 \pm 2 \mu\text{g g}^{-1}$ ) and fluxes (average,  $30 \pm 2 \mu\text{g cm}^{-2} \text{ yr}^{-1}$ ) were relatively constant from  $\sim 1860$  to mid-1980s, and they increased rapidly from the mid-1980s to 1999, peaking in 1999 at  $65 \mu\text{g g}^{-1}$  for the concentration and  $61 \mu\text{g cm}^{-2} \text{ yr}^{-1}$  for the flux before decreasing sharply to  $36 \mu\text{g g}^{-1}$  and  $31 \mu\text{g cm}^{-2} \text{ yr}^{-1}$ , respectively, in 2002.

It is reasonable to assume the average Pb concentration ( $32 \pm 3 \mu\text{g g}^{-1}$ ,  $n=4$ ) from  $\sim 1860$  to 1900 to be a baseline Pb value since China had an agricultural economy during that time. The anthropogenic Pb can then be estimated by subtracting the baseline value from the total concentration (Lima et al., 2005), and the maximum anthropogenic Pb concentration calculated in the core samples thus was  $\sim 33 \mu\text{g g}^{-1}$  in  $\sim 1999$ . It is common to use enrichment factor (EF) to underscore the influence of anthropogenic pollution (Kylander et al., 2005). In the present work, the reference concentrations of Ti and Pb in crust were taken from Taylor (1964). The average EF of Pb was 2.9 from  $\sim 1860$  to 1900 and could be considered to be the baseline EF value in the coastal ECS as discussed earlier. The down-core EFs were constant until the mid-1980s (Fig. 3), indicating that the Pb in the core was mostly from natural sources during this period.

The doubling of the Pb concentration and flux from the mid-1980s to 1999 with the sharp increase of EFs reflected the rapid economic development in China from  $\sim 1980$ , and in particular the large increase in automobiles using leaded gasoline from  $\sim 1990$  (Fig. 4) (Lin, 1998). In 1978, China ushered in the "Reform and Open" policy, and the economy increased at a very rapid pace ever since. The decrease in Pb concentration and EF after 1999 could be attributed to the start of the phase-out of leaded gasoline in July 1999 in China, the decrease of coal consumption (Fig. 4), and better

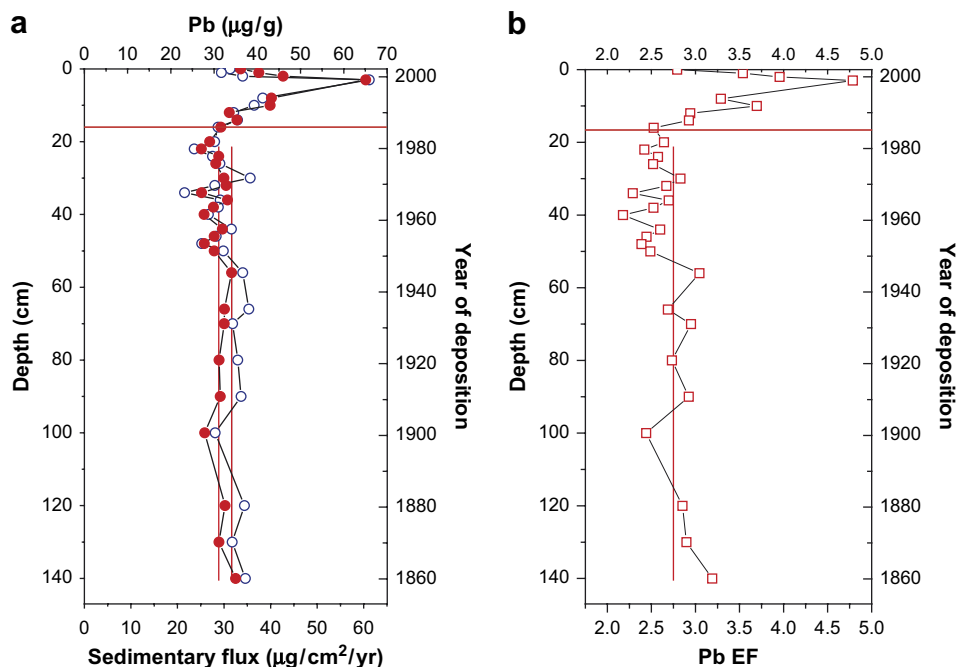


Fig. 3. (a): Pb concentration (solid circles), sedimentary flux (open circles), and (b): enrichment factors of the core S5.

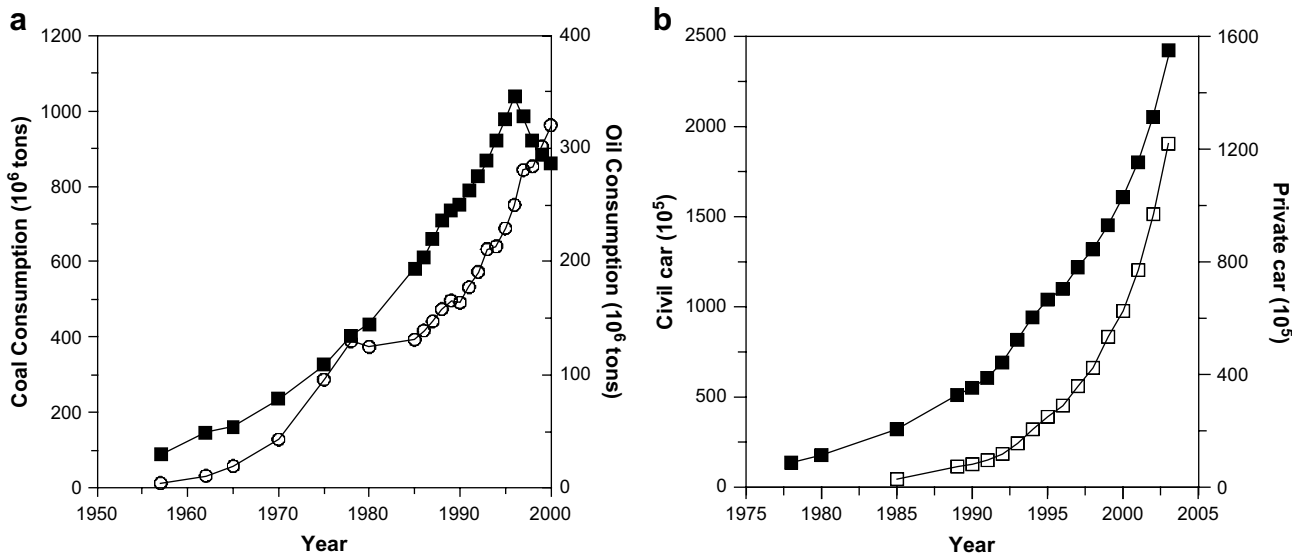


Fig. 4. (a): Annual coal (square) and oil (circle) consumption data (in million tons) in China from 1956 to 2000 (National Bureau of Statistics of China, 2005) and (b): numbers of civil (black square) and private (white square) car in China from 1978 to 2003 (<http://www.stats.gov.cn/tjsj/ndsj/>).

environmental control in China (Fig. 5) (Wang et al., 2006). The high peak of Pb flux and EF in 1999 could be partly attributed to the severe flood of the Yangtze River in July and August of 1998. The concentrations and fluxes of polycyclic aromatic hydrocarbons (PAHs) in S5 had a similar peak in 1999 (Guo et al., 2007). The excessive stripping and leaching by the flood water transported more anthropogenic Pb accumulated in soil and water system (e.g., reservoirs and lakes) in the drainage basin of the Yangtze River into the ECS. Higher amounts of PAHs, polychlorinated biphenyls and organochlorine pesticides, mainly in the particulate phase, have been detected during high tidal flow events in the Anacostia River, Chesapeake Bay, USA (Foster et al., 2000). Flood events also resulted in a noticeable increase of the particle-associated metals, such as As, Pb, Cu and Zn, in the Dese River Estuary, Venice Lagoon, Italy (Zonta et al., 2005).

3.2. Pb isotopic composition profile

Down-core profile of the <sup>206</sup>Pb/<sup>207</sup>Pb ratios is shown in Fig. 6. <sup>206</sup>Pb/<sup>207</sup>Pb was constant from ~1860 to mid-1960s (average, 1.195), and decreased from the mid-1960s to 1.165 in 2000 before

rising to 1.185 in 2002. It was reported that the Pb concentration in the Antarctic seawater was extremely low, but the Pb isotopic composition revealed a significant contribution of Pb from various industrial sources (Flegal et al., 1993), indicating that the stable Pb isotopic composition was more sensitive to indicate the anthropogenic Pb inputs, thus, it was reasonable that the change time of Pb isotopic composition was earlier than that of Pb concentration in the coastal ECS.

The correlations between <sup>206</sup>Pb/<sup>207</sup>Pb and <sup>208</sup>Pb/<sup>207</sup>Pb in S5 are shown in Fig. 7. The data sets changed very little from ~1860 to

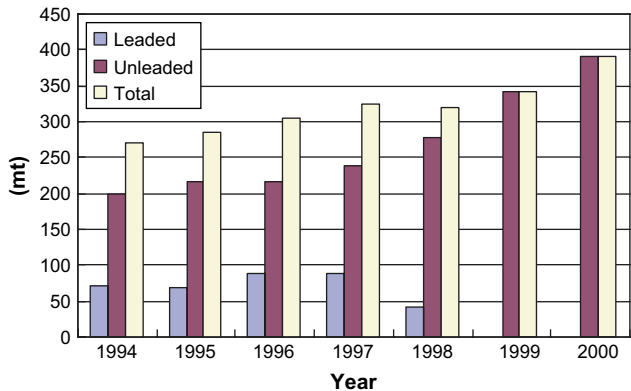


Fig. 5. Time series of annual production of two major petrol producers in China, China Petroleum Corporation (PetroChina) and China Petro-Chemical Corporation (Sinopec) from 1994 to 2000 (Courtesy of Wang et al., 2006).

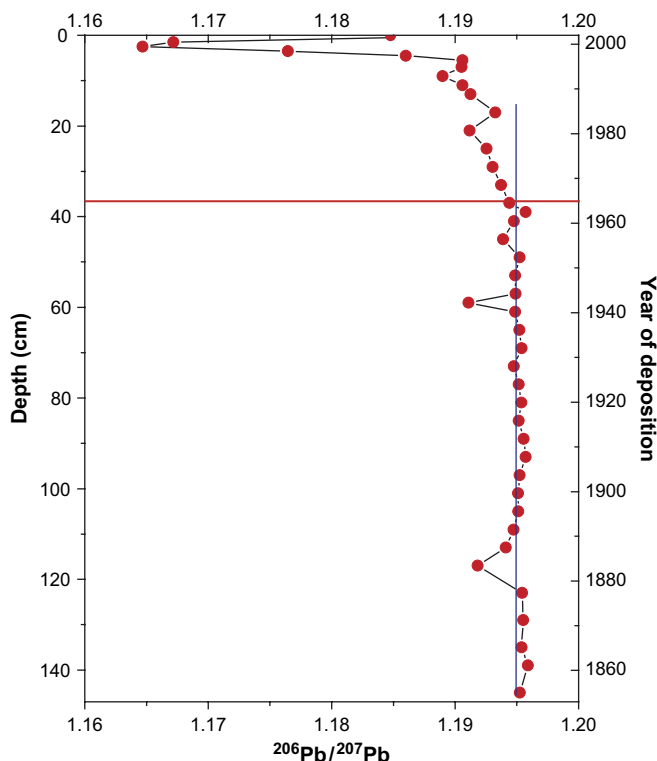


Fig. 6. <sup>206</sup>Pb/<sup>207</sup>Pb profile of the core S5.

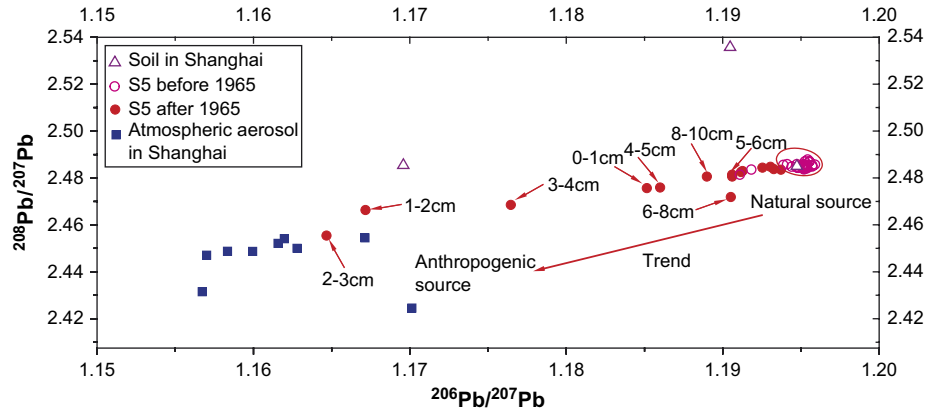


Fig. 7. Correlations between  $^{208}\text{Pb}/^{207}\text{Pb}$  and  $^{206}\text{Pb}/^{207}\text{Pb}$  of the core S5 (this work), and the soils and atmospheric aerosols in Shanghai (Courtesy of Zheng et al., 2004).

mid-1960s. However, there was a distinct shift to anthropogenic sources from the mid-1960 to 2002. This suggested that Pb input from ~1860 to mid-1960s was mainly from natural origins, and anthropogenic Pb increased since ~1960s, especially after the 1980s. The  $^{206}\text{Pb}/^{207}\text{Pb}$  average value of 1.195 from ~1860 to 1900 could be used as a baseline value before industrialization of China.

There are several possible anthropogenic Pb sources in these sediments, including atmospheric deposition, soil erosion, vehicle exhaust, the emission of coal and industrial discharges, and so on. The correlations between  $^{206}\text{Pb}/^{207}\text{Pb}$  and  $^{208}\text{Pb}/^{207}\text{Pb}$  of the S5 sediments, atmospheric aerosols, and soils in the YRD are shown in Fig. 7. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of the exhaust of leaded gasoline vehicles is 1.099–1.117 (Chen et al., 2005). For Shanghai, this ratio is ~1.163 for the coal used which is mainly from Shanxi Province, 1.170–1.195 for soils, 1.157–1.164 for some top soils around industrial boilers, 1.151–1.180 for some metallurgic dust, and this ratio for the atmospheric aerosols collected from 1 November 2000 to 27 January 2001 is 1.156–1.170 (Zheng et al., 2004; Chen et al., 2005). The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of the S5 bottom samples was similar to that of some soils in Shanghai, however, the  $^{206}\text{Pb}/^{207}\text{Pb}$  of the S5 upper layer samples was closer to that of various anthropogenic sources.

Pb emitted from automobiles increased drastically from 1990 due to the marked increase in road vehicles in China (Fig. 4). Leaded gasoline was banned for sale in most cities in July 1999 and was totally banned all over the country in July 2000, triggering the

pronounced reduction in Pb emission from vehicles (<http://www.jxepb.gov.cn/ZCFG/fgtl/gyxq.htm>). Chen et al. (2005) reported that the value of  $^{206}\text{Pb}/^{207}\text{Pb}$  increased from 1.147 to 1.161 in the atmospheric aerosols of Huangpu and Putuo districts of Shanghai, from 1995 to 2003 due to the phasing out of leaded gasoline, and Wang et al. (2006) reported a similar gradually increase from 1994 to 2001 in Tianjin. These evidences suggested that the Pb contribution from vehicle emission was a very important source before the ban of leaded gasoline in China.

The Pb from coal burning had decreased somewhat from 1996 (Fig. 4). The Pb concentration in S5 increased until 1999, and the corresponding  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio decreased until 2000. Considering above results, it should be concluded that the Pb concentrations, fluxes and  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios in S5 (Figs. 3 and 4) followed the phasing out of leaded gasoline in China in July 1999. Consequently, this led to a possibility that the Pb peak in 1999 in S5 was mainly caused by leaded gasoline, and coal burning was a second important source. The proportion of several Pb sources in S5, such as the leaded gasoline, coal, and industrial Pb, was difficult to be quantified due to more than two sources of Pb in the study area.

### 3.3. Comparison with European and North American data

The Pb peaks in sediments and peat in North America and most European countries in the 1970s were also largely due to the Pb additives in the gasoline and industrial emissions (Moor et al., 1996; Shotyk et al., 1998; Brännvall et al., 1999; Renberg et al., 2001;

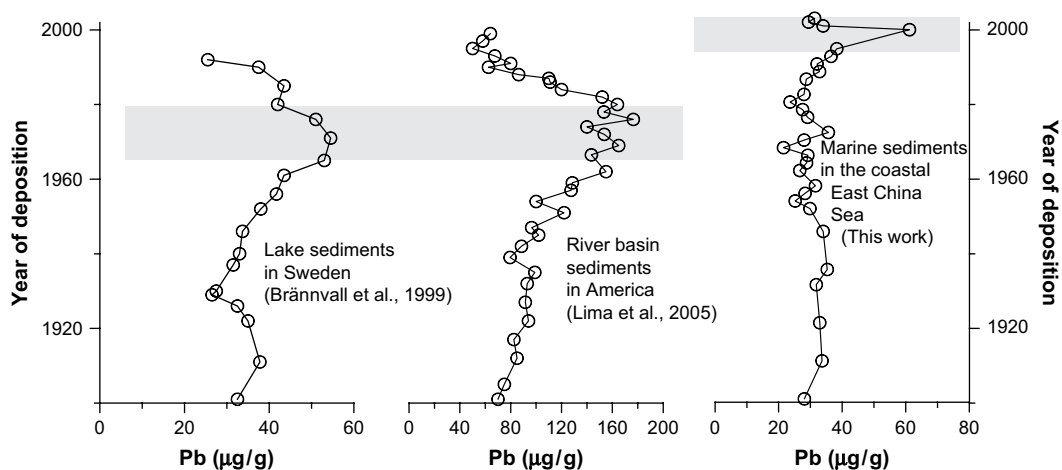


Fig. 8. Records of Pb concentration in sediment cores in Sweden, America and China.

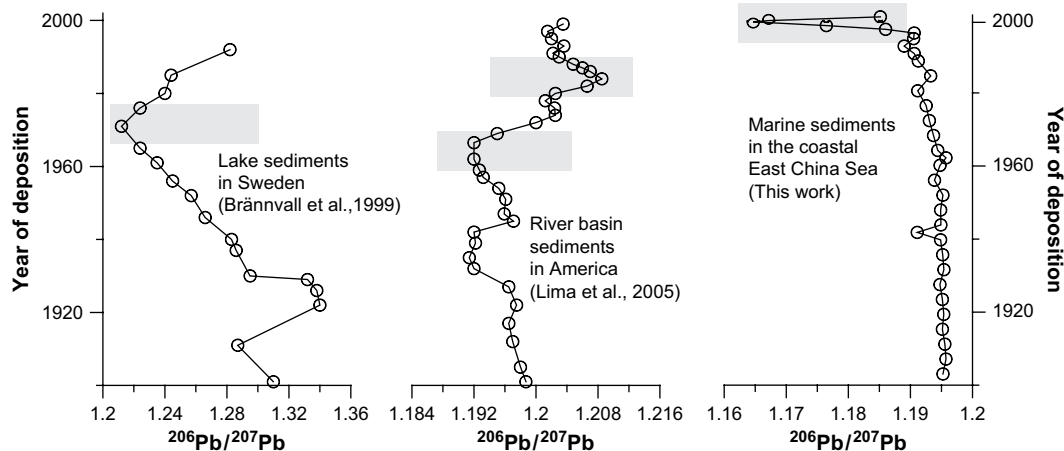


Fig. 9. Records of  $^{206}\text{Pb}/^{207}\text{Pb}$  in sediments cores in Sweden, America and China.

Lima et al., 2005; Soto-Jimenez et al., 2006). Following the implementation of stricter industrial emission standards and the phasing out of leaded gasoline in the 1970s, Pb pollution began to decline in most European countries (Moor et al., 1996; Shotyk et al., 1998; Brännvall et al., 1999; Bindler et al., 2001) and North America (Lima et al., 2005) (Fig. 8). In China, this trend lagged by about 20 years in the phasing out of leaded gasoline.

The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio from the present work and those of the sediments in Europe decreased with the increasing use of leaded gasoline, while in the US, it firstly decreased (1900–1965), but increased rapidly from the late 1960s to the late 1980s (Fig. 9). The Pb anti-knock additives used in Europe and China were mostly from Associated Ocel Company (AOC) (Wang et al., 2006). AOC used the more  $^{206}\text{Pb}$ -depleted ores from geologically old mines in Broken Hill, Australia, whose  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio ranged from 1.04 to 1.10 (Wang et al., 2006). In the US, there was also the same strong  $^{206}\text{Pb}/^{207}\text{Pb}$  decreasing signature of the alkyl-Pb additives from 1900 to 1965. However,  $^{206}\text{Pb}/^{207}\text{Pb}$  increased from the late 1960s to the late 1980s because of the introduction of southeastern Missouri Pb ores, with a higher  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio (Lima et al., 2005).

#### 4. Conclusion

The baseline Pb concentration in the coastal ECS adjacent to the YRE was  $32 \mu\text{g g}^{-1}$ , with the corresponding  $^{206}\text{Pb}/^{207}\text{Pb}$  of 1.195. The variations of Pb flux and stable Pb isotopic composition in the study area reflected the impacts of the economic development, in particular the leaded gasoline usage in China. The severe flood of the Yangtze River could contribute more anthropogenic Pb to the coastal ECS. Due to the different time of economic development, especially about 20 years lag of the phasing out of leaded gasoline in China, the historical records of Pb in the environment of China were obviously different from those of most European countries and United States.

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