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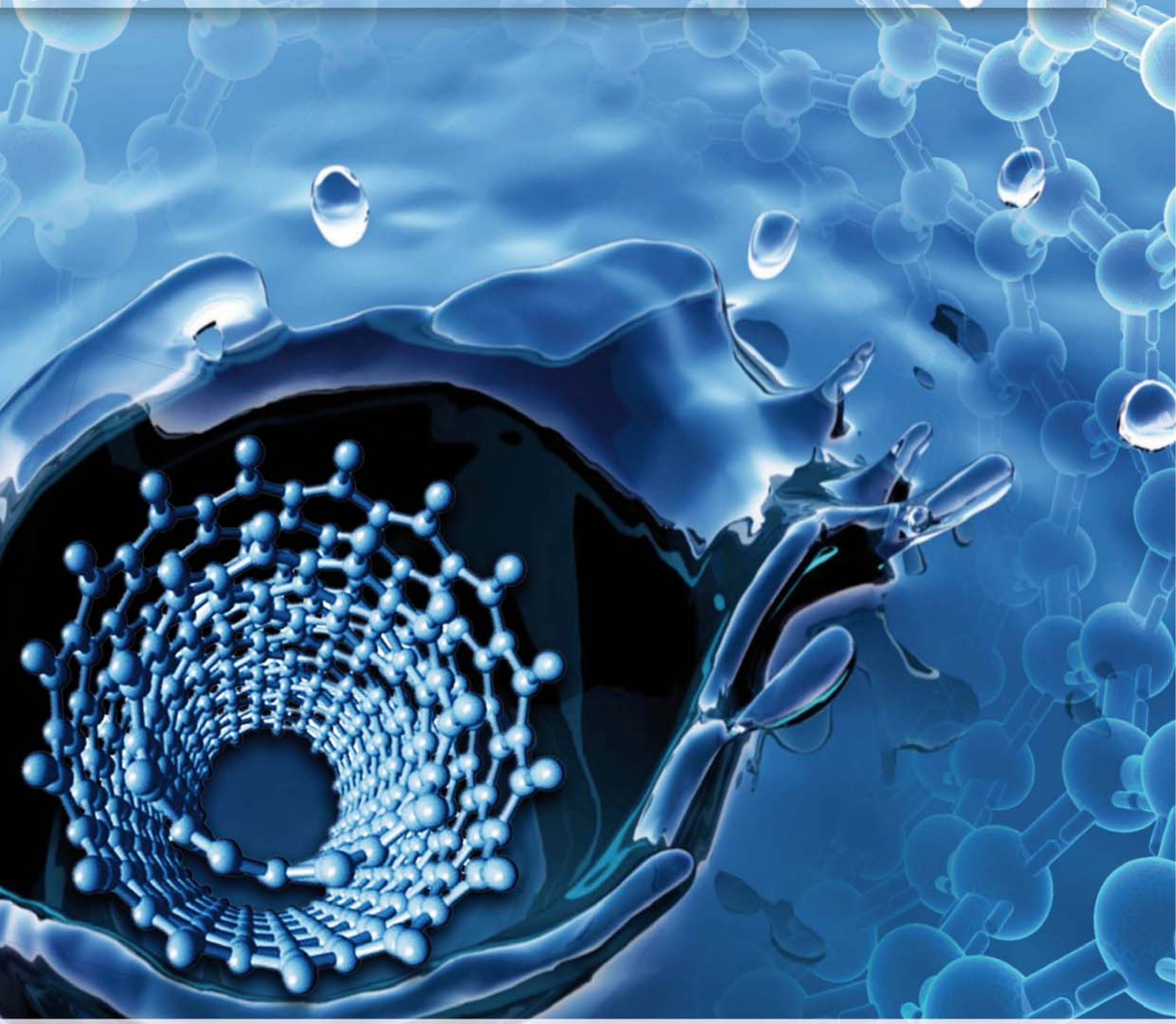

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Historical trends in the anthropogenic heavy metal levels in the tidal flat sediments of Lianyungang, China

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Abstract

The sedimentation of metals can preserve the historical record of contaminant input from local and regional sources and provide information on the historical changes in regional water and sediment quality. We report the ^{210}Pb activities and the heavy metal (Cd, Cr, Cu, Mn, Pb and Zn) depth profiles from sediment cores retrieved in 2010. The mean sedimentation rates of 0.85–1.5 cm/yr are determined by ^{210}Pb dating. The sediments in the tidal flat have recorded heavy metal deposition and thus allow the establishment of a connection between the temporal evolution of the heavy metal pollution and the historical changes in the economic development of Lianyungang. The enrichment factors (EF) are calculated to estimate the level of contamination stored in these sediments. The results show that in the studied sites, Cr and Cu display low EF values and are mainly from lithogenic origin. For the other studied trace metals, a great variability in the sedimentary record is observed. Significant anthropogenic enrichment over the last 50 years is revealed at the tidal flat that receives fluvial inputs. Zinc is the element with the highest EF values, followed by the order of Pb > Cd > Mn > Cu and Cr. The temporal variations of the heavy metals peak during the late 1980s to the early 2000s and show a decreasing trend afterward. The pollution intensity of the tidal flat is determined by using EF and the geo-accumulation index (I_{geo}), which show that, based on the I_{geo} scale, the tidal flat of Haizhou Bay is unpolluted to moderately polluted.

Key words: sediment rates; heavy metals; pollution; anthropogenic input; tidal flat

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Introduction

Due to their toxicity, bioaccumulation capacity and persistence, the cycling of heavy metals is a serious concern that has been the focus of many studies of coastal wetland environments (Eisenrhcil et al., 1986; Lacerda et al., 1988; Benoit et al., 1999; Li et al., 2007; Niu et al., 2009). Coastal tidal marshes play critical buffering roles in slowing shoreline erosion and in absorbing nutrients and contaminants before they reach the oceans and estuaries, which can result in the accumulation of excess levels of contaminants in marshes near urban and industrial areas. As a result of rapid industrial development over the past century, heavy metals have been discharged into coastal environments (Lee and Cundy, 2001; Bellucci et al., 2002; Munksgaard et al., 2003; Spencer et al., 2003; Maanan, 2008). Increased urbanization and industrialization have led to increased marine discharges, in turn, the total load of pollutants, including heavy metals, being delivered to the sea has increased (Natesan et al., 2010). Because of their variable physical and chemical properties, coastal zones may act as sinks or sources of heavy metals (Harbison, 1986). The heavy metals discharged into water are generally adsorbed onto suspended particles and then deposited on the bottom as sediments. Consequently, the heavy metals in sediment cores can be used to reconstruct historical fluctuations in regional water and sediment quality (Hartmann et al., 2005; Cantwell et al., 2007). With this information, the efficiency of pollution management strategies can be evaluated, and regulatory actions can be implemented to reduce potential health risks.

Haizhou Bay lies on the western margin of the South Yellow Sea, near the city of Lianyungang, and receives water from the Linhong River (**Fig. 1**). The bay, which is shaped like a trumpet, has an area of approximately 876.39×10^6 m^2, a coastline length of 86.81×10^3 m, and a maximum width of approximately 42×10^3 m. Over the last 80 years, the bay has been affected by anthropogenic

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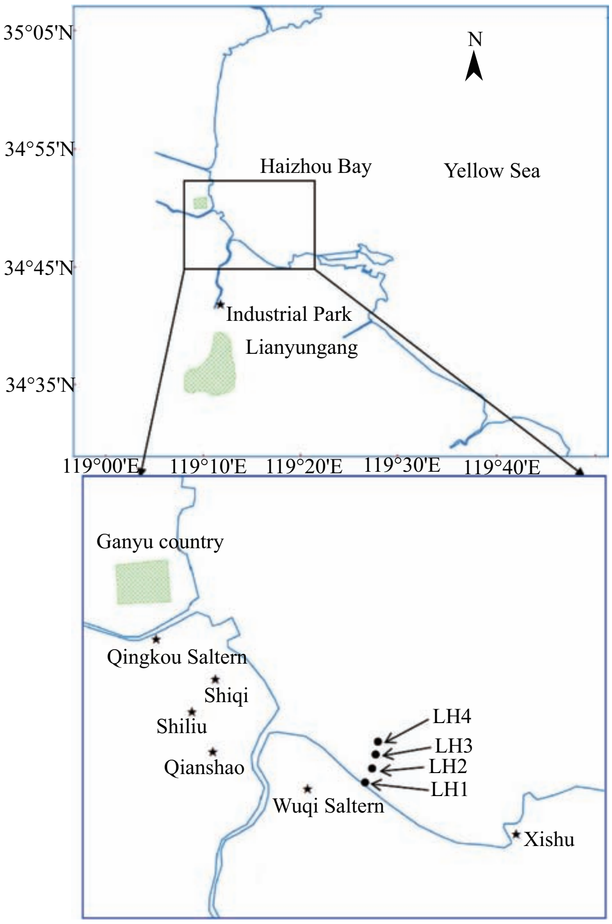

Fig. 1 Sampling site map.

activities in the surrounding area, including reclamation, the construction of harbors and dikes, the dredging of a channel, maritime transport and aquaculture (Chen, 2006). However, the urban infrastructure needed to support the population has not been developed, and substantial amounts of sewage are discharged into the sea or the Linhong River without treatment.

For more than 40 years, various industrial activities near Haizhou Bay have significantly contaminated the coastal tidal flats. A few studies have reported the geochemical composition and the characteristics of both major and trace elements in sediment cores (Zhang et al., 2008). However, there are limited data on historical toxic metals inputs to the tidal flats of Haizhou Bay. This gap needs to be filled because this information will help to predict future trends in the marsh sediment quality and to decide if further action is required to accelerate the recovery of the ecosystem in Haizhou Bay. Moreover, the levels and the orientation of industrialized development near Haizhou Bay in Lianyungang provide an interesting contrast for understanding pollutant sources and distribution mechanisms in similar aquatic environment. This work aims to reconstruct historical changes in terms of heavy metal (Cd, Cr, Cu, Mn, Pb, and Zn) pollution in the tidal flat of Haizhou Bay.

1 Materials and methods

1.1 Sampling and chemical analysis

Four sediment cores were manually collected from the south tidal flat of Haizhou Bay, near the Linhong River estuary, in April 2010. The cores were labeled LH1 (34.774°N, 119.247°E), LH2 (34.778°N, 119.249°E), LH3 (34.782°N, 119.251°E) and LH4 (34.787°N, 119.252°E), and their locations are shown in **Fig. 1**. In the laboratory, these cores, which ranged from 12 to 65 cm in length and which were 7.5 cm in diameter, were subsampled at 1 cm intervals. Each layer was divided for analysis of the metals, the grain size and the ^{210}Pb radionuclide.

The analytical procedures for the measurement of metal concentrations in core sediments are similar to those for soils and sediments. The samples were completely digested by a micro-wave assisted digestion method (1200 W, 180°C, 1 MPa during 30 min) with concentrated HF, HNO_3 and HCl acids in Teflon bottle, and then the samples were placed on a hot plate to evaporated until dry. The residue was solubilised by 5% HNO_3 and diluted to volume. The metal concentrations were determined by an iCAP 6300 inductively coupled plasma optical emission spectrometer (ICP-OES, Thermo Electro Co., USA). The accuracy of the analytical method was established with the standard reference material aquatic sediments GBW07312 (GSD-12). Based on the analysis of the standard reference material, the recovery for each of the analyzed elements were as follows: Al (97%–105%), Cd (91%–110%), Cr (95%–103%), Cu (95%–105%), Mn (98%–106%), Pb (93%–108%) and Zn (99%–105%). Three replicates of each sample were run, and the following coefficients of variation were obtained: Al, 0.44%; Cd, 7.02%; Cr, 5.97%; Cu, 0.95%; Mn, 0.60%; Pb, 3.04%; and Zn, 2.22%.

In addition, a grain size analysis was performed on a part of each homogenized sample using a laser particle size analyzer (Mastersize 2000, Malvern Instruments Ltd., UK), after the removal of carbonate with 1 mol/L HCl and of organic matter using 30% H_2O_2.

The activities of ^{210}Pb were determined by alpha counting of ^{210}Po deposited onto Ag discs (Flynn, 1968) using ^{209}Po as a yield tracer. The ^{209}Po spike had previously been calibrated against the standard reference material (IAEA-257). The sediment was dissolved by adding a solution of 1:1:0.5 (*V*/*V*/*V*) HNO_3+HCl+HF to 2 g sediment and heating the mixture to 200°C overnight in a closed Teflon bottle. Counting was conducted by computerized multichannel α-spectrometry with gold-silicon surface barrier detectors (Model Octete PLUS, ORTEC Co., USA).

1.2 Statistic analysis

Statistic analysis was carried out using Microsoft Office Excel and the Statistical Package for Social Science (SPSS 16.0 for Windows, SPSS Inc., USA). Nonparametric test was used to determine any significant difference in metal concentrations among different sample sites at the level of $p < 0.05$. Correlations among metal concentrations in the water, soils and plants samples were evaluated using Pearson correlation coefficients.

2 Results and discussion

2.1 Sediment characteristics

The sediments in cores LH3 and LH4 are primarily yellow-brown mud; and then towards the bottom of the cores, the sediments change to a dark grey mud with a few black stripes that contains some roots and some shell fragments. The sediments of cores LH1 and LH2 are brown and dark brown without any observable change in the visual or physical structures. The four cores consist of relatively uniform fine-grained materials, with the percentage of silt (4–63 μm) and clay (< 4 μm) exceeding 95% (**Fig. 2**). The silt fraction ranges from 62.5% to 95.9%, and the clay fraction ranges from 2.0% to 27.7%. The percentage of sand is very small in these cores.

2.2 Sedimentation rate and age of cores LH3 and LH4

Figure 3 shows the vertical distributions of the excess ^{210}Pb for the cores LH3 and LH4. The supported ^{210}Pb activity (derived from the decay of its effective parent, ^{226}Ra, in the seabed) is generally determined by measuring the ^{210}Pb activity at the bottom of a core that exhibits low accumulation rates, such that sediment is greater than 100 years old. However, the bottom layers of the four cores do not approach the depth of supported ^{210}Pb activity, thus the supported ^{210}Pb activity was approximated to be 1 dpm/g (decays per minute per gram), as taken from previous studies (Zhang et al., 2008; Chen, 2006). The excess ^{210}Pb activity (^{210}Pb$_{ex}$, the activity particles attain while sinking through the water column) was calculated by subtracting the supported activity from the total activity. The excess

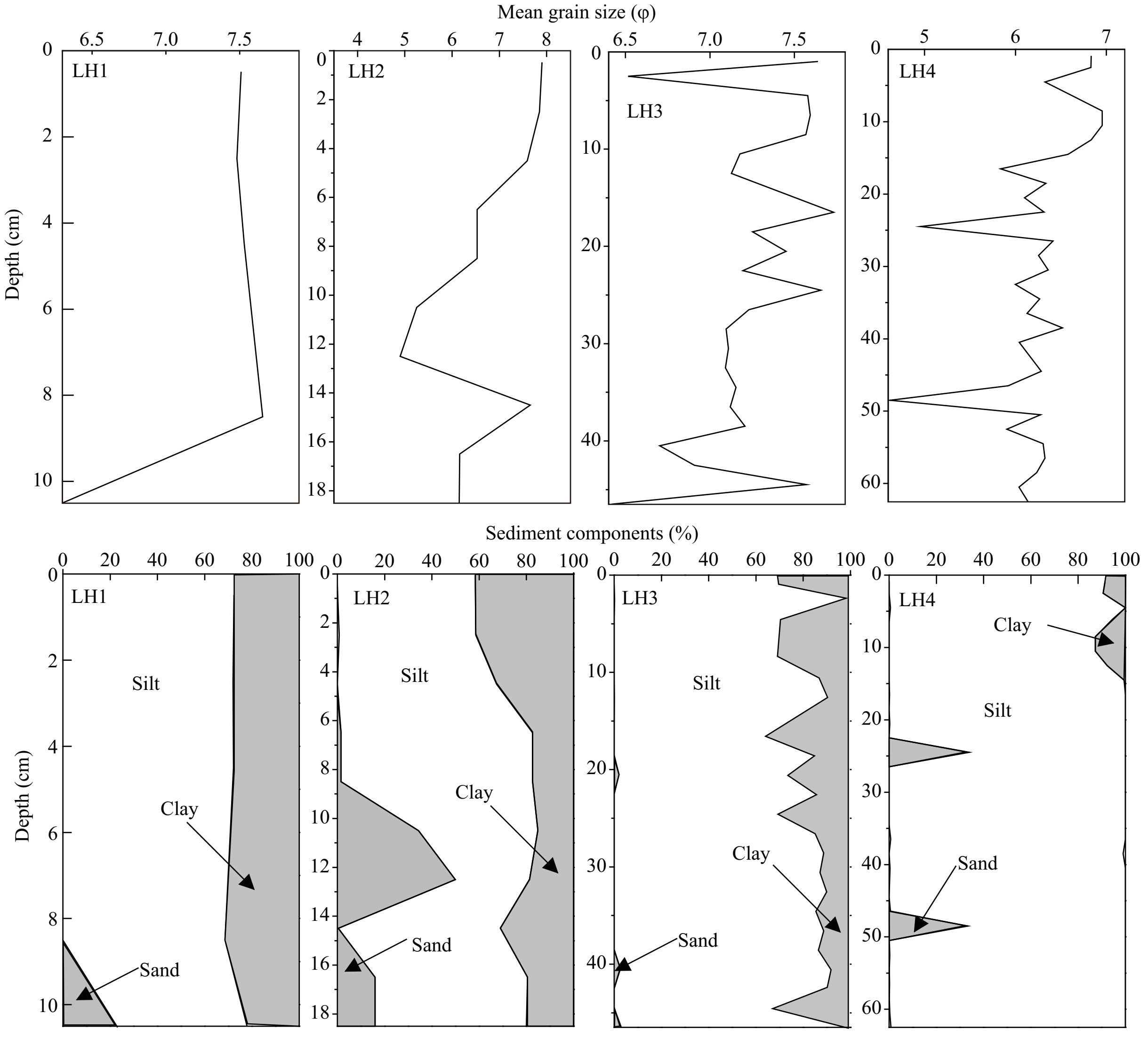

Fig. 2 Profiles of mean grain size and sediment components in sediment cores from tidal flat in Haizhou Bay. $\varphi = -\log_2 d$, where, d (mm) is the diameter of grain.

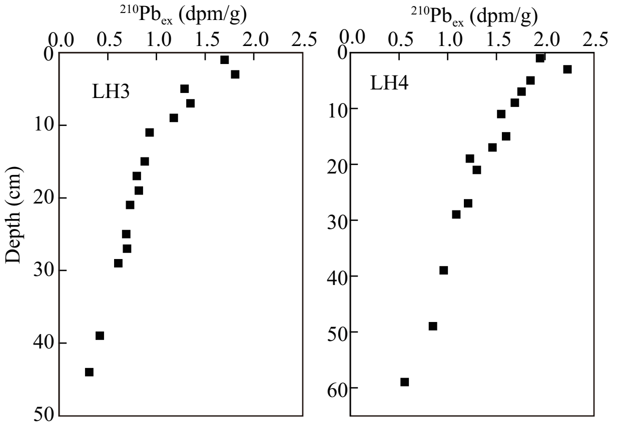

Fig. 3 Depth profiles of excess ^{210}Pb ($^{210}Pb_{ex}$) of cores LH3 and LH4, an exponential decreasing trend with depth of excess ^{210}Pb. dpm/g: decays per minute per gram

^{210}Pb activities exhibit an approximately exponentially decreasing trend with depth (**Fig. 3**), which suggest a constant deposition of excess ^{210}Pb over time at these sites. On the basis of the $^{210}Pb_{ex}$ activities, the constant initial concentration (CIC) model was used to calculate sedimentation rates. The CIC model has been successfully applied in other studies of estuaries and intertidal mudflats (Andersen et al., 2000; Sanders et al., 2010). For this model, the $^{210}Pb_{ex}$ activity (A_z) at any sediment layer (z) with age (t) is expressed as:

$$A_z = A_0 \times e^{-\lambda t} = A_0 \times e^{-\lambda z/S} \quad (1)$$

where, A_0 (dpm/g) is the $^{210}Pb_{ex}$ at the sediment-water interface, λ is the radioactive decay constant for ^{210}Pb (0.03114 yr^{-1}), and S (cm/yr) is the sedimentation rate. From Eq. (1), S was determined by the slope of the $^{210}Pb_{ex}$ profiles using least squares regression. However, a number of studies have shown that the compaction on sediment layers may cause an incorrect depth (z) (Lu, 2007). An alternative method, which expresses $^{210}Pb_{ex}$ as a function of massdepth removes this compaction effect (Lu, 2007). Thus, Eq. (1) can be rewritten as:

$$A_m = A_0 \times e^{-\lambda t} = A_0 \times e^{-\lambda m/r} \quad (2)$$

where, m (g/cm^2) is the mass depth of the cumulative dry weight at the sediment layer (z), r (g/(cm^2·yr)) is the sediment accumulation rate, A_m (dpm/g) is the $^{210}Pb_{ex}$ at the sediment layer (m), and A_0 is the $^{210}Pb_{ex}$ at the sediment-water interface layer. From Eq. (2), r was determined by the slope of the $^{210}Pb_{ex}$ profiles using least squares regression.

Finally, the ^{210}Pb chronologies of the two methods were compared to derive the final chronology. The results by the two methods agreed well, it means that the compaction did not have a significant effect on the ^{210}Pb chronologies in the two sediment cores. Therefore, the result of the sediment chronology was unique and was reported by the least squares regression from Eq. (1). The mean sedimentation rate was calculated to be 1.5 cm/yr for core LH4, and 0.85 cm/yr for core LH3. The sediments in these cores have accumulated over approximately the last 50 years at a constant sedimentation rate. The deepest layer in core LH4, at a depth of 63 cm, was estimated to have been deposited in 1965. For core LH3, the layer at a depth of 47 cm corresponds to 1950. Therefore, these cores should contain records related to the anthropogenic impacts of the pollutants from Lianyungang City. There have been several reports of ^{210}Pb dating of cores from Haizhou Bay. One core from the northern area near the estuary of the Linhong River was determined to have a sedimentation rate of 0.5 cm/yr (Zhang et al., 2008), while another core near Xishu Cape had a rate of 1.15 cm/yr (Peng and Chen, 2010). The variability of the three cores reflects the fact that the sedimentation rate is relatively high in the southern area of Haizhou Bay and decreases gradually moving northwards. According to a previous study (Wang et al., 1980), the southern tidal flat of Haizhou Bay is a rapid sedimentation area with a high average sedimentation rate between 1.0 and 2.5 cm/yr, which is due to the sediment loads from the Linhong River, as well as the abundant sediment from the Old Yellow Delta carried by northward coastal currents.

2.3 Metal contents in the deposits

Figure 4 shows the vertical distribution of heavy metals in the cores. The concentration of these elements in the deepest layer of core LH4 is almost equal to that of sediment without anthropogenic contamination. Except for core LH1, the heavy metal concentrations increased moving from the bottom to the top, and the maximum values were observed at depths of 5–20 cm. In the upper layer, the concentrations decreased at the surface. The vertical profiles of the heavy metals reflect the historical trend of the load produced by human activity in the coastal area of Haizhou Bay, as will be discussed in the following sections.

The correlation matrices for samples were calculated to examine the relationships between the elements analyzed (**Table 1**). Anthropogenic Cd, Cr, Cu, Mn and Zn show statistically significant ($p < 0.05$) correlation among them. They most likely have the same origin and are influenced by the same mechanisms and processes. The main sources of heavy metals in soils from anthropogenic activities are atmospheric deposition, the waste from industrial and domestic activities and traffic-related emissions in urban areas (Zourarah et al., 2007; Liu et al., 2003). There are positive correlations between Pb and the other elements, with coefficients ranging from 0.34 to 0.46. These relationships indicate that Pb originates from a different source than the other elements. Anthropogenic Pb comes mainly from non-point sources such as atmospheric transport and precipitation, which incorporate Pb that originated from coal burning in power generation plants and other industri-

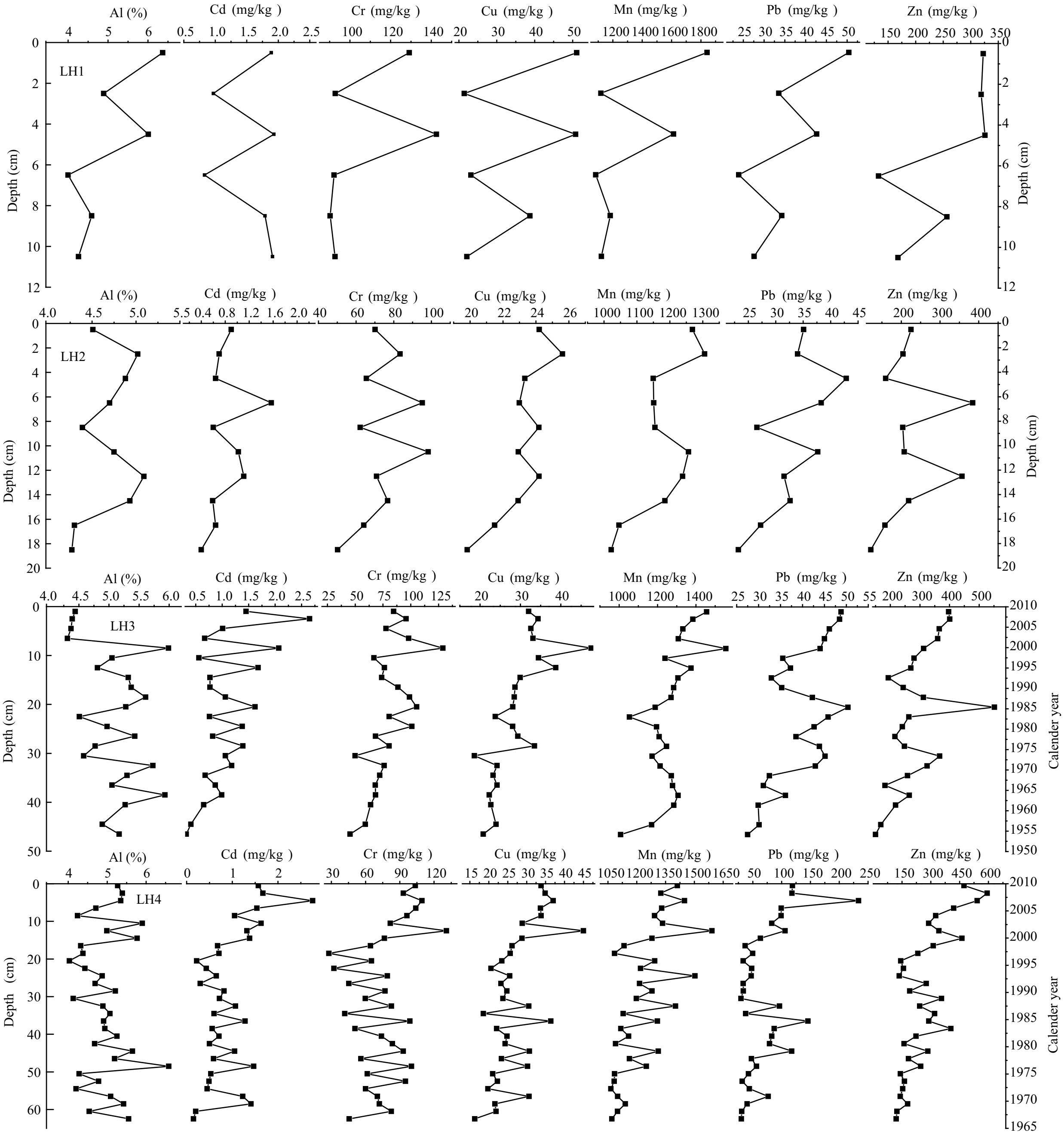

Fig. 4 Profiles of elements in sediment cores from tidal flat in Haizhou Bay.

al activities (Li et al., 2000; Liu et al., 2003; Ip et al., 2004). It is well known that trace metals tend to be adsorbed onto the surfaces of particles and that higher concentrations of metals are usually associated with smaller particles (Schiff and Weisberg, 1999; Tang et al., 2008). In the tidal flat of Haizhou Bay, the trace metals, including Cd, Cr, Cu, Mn, Pb and Zn, showed no significant correlation with the percentage of fine-grained particles in the sediments ($p > 0.05$) (**Table 1**). Therefore, anthropogenic inputs were likely a more significant control on the distribution of trace metals in the tidal flat sediments than natural geochemical processes.

2.4 Enrichment of anthropogenic heavy metals

Since the 1970s, the tidal flats have been at risk of heavy metal pollution from industrial and agricultural discharge and from domestic wastewater due to rapid economic development in Lianyungang City near Haizhou Bay. In previous studies, the heavy metals contamination of the Haizhou Bay sediments has been demonstrated by increases in their concentrations (Zhang et al., 2008). The possibility of sediment contamination was assessed through the calculation of the metal enrichment factor (EF), which is the observed metal/normalizer ratio in the sample divided by the metal/normalizer ratio reported for

Table 1 Comparisons of concentrations of metals (mg/kg) in surface and cores in tidal flat from Haizhou Bay with background values

		Cd	Cr	Cu	Mn	Pb	Zn
Surface	LH1	1.89	129.18	51.07	1841.84	50.41	322.76
	LH2	0.90	70.10	24.20	1268.59	35.08	225
	LH3	1.45	84.63	32.04	1455.72	48.81	396.97
	LH4	1.57	103.58	33.82	1411.27	118.82	472.75
Average in sediment cores	LH1	1.55	107	34.6	1327	35.4	254
	LH2	0.83	73.66	23.17	1177	32.9	224.06
	LH3	1.08	80	28.8	1260	39.4	284
	LH4	0.93	75.4	27.5	1246	73	286
CS[a]		0.365	60.28	15.84	–[e]	24.7	64.68
SYS[b]		0.088	64	18	570	22	67
MS[c]		0.06	15	8	–[e]	21	51
PB[d]		1.0	90	50	–[e]	70	175

[a] Geochemical natural background of coastal soil in Jiangsu (Xia et al., 1987); [b] background of sediments in Yellow Sea (Chi and Yan, 2007); [c] background of marine sediments in Hong Kong (EPD, 1992); [d] natural, preindustrial background values (Hakanson, 1980); [e] no data.

a reference material (Ip et al., 2004; Li et al., 2010). The EF was calculated as the ratio of the elemental concentration in the sediment to its natural background concentration (Tanner et al., 2000; Zhang and Liu, 2002; Feng et al., 2004; Selvaraj et al., 2004; Shi et al., 2010). A reference element was employed in the calculation to account for the lithogenic influence of granulometric and mineralogical variations of the sediment. Most commonly, Fe and Al are used (Laluraj and Nair, 2006; Aksu et al., 2010). The EF in this study is defined as

$$\mathrm{EF} = (C_{\text{M-sed}}/C_{\text{R-sed}})\Big/(C_{\text{M-bk}}\Big/C_{\text{M-bk}}) \qquad (3)$$

where, $(C_{\text{M-sed}}/C_{\text{R-sed}})$ is the ratio of the heavy metal concentration $(C_{\text{M-sed}})$ to concentration of the reference element $(C_{\text{R-sed}})$ in the sediment, and $(C_{\text{M-bk}}/C_{\text{R-bk}})$ is the concentration ratio for the natural background. The natural geochemical background refers to the elemental concentrations in sediments without any anthropogenic enrichment; i.e., the elemental concentrations that are fully derived from natural sources such as terrigenous and biogenic sources (Rubio et al., 2000; Liaghati et al., 2003; Cobelo-Garcia and Prego, 2003; Mil-Homens et al., 2006; Shi et al., 2010). In previous studies, the natural geochemical background has not been extensively reported for Haizhou Bay, because a short core is not sufficient to reconstruct a geochemical history unaffected by any human influence. Therefore, the natural geochemical background of the coastal soil in Jiangsu (Xia et al., 1987) was taken as a background value for Haizhou Bay and is listed in **Table 2**.

In general, EF values from 0.5 to 2.0 can be considered to be in the range of natural variability, indicating that the sediment is unaffected by human activities, and EF values greater than 2.0 represent heavy metal enrichment and contamination of the sediment as a result of significant anthropogenic input (Sutherland, 2000; Shumilin et al., 2002; Liaghati et al., 2003).

Figure 5 summarizes the EFs calculated in this study, and trace metals are divided here in two categories. For the first group, the EFs of Cr and Cu range from 1.5 to 2.3, suggesting no or minimal anthropogenic enrichment. Both EFs present relatively low and constant values over the entire profiles. These results underline the facts that these elements have primarily lithogenic origins and that no significant anthropogenic source was detectable in all of the studied sites. For the other metals, the EF values range between 2 and 13 and follow the order Zn > Pb > Cd > Mn, indicating predominantly anthropogenic input of these elements. The enrichment in sediments of the tidal flat is most likely due to sewage disposal (Liu et al., 2003; Shi et al., 2010). Domestic waste could be a major source of pollution in the sediments. In addition, the emissions from the industrial activities in Lianyungang City significantly influenced the environment of the study area. The sediment cores are located close to the estuary through which the Linhong River flows (**Fig. 1**). This river runs through Lianyungang City, which produces large amounts of industrial and domestic waste that discharge

Table 2 Pearson correlation coefficients between contents of heavy metals and mean grain size in tidal flat sediment from Haizhou Bay

	Mean grain size	Cd	Cr	Cu	Mn	Pb	Zn
Mean grain size	1						
Cd	0.15	1					
Cr	0.21	0.68*	1				
Cu	0.32	0.67*	0.75*	1			
Mn	0.222	0.56*	0.65*	0.81*	1		
Pb	–0.12	0.46*	0.36*	0.40*	0.34*	1	
Zn	–0.20	0.61*	0.41*	0.46*	0.42*	0.52*	1

* Coefficients at 0.05 significance level (2-tailed).

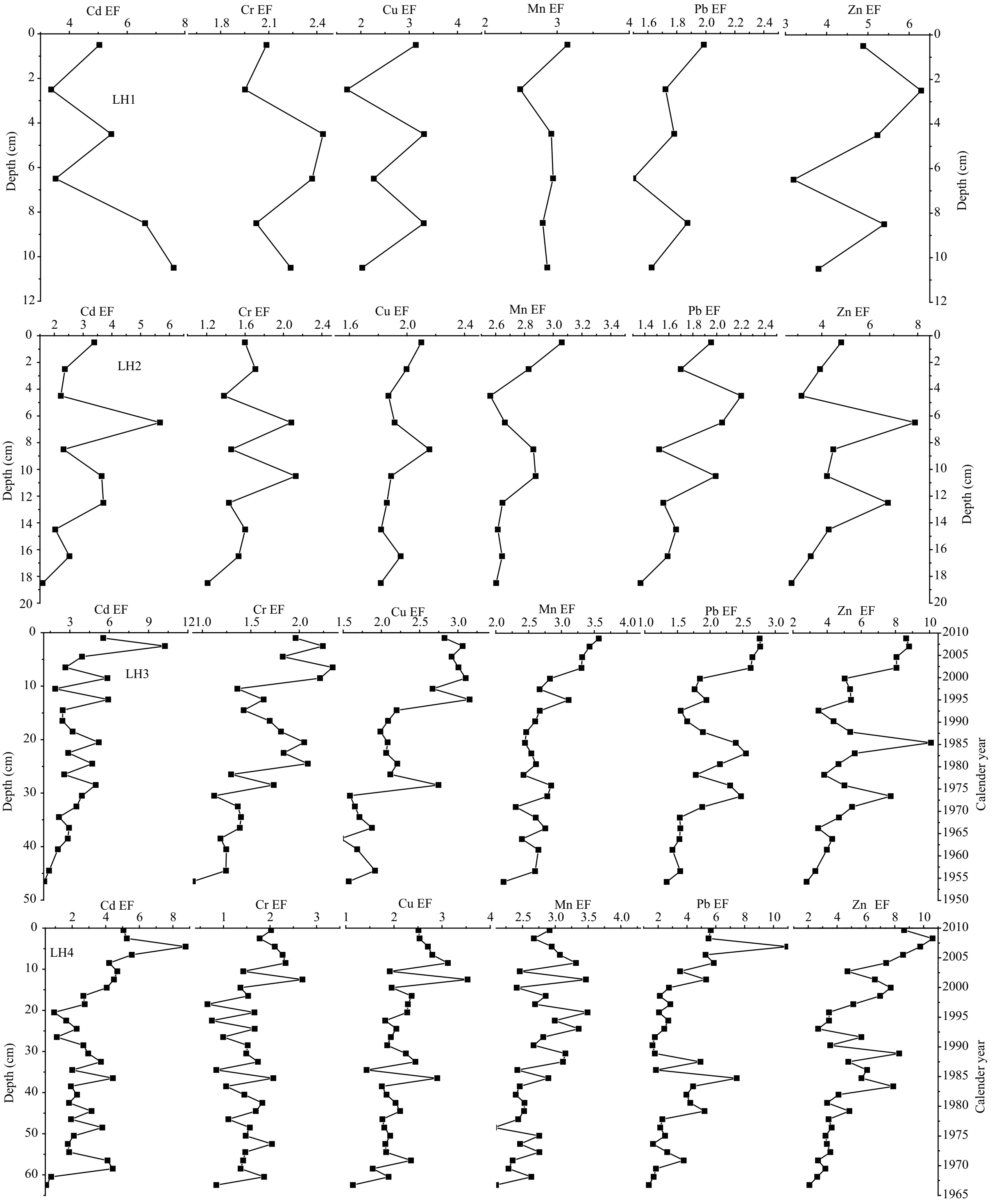

Fig. 5 Profiles of enrichment factors (EF) of heavy metals in sediment cores from tidal flat in Haizhou Bay.

into the river and cause serious heavy metal pollution in the river water, sediment and soil (Zhang et al., 2008). For example, there are many chemical and pharmaceutical factories near the Linhong River and the coastal zone of Haizhou Bay that heavily pollute the soil with heavy metals. In particular, anthropogenic Pb is considered to be primarily distributed by non-point sources such as atmospheric transport and precipitation, which carry Pb that originated from coal burning in power generation plants and emission from ships and traffic (Li et al., 2001; Liu et al., 2003; Ip et al., 2004; Shi et al., 2010). Additionally, these enriched metals may have originated from non-point sources such as agricultural pollution (e.g., fertilizers and livestock manure) and atmospheric transport and precipitation, which contain heavy metals as a result of coal burning in power generation plants, the use of leaded gasoline and other industrial activities (Tug and Duman, 2010). Moreover, domestic and industrial garbage has accumulated in the coastal zone; these deposits then act as point sources of metals (Pb, Zn and Cu) in soils. In addition, the burning of fuel results in widespread lead contamination (Santos et al., 2005)

2.5 Pollution assessment

The index of geoaccumulation (I_{geo}) was calculated to determine the level of pollution using the equation (Muller, 1969):

$$I_{geo} = \log_2(C_n/1.5B_n) \quad (4)$$

where, C_n (mg/kg) is the measured concentration of the element n, and B_n (mg/kg) is the geochemical background value of element n in the coastal soil of Jiangsu (Xia et al., 1987). The factor 1.5 is introduced to include possible variations in the background values due to lithogenic effects. The following classification is given for I_{geo} (Muller, 1969): $I_{geo} \leqslant 0$ essentically unpolluted, $0< I_{geo} < 1$ unpolluted to moderately polluted, $1 < I_{geo} \leqslant 2$ moderately polluted, $2 < I_{geo} \leqslant 3$ moderately to strongly polluted, $3 < I_{geo} \leqslant 4$ strongly polluted, $4 < I_{geo} \leqslant 5$ strongly to extremely polluted, and $I_{geo} > 5$ extremely polluted. The I_{geo} values for Cr, Cu and Mn indicate that the study areas are practically unpolluted by these metals. The high values for Cd, Pb, and Zn in the tidal flat show that the area is unpolluted to moderately polluted by these metals (**Fig. 6**). The I_{geo} values for all of these metals are rated as not significantly harmful, but the tendency for pollution has already been established. Overall, the I_{geo} values reflect the general contamination tendency already showed by the EF values, and hence, the area can be considered to exhibit moderate contamination by Cd, Pb and Zn and weakly contaminated or uncontaminated with respect to Cr, Cu and Mn.

2.6 Modern industrial impact

Dated sediment cores were used to reconstruct the historical metal input to coastal sediments. In this study, we used heavy metal elements of anthropogenic origin to reconstruct the effects of modern industry on sediment quality. **Figures 4** and **5** show the historic profiles of the concentrations and EF values of heavy metals in the cores. In these figures, the vertical axes of cores LH3 and LH4 were expressed as the estimated age of the sediment as determined by the ^{210}Pb method. The surface sediment was from 2010, and the bottom layers were deposited in 1952 and 1967 for cores LH3 and LH4, respectively. The layer with the highest concentration of heavy metals, at a depth of 5–20 cm, was from the period 1990–2005.

In general, the EFs of heavy metals in the core samples tended to increase from the bottom upward (**Fig. 5**). This trend points to the deposition of anthropogenic heavy metals related to the increase of agricultural activity, urbanization and industrialization near Haizhou Bay in the time period spanned by the sediment cores. The history of heavy metal pollution in the tidal flat of Haizhou Bay correlates with the economic growth of Lianyungang City. From 1949 to 1990, the gross industrial output value (GIOV) increased only from 50 million to 559 million RMB Yuan (**Fig. 7**) (SBJ, 2010). This period of slow economic development in Lianyungang is reflected by the slowly rising metal contents in the core samples; in 1978, the “Reform and Open” policy was initiated, which resulted in the higher levels of heavy metals in the 1990s.

The increasing trend of heavy metal pollution continued with the rapid economic growth in the 1990s. The peak heavy metal contamination occurred from the 1990s to the 2000s, and this time period corresponds to the increase in gross domestic product (GDP) and in heavy metal waste output of Lianyungang City (**Fig. 6**). Since the 1990s, Lianyungang has experienced its most rapid industrialization (urbanization). In this period, the GDP has dramatically increased from 4930 million to 45,600 million RMB Yuan (**Fig. 7**) (SBJ, 2010), and at the same time, the demand for energy from fossil fuels for power generation, car and shipping transportation, as well as the corrosion of building materials, urban traffic-related emissions and industrial and domestic waste, have drastically increased the contaminant level and further compromised the environment.

As a consequence of legal restrictions and improvements in waste processing technology, the concentration of heavy metals in the sediment is assumed to have decreased. However, as shown in **Fig. 4** and **Table 2**, the current heavy metal pollution in the tidal flat of Haizhou Bay is still higher than in other sediments, such as local and global background sediments, marine sediments in Hong Kong and sediments in the Yellow Sea, and is higher than the maximum levels in sediments of the pre-industrial period (Xia et al., 1987; Taylor, 1964; EPD, 1992; Chi and Yan, 2007). These trends indicate that the contaminated environment could continue to be a hazard to the health of wildlife dwelling in the tidal flat of Haizhou Bay.

It is not sufficient to reconstruct the heavy metal enrichment and pollution history based on only a few sediment cores and limited dating in such a complicated depositional environment as an estuarine and coastal zone. However, this study clearly demonstrates that heavy metal concentrations have increased over the last several decades and have resulted in significant heavy metal enrichment and pollution in the sediments of the tidal flat of Haizhou Bay in recent years.

3 Conclusions

The short core sediments from the intertidal flat of Haizhou Bay consisted of fine-grained materials. The high mean

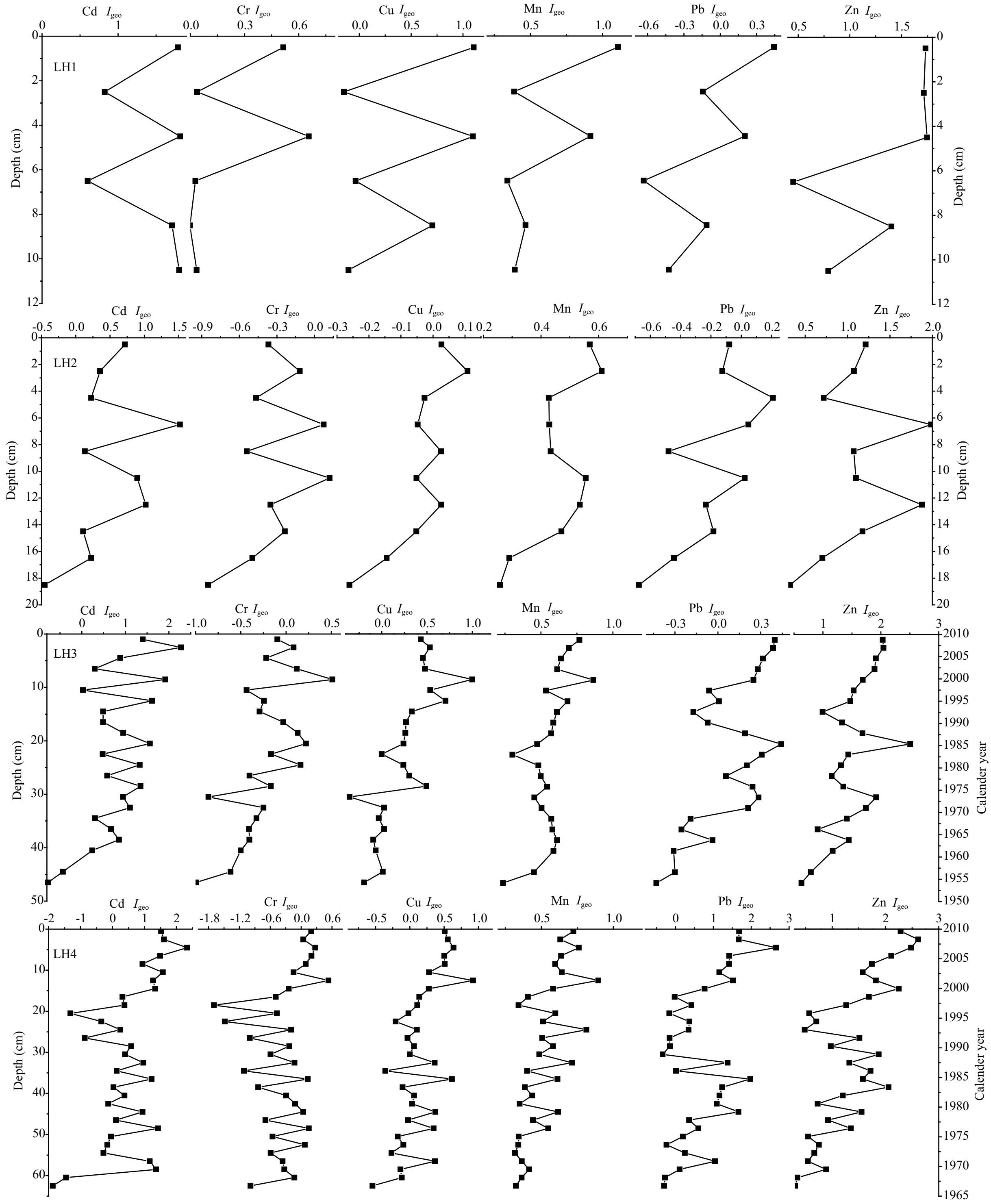

Fig. 6 Index of geoaccumulation for heavy metals in sediment cores.

sedimentation rates of 0.85 and 1.5 cm/yr were calculated for cores LH3 and LH4, respectively, and with the aid of with ^{210}Pb dating, these sediments were used to reveal the recent evolution of heavy metal pollution. This study of trace metal abundances and distributions in the sediment cores shows that the historic (< 50 years) metal pollution, which originated from anthropogenic activities rather than lithogenic geochemical sources, can be directly related to the socio-economic development of the area.

This study has confirmed that the heavy metal pollution in Haizhou Bay has been and continues to be strongly influenced by human activity in the coastal area.

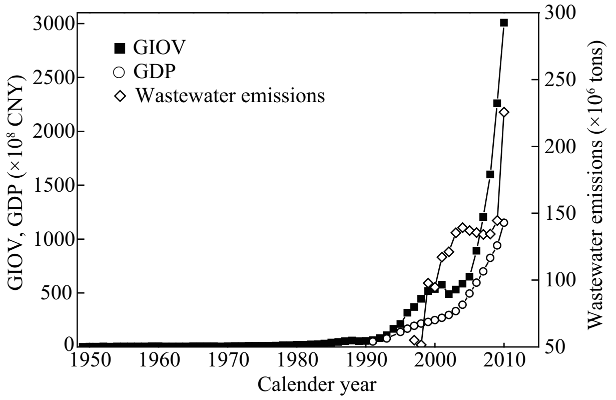

Fig. 7 Historical economic development and wastewater emissions of Lianyungang in past 50 years. GIOV and GDP represent gross industrial output value and gross domestic product, respectively.

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