

Vertical profile, contamination assessment of mercury and arsenic in sediment cores from typical intertidal zones of China

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Abstract The vertical profiles, contamination levels, and potential ecological risks of mercury and arsenic were studied from the sediment cores of seven typical intertidal zones, including the Liaohe River Estuary, the Jianhe River Estuary, the Dagu River Estuary, Yancheng Shoal, the Dongtan Yangtze River Estuary, Hangzhou Bay, and the Pearl River Estuary. Marine sediment quality standards, the threshold effect level (TEL), and the probable effect level (PEL) were used as guidelines to evaluate sediment quality. In addition, the geo-accumulation index (I_{geo}) and potential ecological risk index (E_r^i) were used to assess contamination and potential ecological risks from mercury and arsenic. The results showed that the Pearl River Estuary was moderately polluted by mercury and represented a high potential ecological risk, while other areas were uncontaminated or mildly contaminated with low or moderate potential ecological risks. The Pearl River Estuary was mildly polluted by arsenic

and represented a mild potential ecological risk, while other areas were unpolluted and also posed a mild potential ecological risk.

Keywords Mercury · Arsenic · Sediment cores · Contamination assessment · Intertidal zones

Introduction

Mercury, as an important trace metal, has generated widespread concerns due to its high toxicity, persistence, bio-accumulation, and mobility, and it may produce adverse biological effects and toxicity in the environment (Zhang et al. 2017). Due to its particular chemical and physical characteristics, mercury can go through a variety of environmental reactions and processes, resulting in a complicated geochemical cycle. Mercury originating from anthropogenic and natural sources finally reaches marine environment (Araujo et al. 2017). Arsenic, as a toxic metalloid, has acute and chronic toxic carcinogenic effects on both aquatic and humans (Wang et al. 2016a). Elevated levels of arsenic are of particular concern because of their potential toxicity and prevalence (Gao et al. 2017). Arsenic is discharged into the environment through natural and anthropogenic activities, including emissions and wastewater from ore-mining and ore-processing industries, dye manufacture, tanneries, thermal power plants, and the application of certain insecticides and herbicides (Benzer 2017). Heavy metals, including mercury and arsenic released into environment, are eventually

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distributed between the aqueous phase and marine sediments through adsorption, hydrolysis, and coprecipitation (Delshab et al. 2017). Sediments are important sinks for heavy metals, which makes them useful in the assessment of metallic contamination (Sarasiab et al. 2014). For this reason, the evaluation of metal distribution and contamination in surface and core sediment is of vital importance to evaluate marine environments (Kumar et al. 2013).

As the interface of marine and terrestrial environments, intertidal zones have important hydrological and ecological functions and play crucial roles as habitats for tidal flat organisms and in the removal of aquatic pollution due to intensive industrial and agricultural activities and highly populated areas close to the coast (Qian et al. 2016). Pollutants such as surfactants, heavy metals, nutrients, oils, and persistent compounds discharged into intertidal zones can lead to serious environmental hazards (Traverso-Soto et al. 2015). Core sediments have proven to be an effective tool for establishing the effects of anthropogenic and natural processes on depositional environments, and they can be used to study the contamination history of aquatic ecosystems (Li and Li 2017). Metals that enter aquatic systems are usually transported into sediments and can later be released to represent a secondary contamination source under changing environmental conditions (Gu and Lin 2016). Therefore, sediments are both carriers and sources of heavy metal pollutants in marine systems. It is of vital importance to study the contamination levels and potential ecological risks of trace metals using sediment cores from intertidal zones.

To date, there have been many studies evaluating metal contamination and potential ecological risks in sediment cores from certain region, and they have mainly concentrated on the distribution, contamination, and chronology of heavy metals (Chen et al. 2016; Duan et al. 2013; Gu and Lin 2016; Wang et al. 2016b). However, few studies have focused on mercury and arsenic levels in sediment cores from typical intertidal zones of China, including the Liaohe River Estuary, the Jianhe River Estuary, the Dagu River Estuary, Yancheng Shoal, Hangzhou Bay, the Dongtan Yangtze River Estuary, and the Pearl River Estuary. Comprehensive research on the heavy metal pollution in sediment cores from these zones is still needed.

The following indices have been broadly applied to assess the degree of contamination of heavy metal contamination in sediments derived from human-made or natural sources and its potential biological effects:

enrichment factor (EF), geo-accumulation index (I_{geo}), pollution load index (PLI), potential ecological risk factor (E_r^i), modified pollution index (MPI), and more (Duodu et al. 2017). In this study, I_{geo} and E_r^i were used to assess heavy metal contamination of heavy in view of the strengths and weaknesses of the two methods (Liu et al. 2016). The primary objectives of this study were to (1) estimate heavy metal contents (Flemming and Delafontaine 2000) and evaluate their contamination levels in typical intertidal sediment cores in China, (2) evaluate the potential ecological risks of heavy metals in sediment cores, and (3) analyze the distribution and possible sources of heavy metals in intertidal zones.

Materials and methods

Study area

China's eastern coastline is 18,000 km long with complex geology. The coastline spans various climate zones and is affected by different levels of economic development. Therefore, seven typical intertidal zones were selected as the study areas to explore the quality of sediments along China's eastern coastline. The intertidal zone of the Liaohe River Estuary, located in the southwest of Panjin, Liaoning Province, is the world's second largest coastal reed wetland area, belonging to a typical alluvial plain. The region is the home to the Liaohe oilfield and is affected by human activities, such as oil production and reclamation, causing serious environmental pollution and wetland degradation. The Jianhe River Estuary is located in the northeast of Tianjin, and is a typical intertidal zone under intensive influence of human activities due to the rapid development and large-scale reclamation. The Dagu River intertidal zone is located along the northern shore of Jiaozhou Bay in Qingdao, and is seriously affected by industrial and agricultural pollution. Yancheng Shoal in Jiangsu belongs to China's largest plain silt muddy tidal flat and exhibits clear zonal characteristics, making it convenient for comparing different sediment deposition areas and environmental quality. The Dongtan Yangtze River in Chongming is the largest and the most developed river mouth tidal mudflat wetland, with a complete distribution of tidal flat vegetation distribution, and is a typical river muddy intertidal zone in China. Hangzhou Bay, the most typical type of strong tidal bay silt mass

intertidal zone, is located in the Qiantang River Estuary, a strong tidal estuary type. The Pearl River Estuary is one of China's earliest areas for industrialization, urbanization, and development, and is the area most affected by the human activities. Research on mercury and arsenic of sediment cores from typical intertidal zones can provide data support for the protection of the environment in intertidal zones.

Sediment sampling and pretreatment

Sediment cores were collected using a sediment corer with a length of about 100 cm and sliced at 2-cm interval immediately after collection with a plastic cutter, and

then stored in clearly labeled polyethylene bags that were refrigerated at $-20\text{ }^{\circ}\text{C}$ until laboratory analysis (Fig. 1). Of the cores from the seven typical intertidal zones, the sediment cores from the Liaohe River Estuary, the Jianhe River Estuary, and Yancheng Shoal were collected by the First Institute of Oceanography; sediment cores from the Dagu River Estuary was collected by the Ministry of Land and Resources of the Qingdao Institute of Marine Geology; sediment cores from Hangzhou Bay and the Dongtan Yangtze River Estuary were collected by the East China Normal University; and those from the Pearl River Estuary were collected by the Yantai Institute of Coastal Zone Research Chinese Academy of Sciences.

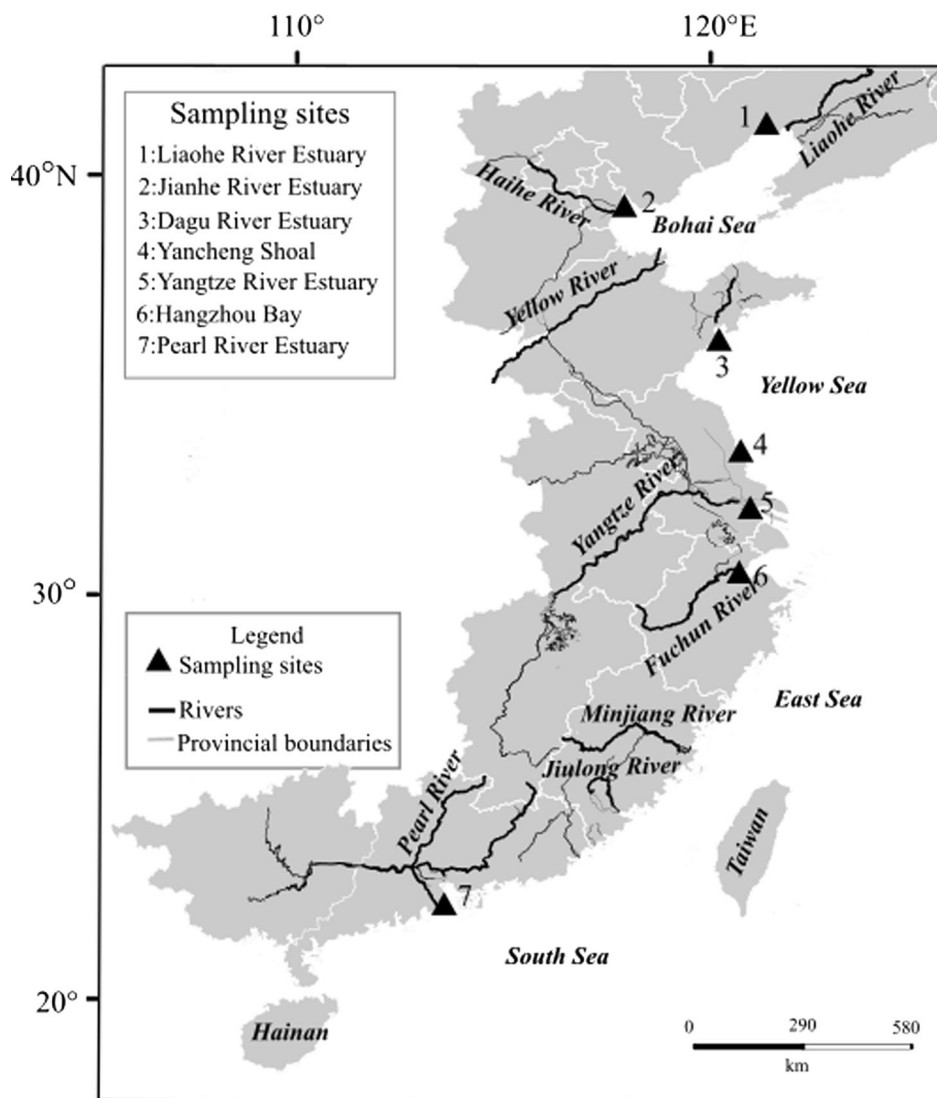


Fig. 1 Location of the study areas

The frozen samples were dried in a lyophilizer (Germany Christ) and ground using an agate pestle and mortar, and then sieved through 95- μm mesh for homogenization (AQSIQ 2007). The sediment samples were then digested using an acid mixture ($\text{HCl}:\text{HNO}_3:\text{H}_2\text{O} = 1:3:5$) in a boiling water bath and were finally analyzed using atomic fluorescence spectrometry (Beijing Ji Tian Instrument Co., AFS-930) for mercury and arsenic determination. A standard reference material (GBW07309) was used to assess measurement accuracy and the results were in accordance with reference values, and the results were listed in Table 1. Reagent blanks and sample replicates were also used for background correction and the verification of analysis precision. Due to the large quantity of samples, replicates were performed for every 10 sediment samples, which demonstrated that the analysis method was adequate. The basic data of sediment grain size, organic carbon contents, and sediment accumulation rates were shown in the supporting information (Tables S1 and S2). All glass and plastic utensils had been soaked overnight in HNO_3 (10% v/v) and rinsed thoroughly with Milli-Q water prior to use. All reagents were guaranteed analytical reagent or at a higher level.

Results and discussion

Contents and vertical profiles of mercury and arsenic of sediment cores

The mercury contents of the intertidal sediment core from the Liaohe River Estuary ranged from 0.02 to 0.13 mg/kg and the average concentration was 0.07 mg/kg (Fig. 2 and Table 3), which was lower than the class I sediment category and the threshold effect level (TEL) (Table 2). This demonstrated that the mercury content of the intertidal sediment core from the Liaohe River Estuary was low and would be unlikely to cause negative biological effects. From the top to the bottom of the sediment core, the content of mercury had

a tendency to increase at a depth of 0–38 cm, reaching the highest in the depth of 38 cm (0.13 mg/kg), and the concentration of mercury decreased in the depth of 38–110 cm. Arsenic contents ranged from 5.79 to 13.48 mg/kg and the average concentration was 9.98 mg/kg (Fig. 3 and Table 3), which was lower than the class I sediment category and higher than the TEL. These results implied that arsenic would have negative biological effects on the aquatic system. The arsenic content at 0–60-cm depth was higher than that at 62–110-cm depth, and contents varied little in both layers. These results revealed that the human-caused contributions of mercury and arsenic in the Liaohe River Estuary declined recently.

Mercury contents of the intertidal sediment core from Hangu the Jianhe River Estuary ranged from 0.03 to 0.08 mg/kg and the mean concentration was 0.03 mg/kg (Fig. 2 and Table 3), which was lower than the class I sediment category and TEL. These results implied that mercury would not cause negative biological effects on the surrounding aquatic systems. There was no clear change in mercury content except for the high value of 0.08 mg/kg at a depth of 24 cm. Arsenic contents of the intertidal sediment core from the Jianhe River Estuary ranged from 8.60 to 15.76 mg/kg (Fig. 3 and Table 3) and the average concentration was 12.21 mg/kg, which was lower than the class I sediment category and higher than the TEL. From the top to the bottom of the sediment core, the arsenic content tended to decrease and changed little within the depth range of 10 and 86 cm. The vertical profiles of the mercury contents were relatively constant, whereas the concentration of arsenic decreased from the upper to lower layers in the depth of 0–14 cm, indicating that As had continuously accumulated in sediment in recent years.

The mercury contents of the intertidal sediment core in the Dagu River Estuary of Qingdao ranged from 0.01 to 0.06 mg/kg with an average concentration of 0.04 mg/kg (Fig. 2 and Table 3), far lower than the class I sediment category and the TEL. From the top to the

Table 1 Results of an analysis of certified reference materials (GBW07364)

Element	GBW07364			
	Measured values	Certified values	Standard deviation (SD)	Coefficient of variation (CV%)
As	19.20 \pm 1.90 mg/kg	18.85 mg/kg	0.53 mg/kg	2.79
Hg	25.00 \pm 5.00 $\mu\text{g}/\text{kg}$	24.00 $\mu\text{g}/\text{kg}$	2.00 $\mu\text{g}/\text{kg}$	9.44

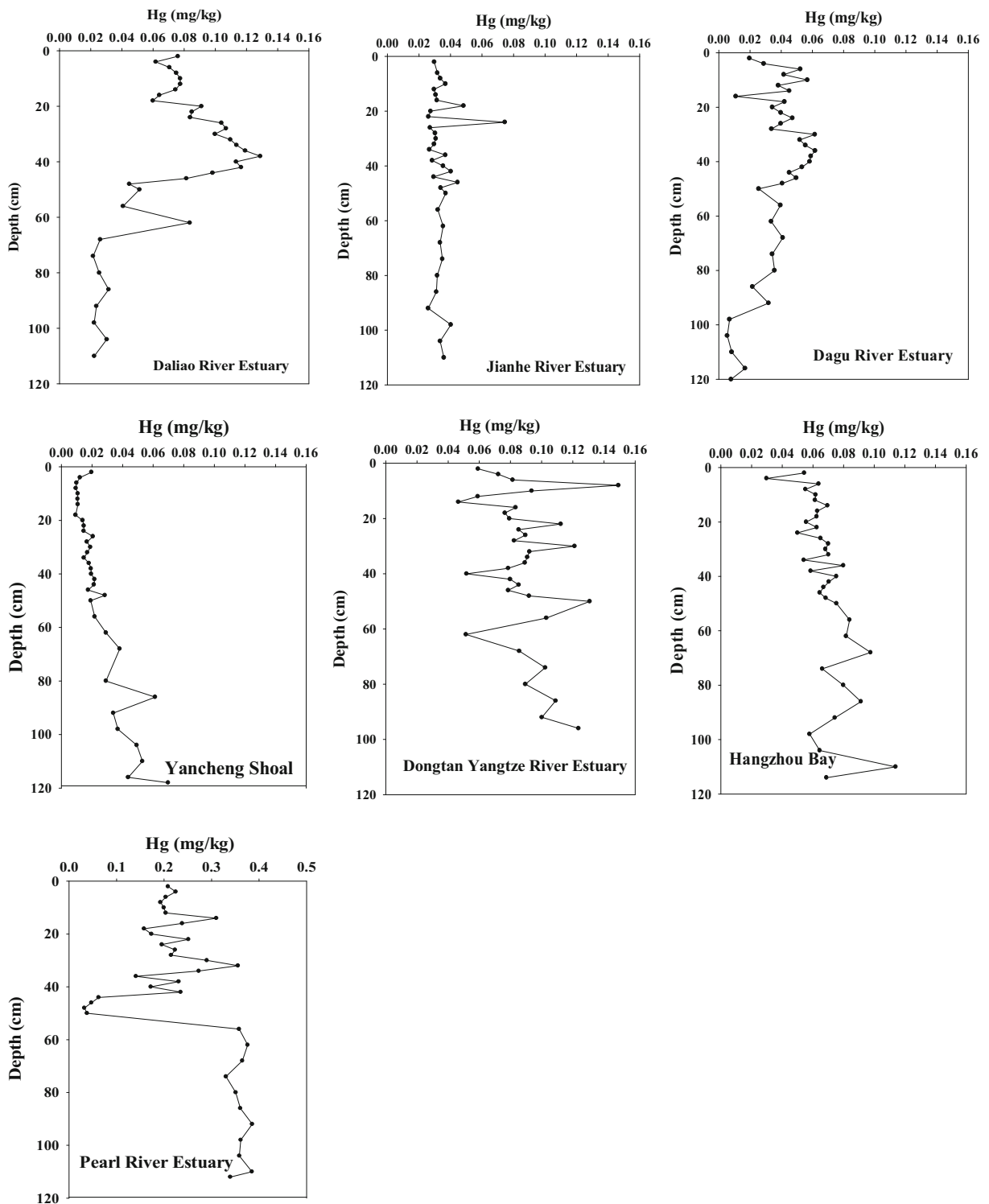


Fig. 2 Vertical profiles of mercury contents of sediment cores in seven typical intertidal zones

bottom of the sediment core, the mercury content tended to decrease and exhibited small variations in the depth of

0–40 cm. Arsenic contents ranged from 5.40 to 12.19 mg/kg with an average concentration of

Table 2 Sediment quality guidelines

Sediment quality guidelines (SQDS)	As (mg/kg)	Hg (mg/kg)	Description	References
Marine sediment quality standards	Class I 20.00	0.20	Suitable for mariculture, nature reserve, endangered species reserve, and recreational activities	SEPA (2002)
	Class II 65.00	0.50	Suitable for industry and tourism sites	
	Class III 93.00	1.00	Suitable for harbors	
Threshold effect level (TEL)	7.30	0.13	Metal concentrations in sediments below which adverse effects on biota are rarely observed	R. Long and D. Macdonald (1995)
Probable effect level (PEL)	41.60	0.70	Metal concentrations in sediments above which adverse effects on biota are frequently observed	

9.04 mg/kg (Fig. 3 and Table 3), which was lower than the class I sediment category and higher than the TEL. The overall arsenic concentration decreased from the upper to lower layers, with small variability in the layer at a depth of 0–30 cm. The vertical profiles of mercury and arsenic indicated that recent anthropogenic activities posed less effect on the study area.

Unlike the three prior sediment cores, the overall mercury contents of the intertidal sediment core from Yancheng Shoal tended to increase from the top to the bottom layers. Mercury content ranged from 0.01 to 0.07 mg/kg with an average value of 0.02 mg/kg (Fig. 2 and Table 3), which indicated an absence of mercury pollution. Arsenic contents in this study area ranged from 8.08 to 16.07 mg/kg with an average concentration of 10.66 mg/kg (Fig. 3 and Table 3), which was lower than the class I sediment category and higher than the TEL. Similar to the mercury concentration trend, the arsenic content tended to increase dramatically from the upper to lower layers. These results indicated that human activities hardly affected the environment of Yancheng Shoal and that pollution control work may have been more successful than in other areas.

Mercury contents from the intertidal sediment core in the Dongtan Yangtze River Estuary ranged from 0.05 to 0.15 mg/kg with an average concentration of 0.09 mg/kg (Fig. 2 and Table 3), which was lower than the class I sediment category and the TEL except for several samples. From the top to the bottom of the sediment core, the mercury content tended to increase, and the overall mercury contents were widely variable. Arsenic contents ranged from 5.10 to 11.74 mg/kg with an average concentration of 9.43 mg/kg (Fig. 3 and Table 3), which was lower than the class I sediment category and higher than the TEL. Arsenic content levels at depths of 14, 40, 62, and 92 cm were low (6.01, 5.10, 6.47, 7.17 mg/kg); otherwise, the overall content of arsenic changed little across the samples.

Mercury contents of the intertidal sediment core from Hangzhou Bay ranged from 0.03 to 0.11 mg/kg with an average concentration of 0.07 mg/kg (Fig. 2 and Table 3). The contents of the samples increase with depth, which is not the expected variation tendency. That may be due to the different hydrodynamic mechanisms caused by the discharges of heavy metals from different rivers and tides and varied characteristics of sediment cores (Chen et al. 2016). Arsenic contents ranged from 5.72 to 12.87 mg/kg with an average concentration of 10.03 mg/kg (Fig. 3 and Table 3), which

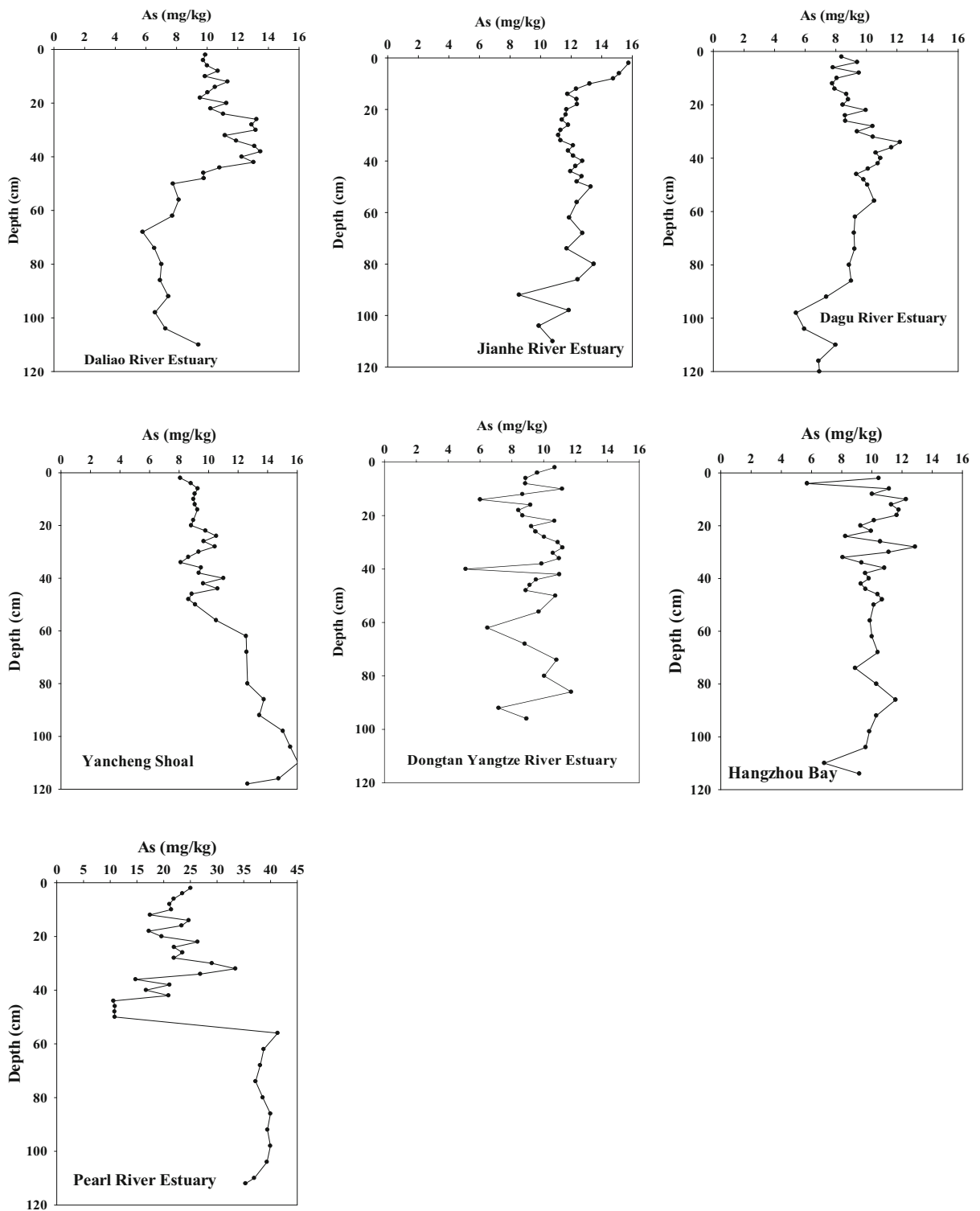


Fig. 3 Vertical profiles of arsenic contents of sediment cores in seven typical intertidal zones

Table 3 Concentrations and background values of Hg and As in sediment cores of this study

Sampling sites	This study				Background value		
	As(mg/kg)		Hg(mg/kg)		As (mg/kg)	Hg (mg/kg)	Reference
	Range	Mean	Range	Mean			
Liaoh River Estuary	5.79–13.48	9.98	0.02–0.13	0.07	9.00	0.19	Ma et al. (2008)
Jianhe River Estuary	8.60–15.76	12.21	0.03–0.08	0.03	15.00	0.05	Jiang et al. (2012)
Dagu River Estuary	5.40–12.19	9.04	0.01–0.06	0.04	9.30	0.02	Centor (1990)
Yancheng Shoal	8.08–16.07	10.66	0.01–0.07	0.02	8.59	0.02	Zuo et al. (2010)
Dongtan Yangtze River Estuary	5.10–11.74	9.43	0.05–0.15	0.09	7.57	0.13	Tao et al. (2014)
Hangzhou Bay	5.72–12.87	10.03	0.03–0.11	0.07	9.99	0.04	Chai et al. (2015)
Pearl River Estuary	10.62–41.35	26.11	0.03–0.39	0.25	10.00	0.06	Gan et al. (2010)

was lower than the class I sediment category and higher than the TEL. The arsenic content of the sediment core changed little.

Mercury contents of the intertidal sediment core from the Pearl River Estuary in Guangdong ranged from 0.03 to 0.39 mg/kg with an average concentration of 0.25 mg/kg (Fig. 2 and Table 3), which was higher than the class I sediment category and the TEL. This indicated that the Pearl River Estuary sediment was polluted by mercury and would pose negative biological effects to the aquatic system. The mercury content was about 0.20 mg/kg in the depth of 0 to 42 cm, it was about 0.04 mg/kg in the depth of 42 to 50 cm, it was about 0.30 mg/kg in the depth of 50 to 112 cm, and it changed little within each of the three depth ranges. Arsenic contents ranged from 10.62 to 41.35 mg/kg with an average concentration of 26.11 mg/kg (Fig. 3 and

Table 3), higher than the class I sediment category and the TEL. The arsenic content was low (10.62 to 33.44 mg/kg) within the depth range of 0 to 56 cm and higher (35.32 to 41.35 mg/kg) within the depth range of 56 to 112 cm. Similar to the previous studies (Chen et al. 2016; Wang et al. 2015c; Ye et al. 2012), the content of Hg and As elevates from 60 cm towards the bottom of the core, which might result from the more environment protection activities and less pollution from human and industrial activities during these years.

This study also compared mercury and arsenic contents from sediment samples across seven typical intertidal zones in China with results from other regions worldwide (Table 4). Mercury and arsenic contents were similar for the Liaoh River Estuary both in this study and the previous study, as the reference values have shown. Contents differed slightly from this study and

Table 4 Concentrations of Hg and As in different regions from other studies

Sampling site	As (mg/kg)		Hg (mg/kg)		Reference
	Range	Mean	Range	Mean	
Liaoh River Estuary	2.76–24.50	8.13	0.01–0.80	0.07	Bi et al. (2017)
Jiaozhou Bay	10.00–20.80		0.12–0.58		Liu et al. (2017)
Yellow River Delta	6.20–17.20	9.47	0.02–0.10	0.04	Cheng et al. (2017)
Yangtze River mouth	4.66–65.60	13.51	0.01–0.09	0.03	Hu et al. (2015)
Minjiang River Estuary	2.39–14.48	8.83	0.00–0.22	0.05	Bi et al. (2017)
Pearl River Estuary	1.93–39.49	21.90	0.01–0.26	0.14	Bi et al. (2017)
Asalyeh part	1.50–9.30	3.70	0.03–0.37	0.12	Delshab et al. (2017)
Brisbane River Estuary	2.40–5.20	3.90 ± 0.90	0.01–0.90	0.40 ± 0.02	Duodu et al. (2017)

the previous one for the Yangtze River Estuary and the Pearl River Estuary, however, which may be due to the different sampling sites and times. Concentration ranges observed in this study were similar to those from Jiaozhou Bay, the Yellow River Delta, and the Minjiang River Estuary, but were clearly higher than those from Asalyeh and the Brisbane River Estuary.

Contamination levels of heavy metals

Numerous indices have been proposed for quantifying the degree of heavy metal contamination in sediment. This study selected two types of indices (I_{geo} and EF) to assess the degree of heavy metal contamination in the sediment cores.

The geoaccumulation index (I_{geo}) is a common parameter used to assess the metal contamination in sediments with a corresponding natural background level as a reference (Müller 1969; Najamuddin et al. 2016; Williams and Block 2015):

$$I_{geo} = \log_2[C_n / (1.5 \times B_n)] \tag{1}$$

where C_n is the concentration of metal (n) in the sample and B_n is the geochemical background concentration of metal (n). (The B_n of Hg and As in different areas are listed in Table 3.) The factor (1.5) is a background matrix correction factor attributed to the lithogenic effect. I_{geo} is classified into seven levels: unpolluted ($I_{geo} \leq 0$), unpolluted to moderately polluted ($0 < I_{geo} \leq 1$), moderately polluted ($1 < I_{geo} \leq 2$), moderately to strongly polluted ($2 < I_{geo} \leq 3$), strongly polluted ($3 < I_{geo} \leq 4$),

strongly to extremely polluted ($4 < I_{geo} \leq 5$), and extremely polluted ($I_{geo} > 5$).

As shown in Fig. 4, the I_{geo} values for Hg were below zero in the sediments from the Daliao River Estuary, the Jianhe River, Yancheng Shoal, and the Dongtan Yangtze River, which indicated that the above areas were uncontaminated by Hg. The I_{geo} values for Hg ranged between 0 and 1 in sediments from the Dagu River Estuary and Hangzhou Bay, indicating mild levels of pollution. The I_{geo} values for Hg were in the range of 1 and 2 in the Pearl River Estuary, which demonstrated moderate levels of contamination.

As shown in Fig. 4, the I_{geo} values for As in the Pearl River Estuary were between 0 and 1, which revealed mild pollution levels; I_{geo} values for As in the other study areas were lower than 0, demonstrating that they were unpolluted.

The potential ecological risk factor (E_r^i) was used to assess the potential ecological risk of heavy metals in sediment and was initially introduced by Håkanson (1979). This method not only assesses the pollution status in sediments but also combines potential ecological and environmental effects with toxicology, providing a better evaluation of the potential risk of heavy metal contamination with the index level. The potential ecological risk factor of a given metal (E_r^i) is defined as

$$E_r^i = T_r^i \times C_r^i = T_r^i \times (C_0^i / C_n^i) \tag{2}$$

where E_r^i is the potential ecological risk for a given element i; T_r^i is the toxic response factor (the T_r^i of As

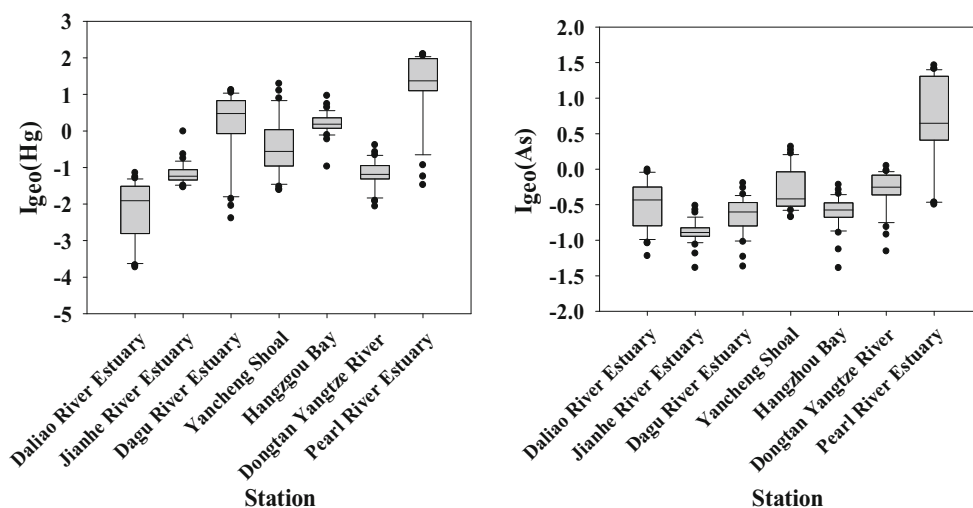


Fig. 4 The I_{geo} values of Hg and As in sediment cores from seven typical intertidal zones

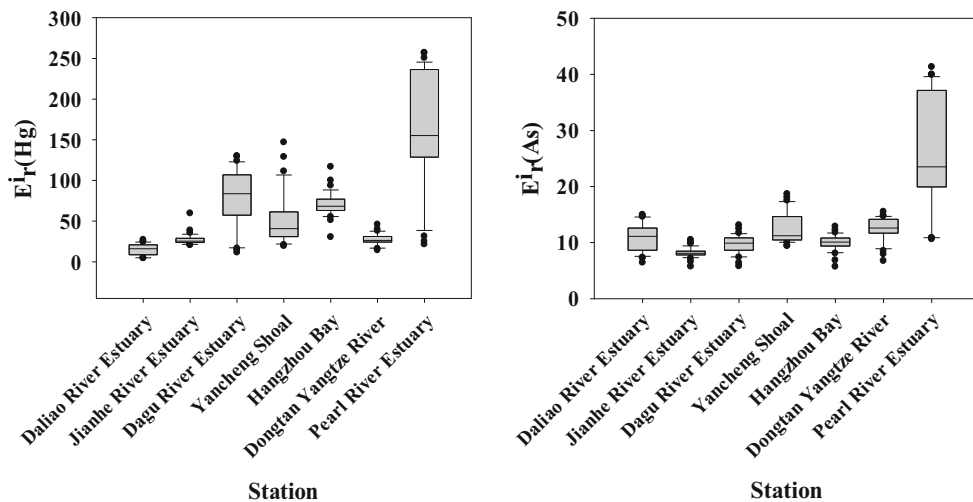


Fig. 5 The E_r^i values of Hg and As in sediment cores from seven typical intertidal zones

is 10; the T_r^i of Hg is 40); and E_r^i , E_{θ}^i , and E_n^i are the contamination factor, the concentration in sediment, and the background reference level for element i , respectively. E_r^i is classified into five levels: low risk ($E_r^i < 40$), moderate risk ($40 \leq E_r^i < 80$), considerate risk ($80 \leq E_r^i < 160$), high risk ($160 \leq E_r^i < 320$), and very high risk ($E_r^i \geq 320$).

As shown in Fig. 5, the values of E_r^{Hg} in the Daliao River Estuary, the Jianhe River Estuary, and the Dongtan Yangtze River ranged between 0 and 40, which showed that there was a low environmental risk; values of E_r^{Hg} in the Dagu River Estuary and Yancheng Shoal ranged from 40 and 80, representative of a moderate potential ecological risk. Values of E_r^{Hg} in the Pearl River Estuary ranged between 80 and 160, indicating a strong potential ecological risk. The evaluation results of E_r^{Hg} were similar to those of I_{geo} .

Values of E_r^{As} in the seven study areas were all less than 40, indicating mild potential ecological risks and posing fewer negative environmental effects when compared with Hg.

Conclusions

The vertical profiles, contamination levels, and potential risks of mercury and arsenic from seven typical intertidal sediment cores in China were presented in this study. Results showed that contents of mercury and arsenic in sediment cores from the Pearl River Estuary were greater than the class I sediment category, whereas

the content from other study areas were all lower than the class I sediment category, which indicated that the Pearl River Estuary was more likely influenced by nearby industrial and tourism impacts. The mercury contents from the study areas were all lower than the TEL except for the Pearl River Estuary, indicating that some adverse effects on biota due to mercury would probably be observed there. Arsenic contents were all greater than the TEL, indicating that negative biological effects due to arsenic may be observed in the study areas. The Pearl River Estuary was moderately polluted by mercury and had a strong potential ecological risk; other areas were uncontaminated or mildly contaminated, and had a low or moderate potential ecological risk. The Pearl River Estuary was mildly polluted by arsenic and had a mild potential risk, while other areas were unpolluted and had a mild potential risk. This study provided some baseline information for further research in the study areas.

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