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Assessment of heavy metal contamination in urban river sediments in the Jiaozhou Bay catchment, Qingdao, China

Fangjian Xu [a,b,c,*], Zhaoqing Liu [a], Yingchang Cao [a], Longwei Qiu [a], Jianwei Feng [a], Feng Xu [a], Xu Tian [d]

[a] School of Geosciences, China University of Petroleum, Qingdao 266580, China
[b] Key Laboratory of Marine Geology and Environment, Institute of Oceanology, Chinese Academy of Sciences, Qingdao 266071, China
[c] Laboratory for Marine Mineral Resources, Qingdao National Laboratory for Marine Science and Technology, Qingdao 266071, China
[d] National Deep Sea Center, Qingdao 266237, China

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ABSTRACT

Selected heavy metals (Cu, Pb, Zn, Cr, Cd, and As) in 47 river sediment samples collected in the Jiaozhou Bay (JZB) catchment were evaluated to assess their spatial distributions and the potential ecological risks. The heavy metal concentrations in the sediments ranged from 4.5–178.7 mg kg^{-1} for Cu, 8.2–65.8 mg kg^{-1} for Pb, 8.2–325.7 mg kg^{-1} for Zn, 12.2–185.5 mg kg^{-1} for Cr, 0.013–1.486 mg kg^{-1} for Cd, and 1.2–20.6 mg kg^{-1} for As. The results showed that the overall sediment quality in the area generally met the China Marine Sediment Quality criteria. Based on the effect-range classification (the threshold effect level (TEL)/probable effect level (PEL) Sediment Quality Guidelines, Cu, Cr, and As were likely to have adverse biological impacts on local aquatic ecosystems. The geoaccumulation index (I_{geo}), enrichment factor (EF), contamination factor (CF), and pollution load index (PLI) values suggested that elevated levels of Cd, As, and Pb contamination occurred in the eastern JZB catchment (i.e., Hongjiang River, Moshui River, Baisha River, Loushan River, Licun River, and Haipo River). This study presents the current state of the sediment quality in the JZB catchment. The results may assist in the definition of future coastal and river management measures specifically targeted at monitoring heavy metal contamination.

1. Introduction

Heavy metals in aquatic sediments originate from both natural sources (mainly erosion and rock weathering) and anthropogenic activities (e.g., industrial discharge, mining, agriculture, transportation, damming, sewage disposal, and wastewater runoff) (Çevik et al., 2009; Feng et al., 2011; Hernández-Crespo and Martín, 2015; Keshavarzi et al., 2015; Sun et al., 2015; Xu et al., 2016a). Heavy metals are recognized as a group of pollutants with high ecological significance because they are not removed from water via self-purification (Ghrefat and Yusuf, 2006). They can accumulate in suspended particulates and sediments, be released back into aquatic systems under favorable conditions, enter the food web, and cause health problems (Ghrefat and Yusuf, 2006; Varol, 2011; da Silva et al., 2015; Keshavarzi et al., 2015; Morina et al., 2015).

Over 50% of the global population lives in urban centers. Therefore, understanding the processes that affect urban systems is a global issue (Taylor and Owens, 2009). Expanding urbanization and industrialization has resulted in enormous increases in the volumes of heavy metals being discharged into urban rivers. Therefore, people living in and around urban centers are often exposed to unhealthy environments (Taylor and Owens, 2009; Lin et al., 2012; Sibanda et al., 2015; Xu et al., 2016a). Heavy metal contamination in urban river sediments has become the most serious problem in urban environments, and it has attracted increased attention from researchers (Gaur et al., 2005; Lin et al., 2012; Keshavarzi et al., 2015; Kadhum et al., 2016). Studying the distributions of heavy metals in urban river sediments can aid in assessing anthropogenic contamination in rivers and provide evidence of the anthropogenic impacts on ecosystems.

Qingdao is a coastal city situated at the southern tip of the Shandong Province in northern China with an area of 11,282 km^2 and a population of 9.0 million. Jiaozhou Bay (JZB) is a typical semi-enclosed coastal embayment located within the territory of Qingdao, and it connects to the Yellow Sea through a 3 km-wide channel (Fig. 1). Due to rapid economic and social developments in this region, JZB is influenced by human activities, leading to increased industrial, agricultural, and aquacultural inputs. Previous studies have indicated that heavy metal levels have increased in JZB in recent years (State Oceanic Administration (SOA) of China, 2004; Dai et al., 2007; Wang et al., 2007; Xu et al., 2016a). However, data were only available in the river outlet, intertidal, and/

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* Corresponding author at: School of Geosciences, China University of Petroleum, Changjiang West Road 66#, Qingdao, Shandong 266580, China.
E-mail address: xufangjiangg@163.com (F. Xu).

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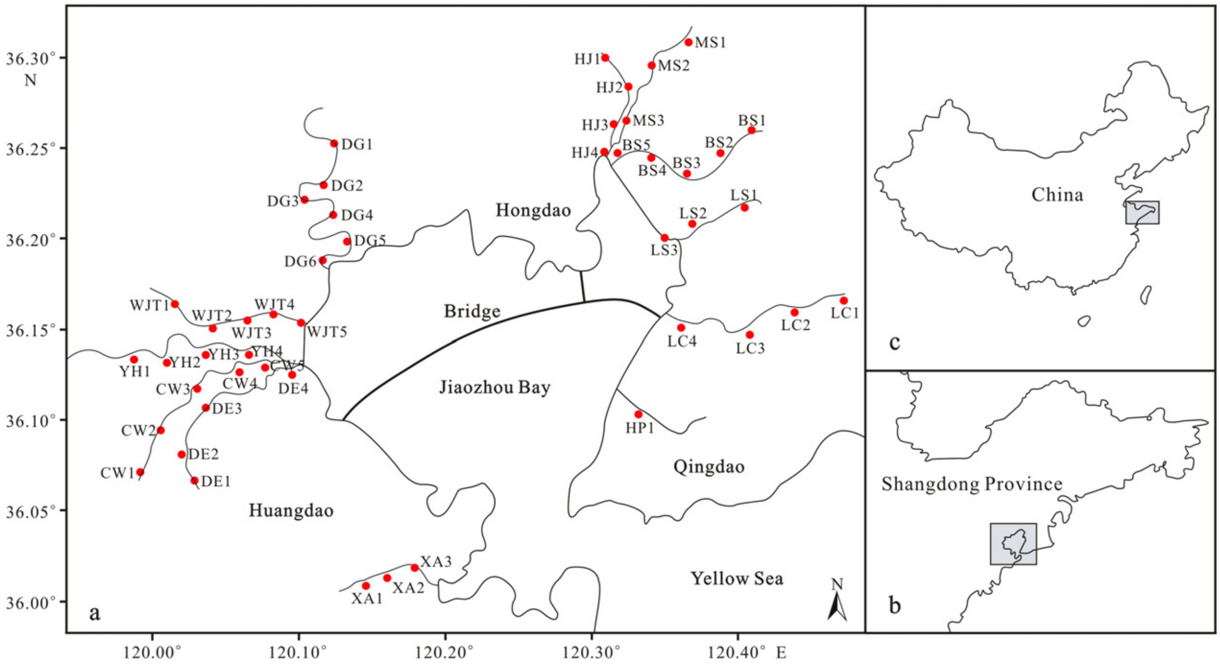

Fig. 1. Locations of the study area and river sampling sites in the Jiaozhou Bay catchment, Qingdao (a), Shandong Province (b), China (c). XA-Xin'an River; DE-Daoer River; CW-Caowen River; YH-Yanghe River; WJT-Wangjiatan River; DG-Dagu River; HJ-Hongjiang River; MS-Moshui River; BS-Baisha River; LS-Loushan River; LC-Licun River; HP-Haipo River.

or deep water areas (Dai et al., 2007; Wang et al., 2007; Deng et al., 2010; Ye et al., 2011; Xu et al., 2016a). Thus, studies of the urban rivers around the bay are lacking, limiting our understanding of the potentially adverse environmental impacts associated with heavy metal pollution in the area.

In recent decades, various indices have been developed to assess heavy metal contamination in sediments and the associated ecological risk. Geochemical normalization approaches such as the geoaccumulation index (I_{geo}), enrichment factor (EF), contamination factor (CF), and pollution load index (PLI) methods have been commonly used for this purpose (Müller, 1979, 1981; Feng et al., 2011; Hu et al., 2013; Zhao et al., 2015). This study addresses existing research gaps and provides valuable information regarding the spatial distributions of selected heavy metals in the urban rivers around the JZB catchment. The goals of this paper are to (1) determine the concentrations of heavy metals (Cu, Pb, Zn, Cr, Cd, and As) in the surface sediments of twelve rivers around the JZB catchment, (2) assess the potential ecological risks of these metals using the Sediment Quality Guidelines (SQGs), and (3) assess the heavy metal contamination using the I_{geo}, EF, CF, and PLI methods.

2. Materials and methods

2.1. Study area and sampling

Qingdao is located in the warm temperate monsoon climate zone. The annual mean temperature is 12.7 °C, and the annual average rain precipitation is 662.1 mm (Chen and Wang, 2012). From 1949 to 2013, the population of Qingdao City increased from 4.0×10^6 to 9.0×10^6 (Qingdao Municipal Statistics Bureau, 2014). The wastewater discharge in urban districts was 84.6×10^6 t yr^{-1} in 1980 (Shen, 2001), and it increased to 472×10^6 t yr^{-1} in 2013 (Qingdao Municipal Statistics Bureau, 2014). The Jiaozhou Bay Bridge (Fig. 1), which stretches 42 km and is the longest sea bridge in the world, was opened in July 2011.

More than 10 small seasonal rivers with varying water and sediment loads discharge into the bay, notably, the Xin'an (XA), Daoer (DE), Caowen (CW), Yanghe (YH), Wangjiatan (WJT), Dagu (DG), Hongjiang (HJ), Moshui (MS), Baisha (BS), Loushan (LS), Licun (LC), and Haipo (HP) Rivers (Fig. 1; Table 1). However, most of these rivers have become channels for industrial and domestic waste discharge due to increased

Table 1
Lengths, drainage areas, water discharges, and sediment loads in small rivers of the Jiaozhou Bay catchment, Qingdao.

Rivers	Area	Length (km)	Drainage area (km^2)	Water discharge (10^4 m^3 a^{-1})	Sediment load (10^4 t a^{-1})	References
Xin'an River	Western Jiaozhou Bay	12	20.96	ND	ND	Qingdao Daily (2012)
Daoer River		25	82.9	413	0.431	Zhao (2007); Sheng et al. (2014)
Caowen River		25.35	128.75	1089	1.136	Zhao (2007); Sheng et al. (2014)
Yanghe River		49	303	2106	2.215	Zhao (2007); Sheng et al. (2014)
Wangjiatan River		12.7	37.3	315	0.329	Zhao (2007); Sheng et al. (2014)
Dagu River		179.9	6131.3	50,366	36.590	Zhao (2007); Sheng et al. (2014)
Hongjiang River	Eastern Jiaozhou Bay	25.5	56	558	1.027	Zhao (2007); Sheng et al. (2014)
Moshui River		42.3	317.2	3734	6.867	Zhao (2007); Sheng et al. (2014)
Baisha River		33	215	3133	1.280	Zhao (2007); Sheng et al. (2014)
Loushan River		5.1	26.5	721	1.057	Zhao (2007); Sheng et al. (2014)
Licun River		22.5	131.5	3576	5.245	Zhao (2007); Sheng et al. (2014)
Haipo River		7	14	381	0.559	Zhao (2007); Sheng et al. (2014)

ND: no data.

economic activities and population in the region (Zhang, 2007; Shi et al., 2011).

Surface sediment samples (top 2 cm) were scraped carefully from 47 sites using plastic spoon in the urban rivers in December 2015. These samples were distributed evenly across the study area (Fig. 1). During sample collection, a hand-held global positioning system (GPS) was used to identify the locations of the sites. After sampling, the sediment samples were sealed in clean polyethylene bags and transported back to the laboratory within a few hours. They were kept frozen until further analysis. The sediment samples were then oven-dried at 60 °C, and large calcareous debris as well as rock and plant fragments were removed.

2.2. Analytical methods

Grain size measurements were performed using a Bettersize-2002 laser particle analyzer according to Xu et al. (2016a). This facility can measure grains in the 1 to 2600 μm range, with a measurement repeatability error of <3%. The textural classification of the sediment samples was based on the relative percentages of clay (<4 μm), silt (4–63 μm) and sand (63–2000 μm) (Wentworth, 1922).

The samples that were used for the elemental analysis were pretreated with 1 mol L^{-1} HCl in a water bath at 60 °C for 1 h to remove calcium carbonate. This process was similar to those used by Wan et al. (2015) and Xu et al. (2016a). The suspension was centrifuged (3500 rpm, 6 min) 3 times with distilled water, and the upper clear liquid was discarded. The residues of the leached samples were heated to dryness at 60 °C and then ground into powders using an agate mortar and pestle. The major (Al) and heavy metal (Cu, Pb, Zn, Cr, Cd, and As) concentrations were determined using a Thermo IRIS Intrepid II XSP and a Perkin Elmer ELAN 9000 ICP-MS, respectively. The analytical accuracy was assessed by analyzing United States Geological Survey (USGS) and Chinese certified reference materials (BHVO-2, GBW07315, and GBW07316). Overall, the measured values of the reference material were within the range of the certified values (Table 2).

2.3. Sediment quality guidelines (SQGs)

The Marine Sediment Quality Standards (GB 18668-2002) were established to prevent and control marine sediment pollution, protect marine life and resources, encourage the sustainable use of marine resources, maintain marine ecological equilibrium, and protect human health (CSBTS, 2002). The threshold effect level (TEL)/probable effect level (PEL) SQGs were also applied to assess the degree to which the sediment-associated metals might adversely affect aquatic organisms (Long et al., 1995; MacDonald et al., 2000). The TELs represent chemical concentrations below which adverse biological effects rarely occur, and the PELs represent chemical concentrations above which adverse biological effects frequently occur (Long et al., 1995; MacDonald et al., 2000).

Table 2
Results of an analysis of certified reference materials (GBW07315, GBW07316, and BHVO-2). The concentrations are in mg kg^{-1}.

Element	GBW07315		GBW07316		BHVO-2	
	Measured values	Certified values	Measured values	Certified values	Measured values	Certified values
Al	11.31	11.41 ± 0.22	7.92	7.7 ± 0.3	13.41	13.5 ± 0.2
Cu	289	357 ± 20	207	231 ± 10	118	127 ± 7
Pb	33.8	37 ± 4	18.9	22 ± 5	1.96	NA
Zn	127	137 ± 15	140	142 ± 22	116	103 ± 6
Cr	63.9	59 ± 6	42.8	38 ± 2	290	NA
Cd	0.292	0.250	0.2	0.3	0.159	NA
As	6.94	7.1 ± 0.6	5.2	4.6 ± 0.5	1.35	NA

NA: not available.

2.4. Assessment of sediment contamination

The I_{geo}, EF, CF, and PLI methods are commonly used to estimate anthropogenic inputs (Müller, 1979, 1981; Feng et al., 2011; Hu et al., 2013; Zhao et al., 2015). According to these methods, the metal concentrations were normalized to the metal concentrations of average shale (Ghrefat and Yusuf, 2006; Çevik et al., 2009) or average crust (Hu et al., 2013; Xu et al., 2015). However, these metal levels tend to be very general and may be misleading in certain areas (Gibbs, 1993). Therefore, several researchers have recommended the use of regional background values (Rubio et al., 2000; Christophoridis et al., 2009; Xu et al., 2016a). In this study, the elemental abundances in the upper crust of East China (Gao et al., 1998) were used as background references.

The I_{geo} values were calculated using the following equation: $I_{geo} = \log_2(C_n / 1.5B_n)$, where C_n is the metal concentration and B_n is the background concentration of that metal. The factor 1.5 represents a

Table 3
Aluminum (%), heavy metal concentrations (mg kg^{-1}) and mean grain sizes (φ) in the river sediments of the Jiaozhou Bay catchment.

Sampling site	Area	Mean grain size	Al	Cu	Pb	Zn	Cr	Cd	As
XA1	Western Jiaozhou Bay	6.5	13.9	12.3	15.7	39.2	56.3	0.030	3.0
XA2		5.5	13.6	10.5	15.7	32.7	36.5	0.070	3.0
XA3		5.8	14.3	73.1	26.9	162.9	129.0	0.282	5.9
DE1		5.6	13.1	13.0	15.7	34.1	48.9	0.048	7.9
DE2		6.6	14.6	14.0	12.6	52.4	70.0	0.067	6.1
DE3		4.6	13.3	14.7	11.6	37.5	59.0	0.065	5.0
DE4		4.7	12.8	17.1	13.1	48.0	74.1	0.057	9.4
CW1		5.6	12.0	11.2	12.6	30.3	52.9	0.045	7.9
CW2		5.6	11.6	10.3	12.9	34.7	49.8	0.062	4.6
CW3		5.8	12.0	11.1	14.6	38.2	52.9	0.073	4.7
CW4		4.6	13.7	13.1	15.0	46.1	62.4	0.060	8.5
CW5		5.8	13.2	16.1	17.1	47.1	68.6	0.043	8.6
YH1		6.1	12.5	14.6	18.3	44.3	59.6	0.084	7.6
YH2		5.3	10.1	7.5	11.9	23.7	41.4	0.024	3.6
YH3		5.8	12.1	12.0	15.6	41.8	61.4	0.045	5.1
YH4		6.5	15.1	17.0	14.8	52.7	73.9	0.056	9.7
WJT1		4.2	13.6	19.0	14.9	52.6	86.8	0.063	11.8
WJT2		3.4	7.7	4.5	8.2	8.2	12.2	0.013	1.2
WJT3		6.4	11.7	10.6	13.3	34.6	60.5	0.043	5.7
WJT4		4.3	10.0	6.9	12.9	20.5	43.0	0.037	3.1
WJT5		3.8	10.8	6.1	13.0	23.8	29.9	0.029	4.2
DG1		6.3	14.8	27.3	15.4	59.0	79.9	0.097	11.4
DG2		6.7	14.9	17.8	15.2	56.0	73.3	0.069	9.6
DG3		6.7	16.2	21.9	14.5	63.6	83.0	0.052	12.4
DG4		5.1	13.0	10.4	15.5	32.7	53.0	0.051	6.0
DG5		3.2	14.0	16.4	13.9	47.5	68.7	0.058	10.4
DG6		5.2	13.8	20.9	14.0	48.2	75.9	0.067	9.9
HJ1	Eastern Jiaozhou Bay	5.5	10.7	13.4	13.9	37.5	84.5	0.066	5.5
HJ2		6.2	15.4	29.2	16.0	71.2	91.2	0.125	10.7
HJ3		6.5	15.8	26.4	15.9	68.5	91.0	0.129	12.1
HJ4		6.7	15.1	22.2	15.5	62.6	80.9	0.084	9.4
MS1		2.0	10.8	25.0	13.4	26.3	63.0	0.151	5.2
MS2		5.5	14.8	52.3	20.8	107.5	97.4	0.305	8.2
MS3		6.2	14.3	29.0	16.2	66.4	85.1	0.130	8.8
BS1		5.7	13.1	15.4	13.6	47.2	58.3	0.082	4.9
BS2		5.6	12.5	7.5	13.3	27.4	31.3	0.050	2.7
BS3		6.7	18.6	19.4	49.4	178.9	38.6	0.443	4.1
BS4		5.3	11.5	14.5	13.3	40.8	45.8	0.088	6.9
BS5		6.0	15.7	23.5	18.7	70.7	92.1	0.113	9.7
LS1		6.1	13.4	23.3	22.3	70.8	87.5	0.151	20.6
LS2		6.7	16.0	29.9	63.5	133.5	185.5	0.568	7.5
LS3		6.1	20.2	47.6	54.2	105.9	106.7	0.387	7.9
LC1		5.8	14.8	23.0	27.2	59.1	49.9	0.198	5.4
LC2		5.8	12.3	25.6	32.3	38.8	60.9	0.157	19.4
LC3		5.1	11.6	15.6	18.1	49.0	54.9	0.125	3.8
LC4		6.1	15.1	58.6	48.7	325.7	70.9	0.936	10.8
HP1		5.9	14.4	178.7	65.8	233.4	117.0	1.486	11.6
Minimum		2.0	7.7	4.5	8.2	8.2	12.2	0.013	1.2
Maximum		6.7	20.2	178.7	65.8	325.7	185.5	1.486	20.6
Mean		5.6	13.5	23.6	20.2	64.6	69.3	0.159	7.7

Table 4
A summary of heavy metal concentrations in river sediments of the Jiaozhou Bay catchment. The average upper crust values in East China and the related values reported for surface sediments in other estuaries and coastal areas are shown for comparison. The concentration units are mg kg^{-1} dry weight for all the elements.

Location		Cu	Pb	Zn	Cr	Cd	As	References
Jiaozhou Bay rivers, China	Range	4.5–178.7	8.2–65.8	8.2–325.7	12.2–185.5	0.013–1.486	1.2–20.6	This study
	Mean	23.6	20.2	64.6	69.3	0.159	7.7	
Intertidal Jiaozhou Bay, China	Mean	38.8	55.2	107.4	69.9	0.42	9.2	Xu et al. (2016a)
Yellow River Estuary, China	Mean	22.85	43.05	54.12	44.09	NA	NA	Sun et al. (2015)
Yangtze River Estuary, China	Mean	24.7	23.8	82.9	79.1	0.19	9.1	Wang et al. (2015)
Pearl River Estuary, China	Mean	45.7	57.9	177	106	NA	NA	Yu et al. (2010)
Masan Bay, Korea	Mean	43.4	43.97	206.26	67.07	1.24	NA	Hyun et al. (2007)
Gironde Estuary, France	Mean	24.5	46.8	168	78.4	0.48	18.7	Larrose et al. (2010)
Average crust of East China	Mean	32	18	70	80	0.079	4.4	Gao et al. (1998)
MSQ-1[a]		35	60	150	80	0.5	20	CSBTS (2002)
MSQ-2[a]		100	130	350	150	1.5	65	CSBTS (2002)
MSQ-3[a]		200	250	600	270	5.0	93	CSBTS (2002)

NA: not available.
[a] The MSQ-1, MSQ-2, and MSQ-3 are the Marine Sediment Quality Standards (GB 18668–2002) issued by the China State Bureau of Quality and Technical Supervision (CSBTS, 2002).

background matrix correction factor that includes possible variations of the background values due to lithogenic effects (Müller, 1979). Müller (1981) established seven classes of geoaccumulation index values: Class 0 (practically unpolluted), $I_{geo} \leq 0$; Class 1 (unpolluted to moderately polluted), $0 < I_{geo} < 1$; Class 2 (moderately polluted), $1 < I_{geo} < 2$; Class 3 (moderately to heavily polluted), $2 < I_{geo} < 3$; Class 4 (heavily polluted), $3 < I_{geo} < 4$; Class 5 (heavily to extremely polluted), $4 < I_{geo} < 5$; and Class 6 (extremely polluted), $I_{geo} > 5$.

The EFs of heavy metals have commonly been used to assess anthropogenic contamination. To identify anomalous metal contributions, Al was chosen as the normalizing element (Yang et al., 2003; Wan et al., 2015). The EFs were calculated using the following equation: $EF = (C_M / Al)_{sample} / (C_M/Al)_{background}$, where $(C_M/Al)_{sample}$ and $(C_M/Al)_{background}$ represent the heavy metal to Al ratios in this study and in the background sample, respectively. Generally, EFs < 1.5 (or 2) are typical of heavy metal levels, reflecting the regional rock composition, whereas EFs > 1.5 (or 2) indicate non-crustal contributions and/or non-natural weathering processes (e.g., anthropogenic influences) (Sutherland, 2000; Zhang and Liu, 2002; Andrews and Sutherland, 2004; Wang et al., 2007; Xu et al., 2016a; Xu et al., 2016b). In the studied region, EF = 1.5 is adopted as the assessment boundary, according to Wang et al. (2007) and Xu et al. (2016a).

The CF and PLI are also used to evaluate the status of heavy metal contamination (Håkanson, 1980; Varol, 2011; Li et al., 2013; Zhao et al., 2015). The CF of each metal is the ratio obtained by dividing the concentration of the metal in the sediment by the background value, which is defined as follows:

$$CF = C_{heavy\ metal}/C_{background}$$

CF values were interpreted as follows: CF < 1 indicates no or low contamination; $1 \leq CF < 3$ is moderate contamination; $3 \leq CF < 6$ is considerable contamination; and $CF \geq 6$ is very high contamination (Håkanson, 1980; Kadhum et al., 2016).

Table 5
Comparison between the heavy metal concentrations (mg kg^{-1}) in the Jiaozhou Bay catchment and sediment quality guidelines (SQGs), with the percentage of samples in each guideline.

Sediment quality guidelines	Metal concentration (mg kg^{-1})					
	Cu	Pb	Zn	Cr	Cd	As
TEL	18.7	30.2	124	52.3	0.68	7.2
PEL	108.2	112.2	271	160.4	4.2	41.6
Compared with TEL and PEL (% of sample in each guideline)						
<TEL	58	87	89	23	96	47
≥TEL < PEL	40	13	9	75	4	53
≥PEL	2	0	2	2	0	0

The PLI was determined as the nth root of the n contamination factors (CF_n) multiplied using the following equation:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \cdots \times CF_n)^{1/n}$$

PLI > 1 indicates a polluted sediment condition, whereas PLI < 1 indicates no metal contamination (Tomlinson et al., 1980).

3. Results and discussion

3.1. Sediment types and geochemistry

All 47 sediment samples were identified as sandy silt according to Folk's classification (Folk et al., 1970). The mean grain size of the sediments varied from 2.0 to 6.7 φ, with an average of 5.6 φ (Table 3).

The concentrations of heavy metals in the sediments of the JZB catchment are summarized in Table 3. The elemental concentrations ranged from 7.7 to 20.2% for Al (mean 13.5%), 4.5–178.7 mg kg^{-1} for Cu (mean 23.6 mg kg^{-1}), 8.2–65.8 mg kg^{-1} for Pb (mean 20.2 mg kg^{-1}), 8.2–325.7 mg kg^{-1} for Zn (mean 64.6 mg kg^{-1}), 12.2–185.5 mg kg^{-1} for Cr (mean 69.3 mg kg^{-1}), 0.013–1.486 mg kg^{-1} for Cd (mean 0.159 mg kg^{-1}), and 1.2–20.6 mg kg^{-1} for As (mean 7.7 mg kg^{-1}).

In this study, the heavy metal concentrations of the river sediments in the JZB catchment were compared with those of other estuaries and coastal areas. The heavy metal concentrations in the study area were

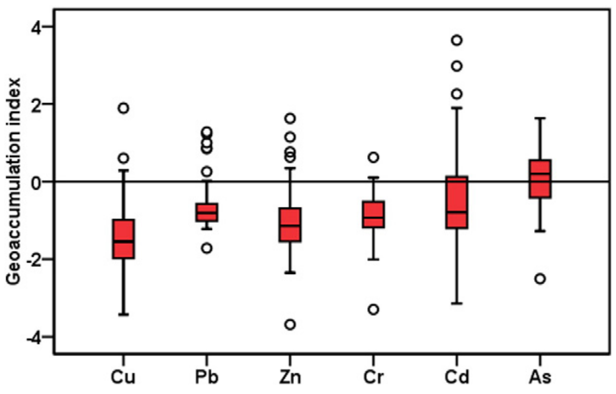

Fig. 2. Box-and-whisker plots for the geoaccumulation index of heavy metals (Cu, Pb, Zn, Cr, Cd, and As) in the river sediments of the Jiaozhou Bay catchment.

Table 6
Sediment contamination assessed by I_{geo}, EF, CF, and PLI in the Jiaozhou Bay catchment.

Samples	Area	$I_{geo\text{-}Pb}$	$I_{geo\text{-}Cd}$	$I_{geo\text{-}As}$	EF_{Pb}	EF_{Cd}	EF_{As}	CF_{Pb}	CF_{Cd}	CF_{As}	PLI
XA1	Western Jiaozhou Bay	−0.79	−1.97	−1.16	0.86	0.38	0.66	0.87	0.38	0.67	0.57
XA2		−0.78	−0.76	−1.13	0.88	0.89	0.69	0.87	0.88	0.69	0.58
XA3		−0.01	**1.25**	−0.17	1.42	**3.40**	1.27	**1.49**	**3.57**	**1.34**	**1.98**
DE1		−0.78	−1.31	**0.25**	0.91	0.63	**1.87**	0.87	0.60	**1.79**	0.70
DE2		−1.10	−0.83	−0.12	0.65	0.79	1.29	0.70	0.84	**1.38**	0.78
DE3		−1.22	−0.87	−0.40	0.66	0.84	1.16	0.64	0.82	**1.14**	0.69
DE4		−1.04	−1.05	**0.51**	0.78	0.77	**2.28**	0.73	0.72	**2.14**	0.85
CW1		−1.10	−1.40	**0.25**	0.79	0.65	**2.04**	0.70	0.57	**1.79**	0.64
CW2		−1.07	−0.92	−0.53	0.84	0.93	1.23	0.72	0.79	**1.04**	0.62
CW3		−0.89	−0.70	−0.49	0.92	1.05	1.21	0.81	0.92	**1.07**	0.68
CW4		−0.84	−0.98	**0.37**	0.83	0.76	**1.93**	0.84	0.76	**1.93**	0.80
CW5		−0.66	−1.46	**0.39**	0.98	0.56	**2.03**	0.95	0.54	**1.96**	0.82
YH1		−0.56	−0.49	**0.20**	1.11	1.17	**1.89**	**1.01**	**1.07**	**1.73**	0.86
YH2		−1.18	−2.30	−0.87	0.89	0.41	1.11	0.66	0.30	0.82	0.44
YH3		−0.79	−1.39	−0.38	0.98	0.65	1.30	0.87	0.57	**1.15**	0.68
YH4		−0.87	−1.08	**0.55**	0.74	0.64	**1.99**	0.82	0.71	**2.20**	0.88
WJT1		−0.86	−0.91	**0.84**	0.83	0.80	**2.70**	0.83	0.80	**2.69**	0.98
WJT2		−1.72	−3.14	−2.51	0.81	0.30	0.47	0.46	0.17	0.26	0.19
WJT3		−1.03	−1.45	−0.21	0.86	0.64	**1.50**	0.74	0.55	**1.29**	0.63
WJT4		−1.06	−1.68	−1.08	0.98	0.64	0.97	0.72	0.47	0.71	0.45
WJT5		−1.06	−2.01	−0.66	0.91	0.47	1.20	0.72	0.37	0.95	0.43
DG1		−0.81	−0.29	**0.78**	0.79	1.13	**2.38**	0.86	**1.22**	**2.58**	**1.12**
DG2		−0.83	−0.79	**0.53**	0.77	0.80	**1.99**	0.84	0.87	**2.17**	0.93
DG3		−0.90	−1.18	**0.91**	0.68	0.56	**2.36**	0.81	0.66	**2.81**	0.99
DG4		−0.80	−1.21	−0.15	0.90	0.68	1.42	0.86	0.65	**1.35**	0.65
DG5		−0.95	−1.03	**0.66**	0.76	0.72	**2.32**	0.77	0.74	**2.37**	0.86
DG6		−0.95	−0.82	**0.59**	0.77	0.85	**2.24**	0.78	0.85	**2.25**	0.93
HJ1	Eastern Jiaozhou Bay	−0.96	−0.83	−0.26	0.99	1.08	**1.60**	0.77	0.84	**1.25**	0.76
HJ2		−0.76	**0.08**	**0.69**	0.79	1.40	**2.15**	0.89	**1.58**	**2.43**	**1.24**
HJ3		−0.76	**0.12**	**0.87**	0.76	1.41	**2.37**	0.88	**1.63**	**2.74**	**1.24**
HJ4		−0.80	−0.49	**0.51**	0.78	0.97	**1.94**	0.86	**1.07**	**2.14**	**1.04**
MS1		−1.01	**0.35**	−0.34	0.95	**2.42**	**1.50**	0.75	**1.91**	**1.19**	0.86
MS2		−0.38	**1.37**	**0.32**	1.06	**3.56**	**1.72**	**1.16**	**3.86**	**1.87**	**1.72**
MS3		−0.74	**0.14**	**0.42**	0.86	**1.57**	**1.92**	0.90	**1.65**	**2.00**	**1.18**
BS1		−0.99	−0.53	−0.42	0.79	1.08	1.17	0.75	**1.04**	**1.12**	0.77
BS2		−1.02	−1.24	−1.28	0.81	0.69	0.67	0.74	0.63	0.62	0.47
BS3		**0.87**	**1.90**	−0.67	**2.01**	**4.12**	0.69	**2.74**	**5.61**	0.94	**1.49**
BS4		−1.02	−0.43	**0.07**	0.88	1.33	**1.87**	0.74	**1.11**	**1.57**	0.76
BS5		−0.53	−0.07	**0.56**	0.91	1.24	**1.92**	**1.04**	**1.43**	**2.21**	**1.19**
LS1		−0.27	**0.35**	**1.64**	1.27	1.95	**4.78**	**1.24**	**1.91**	**4.68**	**1.44**
LS2		**1.23**	**2.26**	**0.18**	**3.01**	**6.13**	1.45	**3.53**	**7.19**	**1.70**	**2.37**
LS3		**1.01**	**1.71**	**0.27**	**2.04**	**3.32**	1.22	**3.01**	**4.90**	**1.81**	**2.08**
LC1		**0.01**	**0.74**	−0.30	1.40	**2.32**	1.12	**1.51**	**2.51**	**1.22**	**1.10**
LC2		**0.26**	**0.41**	**1.56**	**2.00**	**2.22**	**4.92**	**1.80**	**1.99**	**4.42**	**1.32**
LC3		−0.58	**0.07**	−0.79	1.18	**1.85**	1.02	**1.01**	**1.58**	0.87	0.83
LC4		**0.85**	**2.98**	**0.71**	**2.44**	**10.71**	**2.22**	**2.70**	**11.85**	**2.45**	**2.90**
HP1		**1.29**	**3.65**	**0.81**	**3.46**	**17.78**	**2.49**	**3.66**	**18.81**	**2.64**	**4.13**

The underlined bold values denote the contaminated samples sites.

comparable to or lower than those in the intertidal JZB (Xu et al., 2016a), Yellow River Estuary (Sun et al., 2015), and Yangtze River Estuary (Wang et al., 2015) in China, as listed in Table 4. In addition, the metal concentrations were significantly lower than those found in larger industrialized/urban ports and estuaries around the world (Table 4), such as the Pearl River Estuary in China (Yu et al., 2010), Masan Bay in Korea (Hyun et al., 2007), and the Gironde Estuary in France (Larrose et al., 2010).

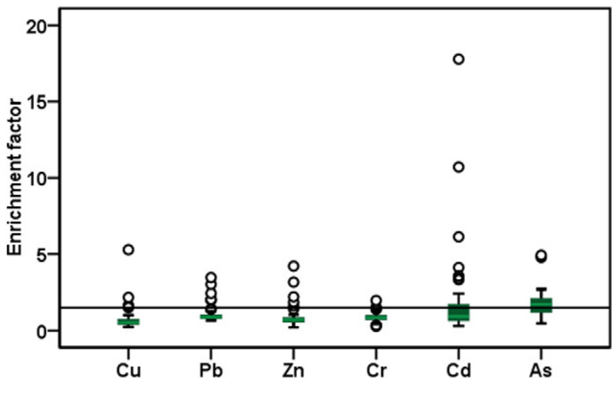

Fig. 3. Box-and-whisker plots for the enrichment factor of heavy metals (Cu, Pb, Zn, Cr, Cd, and As) in the river sediments of the Jiaozhou Bay catchment.

3.2. Assessment of the potential ecological risk

When compared with GB 18668-2002 (Table 4), 42 of the 47 sampling sites met MSQ-1 for Cu, and 4 and 1 sites met MSQ-2 and MSQ-3, respectively; 45 sites met MSQ-1 for Pb, and 2 sites met MSQ-2; 43 sites met MSQ-1 for Zn, and 4 sites met MSQ-2; 33 sites met MSQ-1 for Cr, and 13 and 1 sites met MSQ-2 and MSQ-3, respectively; 44 sites met MSQ-1 for Cd, and 3 sites met MSQ-2; and 46 sites met MSQ-1 for As, and 1 site met MSQ-2. In general, the concentrations of the six heavy metals in this study suggest that the overall sediment quality in the JZB catchment has not been significantly impacted by heavy metal contamination.

A comparison of the TEL and PEL SQGs indicates that the concentrations of Cu, Pb, Zn, Cr, Cd, and As were below the TEL for 58%, 87%, 89%, 23%, 96%, and 47% of the sample sites, respectively, while 40%, 13%, 9%, 75%, 4%, and 53% of the samples fell in the range between the TEL and PEL (Table 5). For Cu, Zn, and Cr, 2% of the samples slightly exceeded the PEL. Therefore, Cu, Cr, and As in the JZB catchment are likely to have adverse biological effects on local aquatic ecosystems.

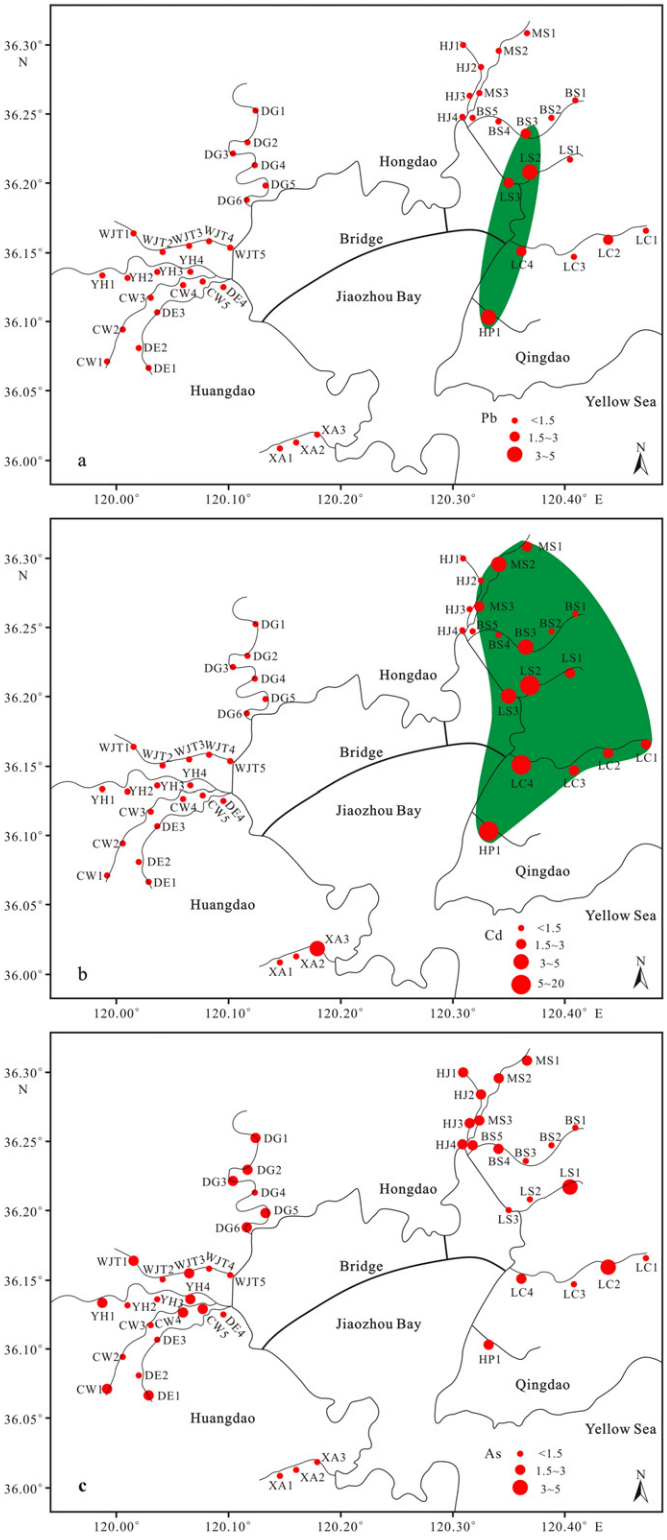

Fig. 4. Enrichment factor distributions of the heavy metals Pb (a), Cd (b) and As (c). The green shaded area indicates heavy contamination in the river sediments of the Jiaozhou Bay catchment.

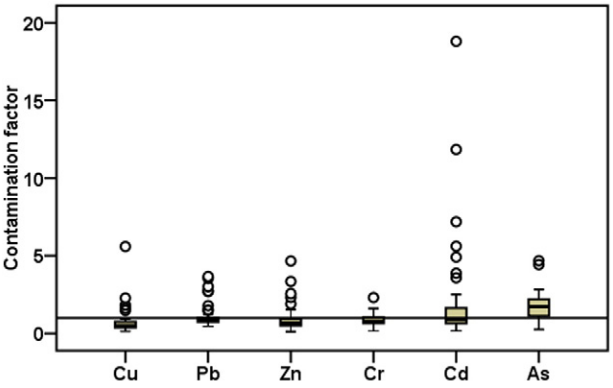

Fig. 5. Box-and-whisker plots for the contamination factor of heavy metals (Cu, Pb, Zn, Cr, Cd, and As) in the river sediments of the Jiaozhou Bay catchment.

3.3. Assessment of sediment contamination

The average I_{geo} values below zero that were observed for Cu (−1.42), Pb (−0.62), Zn (−1.04), Cr (−0.91), and Cd (−0.39) suggest a lack of contamination by these metals in the region (Fig. 2). However, the I_{geo} value of As (0.03) suggests moderate As contamination in this area. Although the I_{geo} values of Cd (−0.39) and Pb (−0.62) were less than zero, 15 and 7 sites, respectively, had positive values, indicating minor to moderate metal contamination (Table 6).

The EFs of the river sediments in the study area are shown in Fig. 3, and they exhibit the following order: Cd > As > Pb > Zn > Cr > Cu. The average EFs of Pb (1.1), Zn (0.9), Cr (0.9), and Cu (0.7) were <1.5 in the JZB catchment, suggesting that these metals are not major concerns. However, moderate enrichment of Pb was found at 6 sites in the eastern JZB catchment (2.0 to 3.5, mean of 2.5, Fig. 4a; Table 6). By contrast, the average EFs of Cd (1.9) and As (1.8) were >1.5, suggesting that Cd and As contamination occurs in the study area (Fig. 4b and c; Table 6). Similar to the distribution of Pb, most of the heavy Cd contamination occurred in the eastern part of the JZB catchment (Fig. 4b). Among the sampling sites in the eastern JZB catchment, 11, 13, and 6 of 20 EF values were >1.5 (Table 6), suggesting that 55.5%, 65%, and 30% of the sites are contaminated by Cd, As, and Pb, respectively.

As discussed above, when compared with the Marine Sediment Quality Standards (GB 18668-2002), 45, 44, and 46 sites in the study area met MSQ-1 for Pb, Cd, and As, respectively. When compared with the TEL-PEL SQGs, 87%, 96%, and 47% of the sample sites were below the TEL for Pb, Cd, and As, respectively. However, the EF and I_{geo} values of Pb (1.1 and −0.62, respectively), Cd (1.9 and −0.39, respectively), and As (1.8 and 0.03, respectively) clearly show that elevated concentrations of Cd and As, as well as partial elevation of Pb, occurred in the region. Therefore, both I_{geo} and EF are more suitable tools for assessing heavy metal contamination in the region than the Marine Sediment Quality Standards (GB 18668-2002) and the TEL-PEL SQGs.

The CF values of sediments in the JZB catchment were 0.1 to 5.6 (mean of 0.7) for Cu, 0.5 to 3.7 (mean of 1.1) for Pb, 0.1 to 4.7 (mean of 0.9) for Zn, 0.2 to 2.3 (mean of 0.9) for Cr, 0.2 to 18.8 (mean of 2.0) for Cd, and 0.3 to 4.7 (mean of 1.7) for As (Fig. 5), decreasing in the same order as the EFs as follows: Cd > As > Pb > Zn > Cr > Cu. Among the sampling sites in the eastern JZB catchment, 18, 17, and 11 of 20 CF values were >1 (Table 6), suggesting 90%, 85%, and 55% of the sites were contaminated by Cd, As, and Pb, respectively.

The PLI values were between 0.2 and 4.1, with a mean of 1.1. Among all sampling sites, 16 of 47 values were >1 (Table 6), suggesting that 34% of the sites were contaminated by metals to some extent. As shown in the spatial distribution map of the PLI values, the most substantial contamination occurred in the eastern part of the JZB catchment (Fig. 6), which is similar to that observed for Cd (Fig. 4b). Among the sampling

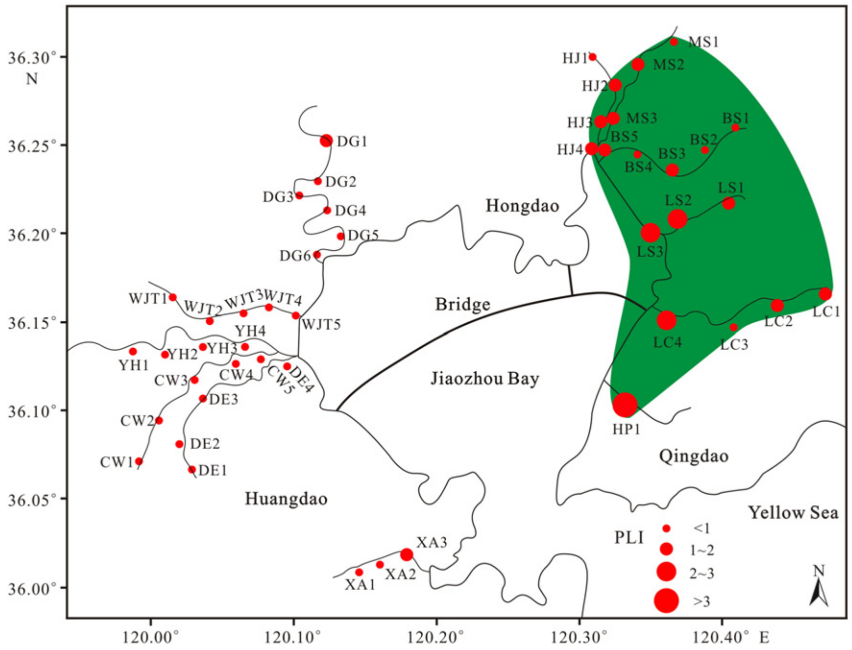

Fig. 6. Spatial distributions of the pollution load index. The green shaded area indicates heavy contamination in the river sediments of the Jiaozhou Bay catchment.

sites in the eastern JZB catchment, 14 of 20 PLI values were > 1 (Table 6), suggesting that 70% of the sites were contaminated by metals.

The I_{geo}, EF, CF, and PLI values showed that considerable contamination occurred in the eastern part of the JZB catchment (i.e., Hongjiang River, Moshui River, Baisha River, Loushan River, Licun River, and Haipo River, Fig. 6; Table 6). Rapid urbanization within the JZB area, especially in the eastern part due to changes in economic policies, involved changes in land use activities. These changes caused rivers to be more susceptible to different contamination issues, such as industrial and domestic sewage and agrochemical inputs (fertilizers and herbicides). Hundreds of enterprises exist around the JZB coast, including dozens of chemical plants. Since Qingdao was selected as the co-host of the 2008 Beijing Olympic Games, measures have been adopted to manage the JZB and the river inputs, such as wastewater treatment and improved effluent treatment projects in the eastern JZB (Wang et al., 2007). However, millions of tons of untreated wastewater are discharged into the bay annually via the rivers (Li et al., 2009). Furthermore, these rivers do not have significant fresh water discharges to the bay throughout the majority of the year, instead serving as the main pathways for urban runoff and sewage and industrial waste water discharge to the JZB (Wang et al., 2007). Consequently, the rivers around the JZB, especially in the eastern JZB, must be comprehensively studied.

The rivers in the eastern part of the catchment were the most seriously polluted rivers around the JZB. However, this is not the case for other watersheds or regions. A recent study noted considerable contamination in the northeastern JZB, which is likely related to the construction of the Jiaozhou Bay Bridge (Xu et al., 2016a). Model results showed that the Jiaozhou Bay Bridge had a relatively large impact on the hydrodynamic environment of the bay, which was most obvious in decreasing the tidal flow and tidal flux (Li et al., 2014). Therefore, heavy metal contamination in the eastern and northeastern JZB may increase in the future due to increasing heavy metal inputs and decreasing tidal flow and tidal flux outputs. Under the current government plan, each watershed region in the JZB was forced to reduce the same proportion of their current annual discharge of pollutants without considering differences in regional capacities (Qingdao Municipal Government, 2013). Therefore, the Qingdao government must strengthen integrated river and coastal zone management and create regional reduction targets for pollutant discharge.

4. Conclusions

In this study, the concentrations and spatial distributions of heavy metals (Cu, Pb, Zn, Cr, Cd, and As) were analyzed in the river sediments of the JZB catchment. The concentrations of the six heavy metals in this study suggested that the overall sediment quality has been impacted by heavy metal contamination. The I_{geo}, EF, and CF values indicated that no Cu, Cr, and Zn contamination occurred in the study area on the whole, while some areas were polluted by Cd, Pb, and As. The I_{geo}, EF, CF and PLI results suggested that the contamination in the eastern part of the JZB catchment (i.e., Hongjiang River, Moshui River, Baisha River, Loushan River, Licun River, and Haipo River) was higher than contamination in other areas. This study presents the current state of sediment quality in the JZB catchment, and the results may assist in the definition of future coastal and river management measures that specifically target the monitoring of heavy metal contamination. With increased industrialization and economic development in the study region, additional attention should be paid to the eastern part of the JZB catchment. The Qingdao government must strengthen integrated river and coastal zone management and create regional reduction targets for pollutant discharge.

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