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Source: Journal of Coastal Research, 32(2) : 374-386

Published By: Coastal Education and Research Foundation

URL: https://doi.org/10.2112/JCOASTRES-D-14-00222.1

Journal of Coastal Research	32	2	374–386	Coconut Creek, Florida	March 2016

Assessment of Heavy Metal Contamination and Wetland Management in a Newly Created Coastal Natural Reserve, China

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ABSTRACT

Xie, Z.; Zhang, H.; Zhao, X.; Du, Z.; Xiang, L., and Wang, W., 2016. Assessment of heavy metal contamination and wetland management in a newly created coastal natural reserve, China. *Journal of Coastal Research,* 32(2), 374–386. Coconut Creek (Florida), ISSN 0749-0208.

The investigation of the content and distribution of heavy metals in coastal soils is useful for improving our understanding of biogeochemical cycles and their potential feedback to global environmental change. The accumulation of heavy metals in the Yellow River Delta (YRD) was investigated using 43 sampling sites to determine the concentrations and vertical distribution of heavy metals. Factor analysis, contamination factors, and the geoaccumulation index were applied to evaluate the contamination condition. The contamination factors and geoaccumulation index clearly indicated that the coastal ecosystems are still in their pristine state with respect to metal pollution. Factor loadings reveal that the first principal component was strongly and positively related to arsenic, chromium, copper, nickel, lead, and zinc, while the second showed highly positive factor loading on cadmium. The study demonstrated that sediment quality in the Yellow River Delta generally met the primary sediment criteria of Chinese marine sediment quality (standard no. GB 18668-20020). The YRD could still be regarded as a "clean site" because several typical heavy metals were found to have accumulated less in this region compared with other regions, *e.g.*, the Pearl River Delta, which was heavily contaminated by heavy metals during the past decades. There is a critical need for a holistic approach to monitor heavy metal concentrations and distributions, as well as a comprehensive strategy to prevent health risks. The findings of this study could contribute to wetland conservation and management in coastal YRD regions.

ADDITIONAL INDEX WORDS: *Heavy metals content, new wetland, environment monitor, Yellow River Delta.*

INTRODUCTION

Heavy metals have been widely used as environmental monitoring factors, and their toxicity in humans, animals, and plants is well known (An *et al*., 2010; Arik and Yaldiz, 2010; Naser, 2013; Xiao *et al*., 2013; Bai *et al*., 2015). Heavy metals, which may result from chemical leaching of bedrock, water drainage, and runoff from banks, as well as discharge of urban industrial and rural agricultural wastewaters, are widely present in rivers and serve as important indicators of environmental water quality (Xiao *et al*., 2013; Zhan *et al*., 2010). Heavy metals from anthropogenic sources can impact estuarine areas, and they are considered to be one of the most common types of pollutants (Bai *et al*., 2011a; Bai *et al*., 2011b; Li *et al*., 2007; Pan and Wang, 2012; Tang *et al*., 2008; Zhang *et al*., 2009; Zhang *et al*., 2013). Many researchers found that heavy metals could be released to water bodies from sediments, which would increase the potential ecological risk and toxicity to aquatic organisms (Kumar, Solanki, and Kumar, 2013). Thus, it is crucial to clarify the potential risks of heavy metals in coastal areas.

Various methods have been provided for assessing heavy metal contamination (Xiao *et al*., 2013). The toxic unit (TU), which is defined as the ratio of the measured concentration to the probable effect level (PEL), is usually employed to evaluate the toxicities of various metals in coastal areas (Lu *et al*., 2014; Xiao *et al*., 2012, 2013). Application of risk indices, such as the enrichment factor (EF) and geoaccumulation index (I_{geo}), also supply crucial information regarding the quantification of metal contamination (Panda *et al*., 2010). Li *et al*. (2013) selected heavy metals in surface sediments from the coastal Shandong Peninsula (Yellow Sea) to determine the spatial distribution and potential ecological risk by applying the EF, I_{geo}, pollution load index (PLI), and the mean PEL-quotient index. Xiao *et al*. (2013) analyzed TUs and contamination factors (CFs) to assess pollution levels, toxicity, and ecological risk levels and compared the characteristics of heavy metal pollution between the two rivers. The potential ecological risk indices of rural river sediments were equal to those of rural river sediments. Bai *et al*., 2011a; investigated heavy metal contamination using the contamination index in wetland soils near tidal ditches and their main sources of water. Bai *et al*. (2014) applied TUs, correlation analysis, and principal component analysis to investigate temporal variations in soil profiles

DOI: 10.2112/JCOASTRES-D-14-00222.1 received 7 November 2014; accepted in revision 13 April 2015; corrected proofs received 17 June 2015; published pre-print online 28 August 2015.
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of three salt marshes, assess the enrichment levels and ecological risks of these heavy metals in three sampling seasons, and identify their influencing factors. Xiao *et al*. (2011) applied the contamination index, integrated contamination index, I_{geo}, TUs, and sum of TUs to assess heavy metal contamination levels and ecotoxicity. Bai *et al*. (2012) sampled soil in tidal freshwater and salt marshes in the Yellow River Delta (YRD) to investigate the characteristics of heavy metal pollution in tidal wetlands before and after the flow-sediment–regulation regime. The coastal wetland of the YRD is not only the most complete estuary wetland but also the youngest wetland ecosystem in the warm-temperate zone in China, with immature, fragile, and unstable characteristics (Bai *et al*., 2012). Many previous studies showed that the accumulation of heavy metals is significantly affected by anthropogenic activities and severely threatens coastal ecosystems (Feng *et al*., 2011; Hu *et al*., 2013a; Hu *et al*., 2013b). However, little information is available on the biogeochemical variability and vertical distribution of heavy metals under different land use types in the newly created wetlands of the YRD.

Rivers clearly reflect the current status of environmental pollution from their upper to lower reaches in highly industrialized regions (Xu *et al*., 2011). The concentrations in the upper reaches of rivers are always lower than those downstream (Bai *et al*., 2009). Xie *et al*. (2014) have compared the heavy metal contamination of existing wetlands with wetlands created by river diversion in the Yellow River Estuary. Previous studies primarily focused on the surface soil; dynamics in deeper soils and the driving factors behind vertical distributions of heavy metals remain poorly understood in the newly created coastal Yellow River Delta Nature Reserve. Knowledge of concentrations of heavy metals across different soil depths is essential to determining whether heavy metals in deep soil layers will continue to increase in concentration (Silva, Haro, and Prego, 2009). The vertical distribution of heavy metals in different land use types may play a crucial role in the fates of these contaminants and can alert estuarine managers to apply reasonable methods to restore wetlands (Liu *et al*., 2010).

In this study, we investigated vertical distributions of heavy metals in a newly created estuarine wetland using 43 soil profiles obtained from a coastal wetland survey in 2010.The primary objectives of this study are (1) to investigate heavy metal concentrations in the soil profiles of different land use types using TUs, CFs, and I_{geo} and (2) to identify the pollution sources of heavy metals using multivariate analysis and provide management suggestions for heavy metal remediation.

METHODS

The methods section includes study areas, sample collection and lab analysis, assessment references and indexes, and statistical analysis.

Study Area

The Yellow River Delta (36.52°–38.12° N, 117.48°–119.45° E) is situated NE of Dongying City, Shandong Province, and on the S bank of the Bohai Sea (Figure 1). The climate is warm-temperate and continental monsoon with an annual precipitation of 596.9 mm, annual evaporation of 1900–2400 mm, and annual average temperature of 12.9°C. The delta was formed by sediment siltation, and the highest elevation in the delta is 37 m above mean sea level in the southern hills; the lowest elevation is less than 1 m at the northern and eastern coasts. The Bohai Sea and YRD provide a unique depositional environment into which approximately 1.1 billion tons of sediments are transported annually. Because of the large amount of deposition associated with the Yellow River, the delta area had an accretion by land extension at a rate of 32.4 km^2/a before 1979; then it had a rate of 2.7 km^2/a because of the channel diversion of the Yellow River and variation of flow and sediment in the estuary. Shengli Oilfield, China's second largest oil field, is located on the delta, and the delta has become an important base of manufacturing and agricultural production. The YRD has become vulnerable to anthropogenic pollutant inputs, such as heavy metals and petroleum-derived hydrocarbons, which affect the YRD coastal environment quality (Chatterjee *et al*., 2009). Within the delta, the large areas of shallow sea and bog, which offer an abundance of wetland vegetation and aquatic biological resources, provide birds with an exceptional habitat for breeding, migrating, and wintering (Wang, Qi, and Zhang, 2012; Xu, Lin, and Fu, 2004).

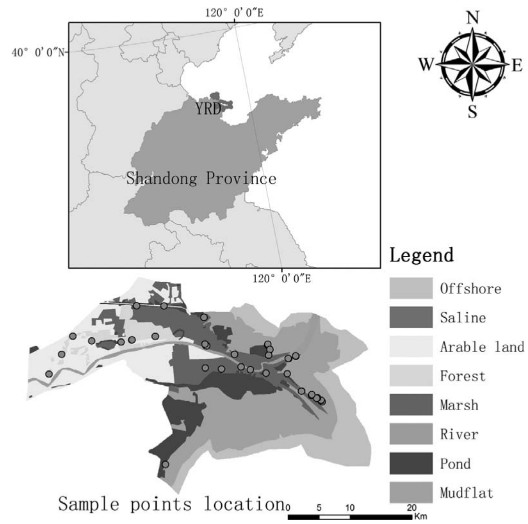

Figure 1. The study area on the Yellow River Delta of the newly created coastal wetland. The sampling points and land use map in 2010 are indicated. Sampling points are located in mudflat, marsh, forest, arable land, and saline areas.

Sample Collection and Lab Analysis

A total of 43 sampling sites were chosen in different land use types, such as mudflat, reed swamp, arable land, and *Suaeda heteroptera* wetland. The mudflat is covered by water at high tide and becomes a beach at low tide. Mudflats are a typical land use type in the YRD. The reed swamp is the main vegetation type, which plays a role in preventing floods and reducing environmental pollution. Generally, arable land is used for planting corn and wheat. Although the natural reserve also has this type of land use, it only exists in the experimentation zone. The *Suaeda heteroptera* wetland is

Table 1. *Main sampling sites of this study.*

	Location	Vegetation
Mudflat	37°36′56.8″ N, 118°57′45.1″ E	No vegetation
	37°47′13.2″ N, 119°9′16.5″ E	
	37°44′35.9″ N, 119°11′20.5″ E	
	37°44′36.1″ N, 119°11′20″ E	
	37°45′54.3″ N, 119°11′30.3″ E	
Arable Land	37°47′49.8″ N, 118°49′57.9″ E	Cotton
	37°47′40.8″ N, 118°53′11.3″ E	
	37°47′50.2″ N, 118°54′21.4″ E	
	37°44′59.9″ N, 118°45′12.4″ E	
Reed Swamp	37°36′43.7″ N, 119°2′26.6″ E	*Phragmites*
	37°45′8.8″ N, 119°4′6.5″ E	
	37°45′17.4″ N, 119°6′16.4″ E	
	37°44′41.4″ N, 119°9′0.2″ E	
	37°46′24.2″ N, 119°5′35.2″ E	
	37°47′13.9″ N, 119°2′26.5″ E	
	37°47′12.3″ N, 119°2′33.3″ E	
	37°43′3.2″ N, 119°12′51.7″ E	
	37°45′56.4″ N, 119°11′28.6″ E	
	37°47′22.3″ N, 119°2′22.4″ E	
	37°47′21.3″ N, 119°2′21.9″ E	
	37°49′41.7″ N, 119°2′18.4″ E	
	38°1′11.3″ N, 118°48′14.2″ E	
	37°50′46″ N, 118°57′57.6″ E	
	37°50′49.8″ N, 118°54′59.4″ E	
Suaeda heteroptera wetland	37°47′9.2″ N, 119°9′15.1″ E	*Suaeda heteroptera*
	37°46′44.9″ N, 119°9′30.1″ E	
	37°46′45.3″ N, 119°9′29.5″ E	
	37°46′15.6″ N, 119°9′19.9″ E	
	37°42′5″ N, 119°15′6.5″ E	
	37°42′10.5″ N, 119°14′56.5″ E	
	37°42′13.4″ N, 119°14′58.1″ E	
	37°46′6.1″ N, 119°12′18.6″ E	
	37°46′7.9″ N, 119°12′17.7″ E	
	37°42′43.6″ N, 119°13′58.8″ E	
	37°43′2.9″ N, 119°12′50.7″ E	

found in coastal beach areas. Soil profiles from 0 to 60 cm depth were sampled and sectioned into three transects at 20 cm intervals. A total of 126 soil samples were collected and taken to the laboratory, where the samples were air dried for 3 weeks. The coarse debris was removed, and all samples were ground and passed through a 0.149 mm nylon sieve (Bai *et al.*, 2011b). The sample information is summarized in Table 1.

For analysis of total concentrations of heavy metals, soil samples were digested using an acid mixture of 2 mL $HClO_4$, 8 mL HNO_3, and 12 mL HF in microwave Teflon vessels. The digests were filtered through Schleicher & Schuell blue-band filters (GE Healthcare Bio-Sciences, Pittsburgh, Pennsylvania, U.S.A.), diluted up to 50 mL with Milli-Q water (EMD Millipore, Darmstadt, Germany), transferred to polyethylene containers, and stored at 4°C until analysis. The contents were transferred, and 10 mL 10% HNO_3 was used to rinse thoroughly for complete transfer of the contents (Chandra *et al.*, 2013). The digested solutions of soil samples were analyzed using inductively coupled plasma atomic emission spectrometry on a Varian Vista Pro (Agilent Technologies, Palo Alto, California, U.S.A.) (Xiao *et al.*, 2012). To assess quality assurance and quality control, every extraction batch of 10 samples included a blank extraction and a reference material (no. GBW 07401) obtained from the Chinese Academy of Measurement Sciences. All samples were analyzed in triplicate with blanks similarly treated for metal analysis. A satisfactory performance of heavy metal determination was achieved depending on the recovery of heavy metals in the certified samples varying from 95% to 110%. The tests were performed in the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences.

Assessment References and Indexes

The CF was applied to evaluate the degree of anthropogenic metal contamination (Li *et al.*, 2013). The CF is the ratio of measured concentration in the soil to the background value, which is defined as follows:

$$CF = \frac{Me_{\text{sample}}}{Me_{\text{baseline}}} \quad (1)$$

The CF was classified into four ranking systems to express the degree of anthropogenic influence on concentrations of metals. The main soil types of the YRD are alluvial soil, fluvo-aquic soil, coastal saline soil, and coastal tidal beach saline soil. If $CF < 1$, no or low contamination exists; $1 < CF < 3$ indicates moderate contamination; $3 < CF < 6$ indicates considerable contamination; and $CF > 6$ indicates very high contamination. After the concentrations of heavy metal were obtained, I_{geo} was used to quantify metal pollution (Long *et al.*, 1995). This index was applied by Müller (1981) to compare present-day heavy metal concentrations with precivilization background values to understand anthropogenic effects (Chandra *et al.*, 2013). I_{geo} is expressed as follows:

$$I_{\text{geo}} = \log_2 \frac{C_n}{1.5 \times B_n} \quad (2)$$

where C_n is the concentration of elements of n, and B_n is the geochemical background value. The factor 1.5 is used to minimize the effect of possible variations in the background values that may be attributed to lithologic variations (Ghrefat, Abu-Rukah, and Rosen, 2011). These geochemical background values were obtained based on environmental background concentrations of the loess materials of the Yellow River. According to the baseline values, the sample can be classified as class 0, pollution free ($I_{\text{geo}} \leq 0$); class 1, pollution free to moderately polluted ($0 < I_{\text{geo}} \leq 1$); class 2, moderately polluted ($1 < I_{\text{geo}} \leq 2$); class 3, moderately to strongly polluted ($2 < I_{\text{geo}} \leq 3$); class 4, strongly polluted ($3 < I_{\text{geo}} \leq 4$); class 5, strongly to extremely polluted ($4 < I_{\text{geo}} \leq 5$); and class 6, very strongly polluted ($I_{\text{geo}} > 5$). The reference background levels were obtained from soil environmental background concentrations in Shandong Province.

To assess the toxicity of the heavy metals, TUs (the ratio of the determined concentration to the PEL) were applied to assess the contamination status.

Statistical Analysis

Multivariate statistics have been frequently applied to quantify the contribution of environmental factors to sediment quality parameters (Han *et al.*, 2006; Singh *et al.*, 2004). In this study, factor analysis, correlation coefficient matrices, and Pearson correlation coefficients were calculated to reveal the relationships between soil properties and heavy metals (Chandra *et al.*, 2013). One-way analysis of variance was conducted to evaluate whether the vertical distribution of each

Table 2. *Soil heavy metal concentrations*.

Soil Layer	As	Cd	Cr	Cu	Ni	Pb	Zn
A (0–20 cm)							
Mean (mg/kg)	8.49	0.14	20.70	17.48	25.21	14.67	58.97
Range (mg/kg)	5.09–13.35	0–0.84	12.33–31.68	8.04–28.58	15.46–35.59	7.28–26.34	28.5–95.32
SD	2.00	0.21	5.04	5.30	5.28	5.23	15.67
CV	4.25	0.68	4.10	3.30	4.78	2.81	3.76
B (20–40 cm)							
Mean (mg/kg)	8.08	0.20	22.01	19.02	28.71	14.47	57.98
Range (mg/kg)	5.27–14.36	0–0.97	10.78–39.15	10.63–34.19	19.14–47.1	6.39–26.83	33.02–102.65
SD	2.00	0.30	5.63	5.51	5.91	4.61	16.21
CV	4.06	0.67	3.91	3.45	4.86	3.14	3.58
C (40–60 cm)							
Mean (mg/kg)	7.97	0.15	20.36	17.34	25.27	14.53	55.83
Range (mg/kg)	5.57–18.66	0–0.67	10.92–38.38	10.44–33.59	16.72–39.81	5.16–58.25	34.05–95.29
SD	2.47	0.17	6.02	5.61	5.46	8.63	15.01
CV	3.23	0.92	3.38	3.09	4.63	1.68	3.72
Background Value	9.3	0.084	66	24	25.8	25.8	63.5

SD denotes standard deviation; CV denotes coefficient of variation.

heavy metal differed significantly between different land use types. Data were processed with SPSS version 17.0 statistical software (IBM, Armonk, New York, U.S.A.).

RESULTS

The heavy metal concentrations and assessment using a correlation matrix, factor analysis, I_{geo}, CFs, and TUs are as follows.

Heavy Metal Concentrations

The mean concentrations of heavy metals are summarized in Table 2. Generally, heavy metal concentration followed this order: zinc (Zn) > chromium (Cr) > nickel (Ni) > copper (Cu) > lead (Pb) > arsenic (As) > cadmium (Cd). The coefficient of variation (CV) of Ni is the largest of all of the heavy metals, and the CV of Cd is the smallest in the first layers. This characteristic is the same for the second and third layers, indicating that heavy metals in all three layers have similar concentrations.

Vertical Distributions of Heavy Metals

Figure 2 illustrates the vertical distributions of heavy metals in different land use types. Generally speaking, heavy metal concentration fluctuated with increasing soil depth. The content of Cd, Cr, Pb, and Zn in all three layers of the four land use types are below the LEL (threshold of lowest effect level), indicating that these heavy metals may not exert adverse effects on soil in this estuarine ecosystem. The content of As and Ni are above the LEL in three layers of the four land use types. In some land use types, Cu contents are above the LEL. Significant accumulation of Cr, Cu, Ni, Pb, and Zn in reed swamps at a depth of 20–40 cm ($p < 0.05$) was indicated. Concentrations of Cr and Zn were higher and well distributed. Concentrations of As, Cr, Cu, Pb, and Zn in the surface layers (0~20 cm) of reed swamps and *Suaeda heteroptera* wetlands were slightly higher than those at other sampling sites. Different soil depths have similar content traits (Cr, Cu, Ni, Pb, and Zn). No significant differences in the vertical distributions of heavy metal concentrations were observed among the different land use types.

Correlation Matrix and Factor Analysis

To analyze the general characteristics of the heavy metals, Pearson correlation and factor analysis were applied. The correlation matrix between these metals exhibits a significant correlation between all the metals, except for Cr, Cu, Ni, Pb,

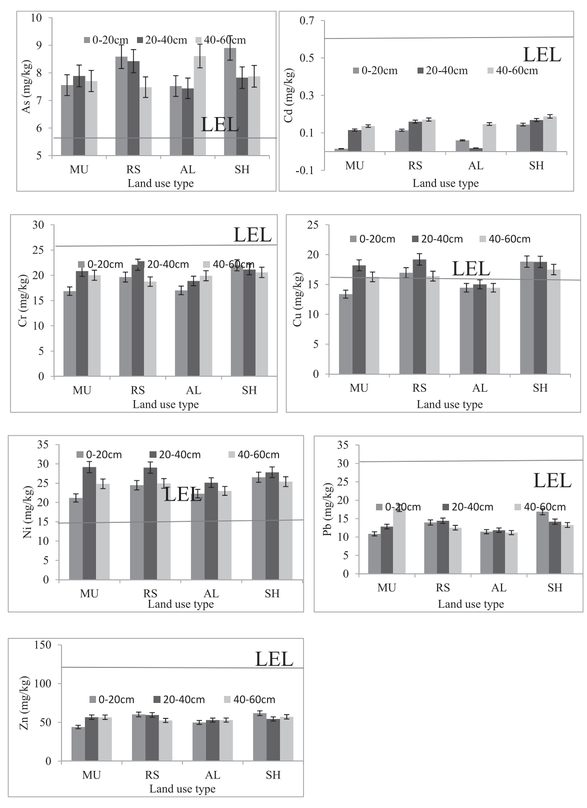

Figure 2. Vertical distributions of heavy metals in mudflat (MU), reed swamp (RS), thin reed (TR), forest (FO), arable land (AL), and *Suaeda heteroptera* wetland (SH). The horizontal line presents the LEL. The vertical distribution includes three soil layers, 0–20 cm, 20–40 cm, and 40–60 cm. Concentrations of As and Ni exceed the LEL, while concentrations of other heavy metals are lower than the LEL.

Table 3. *Correlation matrix between heavy metal concentrations and soil properties for three different layers.*

	As	Cu	Cr	Ni	Cd	Pb	Zn	SOC	Saline	pH	TN
First Layer (0–20 cm)											
As	1										
Cu	0.918**	1									
Cr	0.803**	0.856**	1								
Ni	0.898**	0.973**	0.848**	1							
Cd	0.105	0.102	0.363*	0.148	1						
Pb	0.888**	0.912**	0.803**	0.873**	0.173	1					
Zn	0.867**	0.921**	0.912**	0.915**	0.304	0.867**	1				
SOC	0.208	0.214	0.387*	0.233	0.500**	0.290	0.366*	1			
Saline	0.239	0.249	0.140	0.273	0.015	0.155	0.174	−0.328*	1		
pH	0.092	0.089	0.164	0.080	−0.020	0.141	0.170	0.308	−0.777**	1	
TN	0.192	0.220	0.383*	0.226	0.447**	0.289	0.355*	0.958**	−0.448**	0.432**	1
Second Layer (20–40 cm)											
As	1										
Cu	0.876**	1									
Cr	0.782**	0.814**	1								
Ni	0.865**	0.891**	0.759**	1							
Cd	0.043	0.263	0.382*	0.035	1						
Pb	0.858**	0.868**	0.819**	0.803**	0.213	1					
Zn	0.810**	0.761**	0.786**	0.784**	0.161	0.745**	1				
SOC	0.538**	0.627**	0.653**	0.548**	0.277	0.624**	0.606**	1			
Saline	0.104	0.200	0.217	0.159	−0.074	0.084	0.175	−0.069	1		
pH	−0.016	−0.137	−0.156	−0.014	−0.240	0.001	−0.109	0.007	−0.794**	1	
TN	0.532**	0.577**	0.640**	0.523**	0.259	0.621**	0.590**	0.947**	−0.230	0.149	1
Third Layer (40–60 cm)											
As	1										
Cu	0.904**	1									
Cr	0.892**	0.905**	1								
Ni	0.913*	0.963**	0.893**	1							
Cd	0.052	0.280	0.179	0.232	1						
Pb	0.630**	0.639**	0.678**	0.631**	0.175	1					
Zn	0.894**	0.925**	0.883**	0.919**	0.127	0.597**	1				
SOC	0.623**	0.814**	0.703**	0.744**	0.495**	0.499**	0.693**	1			
Saline	0.004	−0.096	−0.052	−0.122	−0.257	0.152	0.025	−0.152	1		
pH	0.025	0.192	0.121	0.177	0.453**	−0.005	0.086	0.190	−0.715**	1	
TN	0.675**	0.820**	0.726**	0.757**	0.404*	0.414*	0.708**	0.931**	−0.257	0.203	1

SOC denotes soil organic carbon; TN denotes total nitrogen.
**Correlation is significant at the $p < 0.01$ level.
*Correlation is significant at the $p < 0.05$ level.

and Zn with Cd (Table 3). Huang *et al*. (2013) found that the correlation coefficients between Cd and Cu, Pb, Zn, and Cr are 0.638, 0.465, 0.395, and 0.681, respectively. In our study, the correlations with Cd are smaller than those reported by Huang. The correlation coefficients between heavy metals (except Cd) are significant in the three soil layers, while the coefficients between soil properties and heavy metals are not significant in the first layer, except for Cd with soil organic carbon (SOC) and total nitrogen (TN). A factor analysis was carried out to clarify the relationships among heavy metals in the study area (Chandra *et al*., 2013). A Varimax rotation of principal components or factors was applied to clarify the traits for a meaningful representation of the underlying factors (Chandra *et al*., 2013). Factor loadings were calculated using eigenvalues > 1. The factor loadings may be classified as "strong," "moderate." and "weak" based on their significant influence on geochemical processes, corresponding to absolute loading values $>$0.7, 0.7–0.5, and 0.50–0.40, respectively (Liu, Lin, and Kuo, 2003). In the first layer, based on the cumulative proportion, two principle components (PCs) explained 91.91% of the total variance (Table 4). PC1, explaining 77.04% of the total variance, was strongly and positively related to As, Cr, Cu, Ni, Pb, and Zn. PC2, explaining 14.87% of the total variance, showed a highly positive factor loading on Cd (Table 5). The variances and the rotated component matrices of the second and third layers are summarized in Tables 5 and 6. The factor analysis loading plot showed that these metals could be classified into Group 1 (Cd) and Group 2 (As, Cu, Cr, Pb, Ni, Zn) according to similarities in behavior and distribution of the heavy metals (Figure 3) (Huang *et al*., 2013). The negative loading with Cd is similar to that reported by Chandra *et al*. (2013). PC2 may represent the anthropogenic sources of metal pollution; thus, the presence of Cd would be due to both anthropogenic and natural inputs from riverine sources that contain some Cd in the sediment. Group 2 would still be influenced by PC2, indicating that the weathering of soil and runoff are the predominant contribution to their geologic origins (Figure 3).

Assessment of Arsenic and Heavy Metals Using I_{geo}

Heavy metal enrichment was calculated and shown in Figure 4 using I_{geo}, which is based on YRD background values. The I_{geo} values of As, Cr, Cu, Ni, Pb, and Zn are below 0, implying that the soils in the YRD are not polluted by these heavy metals. There are relatively high I_{geo} values for Ni in three layers, suggesting that these layers are severely polluted with Ni.

Table 4. *Total variances and rotation component matrices of the first soil layer (0–20 cm).*

Principal Component	Initial Eigenvalues			Element	Rotation Sum of Squared Loadings			Rotated Component Matrix	
	Total	% Variance	Cumulative %		Total	% Variance	Cumulative %	PC1	PC2
1	5.393	77.042	77.042	Cu	5.357	76.531	76.531	0.987	−0.036
2	1.041	14.867	91.909	Ni	1.076	15.378	91.909	0.972	0.015
3	0.198	2.835	94.744	Zn				0.944	0.096
4	0.153	2.192	96.936	As				0.939	−0.028
5	0.115	1.650	98.585	Pb				0.922	0.071
6	0.083	1.193	99.778	Cr				0.901	0.264
7	0.016	0.222	100.000	Cd				0.047	0.995

Extraction method: principal component analysis; rotation method: Varimax.

Moderate or even no contamination with these five metals (As, Cr, Cu, Pb, and Zn) was observed in deeper soils. Additionally, the degree of heavy metal pollution appeared similar, corresponding to a generally observed decreasing concentration of heavy metals from topmost soil to deeper soil (Xiao *et al.*, 2012).

According to the Müller (1981) scale, the heavy metals can be classified in the unpolluted category for all samples. Cr, Cu, Ni, Pb, and Zn had lower I_{geo} values, averaging less than 0 for three layers, indicating that the soils remain unpolluted. This result was in agreement with the results of pollution assessments using CFs. The averaged degree of pollution of these metals decreased in the following order: As > Ni > Zn > Cu > Pb > Cr for the first layer, Ni > As > Zn > Cu > Pb > Cr for the second layer, and Ni > As > Zn > Cu > Pb > Cr for the bottom layer.

Assessment of Heavy Metal Concentration Using CFs and TUs

The CFs for the heavy metals for all samples are presented in box-whisker plots (Figure 5). The empirical index provides a comparative means for assessing the level of heavy metal pollution (Hu *et al.*, 2013a). The analysis indicates that the soils are polluted with As, Ni, Pb, and Zn and act as a sink for heavy metals contributed by a multitude of anthropogenic sources (Chandra *et al.*, 2013).

The sum of the toxic units ($\sum$TU) of all metals has been used to assess the potential acute toxicity of heavy metals in each soil sample. All samples have $\sum$TU values that do not exceed 1.5 in mudflat and arable land, implying that the soil had no toxicity in the YRD. The TUs, $\sum$TU, and relative contributions of all the heavy metals at each soil increment in the sampling sites are illustrated in Figure 6. The mean TU values of heavy metals in soil samples decreased in the order Ni > As > Cr > Cu > Zn > Pb. This indicated relatively high contributions of Ni (32.13% ±1.18%), As (25.80% ±2.43%), Cr (17.98% ±0.66%), Cu (13.46% ±1.61%), Zn (6.00% ±0.50%), and Pb (4.63% ±0.63%) to $\sum$TU. Zhang *et al.* (2008) reported that $\sum$TU increased away from the source to the lower reaches of a freshwater river, but our results indicated that the $\sum$TU values decreased from inland to the coastal wetlands. Additionally, the sampling sites were less toxic because metal concentrations in these soils were all below severe effect level (SEL) values (Figure 6). This suggests that heavy metal pollution in coastal wetlands has not resulted in serious ecological risk. Generally, the contamination levels of four land use types followed the order *Suaeda heteroptera* wetland > reed swamp > arable land > mudflat. Cr and As had much higher TUs than Zn and Pb in all samples, which is in agreement with the findings of Xiao *et al.* (2012). In the mudflat, all sites had TUs < 1.5 and $\sum$TU < 2, suggesting less contamination with heavy metals at these sites and little potential to produce adverse effects on organisms in the area (Xiao *et al.*, 2012).

DISCUSSION

The discussion section compares the differences in heavy metal concentrations with those in other areas. Factor analysis, CFs, and I_{geo} are applied to evaluate the contamination conditions. Moreover, recommendations for protecting the vulnerable and newly created coastal area have also been provided.

General Comparison of Contamination of Heavy Metals

The statistical results summary and other comparisons of heavy metal concentration are summarized in Table 7. The average concentration of Cr for the YRD was higher than that of Sanya Bay but much lower than in other regions. For the other selected metals (Cu, Ni, Pb, and Zn), the mean concentrations in the YRD were comparable with those in Bohai Bay,

Table 5. *Total variances and rotation component matrices of the second soil layer (20–40 cm).*

Principal Component	Initial Eigenvalues			Element	Rotation Sum of Squared Loadings			Rotated Component Matrix	
	Total	% Variance	Cumulative %		Total	% Variance	Cumulative %	PC1	PC2
1	5.376	76.796	76.796	Cu	5.376	76.796	76.796	0.964	0.079
2	1.058	15.111	91.907	As	1.058	15.111	91.907	0.957	−0.146
3	0.195	2.785	94.691	Ni				0.947	−0.102
4	0.145	2.072	96.763	Pb				0.943	0.059
5	0.106	1.514	98.277	Cr				0.939	0.142
6	0.079	1.123	99.400	Zn				0.930	−0.030
7	0.042	0.600	100.000	Cd				0.001	0.997

Extraction method: principal component analysis; rotation method: Varimax.

Table 6. *Total variances and rotation component matrices of the third soil layer (40–60 cm).*

Principal Component	Initial Eigenvalues			Element	Rotation Sum of Squared Loadings			Rotated Component Matrix	
	Total	% Variance	Cumulative %		Total	% Variance	Cumulative %	PC1	PC2
1	4.995	71.351	71.351	Zn	4.949	70.694	70.694	0.964	0.006
2	1.050	14.998	86.349	Cr	1.096	15.655	86.349	0.964	0.063
3	0.494	7.056	93.406	Ni				0.954	0.156
4	0.289	4.128	97.533	Cu				0.950	0.183
5	0.082	1.172	98.706	As				0.853	−0.203
6	0.062	0.889	99.595	Pb				0.739	0.189
7	0.028	0.405	100.000	Cd				0.070	0.978

Extraction method: principal component analysis; rotation method: Varimax.

Rongcheng Bay, and Liaodong Bay but were lower than those found in the Yangtze and Pear River estuaries, whose surrounding areas are the other two most heavily urbanized zones in China. The present metal concentrations in the YRD are lower than those in Jiaozhou Bay. Moreover, with the exception of Ni, heavy metal concentrations in soils were lower than those in the Pearl River Delta, where industrialization and urbanization rapidly developed after the open and reform policy was launched in 1978. In comparison with Lu *et al.* (2014), we found the As, Cd, Cr, Cu, Ni, Pb, and Zn concentrations in the YRD to be at lower levels. The mean concentrations of these elements from the newly created YRD were generally lower than for other deltas across the world (*e.g.*, Jinzhou Bay, Bohai Bay, Jiaozhou Bay, Yangtze Estuary, and Hong Kong). Bai *et al.* (2014) found that most soil samples in the YRD have been moderately polluted by As and Cd. Compared with other elements, both Cd and As had higher enrichment factors exceeding moderate enrichment levels. The TU values of these elements did not exceed probable effect levels. High metal concentrations and hence ecotoxicity were found in the most urbanized and most densely populated areas (Jennerjahn and Mitchell, 2013; Yang *et al.*, 2012). This implied that metal pollution in the estuary is less serious than in other deltas, due to less industrialization and human activity in the YRD (Lu *et al.*, 2014). Therefore, the YRD could be regarded as a "clean island" because heavy metals in soils of this region are less accumulated compared with those in other regions of the Pearl River Delta (Xiao *et al.*, 2012). Metal concentration has been aggravated in the YRD in recent decades because of rapid exploration of oil and irrigated agriculture; increasing concentrations of Cd, which were associated with anthropogenic activities, might have resulted from rapid development of local petrochemical industries in this study area, and this should be closely followed to sustain the environmental quality of this green island in the future. For comparison, the average upper continental crust values in East China and other regions throughout the world are listed in Table 7. In surface sediments from the YRD, the mean concentrations of Cu, Ni, and Cr were lower than their corresponding average values in the upper continental crust of East China. Though the measured values were below threshold values based on quality guidelines (Long *et al.*, 1995), an accumulation mechanism may lead to concentrations harmful to life even when anthropogenic inputs are low (Koukina and Vetrov, 2013).

Implications for Wetland Management

The YRD, as a newly created wetland, is subjected to anthropogenic activities that make it susceptible to heavy metal contamination. Thus, prevention and management of heavy metal pollution is required. Local government action should establish quality guidelines and standards and environmental regulations and laws, build the capacity for advanced scientific research, and apply technological methods to prevent heavy metal pollution (Naser, 2013).

Establishment of Environmental Management Laws and Coordination

Environmental protection legislation is based on many national laws and regulations as well as regional and

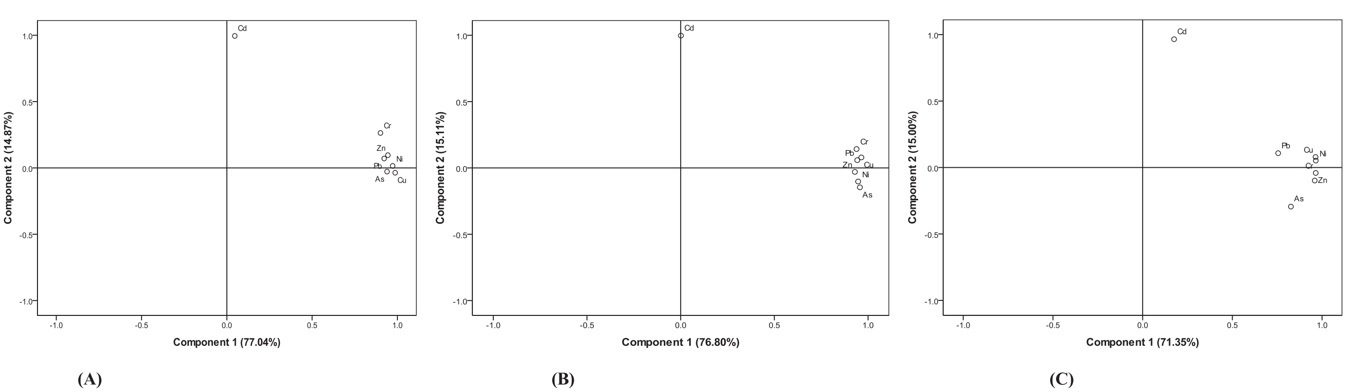

Figure 3. The factor analysis loading plot from the newly created wetland in the Yellow River Delta, which shows that these metals can be classified into Group 1 (Cd) and Group 2 (As, Cu, Cr, Pb, Ni, and Zn) according to similarities in behavior and distribution of the heavy metals.

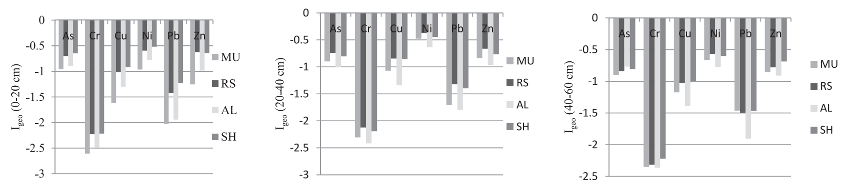

Figure 4. I_{geo} of As, Cr, Cu, Ni, Pb, and Zn of three soil layers. The I_{geo} value of As, Cr, Cu, Ni, Pb, and Zn are below 0, implying that the soils in the YRD are not polluted by these heavy metals. Relatively high I_{geo} values for Ni in three layers suggest that these layers are severely polluted with Ni.

international agreements (Naser, 2013). China has several national laws with respect to protecting the environment, such as the "Sea Area Use" Law, Fishery Law, Marine Environmental Protection Law, regulation law to avoid the pollution of marine environments by coastal engineering projects, and regulation laws for environmental protection management related to petroleum survey exploration. Regional laws include the regulation of Shandong Peninsula Blue Economic Zone, Dongying Modern Marine Fishery Development Plan, and Dongying City Coastal Zone Integrated Management Plan (Shandong Provincial Department of Forestry, 2013). It is confirmed that formulating wetland utilization laws would decrease the levels of heavy metal contamination in estuarine areas (Hosono *et al*., 2011). The government should establish general strategies for marine environmental management and acquire the ability to tackle disputes; the enterprise is one of the main bodies of marine environmental management that provides funding for public marine environmental protection activities. A marine environmental management model, including government, enterprise, and the public, needs to be established.

Failure to identify, recognize, and specifically designate wetlands or wetland communities that should be prioritized for conservation, protection, or restoration enables continued incremental losses of wetland area and function at both local and regional scales (Clare *et al*., 2011). More comprehensive land use planning that identifies high priority wetlands would allow land managers, developers, and individual landholders to make more informed decisions about land acquisition and provide them with the ability to weigh the potential benefits and costs associated with development (Clare *et al*., 2011). From the perspective of land developers, better regional planning and prioritization of high-value wetlands provides increased certainty and decreased risks associated with the existing permit process. This fragmentation of decision making and the general failure to better integrate planning at multiple scales has contributed to the ineffectiveness of the wetland policy.

Land use planning and regulatory decisions are made at different scales by multiple governments and agencies. This fragmentation of decision making, and the general failure to better integrate planning at multiple scales, has contributed to the ineffectiveness of the wetland policy. The conflict between exploration of coastal areas and resource protection is obvious: the rate of resource industry development is too high and disaster reduction and prevention measures are too weak, and integrated coordination needs to be strengthened. Pollutants from land-based sources have not been tackled effectively, and some marine engineering projects would damage marine resources. Laws and regulations for marine ecosystem protection associated with construction need to be strengthened, and a capability for effective coordination by the coastal zone integrated-management committee needs to be established. A general investigation of marine utilization status at a large scale was completed in 2008 and has clarified the status of marine utilization. An emergency plan for preventing storm tides, tsunamis, and disasters related to sea ice, red tide, and oil spill accidents has also been established.

The coordination ability of the coastal zone integrated-management committee needs to be strengthened. Overlap in administrative management and the unified coordination agency should be strengthened. Conflicts of interest and the lack of an integrated development plan for the management department often leads to uncoordinated management. The environmental protection of estuarine and marine environments involves different departments and areas; a regional coordination mechanism for launching pollution control standards, laws, and regulations to avoid pollution should be established. The marine management department has established the coastal zone integrated management committee. The government should establish the policy of compulsory insurance for oil contamination and a compensation fund system,

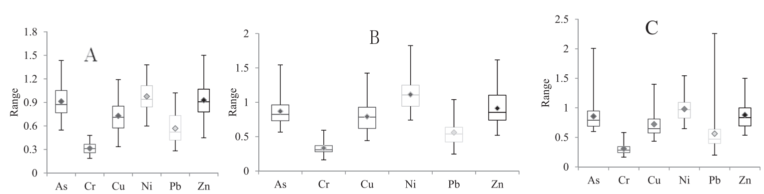

Figure 5. Box-whisker plots showing CFs for heavy metals in the YRD: (A) 0–20 cm, (B) 20–40 cm, and (C) 40–60 cm.

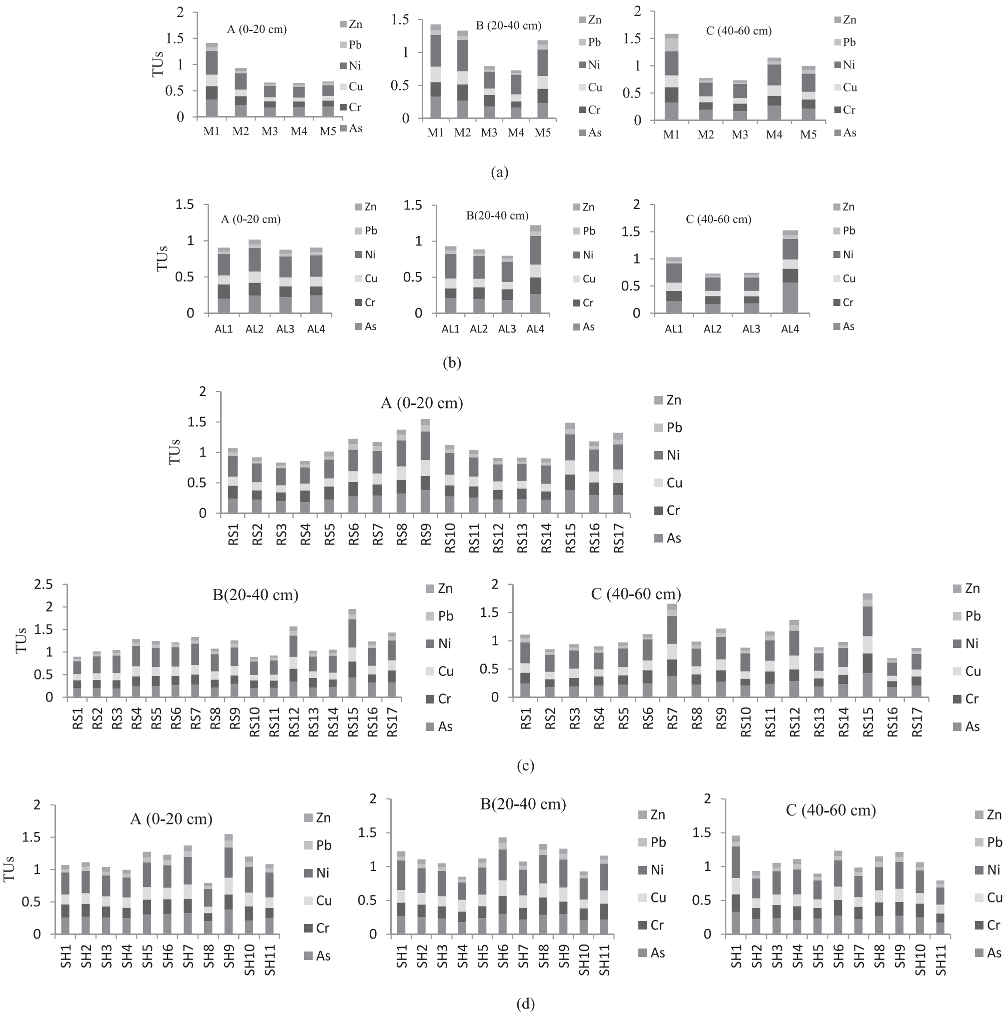

Figure 6. Sum of the toxic units of heavy metals in three soil profiles in the newly created wetland of the Yellow River Delta. The TU values of the samples do not exceed 1.5 in mudflat and arable land, implying that the soil had no toxicity in the YRD. The mean TU values of heavy metals in soil samples decreased in the order Ni > As > Cr > Cu > Zn > Pb. Generally, the toxic levels of four land use types followed the order *Suaeda heteroptera* wetland > reed swamp > arable land > mudflat. Cr and As had much higher TUs than Zn and Pb in all samples.

while formulating an ecological compensation mechanism in the three levels of national government, regional government, and industry, applying economic policies and laws.

Monitoring and Assessment

Investigating heavy metal pathways and the response of estuarine organisms to heavy metal pollution is fundamental to the implementation of assessment and remediation strategies (Naser, 2013). As China's second-largest oil field is located in the YRD, and the delta has become an important base of manufacturing and agricultural production in the country, wetland pollution through groundwater is increasingly serious, particularly with regard to the petrochemical and paper-manufacturing industries (Qi and Luo, 2007; Wang *et al.*, 2013). Additionally, oil industry activities may interfere with underground reservoirs, which increases the risk of heavy metal contamination. Adopting and applying environmentally friendly methods and technologies for soil disposal may minimize the release of pollutants. The wetlands ecosystem of the YRD is becoming more fragile and susceptible to natural hazards. Tidal flats play an important role in hydrologic and ecological processes in the coastal zone, which is also an ideal environment for wildlife, fishing, and recreation (Wang, Qi, and Zhang, 2012). However, with the intense operation of oil exploitation, the buffering and ecological effects of wetlands and salt marshes have greatly degenerated over the last 20 years. Spatial and temporal environmental monitoring is required. Conservation planners must make both tactical and strategic decisions to maintain viable metapopulations given the near-term impacts of habitat fragmentation, and they must make strategic decisions to identify critical habitats under future climates and facilitate potential range adjustments over longer time spans (Wright, 2010).

Table 7. *Comparison of the mean values of soil heavy metal and As concentration in the YRD with other coastal delta and quality guidelines.*

	Cu (mg/kg)	Pb (mg/kg)	Zn (mg/kg)	Cd (mg/kg)	Cr (mg/kg)	Ni (mg/kg)	As (mg/kg)	Reference
YRD Newly Created Wetland	17.48	14.67	58.97	0.14	20.70	25.21	8.49	This study
North China (Liaoning Province)								
Jinzhou Bay	417	753.2	6419		60.6		396.5	Zhang *et al*., 2008
Liaodong Bay, Bohai Sea	19.4	31.8	71.7	1.2	46.4	22.5		Hu *et al*., 2013b
Bohai Bay	28	21.2	102.5	0.2				Zhan *et al*., 2010
North China (Shandong Province)								
Jiaozhou Bay	25.1	21.9	85.0	1.47	42.8			Wang *et al*., 2010
Rongcheng Bay,	19.2 ± 7.3	34.1 ± 12.3	64 ± 15.5	0.15 ± 0.06	33.0 ± 10.4			Huang *et al*., 2013
Coastal Shandong Peninsula	20.0 ± 7.2	28.4 ± 3.7	74.7 ± 22.4		57.8 ± 10.7	31.2 ± 6.5		Li *et al*., 2013
Yellow River Delta	22.26	21.01	82.71	0.68	78.59	35.6	31.03	Lu *et al*., 2014
Yellow River Delta	30.7 ± 5.5	28.56 ± 5.3	95.1 ± 11.0	0.9 ± 0.1			35.8 ± 2.4	Bai *et al*., 2014
East China (Shanghai)								
Yangtze River Estuary	38.0	54.1	90.2	0.11	51.7	50.9		Hu *et al*., 2013
Yangtze River Intertidals	30.7 ± 9.7	27.3 ± 5.6	94.3 ± 23.9	0.261 ± 0.125	78.9 ± 19.7	31.8 ± 7.2		Zhang *et al*., 2009
Yangtze River Estuary	28	21.9	78	0.2	52.1		11.6	
East China (Jiangsu Province)								
Yellow Sea Coast			63.02		54.1	26.41		Fang *et al*., 2010
Haizhou Bay	42.38	90.07	385.33	1.76	116.67			Zhang *et al*., 2013
Southeast China (Fujian Province)								
Xiamen Bay	44.0	50.0	139	0.33	75	37.4		Zhang *et al*., 2007
Fujian Coastal Areas	22	37.1	96	0.08	57.4	27.4	9.1	Zhang *et al*., 2008
Southeast China (Guangdong Province)								
Daya Bay, Guangdong	20	223	108	0.19	63	28		Gao *et al*., 2010
College Town, Pearl River	42.4	36.1	80.3	1.1	95.4	18.8		Xiao *et al*., 2012
Pearl River Estuary	63.9	68.3	172.0		80.7	46.6		Yang *et al*., 2012
Shenzhen Bay	48.8	46.0	135			29.9		Huang *et al*., 2003
Southeast China (Hong Kong)								
Victoria Harbor	171	69	223	1.3	51	23	7.5	Tang *et al*., 2008
Southeast China (Hainan Province)								
Hainan Island	15.0 ± 7.5	27 ± 7.6	73.7 ± 34.8	0.09 ± 0.01	53.1 ± 24.8	23 ± 10.9	9.5 ± 2.6	Hu *et al*., 2013a
Sanya Bay	9 ± 7	18 ± 8	53 ± 28	0.13 ± 0.08	12 ± 3		7 ± 2	Qiu *et al*., 2011
Dongzhai Harbor	18	19	57	0.11	40		13	Qiu *et al*., 2011
Average Upper Crust of East China	28	17	67	0.09	92	47	5.0	Gao *et al*., 1998
Shandong Background	24	25.8	63.5	0.087	66	25.8	9.3	
Southeast China (Guangxi Province)								
Beibu Gulf	79	19	55	0.1	45	18.7		Gan *et al*., 2013
Ontario Guidelines								Ontario Ministry of Environment and Energy Staff, 1993
LEL	16	31	120	0.6	26	16	6	Gao *et al*., 1998
SEL	110	250	820	10	110	75	33	

LEL denotes lowest effect level; SEL denotes severe effect level; empty cells denote no data available.

Many studies have shown that compensatory laws and policies have not been effective in maintaining wetland areas and function (Clare *et al*., 2011). The principal intent of the U.S. Federal Water Pollution Control Act was to "restore and maintain the biological, chemical, and physical integrity of the Nation's water" in part through the establishment of the program. In 1993, Alberta, Canada, introduced a regional wetland policy that was primarily applied to marsh wetlands in the settled areas of the province (Rubec and Hanson, 2008). While the stated policy goal is to "sustain the social, economic, and environmental benefits that functioning wetlands provide, now and in the future," the implementation of the policy has focused on achieving no net loss of wetland area through conserving wetlands in a natural state; mitigating degradation or loss as close to the site as possible; and enhancing, restoring, or creating wetlands in areas where they have been depleted or degraded (Rubec and Hanson, 2008).

Current approaches in China to land use planning do not identify and prioritize wetlands in advance of development (Clare *et al*., 2011). Requirements for compensation are inadequately enforced. Some government agencies also consider their role to be about managing for development, rather than conserving or protecting wetland resources. The pollutant discharge from land-based sources of the Yellow River Basin has not been tackled and has caused increased pollution of estuarine areas. Therefore, the government should monitor anthropogenic activities, such as mudflat exploration for marine engineering projects and dam construction in estuarine areas, that have damaged habitat systems and have caused ecological deterioration. In the harbor, pollution from ships, oil and gas, and aquaculture has increased, and the system remains vulnerable to hazards such as storm tides, tsunamis, and saltwater intrusion. Thus, the management department should monitor the estuarine zone, coastal tourism vocation zone, and typical marine ecological vulnerabilities, and enhance environmental monitoring and integrated management capability. Moreover, we should also monitor the effects of the Xiaolangdi hydrologic engineering construction on the allocation of water resources in the low reaches of the YRD. High standards for storm tide, red tide, and other marine disaster prevention systems have not been established in the YRD, and it lacks modern alarm facilities and the capability to monitor extreme climate events in the event of disaster. Shengli Oilfield, an important oil-producing area in China, is located in the YRD, and there are many oil facilities on land and for offshore oil exploitation. Oil contamination has a deleterious effect on the basic geotechnical properties of coastal soil. When soils are contaminated with oil, the process of remediation is much longer and may have significant environmental and engineering effects. The extent of oil contamination and its influence on coastal soil should be investigated. The effect of drought on estuarine ecosystems is another form of extreme event, and further work is needed to understand the likely impacts on estuaries when freshwater flows are low for long periods of time (Jennerjahn and Mitchell, 2013). Reducing pollution, maintaining hydrologic balance, and protecting wetland biological diversity and integrity are important activities to maintain and improve the resilience of wetland ecosystems so that they continue to provide important functions under changing climatic conditions.

In distinguishing between natural and anthropogenic components of change, a better understanding of the respective processes is necessary (Jennerjahn and Mitchell, 2013). Bearing in mind that estuaries "provide a wider variety of ecosystem services and an increased delivery of societal benefits" on the one hand, but "have more human-induced pressures than many other ecosystems" (Elliott and Whitfield, 2011) on the other, proper management is required to sustain ecosystem services. Therefore, our recommendation is that management efforts of estuarine systems should focus on modeling various heavy metal contamination scenarios and collaboration with decision makers and relative stakeholders. Protection of estuarine wetlands is recommended for ecosystem health and to meet the requirements of tourism development.

CONCLUSIONS

The CF and I_{geo} have demonstrated that the YRD estuarine ecosystems are still in their unaffected state with respect to metal pollution (Wang *et al.*, 2013). The present study indicates that the concentrations of heavy metals in the study areas are lower than average, owing to less anthropogenic activity in the newly created wetland. The YRD may still be considered a "clean wetland" because some heavy metals were found to be less accumulated in this region compared with other regions, *e.g.*, the Pearl River Delta, which has been heavily polluted by heavy metals during recent decades. Factor loading reveals that PC1 was strongly and positively related to As, Cr, Cu, Ni, Pb, and Zn, while PC2 exhibited highly positive factor loading on Cd. The current information certainly justifies the requirements to initiate more detailed research on the impact of higher concentrations of heavy metals in the estuarine ecosystem. The national Shandong Peninsula Blue Economic Zone Development Plan in 2010 compels further understanding of the distribution and potential risk of metal pollution on the east coast of China, where rapid economic development and urbanization has occurred and metal pollution has become an obvious problem. Therefore, current and future impacts on the estuarine ecosystem associated with public health and natural disasters should be evaluated to provide scientific feedback for preventing pollution caused by intensive anthropogenic activities. Although the concentrations of heavy metals did not exceed permissible limits, regular monitoring of heavy metals is necessary to prevent human health risks and to ensure healthy ecosystem conditions. Therefore, there is a critical need for an approach to monitoring heavy metal concentrations and distributions, and for a comprehensive strategy to combat and manage heavy metal pollution.

ACKNOWLEDGMENTS

This research is founded by the National Natural Science Foundation of China (No. 41361018, 41101084); the opening fund (PK2013003) of the Key Laboratory of Poyang Lake Wetland and Watershed Research (Jiangxi Normal University), Ministry of Education; the National Basic Research Program of China (2010CB950900, 2009CB421100); the Collaborative Innovation Center for Major Ecological Security Issues of Jiangxi Province and Monitoring Implementation (No. JXS-EW-00); and the open fund of Jiangsu Key Laboratory of Environmental Change and Ecological Construction. Zongwen Ma has provided useful help during field investigations. We also thank the anonymous reviewers for their valuable comments, which greatly improved the manuscript.

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