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ORIGINAL PAPER

Contamination characteristics of heavy metals in wetland soils along a tidal ditch of the Yellow River Estuary, China

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Abstract Surface soils (0–20 cm) were collected from along a tidal ditch of the Yellow River Estuary in August of 2007. Samples were subjected to a total digestion technique before they were analyzed for total concentrations of As, Cr, Cd, Cu, Ni, Pb, Zn, P and S in order to investigate heavy metal contamination levels in wetland soils nearby the tidal ditches and their main sources. Results showed that the mean concentrations of these heavy metals except for As and Cd were lower than the Class I criteria. Nearly all sampling sites showed lower contamination levels for As and Cd, while no contamination levels for other heavy metals. Cr, Cu, and Ni mainly originated from parent rocks, and Pb and As might originate from tidal seawater and oil field pollution, respectively; while Cd and Zn mainly originated from parent rocks and tidal seawater. Most of heavy metals showed significant correlations with total concentrations of P and S, however, no significant correlations were observed between them and soil pH, slat and soil organic matter.

Keywords Heavy metals · Wetland soils · Assessment · Tidal ditch · Yellow River Estuary

1 Introduction

Tidal wetlands are the important components of coastal wetland systems, which play the important role in preventing erosion of coastal lines and seawater contamination. Meanwhile, they are also the most vulnerable and sensitive ecosystems to global sea-level rise and anthropogenic activities (Hardaway et al. 2002; USGS 1997). With the rapid industrialization and economic development in coastal region, heavy metals are continuing to be introduced to estuarine and coastal environment through rivers, runoff and land-based point sources (Zhang et al. 2007). Salt marshes serve as natural deposits of heavy metals in the estuarine system (Doyle and Otte 1997; Williams et al. 1994), and heavy metals can spread along with the tides and periodic floods (Suntornvongsagul et al. 2007). Therefore, heavy metal contamination in the salt marshes has been widely investigated in worldwide larger river delta (Zhang et al. 2007; Gorenc et al. 2004; Vital and Karl 2000). However, little information is available on heavy metal contamination in wetland soils along the tidal ditch due to seawater intrusion.

The estuary wetland of the Yellow River Delta 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 (Li et al. 2007). Seawater intrusion along tidal ditches is one important factor resulting in coastal wetland degradation, as tidal currents have been identified as the dominant factor controlling wetland evolution (D'Alpaos et al. 2005) due to changes of soil properties. Therefore, the

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primary objective of this study is to investigate contamination levels of heavy metals in wetland soils along a tidal ditch of the Yellow River Estuary.

2 Materials and methods

2.1 Site description

The study area is located in the Yellow River estuary (117°31′–119°18′E, 36°55′–38°16′N) is situated in the northeast of Dongying City, Shandong Province, China. It has a warm-temperate and continental monsoon climate with an annual precipitation of 596.9 mm, annual evaporation of 1,900–2,400 mm and annual average temperature of 12.9°C. Due to the effects of tidal seawater, *Suaeda salsa* is the dominant plant species in the tidal wetlands along the tidal ditch.

2.2 Soil sampling and analysis

One typical tidal ditch nearby the estuary of the Yellow River, which is located the core protection area of the Yellow River Delta National Natural Reserve, was chosen as the study area. Eight sampling sites were randomly selected along the tidal ditch in August of 2007. Surface soils (0–20 cm) were collected from each sampling site. All soil samples were placed into polyethylene bags and brought to the laboratory and air dried at room temperature for 3 weeks. All air-dried soils were sieved through a 2-mm nylon sieve to remove coarse debris and then ground with a pestle and mortar until all particles passed a 0.149-mm nylon sieve for determining soil chemical properties. Physico-chemical properties of the tested soils were listed in Table 1.

For analysis of the total concentrations of soil As, Cd, Cu, Ni, Pb, Zn, P and S, soil samples were digested by $HClO_4$–HNO_3–HF mixture in Teflon tubes. The solution of the digested samples was analyzed by inductively coupled plasma atomic absorption spectrometry (ICP/AES). Quality assurance and quality control were assessed using duplicates, method blanks and standard reference materials (GBW07401) from Chinese Academy of Measurement Sciences with each batch of samples (1 blank and 1 standard for each 10 samples). The recoveries of samples spiked with standards ranged from 95 to 105%. Soil organic matter (SOM) was measured using dichromate oxidation (Nelson and Sommers 1982). The amount of total soil C was determined by CHNOS Elemental Analyzer (Vario EL III, Germany). Soil inorganic carbon (In-C) was estimated by subtracting the amount of soil organic carbon from the total C. Soil pH and salinity were measured in supernatant of 1:5 soil–water mixtures using a pH meter and a salinity meter, respectively.

2.3 Assessment of heavy metal contamination

Assessment of heavy metal contamination in wetland soils were performed by the contamination index (P_i) and integrated contamination index (P_c).

The contamination index (P_i) is expressed by the fuzzy functions (Bai et al. 2010b, 2011):

$$P_i = C_i/X_a \quad (C_i \le X_a) \tag{1}$$

$$P_i = 1 + (C_i - X_a)/(X_b - X_a) \quad (X_a < C_i \le X_b) \tag{2}$$

$$P_i = 2 + (C_i - X_b)/(X_c - X_b) \quad (X_b < C_i \le X_c) \tag{3}$$

$$P_i = 3 + (C_i - X_c)/(X_c - X_b) \quad (C_i > X_c) \tag{4}$$

where C_i is the observed content of the substance; X_a is the no-polluted threshold value; X_b is the lowly polluted threshold value and X_c is the highly polluted threshold value.

Based on Chinese Marine Sediment Quality (GB 18668-2002) (National Standard of PR China 2002), Class I criteria was suitable for fishery, nature, and Class II could be used for industry and tourism site, while Class III could just be used for harbor. Therefore, X_a, X_b and X_c in above functions could be defined according to Class I, Class II and Class III criteria, respectively.

The following terminologies are used to describe the contamination index: $P_i \le 1$ no contamination; $1 < P_i \le 2$ low contamination; $2 < P_i \le 3$ moderate contamination; $P_i > 3$ high contaminations.

Integrated contamination index (P_c) is calculated by the form as follows (Bai et al. 2011; Huang 1987):

$$P_c = \sum_{i=1}^{7} (P_i - 1) \tag{5}$$

Table 1 Soil properties of the tested soils along the tidal ditch of the Yellow River Delta

	pH	Salinity (‰)	SOM (g/kg)	In-C (g/kg)	TP (mg/kg)	TS (mg/kg)
Maximum	8.55	0.48	3.71	13.02	640.37	584.00
Minimum	8.26	1.21	8.60	17.91	842.86	717.00
Average	8.41	0.83	6.88	15.48	710.40	643.59
SD	0.09	0.40	1.34	1.55	55.87	46.16

where if $P_i < 1$, then $P_{i-1} = 0$. For the description of integrated contamination index, the following terminologies are used: $P = 0$ for no contamination; $0 < P \leq 7$ for low contamination; $6 < P \leq 14$ for moderate contamination; $P > 14$ for high contamination.

2.4 Statistical analysis

Data analysis was carried using SPSS 12.0 software package. Pearson correlation was conducted to reveal their relationships between soil properties and heavy metals and to identify pollution sources of soil heavy metals. Difference was considered significant if $P < 0.05$.

3 Results and discussion

3.1 Mean concentrations of soil heavy metals

The mean concentrations of heavy metals in the wetland soils along the tidal ditch were summarized in Table 2. As Table 2 shown, the mean concentrations of As showed higher spatial variations with the variation coefficient of 21.6% compared to those of other heavy metals. In contrast, the mean concentrations of As and Cd exceeded the criterion values of Class I, while other metals were lower than the criterions of Class I. Moreover, the mean concentrations of these heavy metals, especially As and Cd, were greatly higher compared to those in the Yellow River Estuary in 1990s (Rui et al. 2008). This indicated that the wetland soils along the tidal ditch was increasingly contaminated by heavy metals (e.g. As and Cd) due to industrial and agricultural development in the nearby regions (Zhou et al. 2004). However, these concentrations of heavy metals were much lower than those in the Pearl River Delta (Li et al. 2001). This is closely related to the conservation of the National Natural Reserve of the Yellow River Delta.

3.2 Assessment of heavy metal contamination

Figure 1 illustrates the contamination levels of these heavy metals in wetland soils along the tidal ditch. The contamination index values showed low contamination levels for As and Cd, while no contamination levels for Cr, Cu, Ni, Pb and Zn in this region. This indicated that some As and Cd accumulations occurred in the wetland soils along the tidal ditch due to seawater intrusions. Therefore, it is suggested that some measures should be taken to reduce As and Cd concentrations though they showed lowly contaminated levels at current time, since they could be easily assimilated and accumulated by plant (Bai et al. 2011). Moreover, Kupchella and Hyland (1986) presented that the substance containing As might be transformed by the addition of carbon and hydrogen as a methyl group (CH_3) resulting in methylarsines—which is much more toxic to living things than the unmethylated forms. However, Adriano et al. (2004) presented that As is classed together with elements that are essential in animal nutrition.

The integrated contamination index for each sampling site was shown in Fig. 2. The integrated contamination index values showed low contamination levels for all sampling sites. This implied low environmental risks of heavy metals in this region.

3.3 Correlations between soil properties

Correlation analysis was performed between heavy metals and other soil properties. As Table 3 shown, Cd, Cr, Cu, Ni and Zn showed significant correlations among them at the

Table 2 Summary statistics of heavy metal concentrations in top 20 cm wetland soils along the tidal ditch and soil background values of Shandong Province, and guide values of marine sediment in China (mg/kg)

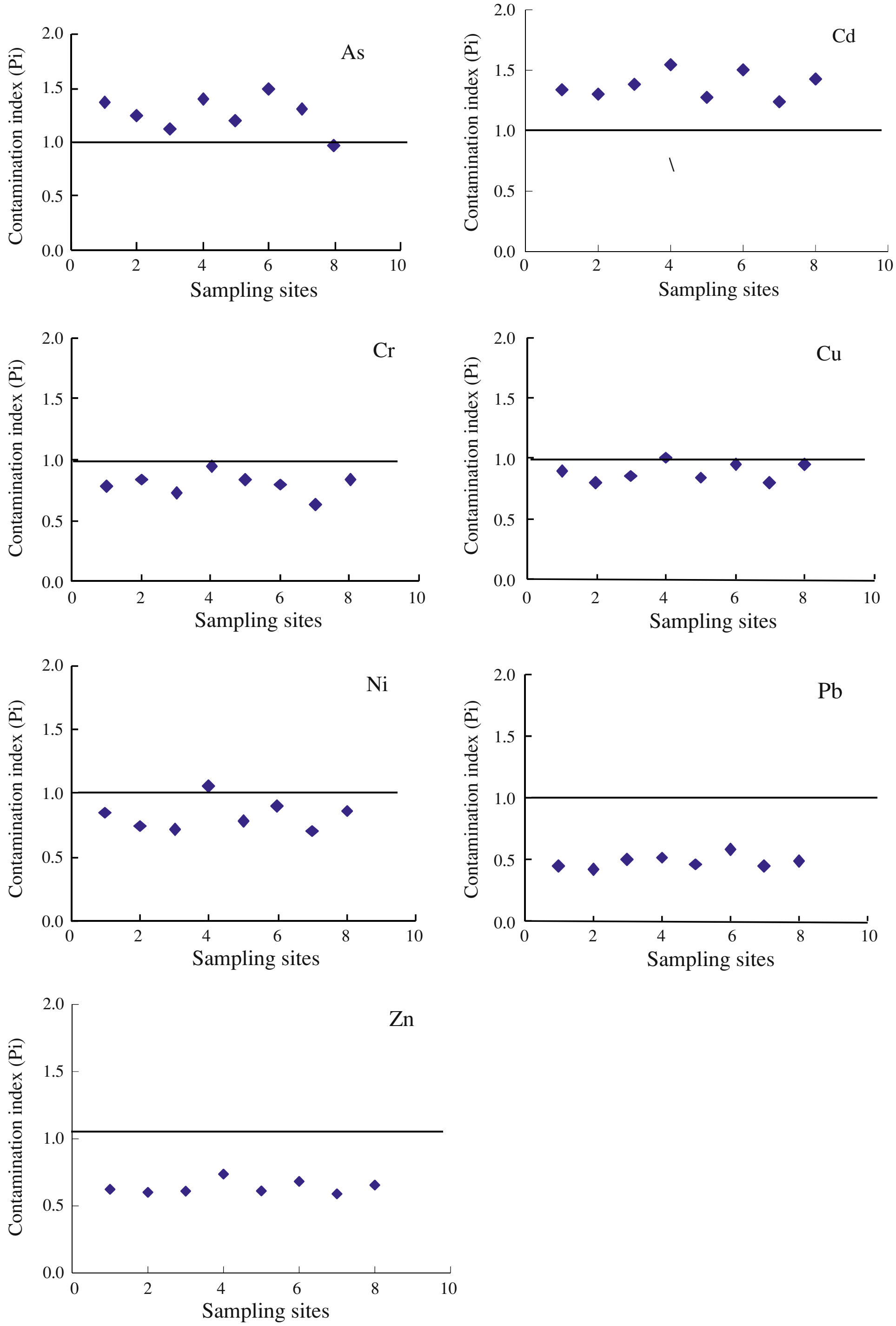
	As	Cd	Cr	Cu	Ni	Pb	Zn
Maximum	42.00	1.05	75.90	36.2	35.70	35.30	102.00
Minimum	19.00	0.74	50.50	28.000	24.10	25.20	88.50
Average	31.66	0.88	64.06	31.39	28.12	29.24	95.79
SD	6.84	0.10	6.96	2.64	3.65	2.93	6.98
Cv (%)	21.60	11.62	10.86	8.41	12.98	10.02	7.29
Sediment quality criteria of China[a]							
Class I	20	0.5	80	35	34[b]	60	150
Class II	65	1.5	150	100	40[b]	130	350
Class III	93	5	270	200	40[b]	250	600

SD standard deviation, *Cv* coefficients of variation

[a] SQGs National Standard of PR China (2002)

[b] Background concentration of marine sediments in Hong Kong (EPDHK 2005)

Fig. 1 Contamination indices of heavy metals in wetland soils along the tidal ditch



0.01 or 0.05 levels, indicated they could mainly originated from parent rocks since Cr and Ni were not identified as mobile elements as they tended to occur in trivalent oxide or co-precipitated with Fe hydrous oxide that had low mobility and bioavailability in soils and retained stable for a long time (Bai et al. 2010a; Kumpiene et al. 2008). However, the significant correlations between Cd, Pb and Zn showed they might originate from another common source such as tidal seawater, this suggested that Cd and Zn originated from two sources. Obviously, no significant correlations were observed between As and other heavy metals or soil properties, this indicated that As might originate from the third source, e.g. oil field pollution. With the exception of As and Pb, all other heavy metals showed

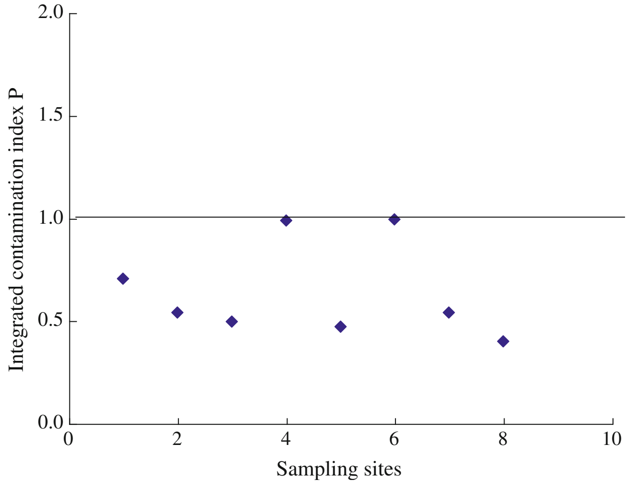


Fig. 2 Integrated contamination indices of sampling sites along the along the tidal ditch

significant correlations with TP. This was because most of them can form their precipitation of phosphorate. Additionally, all heavy metals except for As and Cr showed close correlations with TS due to sulfides. However, we observed all soil heavy metals were not correlated with soil pH, salt and SOM as they had little differences in these sampling sites on a small scale (Table 1). Conversely, numerous studies have demonstrated the importance of pH and SOM in determining the fate of heavy metals in soils (Bai et al. 2010a; Yin et al. 2002), since SOM could act as a major sink for trace elements due to it strong complexing capacity for metallic contaminants (Gonzalez et al. 2006). This is in agreement with the conclusions concluded by Fitz and Wenzel (2002), that no evidence were observed that SOM contributed to the sorption of significant amounts of As in soils (Table 3).

4 Conclusions

The wetland soils along the tidal ditch were lowly contaminated by As and Cd, and no contamination levels for Cr, Cu, Ni, Pb and Zn were observed. However, the integrated contamination index values showed low contamination levels for all sampling sites. These heavy metals such as Cr, Cu and Ni might originate from parent rocks, Pb from tidal seawater and As from oil pollution, while Cd and Zn might deviate from two sources such as parent rocks and tidal seawater. A small-scale effects of soil pH, salt and SOM were weak on heavy metal concentrations. Although soil heavy metals showed low contamination risks in this region, it is necessary to control As and Cd pollution to protect ecological security of the habitats for wild birds.

Table 3 Correlation coefficient matrix between soil heavy metals and other selected soil properties

	As	Cd	Cr	Cu	Ni	Pb	Zn	TS	TP	pH	Salinity	SOM
As	1.000											
Cd	0.279	1.000										
Cr	0.067	0.636	1.000									
Cu	0.236	0.929**	0.646	1.000								
Ni	0.419	0.864**	0.762*	0.939**	1.000							
Pb	0.401	0.807*	0.232	0.701	0.575	1.000						
Zn	0.378	0.943**	0.722*	0.952**	0.968**	0.710*	1.000					
TS	0.344	0.737*	0.389	0.814*	0.773*	0.725*	0.759*	1.000				
TP	0.386	0.735*	0.735*	0.803*	0.926**	0.405	0.908**	0.595	1.000			
pH	0.073	0.096	−0.201	0.050	−0.053	0.207	−0.101	0.407	−0.352	1.000		
Salinity	0.103	0.116	−0.204	0.062	−0.044	0.242	−0.085	0.415	−0.348	0.999**	1.000	
SOM	−0.450	0.331	0.192	0.167	0.030	0.408	0.230	0.145	0.106	−0.203	−0.200	1.000

** Correlation is significant at the $P < 0.01$ level

* Correlation is significant at the $P < 0.05$ level

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