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Arsenic and heavy metal pollution in wetland soils from tidal freshwater and salt marshes before and after the flow-sediment regulation regime in the Yellow River Delta, China

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SUMMARY

Soil samples were collected in tidal freshwater and salt marshes in the Yellow River Delta (YRD), northern China, before and after the flow-sediment regulation. Total concentrations of arsenic (As), cadmium (Cd), copper (Cu), lead (Pb) and zinc (Zn) were determined using inductively coupled plasma atomic absorption spectrometry to investigate the characteristics of heavy metal pollution in tidal wetlands before and after the regulation regime. The results demonstrated that marsh soils in both marshes had higher silt and total P contents, higher bulk density and lower sand contents after the flow-sediment regulation; moreover, soil salinity was significantly decreased in the tidal salt marsh. As and Cd concentrations were significantly higher in both marsh soils after the regulation than before, and there were no significant differences in the concentrations of Cu, Pb and Zn measured before and after the regulation. No significant differences in heavy metal concentrations were observed between freshwater and salt marsh soils, either before or after the regulation. Before the regulation regime, soil organic matter, pH and sulfer (S) were the main factors influencing heavy metal distribution in tidal freshwater marshes, whereas for tidal salt marshes, the main factors are soil salinity and moisture, pH and S. However, bulk density and total P became the main influencing factors after the regulation. The sediment quality guidelines and geoaccumulation indices showed moderately or strongly polluted levels of As and Cd and unpolluted or moderately polluted levels of Cu, Pb and Zn; As and Cd pollution became more serious after the regulation. Factor analysis indicated thatthese heavy metals including As were closely correlated and orginated from common pollution sources before the flow-sediment regulation; however, the sources of As and Cd separated from the sources of Cu, Pb and Zn after the regulation regime, implying that the flow-sediment regulation regime contributed to As and Cd accumulation in the YRD.

1. Introduction

Estuarine and coastal wetlands are complex and important ecosystems in which many critical environmental processes including sediment deposition, fresh water–salt water interaction, delta accretion, pollutant retention, and material-energy exchanges occur. They also provide habitats for a diverse array of flora and fauna (Mitsch and Gosselink, 2007). However, more than 90% of formerly important species and 65% of seagrass and wetland habitats have been depleted and destroyed due to anthropogenic activities, leading to degraded water quality and an increasing number of species invasions in these areas (Lotze et al., 2006).

As one of the most important effects caused by anthropogenic activities in large river/estuary systems, the global regulation of rivers and streams by building reservoirs and dams has altered downstream ecosystems and significantly changed many environment components during the twentieth century (Brandt, 2000). A variety of effects caused by flow regulation, e.g., the alteration and other effects of landscape elements and channel development (Wiens, 2002), river chemistry and sediment movement (Lake et al., 2000), nutrient cycling and decomposition rates (Harris, 2001), the survival of aquatic and terrestrial flora and fauna (Nilsson and Berggren, 2000) and the recruitment and survival of herbaceous community members associated with abuscular mycorrhizal fungi (Osmundson et al., 2002; Valett et al., 2005; Beauchamp et al., 2007; Smedberg et al., 2009; Navarro-Llacer et al., 2010), have been reported. Other studies have focused on

Abbreviations: SOM, soil organic matter; I_{geo}, geoaccumulation indices; YRD, Yellow River Delta; BD, bulk density; PCA, primary component analysis; SQGs, sediment quality guidelines; ISQVs, interim sediment quality values.

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investigating the responses of riparian and aquatic ecosystems and organisms (Bai et al., 2010b; Saintilan and Overton, 2010), in particular, the ecophysiology of riparian *Populus* spp. (cottonwood) and *Salix* spp. (willow) (Johnson, 2000; Hultine et al., 2010), which are the dominant overstory tree species along rivers and streams. The response of plankton and fish that live within the water body has also been studied with respect to flow regulation (Cortes et al., 2002; Osmundson et al., 2002). However, the responses of basic biogeochemical processes in the flow terminal estuarine environment, including hydrological and soil processes and their associated soil pollutant accumulations, need to be further investigated with respect to flow regulation (Li et al., 2009).

Various hydrological and salt conditions significantly influence biochemical processes in wetlands by changing the chemical forms of materials and affecting their spatial movement (Mitsch and Gosselink, 2007; Rai, 2008; Du Laing et al., 2009; Bai et al., 2011). Kumpiene et al. (2008) demonstrated that the variation in hydrological conditions and soil physicochemical properties could change the fixation, mobility and bioavailability of heavy metals in soil. Du Laing et al. (2009) also reported that metal contents were significantly correlated with salinity, clay and organic matter contents in estuarine and riparian floodplain soils. Heavy metals could enter the sediment surface through different biogeochemical processes such as deposition and precipitation onto the water–sediment interface (Liu et al., 2010). Presley et al. (1980) reported that sediment loadings, water discharge and particle size could alter heavy metal inputs to estuarine sediments. Du Laing et al. (2008) found that lower floodwater salinity enhanced the mobility of Cd and its uptake by duckweed in intertidal estuarine, whereas increased floodwater salinity could promote the desorption of metal from floodplain soils in the absence of sulfides (Du Laing et al., 2009). However, little information is available on the effects of flow-sediment regulation on arsenic and heavy metal concentrations in downstream tidal marshes.

The Yellow River has been and remains the second largest river in the world in terms of sediment load over the last few thousand years (Milliman and Meade, 1983). Xiaolangdi Reservoir began storing water in 1999, and considerable siltation occurred in the reservoir after commissioning, with a total sediment trapping of 32.47×10^8 t from 1997 to 2007 (Peng et al., 2010). The flow-sediment regulation scheme has greatly influenced wetland landscape patterns in the lower reach of the Yellow River since 2002 (Li et al., 2009). The variation in hydrological conditions was observed in the YRD (Table 1) due to the implementation of flow-sediment regulation (Cui et al., 2009), which is considered a major limiting factor of soil heavy metal mobilization and transformation in wetland ecosystems (Mitsch and Gosselink, 2007). Before the flow-sediment regulation scheme, lower concentrations of As and heavy metals

Table 1
The monthly average water discharge (WD) and sediment load (SL) at Lijin station (the nearest hydrological monitoring station to this study area) during the periods of 2002 and 2004–2005, since the application of the flow-sediment regulation.

Month	January	February	March	April	May	June	July	August	September	October	November	December
WD	320.70	189.73	124.93	104.83	225.33	890.87	1175.33	811.13	407.07	743.20	399.57	250.17
10^8 m^3	(207.72)	(101.97)	(59.54)	(36.98)	(177.64)	(578.66)	(226.84)	(627.61)	(270.05)	(830.03)	(341.83)	(186.80)
SL	0.73	0.25	0.27	0.18	0.94	12.37	16.87	15.80	5.36	8.75	1.33	0.38
10^8 t	(0.95)	(0.24)	(0.11)	(0.07)	(1.02)	(8.76)	(1.58)	(19.98)	(6.14)	(12.05)	(1.54)	(0.38)

Note: the values in the brackets denote standard deviations.

Table 2
Mean values of soil heavy metal and As concentrations in major world river deltas and the sediment quality guidelines in various countries (mg kg^{-1}).

River	Sampling date	As	Cd	Cu	Pb	Zn	References
Yellow River	1996	13.07	0.02	6.82	8.84	24.87	Rui et al. (2008)
	2006	18.16	0.04	12.48	10.99	39.18	Rui et al. (2008)
	April 2007	27.60	0.57	26.70	27.23	78.10	this study
	August 2007	40.75	0.96	31.94	28.99	91.33	this study
Yangtze River	May 2000	10.09	0.22	27.92	42.85	68.74	Gorenc et al. (2004)
	April 2005	–	0.28	32.13	27.82	98.44	Zhang et al. (2009)
	August 2005	–	0.19	26.49	25.88	82.13	Zhang et al. (2009)
Pearl River	2002	–	0.52	14.7	29.9	50.7	Wong et al. (2002)
	August 2005	–	4.22	68.2	32.3	311.1	Li et al. (2007)
	July 2007	–	1.72	348.0	102.6	383.4	Niu et al. (2009)
Mekong River	March 1997	–	0.4	47	37	144	Cenci and Martin (2004)
	October 1997	–	0.5	53	35	138	Cenci and Martin (2004)
Scheldt River	2003	–	6.4	158	21	1163	Vandecasteele et al. (2003)
Yenisey River	1998	–	1.85	120.8	28.74	193.80	Guay et al. (2010)
Amazon River	2000	–	–	11.44	17.0	78.0	Vital and Stattegger (2000)
Mississippi River	April 1998	–	0.89	19.06	16.37	29.42	Grabowski et al. (2001)
	August 1998	–	1.57	31.77	18.65	45.77	Grabowski et al. (2001)
Danube River	2003	10.4	0.4	21.9	11.9	59.9	Woitke et al. (2003)
Orinoco River	July 2001	–	–	3.81	16.58	81.73	Marchand et al. (2006)
Environmental background concentrations							
The loess materials of the YRD		10.7	0.095	21.1	21.6	64.5	CNEMC (1990)
Sediment quality guidelines in various countries							
Ontario Guidelines	LEL	6	0.6	16	31	120	OMEE (1993)
	SEL	33	10	110	250	820	
Hongkong ISQVs	ISQV-low	8.2	1.5	65	75	200	Chapman et al. (1998)
	ISQV-high	70	9.6	270	218	410	
Sediment Quality Criteria of China	Class I	20	0.5	35	60	150	National Standard of P.R. China (2002)
	Class II	65	1.5	100	130	350	

LEL, lowest effect level; SEL, severe effect level; SQG, sediment quality guideline; ISQV, interim sediment quality value; SQT, soil quality threshold.

were reported in wetland soils of the YRD (Rui et al., 2008) compared to other river deltas in China (Wong et al., 2002; Gorenc et al., 2004; Table 2). However, Grabowski et al. (2001) and Bragato et al. (2009) stated that soil heavy metal concentrations generally remain almost constant between seasons in the natural state. Therefore, the primary objective of this study is to investigate the characteristics of arsenic and heavy metal pollution in tidal freshwater and salt marshes before and after flow-sediment regulation regime in the YRD of China.

2. Materials and methods

2.1. Study area and sample collection

The Yellow River Delta (YRD), geographically spanning 118°07′–119°18′E and 36°55′–38°12′N (Fig. 1), is located on the south side of the Bohai Sea and has a warm-temperate monsoon climate. The flow-sediment regulation scheme is implemented from June to July of each year (since 2002) by the Yellow River Conservancy Commission to control the discharge of waterand sediments from the Xiaolangdi Reservoir (capacity $126.5 \times 108\ m^3$; Yu, 2006) and scour the lower reaches. Prior to the regulation regime, annual runoff and sediment loads into the YRD had been reduced by 50% since 1985 (Yu, 2006) due to climate change and human activities. Since 2002, flow-sediment regulation of the Xiaolangdi Reservoir has alleviated this decreasing trend and resulted in a slight increase (Li et al., 2009). Because of channel scouring in the lower reaches, monthly water discharge and sediment loads have greatly increased since regulation was enacted, especially from June to July, compared to the observed values before regulation (Table 1).

Surface soil (0–20 cm) samples were collected from nine selected sampling plots in tidal freshwater marshes and 15 sampling plots in tidal salt marshes in April of 2007 to represent soil conditions before the flow-sediment regulation. The top 20 cm of soil (three replicates) was collected from the abovementioned sampling plots in August of 2007 to represent soil conditions after the flow-sediment regulation (Fig. 1). In each sampling plot, soil samples (three replicates) were randomly collected using a soil auger (4.8 cm in diameter) and mixed to form a composite sample. Forty-eight soil samples were collected in this study. All soil samples were placed in polyethylene bags and brought to the laboratory, where they were air dried at room temperature. All of the air-dried soil samples were sieved through a 2-mm nylon sieve to remove coarse debris and then ground using a pestle and mortar until all particles passed a 0.149-mm nylon sieve for the determination of soil chemical properties. Additionally, a single 4.8-cm diameter soil core was collected from each site for bulk density (BD) and moisture determination.

2.2. Analytical methods

Soil samples were digested using a mixture of $HClO_4$, HNO_3 and HF in Teflon tubes for analysis of the total concentrations of As, Cd, Cu, Pb, Zn, P and S. The digested sample solutions were analyzed using inductively coupled plasma atomic absorption spectrometry (ICP/AES). Quality assurance and quality control were assessed using duplicates, method blanks and standard reference materials (GBW07401) obtained from the Chinese Academy of Measurement Sciences, which were included in each batch of samples (1 blank and 1 standard for each 10 samples). Accuracy of the analytical method was given as percent recoveries for each of the elements. Results were listed in Table 3.

Soil organic matter (SOM) was measured using dichromate oxidation (Nelson and Sommers, 1982). Soil pH was measured using a Hach pH meter (Hach Company, Loveland, CO, USA) (soil:-water = 1:5). Salinity was determined in the supernatant of 1:5 soil–water mixtures using a salinity meter (VWR Scientific, West Chester, Pennsylvania, USA). The soils were oven dried at 105°C for 24 h and weighed for soil bulk density and moisture. Soil particle size was analyzed using a Laser Particle Size Analyzer (Microtrac Inc., USA).

Table 3
Observed and certified values of elemental concentrations in standard reference material and detection limits for ICP/AES.

Element	Observed ± SD ($\mu g\ g^{-1}$)	Certified ($\mu g\ g^{-1}$)	Recovery (%)	Determination limits ($\mu g\ ml^{-1}$)
As	33.46 ± 1.70	33.5	99.88	0.0012
Cd	4.30 ± 0.17	4.3	100	0.0003
Cu	22.2 ± 0.56	21	105.71	0.001
Pb	97.83 ± 2.48	98	99.83	0.003
Zn	678.33 ± 18.50	680	99.75	0.002

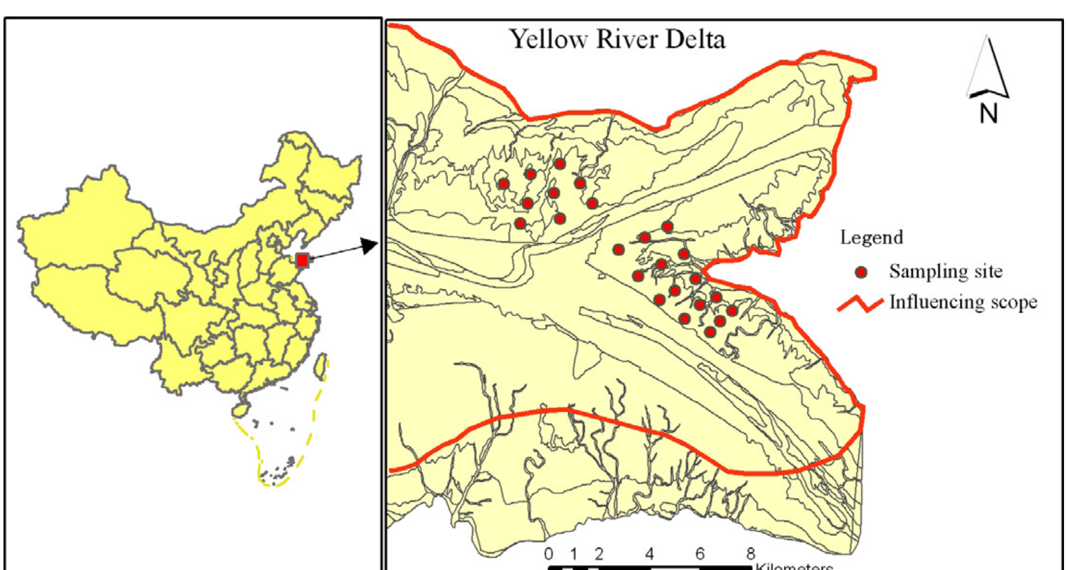

Fig. 1. Location map and sampling sites in the Yellow River Delta. The red line represents the influencing scope of flow-sediment regulation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.3. Geoaccumulation index

The index of geoaccumulation (I_{geo}) was used as a measure of metal pollution in sediments, as given by Muller (1981)

$$I_{geo} = \log_2 \frac{C_n}{1.5 \times B_n} \quad (1)$$

where C_n is the measured concentration of the element n in a sample and B_n is the natural background concentration of this element. These geochemical background values were obtained based on the environmental background concentrations of the loess materials of the Yellow River (CNEMC, 1990; Table 2). According to the baseline values, the samples can be classified as (i) unpolluted ($I_{geo} \leqslant 0$), (ii) unpolluted to moderately polluted ($0 < I_{geo} \leqslant 1$), (iii) moderately polluted ($1 < I_{geo} \leqslant 2$), (iv) moderately to strongly polluted ($2 < I_{geo} \leqslant 3$), (v) strongly polluted ($3 < I_{geo} \leqslant 4$), (vi) strongly to extremely polluted ($4 < I_{geo} \leqslant 5$), and (vii) extremely polluted ($I_{geo} > 5$).

2.4. Statistical analysis

Data analysis was carried out using SPSS 16.0 for Windows (SPSS München, Germany). Correlation analysis was performed to identify the relationships between the concentrations of trace elements and soil properties in both tidal freshwater and salt marshes. Primary component analysis (PCA) was conducted using Canoco 4.5 for Windows to identify the main influencing factors of heavy metal distribution. Factor analysis was performed to identify the sources of various heavy metals. ANOVA was implemented to test the differences between these trace elements and soil properties before and after the regulations were implemented and between tidal freshwater and salt marshes. Differences were considered significant when $P < 0.05$.

3. Results and discussion

3.1. Soil properties in tidal freshwater and salt marshes before and after regulation

The mean values of soil properties and metal concentrations in tidal freshwater and salt marsh soils before and after flow-sediment regulation are summarized in Table 4. After regulation, both marsh soils contained higher BD and silt contents and lower sand contents than those obtained before the flow regulation regime (Table 4). Tidal freshwater soils contained much higher sand and lower silt contents than those in tidal salt marsh soils (Table 4), and there were decreases in sand contents and increases in silt contents in the seaward direction. This might have been caused by the increase in downstream sediment loadings originating from quaternary loess (silt, 66.36%; clay, 14.73%; CNEMC, 1990) and the increase in the ratio of small sediment particles present (Table 1), which results from the carrying of much smaller suspended particles to the river estuary/delta than those occurring during natural floods when the bottom gate of the Xiaolangdi Reservoir, upstream of the study area, is opened. Most researchers have reported that the increased hydrodynamic forces that result prevent particle deposition in channels (Brandt, 2000; Doneker et al., 2004; Ahfir et al., 2007); thus, a much greater quantity of pollutants could be transferred to and settle in the YRD, as demonstrated by the preferential association of heavy metals with the fine fractions (Bai et al., 2010a, 2011). Teuchies et al. (2008) presented that a higher organic matter decomposition rate could occur in the oxic surface layer; however, the suspended organic matter-enriched tidal water in salt marshes could compensate for reductions in the accelerated decomposition of SOM, thereby alleviating differences in the SOM contents between both marshes, before and after regulation. Similarly, total S contents were not significantly decreased after regulation in both types of marsh soils ($P > 0.05$). However, total P showed a significant increase after regulation ($P < 0.05$). Because the abundant freshwater discharge inputs from the upper stream diluted the soil salinity of the tidal freshwater marshes after regulation, soil salinity levels were significantly reduced in both types of wetlands after flow-sediment regulation ($P < 0.05$). The average levels of soil salinity in this delta were significantly reduced (by approximately 36%) after the regulation due to abundant freshwater discharge inputs ($P < 0.05$; Table 4). Cui et al. (2009) also reported that freshwater input and flooding significantly reduced soil salinity in salt marshes in this region after a long-term period of monitoring from 2001 to 2007. Except for soil texture, there were no significant differences in other soil properties between tidal freshwater and salt marsh soils, indicating significant and similar influences of the flow-sediment regulation on downstream marsh soils.

Table 4
Mean soil properties and heavy metals in the wetland soils of tidal freshwater marsh and tidal salt marsh before and after flow-sediment regulation.

	Tidal freshwater marsh		Tidal salt marsh		Average	
	Before	After	Before	After	Before	After
Moisture (%)	23.15 ± 1.22^{a1}	24.41 ± 1.66^{a1}	24.87 ± 2.83^{a1}	27.70 ± 4.34^{a1}	24.01 ± 2.03^{a}	26.69 ± 2.66^{a}
Bulk density (g cm^{-3})	1.48 ± 0.05^{a1}	1.79 ± 0.06^{b1}	1.48 ± 0.08^{a1}	1.79 ± 0.07^{b1}	1.48 ± 0.65^{a}	1.79 ± 0.06^{b}
SOM (%)	5.13 ± 2.30^{a1}	4.93 ± 1.85^{a1}	6.47 ± 1.37^{a1}	6.10 ± 1.24^{a1}	5.8 ± 1.84^{a}	5.52 ± 1.55^{a}
Salinity (‰)	1.22 ± 0.75^{a1}	1.04 ± 0.64^{a1}	2.36 ± 1.05^{a2}	1.19 ± 1.01^{b1}	1.79 ± 0.90^{a}	1.12 ± 0.83^{b}
pH	8.33 ± 0.24^{a1}	8.05 ± 0.30^{b1}	8.38 ± 0.16^{a1}	8.43 ± 0.29^{a2}	8.36 ± 0.20^{a}	8.24 ± 0.30^{a}
Sand (%)	75.14 ± 12.25^{a1}	53.42 ± 8.08^{b1}	56.58 ± 3.74^{a2}	37.40 ± 14.65^{b2}	65.86 ± 7.99^{a}	45.41 ± 11.37^{b}
Silt (%)	23.24 ± 11.15^{a1}	42.37 ± 7.11^{b1}	38.05 ± 3.25^{a2}	52.19 ± 9.92^{b2}	30.65 ± 7.20^{a}	47.28 ± 8.52^{b}
Clay (%)	1.73 ± 1.34^{a1}	4.21 ± 1.27^{b1}	5.36 ± 1.88^{a2}	10.41 ± 4.84^{a1}	3.55 ± 1.61^{a}	7.31 ± 3.06^{b}
S (mg kg^{-1})	549.95 ± 182.32^{a1}	421.52 ± 78.81^{a1}	665.48 ± 127.14^{a1}	584.10 ± 112.30^{a2}	607.72 ± 154.73^{a}	502.81 ± 95.56^{b}
P (mg kg^{-1})	610.09 ± 33.73^{a1}	699.79 ± 61.20^{b1}	584.86 ± 64.85^{a1}	675.88 ± 54.47^{b1}	597.48 ± 49.29^{a}	687.84 ± 57.84^{b}
As (mg kg^{-1})	27.30 ± 4.62^{a1}	44.90 ± 8.77^{b1}	27.77 ± 4.81^{a1}	38.14 ± 5.92^{b1}	27.54 ± 4.72^{a}	41.52 ± 7.32^{b}
Cd (mg kg^{-1})	0.61 ± 0.09^{a1}	1.01 ± 0.16^{b1}	0.56 ± 0.09^{a1}	0.99 ± 0.18^{b1}	0.59 ± 0.12^{a}	1.00 ± 0.14^{b}
Cu (mg kg^{-1})	25.26 ± 5.21^{a1}	32.01 ± 13.15^{a1}	27.56 ± 6.00^{a1}	30.72 ± 6.47^{a1}	26.41 ± 5.61^{a}	31.37 ± 9.81^{a}
Pb (mg kg^{-1})	28.08 ± 6.02^{a1}	28.89 ± 5.79^{a1}	26.72 ± 6.15^{a1}	27.23 ± 4.40^{a1}	27.40 ± 6.09^{a}	28.06 ± 5.10^{a}
Zn (mg kg^{-1})	71.63 ± 8.94^{a1}	77.27 ± 5.14^{a1}	81.99 ± 15.79^{a1}	97.71 ± 8.53^{a1}	76.81 ± 12.37^{a}	87.49 ± 6.84^{a}

[a,b]Different letters indicate significant differences between soil properties and heavy metals in tidal freshwater marsh or salt marsh before and after the flow-sediment regulation.
[1,2]Different numbers indicate significant differences between soil properties and heavy metals in both tidal freshwater and salt marshes before or after the flow-sediment regulation.

3.2. Arsenic and heavy metal concentrations in tidal freshwater and salt marshes before and after regulation

No significant differences were observed for heavy metals such as Cu, Pb and Zn between freshwater and salt marsh soils either before or after regulation ($P > 0.05$). In freshwater and salt marsh soils, As and Cd concentrations were significantly elevated approximately 1.6 times compared with those observed after flow-sediment regulation ($P < 0.05$; Table 4). Cu, Pb and Zn also increased slightly despite the fact that they were not significantly increased in both soils after regulation ($P > 0.05$). This implied that flow-sediment regulation potentially impacts the accumulation of heavy metal and As in marsh soils in the Yellow River Delta. Elevated levels of As and Cd in marsh soils were possibly associated with the decrease in salinity, as salinity can affect the adsorption and desorption of these trace elements; to some extent, the intersection of salt and freshwater will also affect the concentration of heavy metals in water (Tang et al., 2010). Ehlken and Kirchner (2002) reported that higher water content and lower salinity had great potential to increase the absorbance capacity of soil solutions for heavy metals. Moreover, the subalkalinization environment and narrow pH ranges (Table 4) could decrease the mobility of metals and increase the ability of tidal soils to stabilize metals (Ullrich et al., 1999; Kumpiene et al., 2008). Moreover, this hypothesis is supported by historical records pertaining to the Yellow River Delta. Higher soil salt contents were observed in the 1990s (5.3–6.0‰, Wu et al., 1994) than those observed during the study period due to zero flow in the lower reach of the Yellow River; moreover, these soils also exhibited lower heavy metal contents, especially during August (Li et al., 1985; Rui et al., 2008). This verified that the flow-sediment regulation decreased salinity and heavy metal contents in soil due to freshwater discharge input.

In addition, As and Cd concentrations were significantly affected by BD, silt and total P in both types of marsh soil. However, Cu, Pb and Zn were closely related to SOM and total P in tidal freshwater marshes and significantly correlated with soil moisture, total S and total P in tidal salt marshes (Table 5). The specific surface area of soil increases as the particles become finer, leading to an increase in the effective contact area between adsorbent and adsorbate; this results in the adsorption of large amounts of heavy metals (Shim et al., 2003). This fact also supports the conclusion of Huang et al. (1992) and Zhang et al. (1999) that the seaward fluxes of heavy metals are matched by the capacity of riverine suspended matter delivery and sedimentary loads during increasing water discharge. Although some researchers have demonstrated that SOM can act as a major sink for trace elements due to its strong capacity for complexing metallic contaminants (Kalbitz and Wennrich, 1998; Gonzalez et al., 2006; Bai et al., 2010a), heavy metals and As were not significantly correlated with SOM in this study, with the exceptions of Cu, Pb and Zn in the freshwater marsh soils. The low C/N values (<20) observed primarily reflect marine production in both tidal marshes because C/N ratios have been used to distinguish between sedimentary organic matter of marine (4–10) and terrestrial (>20) origins (Mil-Homens et al., 2006). Thus, SOM was not considered to represent the main carrier of contaminants in the studied areas. Fitz and Wenzel (2002) also concluded that there was no evidence of OM contributing to the sorption of significant amount of As in soil. Total P was closely related to Cu, Pb and Zn in all soils because the phosphorate can reduce the mobility of these metals by ionic exchange and precipitation (Kumpiene et al., 2008). In the tidal salt marsh, Cu, Pb and Zn showed significant correlations with total S, which could be explained by the higher S content (sufficient sulfates) and the formation of insoluble sulfides under anoxic conditions due to tidal seawater flooding (Du Laing et al., 2008). This implied that the changed soil properties might be the main factors contributing to higher heavy metal concentrations through the greater potential for sequestering heavy metals from the overlying water during regulation-caused flooding (Spurgeon et al., 2008). Therefore, the increased soil heavy metal concentrations after flow-sediment regulation were caused by the increased input and retention capacity, leading to increased precipitation and deposition of particulate heavy metals to the surface soil (Cabrera et al., 1999; Loska and Wiechu, 2003). Zhang and Liu (2002) reported that concentrations of Pb, Zn and Cu were increased twofold depending upon the water discharge, the sediment origin and content, and the anthropogenic activities over the drainage basin.

PCA analysis was performed to assess the relationship between the spatial distribution patterns of the sampling sites and soil properties associated with the effects of flow-sediment regulation. The factor loading scores in biplots (Fig. 2) showed a clear separation between the sampling sites collected before flow-sediment regulation (green area) and those sites collected after flow-sediment regulation (pink area) along the first and second primary components. The first component accounted for 60.8% and 75.0% of the variances in the tidal freshwater and tidal saltwater marshes, respectively. For the second primary component, the corresponding values are 32.6% and 12.5%, respectively. In Fig. 2, two main groups can be clearly identified. The group identified in green contains soil samples collected before the regulation, and the group identified in pink includes samples collected after the regulation. Flow-sediment regulation played an important role in shaping the spatial distribution pattern of heavy metals in these two types of marsh soils. Site 15 in the freshwater marsh and Site 30 in the salt marsh failed to be grouped into the pink area in August but was grouped in the green area. Additionally, salt marsh Sites 7,

Table 5
Correlations between As, heavy metals and soil properties.

	As	Cd	Cu	Pb	Zn	Moisture	Bulk density	SOM	pH	Sand	Silt	Clay	S	P
Tidal freshwater marsh														
As	1.000	0.857**	0.255	0.265	0.320	0.323	0.831**	−0.157	−0.293	−0.493*	0.489*	0.445	−0.391	0.673**
Cd		1.000	0.500*	0.277	0.448	0.442	0.923**	0.120	−0.454	−0.656*	0.644*	0.668**	−0.155	0.821**
Cu			1.000	0.743**	0.924**	0.221	0.404	0.635**	0.223	−0.425	0.423	0.369	0.024	0.747**
Pb				1.000	0.845**	0.348	0.107	0.646**	0.397	−0.288	0.289	0.206	−0.010	0.494*
Zn					1.000	0.203	0.294	0.589*	0.262	−0.334	0.333	0.271	0.017	0.661**
Tidal salt marsh														
As	1.000	0.742**	0.302	0.225	0.632**	0.102	0.625**	−0.177	−0.087	−0.338	0.371*	0.177	−0.172	0.612**
Cd		1.000	0.205	0.141	0.695**	0.228	0.784**	−0.245	−0.098	−0.335	0.391*	0.128	−0.284	0.479**
Cu			1.000	0.876**	0.722**	0.756**	−0.044	0.331	0.538**	−0.439*	0.452*	0.320	0.479**	0.614**
Pb				1.000	0.647**	0.671**	−0.189	0.272	0.419*	−0.387*	0.393*	0.274	0.458*	0.445*
Zn					1.000	0.481**	0.425**	−0.148	0.476**	−0.401*	0.418*	0.296	0.608**	0.812**

* Significant correlation at the 0.05 level (2-tailed).
** Significant correlation at the 0.01 level (2-tailed).

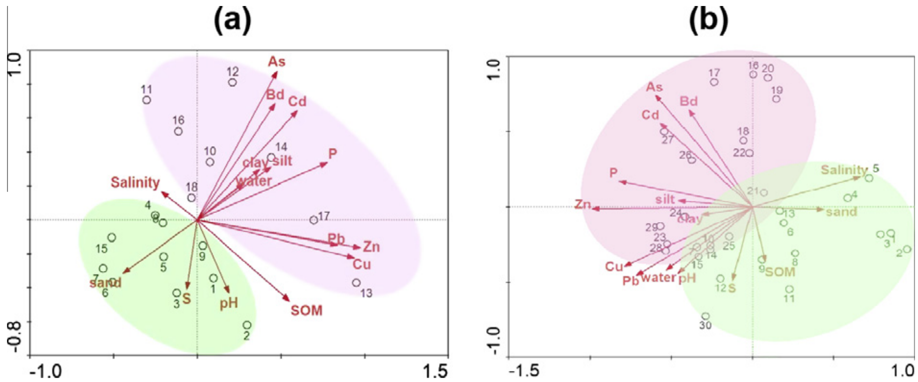

Fig. 2. Ordination plots of the primary component analysis (PCA) of the metal concentrations of tidal freshwater marsh (a) and salt marsh (b) soils. The sampling sites collected before flow-sediment regulation (green area) and the sites sampled after flow-sediment regulation (pink area) are shown. The direction of an arrow indicates the steepest increase in the variable, and the length indicates the strength relative to other variables. BD, bulk density; SOM, soil organic matter; water, water moisture content. Sites 1–9 in (a) and Sites 1–15 in (b) represent sites sampled in April (before regulation), and Sites 10–18 in (a) and Sites 16–30 in (b) represent sites sampled in August (after regulation). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

10, 14 and 15 in April and Sites 21 and 25 in August were grouped into both areas (the transition zone). This result was likely related to the spatial heterogeneity of the marsh microgeomorphology and variation in the environmental conditions (especially salinity changes in the salt marsh) because these factors could affect the distribution of heavy metals (Becker et al., 2006).

From PCA analysis, the strongest determinants of distribution patterns of heavy metals in soil before flow-sediment regulation were SOM, sand content, pH values and S in the freshwater marsh, and the determinants of soil salinity, moisture, pH values and S were most important in the salt marsh. We observed that BD and total P content were the most important factors after regulation. The greater lengths of the five arrows in Fig. 2, representing As, Cd, Cu, Pb and Zn in the August group indicated that As and heavy metals accumulated to a greater extent after regulation.

3.3. Assessment of arsenic and heavy metal pollution

3.3.1. General comparison among river deltas worldwide

The mean concentrations of soil heavy metals in major world river deltas are listed in Table 2. The mean concentrations of Cd, Cu and Zn in the YRD were much lower than those in the Scheldt deltas, which have longer histories of industrial and hydroelectric development (Vandecasteele et al., 2003; Guay et al., 2010). The soil heavy metal concentrations (Cd, Cu, Pb and Zn) found in the YRD in this study were also lower than those found in the Yangtze, Pearl, and Mekong river deltas during the corresponding period. This implied that heavy metal pollution in the YRD is less serious than in deltas of other major rivers (Table 2), probably due to the lower degree of industrialization in the YRD. However, the heavy metal (e.g., Cd, Cu, Pb and Zn) and As concentrations, especially those of As and Cd in the surface soils of the YRD, were close to or even higher than those of deltas where the effects of anthropogenic activities such as industrialization (e.g., the Amazon and Orinoco) or agricultural tillage (e.g., the Mississippi) were not significant. Based on Table 2, As and heavy metal concentrations reached higher levels in 2007 before regulation, indicating that serious accumulation of these elements existed in the soils of the YRD. As and heavy metal pollution increased in this delta in the last decade, probably as the result of intensive human activities and sediment movements resulting from the flow-sediment regulation of Xiaoliangdi Reservoir, the rapid development of petrol oil industries and irrigated agriculture around this delta, which has low levels of urban development (Yu, 2002; Zhou et al., 2004; Nie et al., 2010).

3.3.2. Assessment of arsenic and heavy metal pollution using sediment quality guidelines (SQGs)

The extent of metal pollution was assessed by comparing metal concentrations in surface soils to the sediment quality guidelines (SQGs) developed by China (National Standard of the PR China, 2002), the Ministry of Environment and Energy, Ontario, Canada (MOE, 1993) and the proposed interim sediment quality values (ISQVs) of Hong Kong (Chapman et al., 1998) (Table 2). Based on the Ontario guidelines, all soil samples in tidae firstl freshwater and salt marshes exceeded the lowest effect level (LEL, below which no effects on the majority of sediment-dwelling organisms are expected) for As and Cu, and 30–60% of the soil samples exceeded the LEL for Cd before regulation. After the regulation, all soil samples exceeded the LEL values for As, Cu and Cd, and 26–33% of the soil samples exceeded the LEL for Pb. However, all soil samples in both marshes did not exceed the LEL for Zn before or after regulation. These results demonstrated that adverse toxic impacts to the estuary ecosystem caused by As, Cu, Cd and Pb might be expected. Moreover, it was observed that only 10%-20% of the soil samples exceeded the severe effect levels (SELs), above which seriously adverse effects on the majority of sediment-dwelling organisms are expected) level for As before regulation, and 88–94% of the soil samples had concentrations of As that may cause severe effects on benthic organisms (Table 2). This indicated that wetland soils in this region retained more As and played an important role in water quality purification. With the exception of As, none of the soil samples exceeded the SELs for Cu, Cd, Pb and Zn. Comparison of the data regarding the As and metal levels with the ISQVs of Hong Kong revealed that none of the soil samples in either marshes were polluted by Cd, Cu, Pb or Zn. Despite the fact that As pollution was monitored for all soil samples, the concentration of As was much lower than the ISQV-high value. Based on the Chinese Marine Sediment Quality guidelines (GB 18668-2002) (National Standard of the PR China, 2002), Class I criteria are suitable for fishery and nature uses, and Class II criteria are suitable for industry and tourism locations. None of the soil samples were considered polluted by Zn and Pb before and after the regulation, as their concentrations fell within the scope of Class I, indicating that this region can be considered to contain natural background levels. As and Cd pollution was considered serious because 93–100% and 67–89% of soil samples contained concentrations exceeding the Class I criteria for As and Cd, respectively, before regulation, and As and Cd concentrations in all samples exceeded the Class I criteria after regulation. This region was considered polluted by Cu to some degree because approximately 11–20% of soil samples had

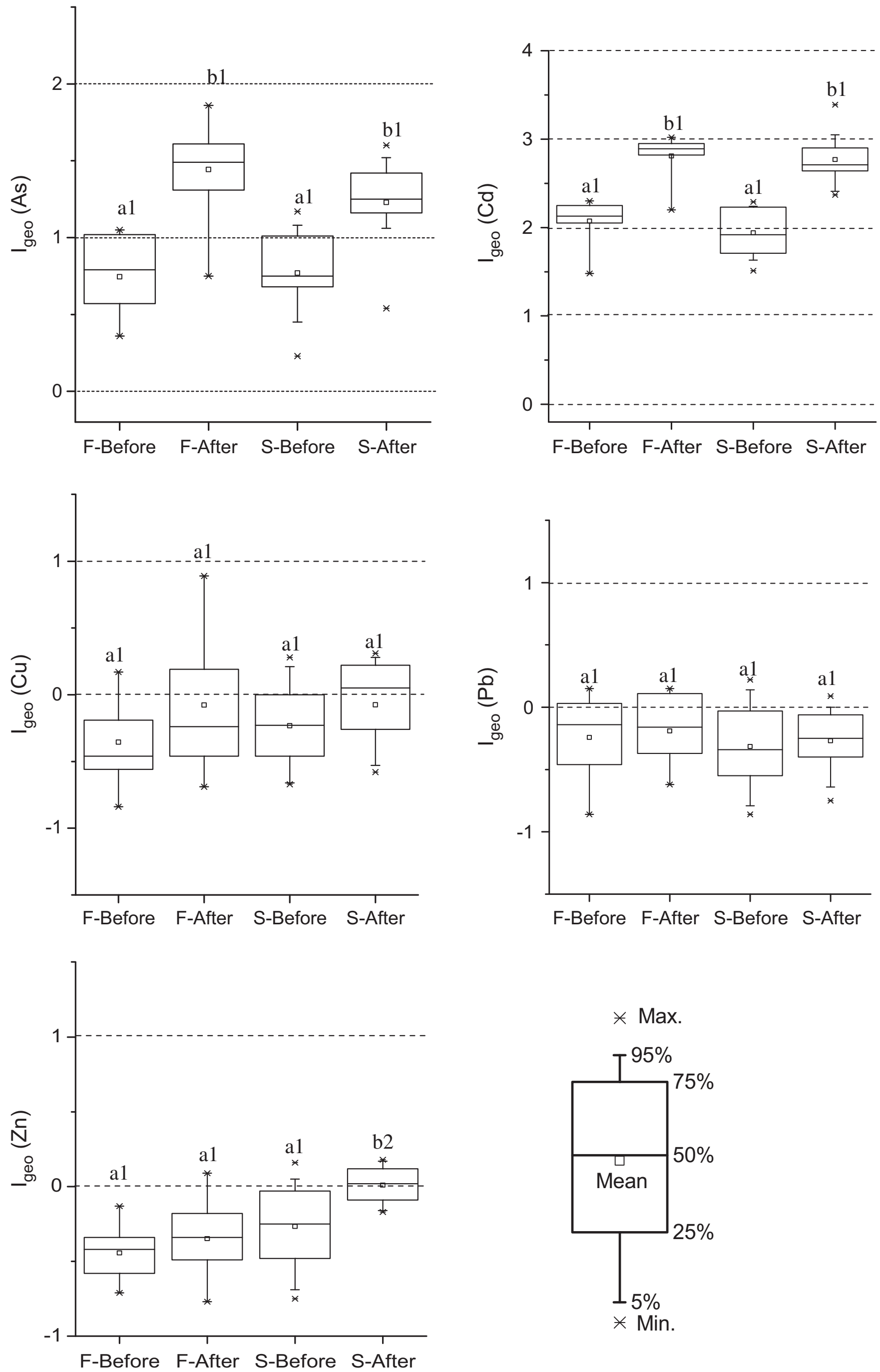

Fig. 3. Box- and- whisker plots of the geoaccumulation indices (I_{geo}) for As and heavy metals in tidal freshwater and salt marshes of the Yellow River Delta before and after flow-sediment regulation. Straight horizontal dotted lines represent the thresholds for unpolluted to moderately polluted ($0 < I_{geo} \leqslant 1$), moderately to strongly polluted ($2 < I_{geo} \leqslant 3$), and strongly polluted ($3 < I_{geo} \leqslant 4$). Different letters above the boxes indicate significant differences ($P < 0.05$) between the I_{geo} values for tidal freshwater marsh or tidal salt marsh before and after flow-sediment regulations. Different numbers above the boxes indicate significant differences ($P < 0.05$) between the I_{geo} values for tidal freshwater and salt marshes before and after regulation.

concentrations exceeding the Class I criteria before regulation, and higher concentrations were observed in 33–40% of soil samples after regulation. However, concentrations of As and the tested heavy metals lay within the scope of Class II criteria. This indicated that the study area was not suitable for fishery and nature uses, but was suitable for use as a tourism site. In fact, this region had been developed as a scenic tourism site in the YRD in recent years.

3.3.3. Assessment of arsenic and heavy metal pollution using I_{geo}

Possible enrichment of heavy metals in wetland soils was evaluated using geoaccumulation indices (I_{geo}) (Muller, 1981). The geoaccumulation indices (I_{geo}) of As and heavy metals in soil are shown in Fig. 3. The I_{geo} values ($0 < I_{geo} \leqslant 1$) for As in both marshes accounted for more than 67% of all I_{geo} values before regulation, whereas approximately 50% -80% of soil samples showed higher I_{geo} values ($1 < I_{geo} \leqslant 2$) after regulation, indicating that As pollution became more serious after the regulation regime. Similarly, Cd pollution was moderate in both marsh soils before regulation ($1.48 \leqslant I_{geo} \leqslant 2.30$) but was measured at moderate to strong levels ($2.20 \leqslant I_{geo} \leqslant 3.39$) after regulation. The mean I_{geo} values for As and Cd in both marsh soils were increased after flow-sediment regulation, in the order $I_{geo(\text{F-after})} > I_{geo(\text{F-before})}$ and $I_{geo(\text{S-after})} > I_{geo(\text{S-before})}$.

However, no significant differences in I_{geo} values were observed for As and Cd between tidal freshwater and tidal salt marshes before or after regulation. Cu, Pb and Zn exhibited much lower I_{geo} values in both marshes, averaging less than 0 before and after regulation, indicating that both marshes were generally not polluted by Cu, Pb and Zn on both sampling dates (except for three sampling sites). Although the mean I_{geo} values for Cu, Pb and Zn in both marshes were elevated after regulation, no significant differences were observed in their I_{geo} values before and after regulation except for the I_{geo} values for Zn in the salt marsh. This result was in agreement with the results of pollution assessments using SQGs (e.g., the SQGs of China and the Ontario guidelines). Therefore, As and Cd pollution could be identified in this region, and the pollution levels in the YRD might be promoted by the flow-sediment regulation of the Xiaolangdi Reservoir (although they might originate from several sources (e.g., river water and atmospheric deposition) in this region).

3.4. Heavy metal sources

Because the factor analysis technique allows a considerable reduction in the number of variables and the detection of the structure of the relationships between metals, it has been considered an effective tool for the identification of heavy metal sources (Han et al., 2006; Mico et al., 2006; Bai et al., 2010a). Factor analysis was performed by evaluating the principal components and computing the eigenvectors of heavy metals in all soil samples. Only the eigenvalues higher than 1 (Kaiser Criterion) and giving a cumulative variance above 85% were retained. The principal components were then rotated using the Varimax normalized method. The results are reported as factor loadings of the rotated matrix in Table 6. Factor loadings that exceed 0.5 have been overstriking.

Only one factor explained the total variance of the total metal concentrations in soils collected before flow-sediment regulation in the YRD (see Table 6). These metals, such as As, Cd, Cu, Pb and Zn, were all associated with Factor 1 (F1). This is an indication of the close relationship between the concentrations of the five metals. It also indicated that all the metals had the same source, which might be directly related to a lithogenic component because the variability of the concentrations appeared to be controlled by loess parent materials. The Yellow River flows over very large loess deposits, which supply 90% of its suspended loads (Gong and Xiong, 1980). The loess materials might contain up to 25 mg kg^{-1} of As, 0.36 mg kg^{-1} of Cd, 55.4 mg kg^{-1} of Cu, 49.0 mg kg^{-1} of Pb, and 221.5 mg kg $^{-1}$ of Zn (CNEMC, 1990) and account for most of the As and heavy metal enrichment observed in both marsh soils in this study.

However, the total variance of the total metal concentrations in marsh soils after regulation was explained by two factors (F1 and F2). The first factor, F1, which explained 46.7% of the total variance, was strongly and positively related to the concentrations of Cu, Pb and Zn. The second factor, F2, which explained 29.8% of the total variance, also showed highly positive factor loadings for As and Cd (Table 6). This implied that the sources of As and Cd might be different than those of Cu, Pb and Zn. Correlation analysis also showed that there were significant correlations between As and Cd, and among Cu, Pb and Zn in both tidal freshwater and salt marshes ($P < 0.01$, Table 5). As and Cd could be defined as exogenous metals because they increased to high levels after flow-sediment regulation. Tang et al. (2010) reported that higher As and Cd concentrations in seawater of the Yellow River Estuary were mainly affected by inputs from the Yellow River. Because of rapid agricultural development upstream of the Yellow River Delta (e.g., in the Yellow River Drainage plain), heavy applications of agrochemicals and fertilizers contributed a large increase in heavy metal concentrations in the YRD (e.g., As and Cd). Moreover, soil P contents were significantly elevated after the regulation and exhibited close correlations with As and Cd, suggesting that As and Cd might originate from upstream agricultural drainage. Cu, Pb and Zn were considered as endogenous metals, although their concentrations were also elevated due to inputs from agricultural sources after the regulation. This result indicated that the sources and composition/structure of heavy metals in surface soils would change with environmental conditions such as water discharge, sediment loads, deposition rates, and grain size (Mico et al., 2006). The flow-sediment regulation regime affected the distribution patterns of heavy metals in the river delta through modifying the hydrodynamic conditions (Zhai et al., 2009). Although Rodriguez et al. (1995) reported that hydrodynamic conditions facilitated the dilution and diffusion of pollutants, the strengthened hydrodynamic condition after regulation in this study increased the pollution loads of soil.

4. Conclusions

This study compared soil heavy metal concentrations before and after flow-sediment regulation the in tidal freshwater and salt marshes of the YRD. The results show that the concentrations of the heavy metals As and Cd in the marsh soils of the YRD are much higher after flow-sediment regulation than before. The strengthened hydrodynamic condition and the change in soil properties may be the major causes for redistribution, deposition and accumulation of heavy metals in the delta. Thus, it is not definite that the increased concentration is directly caused by the flow-sediment regulation. Further studies are required to confirm the changes in the forms of the heavy metals and their accumulation processes before and after the flow-sediment regulation regime.

Table 6
Total variance explained and rotated component matrix (two principal components selected) for heavy metal contents in all soils from tidal freshwater and salt marshes. Factor loadings exceeding 0.5 are expressed in bold font.

Component in the selected sites	Initial eigenvalues			Element	Rotated component matrix	
	Total	% of Variance	Cumulative%		F1	F2
Total variance explained before regulation				*Component matrixes*		
1	3.483	69.667	69.667	As	**0.735**	
2	0.629	12.578	82.245	Cd	**0.749**	
34	0.499	9.983	92.228	Cu	**0.929**	
	0.299	5.979	98.207	Pb	**0.932**	
5	0.090	1.793	100.00	Zn	**0.807**	
Total variance explained after regulation				*Component matrixes*		
1	2.337	46.738	46.738	As	−0.144	**0.864**
2	1.489	29.771	76.509	Cd	0.045	**0.845**
3	0.666	13.319	89.827	Cu	**0.937**	0.014
4	0.2352	7.038	96.886	Pb	**0.882**	0.149
5	0.157	3.134	100.00	Zn	**0.810**	−0.071

Although soil heavy metal pollution is less serious in the YRD than in many other major deltas, an increase of heavy metal concentrations in this region was observed by comparing the concentrations obtained from this research with those obtained during the 1990s. Both As and Cd show moderately or strongly polluted levels in both types of marsh soil after regulation. Additionally, the Yellow River Delta will become a large eco-economic region in China during the coming decade based on the local government development plan. Thus, proactive measures are required to prevent pollution caused by intensive anthropogenic activities from affecting this region.

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