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Potential ecological risk of heavy metal contamination in sediments and
macrobenthos in coastal wetlands induced by freshwater releases: a case
study in the Yellow River Delta, China

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Abstract

We investigated the nine heavy metals contents in the sediments and macrobenthos of the Yellow River Delta Wetlands using three experimental areas that received freshwater releases and one reference area that did not. Heavy metal contents, the single-factor contamination index (*SFCI*), the metal contamination index (*MCI*), and the biota-sediment accumulation factor (*BSAF*) were used to evaluate the potential ecological risk and bioaccumulation. We found that As exceeded the national standard value by more than 50%, and that the ranges of *SFCI* for each metal were generally larger in autumn than in spring. *MCI* showed no clear pattern, but the *BSAF* results suggest that Cd bioaccumulates from sediments to macrobenthos. Pollution-resistant species such as *Corophium sinense*, *Chironomus* sp., and *Einfeldia* sp. became dominant in the areas receiving freshwater releases, and provide direct evidence of ecological risk in the wetlands. Our results provide preliminary information to guide managers for ecological risk assessments.

Key words: Freshwater release, heavy metal contamination, macrobenthos, bioaccumulation, Yellow River Delta Wetlands

1. Introduction

Freshwater release projects have been effective in maintaining and improving the health of ecologically damaged areas (Hou et al., 2007; Orr et al., 2007; Cui et al., 2009). Interventions in these projects have relieved regional water shortages and the shrinking of wetlands (e.g., Colangelo, 2007; Aishan et al., 2013). However, some negative effects have emerged after years of freshwater releases, such as unexpected variations in hydrologic conditions (Hou et al., 2007), unnatural changes in the local biocenosis structure (Casanova and Brock, 2000), and deterioration of water quality

([Hancock and Boulton, 2005](#); [Yang and Yang 2014](#)). Among these problems, heavy metal contamination is a particular concern because of the high toxicity of these substances, their recalcitrance during degradation, and their high enrichment in organisms through bioaccumulation ([Järup, 2003](#); [Feng et al., 2004](#)). Thus, heavy metal contamination is a major threat to the environment.

Organisms in coastal wetlands are highly vulnerable to human activities ([Gordon, 1994](#)). The macrobenthos of wetlands are a particular concern, as they play a crucial role in transferring energy and materials within the food web ([Xie et al., 2010](#)). However, because of this role, they easily transfer heavy metals to consumers at higher trophic levels in the food web, and the resulting bioaccumulation threatens the health of these species. The macrobenthos also strongly influence biogeochemical cycles and are themselves sensitive to heavy metal contamination. Due to their special role in the food web, their restricted habitat, their long life cycle, and their direct contact with sediments, the macrobenthos represent ideal bioindicators of environmental characteristics ([Wardle et al., 1995](#); [Ganesh et al., 2014](#)).

In China's Yellow River Delta wetlands, many scholars have studied the concentration of heavy metals in sediments and have used the method of Hakanson ([1980](#)) to calculate the potential ecological risk created by heavy metal contamination (e.g., [Zhang et al., 2013a](#)). However, those studies neither measured the concentration of heavy metals for macrobenthos or other organisms nor accounted for bioaccumulation of heavy metals in the macrobenthos and in organisms that consume them. Other scholars have paid close attention to the concentrations of heavy metals in the macrobenthos against the background of human activities such as harbor construction ([Chen et al., 2010](#)) and reclamation of irrigation water ([Yang et al., 2011](#)). However, because of the obvious biomagnification effects of Cd, Hg, and Zn moving

through the food web (Cui et al., 2011) and the much higher concentrations of As and Cd after regulation of environmental flows and sediment releases (Bai et al., 2012) in the Yellow River Delta, it has become necessary to pay more attention to the potential ecological risk caused by the heavy metal contamination that can result from freshwater release projects in this area.

In this paper, we used the Yellow River Delta wetlands as a case study to investigate the heavy metal concentrations in the sediments and macrobenthos in both experimental release areas and a reference area before and after freshwater release projects in 2014. We assessed the heavy metal contamination in the macrobenthos using three indices. The results provide insights into the potential ecological risks of heavy metal contamination in the sediments and macrobenthos of these wetlands that are caused by the freshwater releases.

2. Materials and Methods

2.1 Study area

Located on the western coast of the Bohai Sea (Figure 1), the Yellow River Delta Wetlands (37°40'N to 38°10'N, 118°41'E to 119°16'E) are one of China's key national nature reserves. An average of 1.6 Gt of sediment are carried to the sea by the Yellow River and are deposited at the river mouth every year, leading to the formation of new land (Cui et al., 2011; YRCC, 2012). The Yiqianer Management Station is located in the northern part of the wetlands, approximately 4 km from the intertidal zone. This region is in the eastern part of the old Yellow River estuary and lost all of its freshwater inflows after diversion of the Yellow River began in 1976. The region has a temperate, semi-humid, continental monsoon climate, with an average annual temperature of 12.1°C and the highest monthly mean temperature (27.3°C) in July. The annual precipitation averages 552 mm, of which 70% falls during the summer

(May to July). This is much less than the annual mean pan evaporation (1962 mm). To stop continuous erosion by sea waves and restore the coastal wetlands landscape, managers have intervened in several ways, such as the construction of tidal barriers and water diversion canals, as well as the implementation of freshwater releases from the lower reaches of the Yellow River since 2010. Unfortunately, the concentrations of heavy metals in the water released from the Yellow River exceed the limits specified in China's national standard (SEPA, 2002) due to the discharge of domestic sewage and industrial wastewater into the Yellow River (Kaushik et al., 2009; Liu et al., 2013).

Figure 1 approximately here

In this study, we used the area that received freshwater as the experimental area and the adjacent intertidal zone as the reference area. Based on the years when freshwater releases from the Yellow River began to arrive, we divided the experimental area into three sub-areas: I, II, and III. Areas I and II have received freshwater releases since July 2010. They are located on either side of the position that the spring tides originally reached before the construction of tidal barriers. Area III has received freshwater releases since 2012, and is located adjacent to the intertidal zone. Area IV is the relatively undisturbed intertidal area.

2.2 Field sample sites and study design

To determine the magnitude of heavy metal contamination before and after the freshwater releases, which were conducted in July of each year, we sampled the sediments and macrobenthos from areas I to IV during the spring (April to May) and the autumn (September to October) in 2014. The sampling transect runs along the spatial gradient from land (Area I) to sea (Area IV). In total, we used 18 sampling sites (Figure 1). No macrobenthos samples were collected in area III during the spring

because the area had dried out almost completely at this time.

At each sampling site, we used a $0.1 \text{ m}^2 \times 0.3 \text{ m}$ dredge to obtain three macrobenthos samples at three random locations, and mixed them to provide a single bulked sample for the sampling site. In the field, macrobenthos samples were preserved in 300-mL wide-mouth white plastic bottles. At the same time, we collected sediments at three random locations per site by cutting rings (5.0 cm in diameter, 2.54 cm in depth) at a depth from 0 to 0.3 m and stored them in plastic bags. The distance between samples obtained from the three random locations were 3 to 5 m, and the distance between adjacent sampling sites was 500 to 1000 m.

2.3 Laboratory analysis

2.3.1 Macrobenthos samples

To identify and weigh the macrobenthos samples, we passed the samples through 0.5-mm plastic sieves and washed them in the laboratory using water collected near the sample site. We then moved them into a solution of distilled water plus formalin (5% v/v of the formalin; [SOA, 2007](#)) for later analysis and identification. We identified, counted, and weighed all collected organisms to provide the fresh weight of the organisms. We then oven-dried a sample of each organism for 72 h at 80 °C to calculate the dry weight. All the weights were measured using an electronic scale with a precision of 0.001 g (JA1003, Hengping, Shanghai, China).

To determine the concentrations of the target heavy metal elements in the dominant species from areas I to IV, we digested our samples in concentrated HNO_3 and placed them in a hot block digester, initially at an indoor temperature (about 15 °C) for 1 h. We then digested them fully at a high temperature (140 °C) for at least 3 h ([Yap et al. 2004](#)). Next, we used inductively coupled plasma–atomic emission spectrometry (ICP-AES) with a JY-Ultima spectrometer (Horiba Jobin Yvon,

Longjumeau, France) to determine their concentrations. We measured the contents of As, Cd, Cr, Cu, Hg, Mn, Ni, Pb, and Zn in the resulting solution at the wavelengths specified by the manufacturer.

2.3.2 Sediment samples

In the laboratory, we immediately weighed the wet sediment samples, and then air-dried them for 10 days before analysis. We then used a mortar and pestle to grind the sediment sample to a grain size of less than 0.83 mm. Next, we weighed the sediment samples and mixed them with sterile distilled water at a ratio of 1:5 w/w and shook the resulting solution on a rotary shaker (HZQ-F160, Wanhua, Jintan, China) for more than 30 minutes. We then measured the salinity and pH using an HQ 30D portable multi-parameter water quality monitor (Hach, Loveland, Colorado, USA). We then measured the concentrations of the target heavy metals (the same ones measured for the macrobenthos) using ICP-AES. We also weighed 2.000 ± 0.001 g of the air-dried sediments from each sample and put them in a muffle furnace (SG-XL1100, SIOM, Shanghai, China) at 550 °C for 5 hours to calculate the loss of mass on ignition, which represented the total organic carbon content. We measured the sediment grain size distribution by using a laser particle analyzer (HELOS-CUVETTE, Sympatec GmbH, Clausthal-Zellerfeld, Germany); from this data, we calculated the median grain size.

2.4 Statistical analysis

2.4.1 Species dominance

The relative dominance of each species was calculated as follows:

$$Y = (n_j / N) f_j \quad (1)$$

where Y is the relative dominance; n_j is the number of individuals of species j ; N is the total number of macrobenthos individuals; and f_j is the percentage of all sampling sites

where species j was found. When $Y \geq 0.02$, the species is regarded as being one of the dominant species (Shen et al., 2010).

2.4.2 Contamination indices

In this study, we used three indices to represent the degree of heavy metal contamination of the macrobenthos. For the sediments, we will only report the actual concentrations.

We defined the single-factor contamination index (*SFCI*) as the ratio of the heavy metal concentration in the macrobenthos sample to the value defined in the national standard (SAQSIQ, 2001; Yang et al., 2010).

$$SFCI = C_i / S_i \quad (2)$$

where C_i is the concentration of heavy metal i in the macrobenthos sample and S_i is the value in the national standard for living creatures near the seashore (SAQSIQ, 2001).

We defined the metal contamination index (*MCI*) as the geometric mean of all the heavy metal concentrations at the sample site, and used this value to reflect the comprehensive contamination status for all heavy metals and rank the sampling sites (Usero et al., 1997). *MCI* was calculated as follows:

$$MCI = (C_{f1} \times \dots \times C_{fi} \times \dots C_{fn})^{1/n} \quad (3)$$

where C_{fi} is the concentration of the i th heavy metal in the macrobenthos sample (mg/kg dry weight) and n is the number of heavy metals.

We used the biota-sediment accumulation factor (*BSAF*) to quantify bioaccumulation of the target heavy metals by the macrobenthos. It was calculated as follows (Soto-Jiménez et al., 2001):

$$BSAF = C_x / C_s \quad (4)$$

where C_x is the average concentration of a given heavy metal in macrobenthos species

x (mg/kg dry weight) and C_s is the average concentration of the same element in the sediments (mg/kg dry weight).

2.4.3 Statistical analysis

The independent-samples t -test was used to identify statistically significant differences among the sediments and the macrobenthos variables (e.g., the heavy metal concentrations, pH, salinity, water content, total organic carbon content, sediment grain size, and *SFCI*) between spring and autumn samples. These analyses used version 18.0 of the SPSS statistical software (SPSS Inc., Chicago, IL, USA). Differences were considered significant when $p < 0.05$.

We used the non-metric multidimensional scaling (NMDS) method to describe the degree of similarity of the concentrations of the eight heavy metals; Hg was excluded from this analysis because its concentrations were below the detection limit (Clarke and Green, 1988). In the NMDS graph, the degree of similarity of the concentrations of the eight heavy metals between any two sampling sites is measured by the distance between the two points that represent the contamination status at these sites (i.e., shorter distances represent greater similarity). We performed the NMDS analysis using version 5.0 of the Canoco statistical software (<http://www.canoco5.com/>). We also used hierarchical cluster analysis to analyze the degree of similarity among the heavy metal concentrations in the sediments at the 18 sites, using the SPSS software. Comparing the results from the NMDS analysis with those of the cluster analysis provided confirmation of differences among the three experimental area (areas I to III) and between these areas and the reference area (area IV) based on their heavy metal concentrations.

To describe the effect of the heavy metals on the macrobenthos, we also performed redundancy analysis (RDA) using the Canoco software. Based on the

results of the RDA, which shows the correlation between heavy metals in sediments and in the macrobenthos biomass, we can infer the tolerance of typical bio-indicator species for a given metal.

3. Results

3.1 Heavy metal concentrations in the experimental and reference areas

3.1.1 Heavy metal concentrations in the sediments

Figure 2 shows the concentrations of the heavy metals in the sediments along the transect from land (area I) to sea (area IV). All of the As concentrations exceeded the values in the national standard by more than 50%. The concentrations of Mn, Ni, Pb, and Zn in the sediments inside the tidal barriers (areas I to III) were significantly ($p < 0.05$) higher than those in the intertidal area (area IV).

As a result of freshwater releases into areas I to III from Yellow River between 29 June and 6 July 2014, the concentrations of As, Cr, and Ni in the sediments from areas I and II were significantly ($p < 0.05$) higher in the autumn than in the spring, with the increase ranging from 2.1 to 25.1%. In contrast, the concentration of Pb in the sediments from the two areas decreased by 16.6 to 28.9% from the spring to the autumn. Other heavy metals from the three areas that received freshwater releases showed no consistent or significant change from the spring to the autumn. In the reference area that did not receive freshwater releases (area IV), all of the heavy metals except for As were below the value defined in the national standard. Concentrations in area IV in the spring were significantly greater than those in the autumn, except for As, Cr, and Ni ($p < 0.05$).

Figure 2 approximately here

3.1.2 Heavy metal concentrations in the macrobenthos

Tables 1 and 2 present the heavy metal concentrations in the dominant species

collected from the four areas in the spring and the autumn of 2014, respectively. The concentrations of several essential trace elements for living creatures (Cu, Mn, and Zn) in the macrobenthos (Bi et al., 2006) were much higher than those of other metals (such as As, Cd, Cr, Ni, Pb, and Hg), except for Mn in area I.

Table 1 approximately here

Table 2 approximately here

From Tables 1 and 2, we can see that the dominant species composition in a given area differed before and after the freshwater releases, and that the numbers of dominant species in all four areas in the autumn increased compared to that in the spring. In areas I and II, *Chironomus* sp. and *Cladotanytarsus mancus* were the dominant species in the spring and the autumn, respectively, and most of the heavy metal values (except for As in *Chironomus* sp. and for Cd and Hg in *Cladotanytarsus mancus*) were higher in area II than in area I. In the autumn, As, Cu, Mn, Pb, and Zn levels in *Corophium sinense* from area III were obviously lower than those in area IV; in contrast, levels of As, Cu, Mn, and Pb in *Notomastus latericeus* from area III were higher than those of samples from area IV. In *Corophium sinense* from area II, all of the heavy metal concentrations except for As and Zn were higher in the spring than in the autumn. For *Macrophthalmus japonicus* from area IV, all of the heavy metal concentrations except Cu, Mn, and Pb were higher in the autumn than in the spring.

3.1.3 Heavy metal concentrations in the released freshwater

The concentrations of heavy metals in the released freshwater strongly affect the concentrations of heavy metals in sediment and macrobenthos, since this water is the dominant source of inputs of these metals. In addition, the cumulative effect of many years of freshwater releases cannot be ignored when determining the ecological risk of heavy metal contamination in the study area. Figure 3 provides data on the

concentrations of heavy metals in the water diverted from downstream reaches of the Yellow River between 2010 and 2014; wide variation in the heavy metal concentrations are clearly visible, especially for Cr, Cu, and Pb. The magnitude of these fluctuations suggest that it will be complex to predict the cumulative inputs of heavy metals from the Yellow River; complex monitoring will be required to calculate the cumulative inputs and what proportion of each heavy metal is immobilized in the sediments rather than becoming available for uptake by the macrobenthos.

Figure 3 approximately here

3.2 Potential ecological risk of heavy metal contamination

3.2.1 *SFCI* values for the heavy metals

Tables 3 and 4 present the *SFCI* values of the heavy metals in the dominant macrobenthos species in the spring and the autumn, respectively. For all species combined, *SFCI* in the spring ranged from 0 to 0.69 for As, 0 to 11.35 for Cd, 0 to 45.2 for Cr, 1.5 to 5.42 for Cu, 0 to 12.7 for Pb, 0.08 to 1.14 for Zn, and 0 to 27.6 for Hg. In the autumn, the values ranged from 0 to 3.54 for As, 0 to 35.8 for Cd, 0 to 51.8 for Cr, 0 to 5.79 for Cu, 0 to 14.8 for Pb, 0.65 to 4.93 for Zn, and 0 to 4.02 for Hg. The *SFCI* ranges for each metal in the autumn were thus generally larger than those in the spring. However, there was no significant difference between the spring and autumn *SFCI* values in the study area ($p > 0.05$; independent-samples *t*-test).

Table 3 approximately here

Table 4 approximately here

The dominant species with *SFCI* > 1.0 are suffering from contamination by a given heavy metal. From Tables 3 and 4, *SFCI* > 1.0 mainly occurred for As (in the autumn), Cd, Cr, Cu, Pb, and Zn. Based on the average *SFCI* values for all dominant species, the order of severity for heavy metal contamination during the spring was Cr >

Cd > Pb > Cu while the order during the autumn was Cr > Pb > Hg > Cd > Cu.

In the reference area, there were nine dominant species during the spring and the autumn, and for these species, 52 of the 77 *SFCI* values were greater than 1.0. In contrast, 63 of the 112 *SFCI* values in areas I, II, and III, where we found a total of 10 dominant species, had *SFCI* > 1.0. However, there were also some extremely high values, with *SFCI* > 30. In Tables 3 and 4, five *SFCI* values exceeded 30, with two of these values in area IV and the other three in areas I to III. This suggests that some species such as *Chironomus* sp. that are capable of tolerating high levels of certain heavy metals became dominant species in the experimental areas.

The higher *SFCI* values for Cr and Cd in polychaetes such as *Neanthes succinea* and *Nereis* sp. and molluscs such as *Bivalvia* and *Philine* sp. in area IV reveal that these species bioaccumulate Cr and Cd. Similarly, the high *SFCI* values for Cr, Cd, and Hg in some insect species (such as *Einfeldia* sp. and *Chironomus* sp.) in areas I and II shows these species' ability to selectively absorb and tolerate these toxins.

3.2.2 *MCI* values in the sample areas

Figure 4 presents the *MCI* values in the experimental areas (I to III) and the reference area (IV). Figure 4a shows that *MCI* was significantly higher in area II than in the other areas during the spring, indicating worse heavy metal contamination than in the other two areas. Figure 4b shows no significant differences in *MCI* among the areas. The *MCI* value in area II during the spring was 48% greater than the autumn values in any area that received freshwater releases. In contrast, the *MCI* value in area I during the spring was 52% less than the autumn values in any area that received freshwater releases.

Figure 4 approximately here

Because the values obtained in the spring were obtained about 10 months after

the previous freshwater release, we believe that the effect of the freshwater release on heavy metal levels weakened greatly during this period. Thus, it is reasonable to believe that it is the artificial management actions (i.e., the freshwater releases) that led to the difference of heavy metal contaminations between adjacent areas.

We also found that several species could live in areas with different hydrological conditions, such as *Corophium sinense*, *Chironomus* sp., and *Cladotanytarsus mancus*. Figure 5 compares the *MCI* values in a given species that was present in at least two of the four areas. Under different hydrological conditions, the values of *MCI* showed large ranges for all the three species. This suggests that in addition to inherent differences in the ability of a species to take up or exclude certain heavy metals, the hydrological conditions can also affect the availability and uptake of the metals.

Figure 5 approximately here

3.2.3 *BSAF* values for heavy metals in macrobenthos

Tables 5 and 6 present the *BSAF* values for the different species in the three experimental areas and the reference area in the spring and autumn, respectively. The order of bioaccumulation of heavy metals was greatest for Cd, followed by Cu, in both seasons. The higher value of *BSAF* for Cd in some species (such as *Bivalvia*, *Neanthes succinea*, *Macrophthalmus japonicus*, and *Einfeldia* sp.) showed their bioaccumulation of and tolerance for Cd.

Table 5 approximately here

Table 6 approximately here

3.3 Relationship between heavy metals and the macrobenthos communities

3.3.1 Macrobenthos communities in the experimental and reference areas

Figures 6 and 7 shows the differences in the macrobenthos community among the experimental areas (I to III) and the reference area (IV). The structure and

composition of the macrobenthos community in the reference area tended to be more diverse and complicated than those in the experimental areas. The macrobenthos community was dominated by molluscs, crustaceans, and insects in areas I to III, whereas the main members of the macrobenthos communities were Nemertea sp., polychaetes, molluscs, crustaceans, and insects in area IV.

Figure 6 approximately here

Figure 7 approximately here

Table 7 presents population data for the dominant species in the four areas. As a result of the freshwater released into area III, crustacean, insect, and polychaete macrobenthos species could survive until the autumn; in contrast, we found no macrobenthos in this area in the spring because drought eliminated the water in this area. The dominant species in the experimental areas tended to be more diverse after freshwater releases.

Table 7 approximately here

3.3.2 Differences in heavy metal contamination among the four areas

Figure 8 illustrates the results of the NMDS analysis. The geometric distance between two points is proportional to the difference in heavy metal contamination. Both in the spring and the autumn, the NMDS divided the sampling sites into two clear groups: one group mainly comprised sites in area IV, and the other group mostly comprised sites in the experimental areas (I to III). The dendrogram produced using hierarchical cluster analysis confirmed this division (Fig. 9). The results therefore revealed significant differences between the experimental areas (I to III) and the reference area (IV) in both seasons.

Figure 8 approximately here

Figure 9 approximately here

3.3.3 The relationships between the heavy metals and the macrobenthos

Figure 10 shows the RDA results for the relationships between the heavy metal concentrations and the macrobenthos groups. Insects were significantly positively correlated with the concentrations of all heavy metals in the spring and the autumn. The strong association of insects with the heavy metals provides evidence of their pollution resistance.

Figure 10 approximately here

There was a negative correlation between both molluscs and Nemertea sp. and the concentrations of various heavy metals in both seasons, which suggests relatively weak tolerance for heavy metals. In contrast, the relationships between the other macrobenthos and heavy metal values showed seasonal differences; for example, polychaetes were strongly negatively correlated with metal concentrations in the spring, but weakly positively correlated with these concentrations in the autumn. There were no obvious correlations between crustaceans and heavy metal concentrations in either season.

4. Discussion

Freshwater release projects have relieved drought, restored vegetation, and promoted biodiversity in the wetlands of the Yellow River Delta (Cui et al., 2009). However, the low quality of the water source used for these releases (the Yellow River) is creating some potential ecological risks for the wetlands (Tang et al., 2010). We found that the order of the heavy metal concentrations in the sediments and macrobenthos was $Mn > Zn > Cr > As > Ni > Cu > Pb > Cd$, which is consistent with the relative values in the water of the downstream reaches of the Yellow River (Zhang et al., 2013c). Based on the *MCI* and *BSAF* values, the concentrations of Cd were greater than those of Cu, and both were much larger than the values for the other

metals. [Zhang et al. \(2013d\)](#) surveyed the heavy metal concentrations in an irrigation area in which the irrigation water was pumped from the downstream reaches of the Yellow River, and found that the Mn and Zn values exceeded the limits in the national standards. [Cui et al. \(2011\)](#) reported that Cd, Zn, and Hg increased with increasing trophic level (i.e., they bioaccumulated).

We found five *SFCI* values greater than 30, with two of these values in area IV (the reference area) and the other three in areas I to III (which received water releases); these values suggest a strong ecological risk of heavy metal contamination. Three high *SFCI* values that appeared in areas I and II suggested high tolerance of the dominant species in these areas for Cr and Cd. The fact that these resistant species became dominant species suggests a potential ecological risk caused by heavy metal contamination in areas that received freshwater releases. Especially, the *SFCI* value of one new dominant species, *Einfeldia* sp., in area I in the autumn is 35.80 for Cd, showing a contamination risk for heavy metals caused by freshwater releases carried out during the summer. In area IV, the high value of Cr for *Neanthes succinea* appears to be associated with the ability of this species to absorb and tolerate Cr. According to data from China and elsewhere ([Mouneyrac et al, 2003](#); [Zhou et al, 2003](#)), *Neanthes succinea* can survive relatively high heavy metal concentrations. The concentration of Cr in water of the Bohai Sea ranges from 38.2 to 79.8 mg/kg, which is close to the national standard ([Feng et al., 2011](#), [Hu et al., 2013](#)). This input of Cr to Area IV with each tide would be supplemented by the input of up to 2.5 mg/L in the river water (Fig. 3), and this might push Cr above the national standard. Therefore, the relatively high Cr concentration shows *Neanthes succinea*'s selective absorption of this metal. We could not find comparable evidence for Cr in Class Bivalvia because we could not identify these animals to the species level in the present study. However, our literature

search suggested that some species in Class Bivalvia (such as *Bullacta exarata*) show evidence of selective absorption of some heavy metals (Bi et al., 2006; Chen et al., 2010).

We also found that some pollution-resistant species (such as *Chironomus* sp. and *Einfeldia* sp.) in the areas that received freshwater releases became dominant species, and the differences in the species compositions among the four areas provide additional evidence for environmental changes in the wetlands and their consequences. Therefore, it will be necessary for water managers to pay careful attention to both heavy metal pollution and changes in hydrological conditions in the downstream reaches of the Yellow River.

Compared with some other large rivers (Table 8), such as the Mississippi River in the United States (Santschi et al., 2001; Balogh et al., 2009) and the Amazon River in South America (Marchand et al., 2006), the concentrations of some heavy metals in the downstream reaches of the Yellow River are relatively low. In the reference area, the heavy metal concentrations in the autumn (after the freshwater releases) were generally slightly higher than those in the spring (before the freshwater releases), which is consistent with the results in the experimental areas. Thus, we cannot exclude the possibility of a seasonal effect on the heavy metal concentrations based only on the available data from the present study.

Table 8 approximately here

Other environmental factors in addition to the freshwater releases may influence the concentrations of heavy metals in sediments, such as pH, salinity, water content, sediment grain size, and organic matter. To identify such factors, we looked for significant differences between spring and autumn in these five environmental factors in the sediment samples. Supplemental Table S1 shows no significant differences

except for the water content in areas III and IV and the salinity of the sediments in area III. In addition, precipitation from spring to autumn in the study area seems unlikely to have had a major effect on the differences among the areas; precipitation totaled about $0.30 \times 10^7 \text{ m}^3$, which was less than 10% of the $3.17 \times 10^7 \text{ m}^3$ of freshwater that was released in the summer (SBSP, 2014). Thus, the freshwater releases are likely to have a greater effect than precipitation on the concentrations of heavy metals in the study area.

The degree of tolerance of environmental pollution varies among species (Warwick, 1986; Martinez-Garcia et al., 2013). Therefore, sensitive species decrease in abundance when exposed to high levels of pollution, whereas tolerant species may benefit from or not be affected by the polluted conditions. In the present study, the abundance of Crustacea decreased from 96.2% of the specimens in the spring to 70.0% in the autumn in experimental area II, but increased from 10.1% in the spring to 95.6% in the autumn in reference area IV. Tolerant organisms such as polychaetes and insects increased from 0.3% to 4.0% and from 3.5% to 26.0%, respectively, from spring to autumn in experimental area II. Polychaete assemblages are considered good indicators of environmental perturbations (Del-Pilar-Ruso et al., 2010). The presence or absence of the polychaetes in marine sediments provides an excellent indicator of the condition or health of the benthic environment (Martinez-Garcia et al., 2013). Among the insects, the pollution-resistant species *Chironomus* sp. and *Cladotanytarsus mancus* were the dominant species in experimental areas I and II, which provides further evidence of pollution of these areas (Boron and Mirosławski, 2009).

In future research, we will continue to survey the heavy metal concentrations in the freshwater used for the environmental releases, in the sediments, and in the

macrobenthos of our study area with the goal of clarifying the factors responsible for the observed differences. We will also perform some laboratory experiments to clarify whether some key species exhibit selective absorption of one or more heavy metals (such as Cr and Cd) for the five species that showed extremely high *SFCI* values for these metals. We will also study trophic relationships within the macrobenthos community. The results of this analysis will provide scientific support to improve future management of the Yellow River Delta wetlands.

5. Conclusions

We surveyed the concentrations of nine heavy metals in the sediments and macrobenthos of areas in the Yellow River Delta wetlands that received freshwater environmental releases. We found that one of the heavy metals (As) exceeded the value in the national standard by more than 50% and that concentrations of many other metals were close to that limit. The ranges of *SFCI* values for each metal were mostly larger in the autumn than in the spring, with values in the following order: Cr > Pb > Hg > Cd > Cu. *MCI* values showed no clear pattern, but the *BSAF* results suggest bioaccumulation of Cd from the sediments to the macrobenthos in the study area. The pollution-resistant species, including *Corophium sinense*, *Chironomus* sp., and *Einfeldia* sp., became dominant species in the areas that received the freshwater releases, which provides direct evidence of an ecological risk in the wetlands. Further investigations will be required to provide a sound basis for eliminating the natural seasonal variation in heavy metal concentrations in the sediments and macrobenthos of the study area.

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Figure captions

Figure 1. Map of the region around the Yiqianer Management Station in the northern part of the Yellow River Delta. (Experimental areas are I, II, and III; the reference area is IV.)

Figure 2. Variations in the heavy metal concentration in sediments collected along a transect from land (area I) to sea (area IV). The dashed horizontal line represents the concentration limit defined in the national standard. Values are means $\pm$ SD. The concentrations of Hg are not shown because they were below the detection limit.

Figure 3. Concentrations of heavy metals in water from downstream reaches of the Yellow River diverted into the study area during the past five years (2010 to 2014). Values are means $\pm$ SD.

Figure 4. Values of the metal contamination index (*MCI*) in the experimental areas (I to III) and the reference area (IV) during (a) the spring and (b) the autumn. Values are mean $\pm$ SD. Bars labeled with different Greek letters differed significantly in a given season. (No macrobenthos samples were collected in the spring in area III, which dried out completely during this period.)

Figure 5. Values of the metal contamination index (*MCI*) for the species that could live in areas with different hydrological conditions.

Figure 6. The numbers of species in each of the four parts of study area for the main macrobenthos taxa found in the community.

Figure 7. The macrobenthos community structures in the experimental areas (I to III) and the reference area (IV) in (a) the spring and (b) the autumn. (No macrobenthos samples were collected in the spring in area III, which had dried out completely at that time.)

Figure 8. NMDS ordination for heavy metal contamination in the sediments of the

681 experimental areas (I to III) and the reference area (IV) in (a) the spring and (b) the
682 autumn in 2014. The locations of the sample sites in each area are shown in Figure 1.
683 Figure 9. Hierarchical cluster analysis for heavy metal contamination in the sediments
684 of the experimental areas (I to III) and the reference area (IV) (a) in spring and (b) in
685 autumn of 2014. The locations of the sample sites in each area are shown in Figure 1.
686 Figure 10. RDA for the relationships between heavy metal concentrations and the
687 main macrobenthos taxonomic groups in (a) the spring and (b) the autumn of 2014.

Table captions

Table 1. Heavy metal concentrations in of the dominant macrobenthos species in the experimental areas (I to III) and the reference area (IV) in the spring of 2014. (No macrobenthos sample were collected in the spring in area III, which had dried out completely at that time.).

Table 2. Heavy metal concentrations in the dominant macrobenthos species in the experimental areas (I to III) and the reference area (IV) in the autumn of 2014.

Table 3. Values of the single-factor contamination index (*SFCI*) for the dominant species in the experimental areas (I to III) and the reference area (IV) in the spring of 2014. (No macrobenthos sample were collected in the spring in area III, which had dried out completely at that time.)

Table 4. Values of the single-factor contamination index (*SFCI*) for the dominant species in the experimental areas (I to III) and the reference area (IV) in the autumn of 2014.

Table 5. The biota-sediment accumulation factor (*BSAF*) values for the heavy metals in the macrobenthos in the experimental areas (I to III) and the reference area (IV) during the spring of 2014. (No macrobenthos samples were collected in the spring in area III, which had dried out completely at that time.)

Table 6. The biota-sediment accumulation factor (*BSAF*) values for the heavy metals in the macrobenthos in the experimental areas (I to III) and the reference area (IV) during the autumn of 2014.

Table 7. Dominant species from areas I to IV in the spring and the autumn of 2014. *Y* is the relative dominance of each species. (No macrobenthos samples were collected in the spring in area III, which had dried out completely at that time.)

Table 8. A comparison between the heavy metal concentrations in our study area and

713 published results from other big rivers around the world.

Table 1. Heavy metal concentrations in of the dominant macrobenthos species in the experimental areas (I to III) and the reference area (IV) in the spring of 2014. (No macrobenthos sample were collected in the spring in area III, which had dried out completely at that time.).

Area	Species	Concentration (mg/kg)								
		As	Cd	Cr	Cu	Mn	Ni	Pb	Zn	Hg
I	<i>Chironomus</i> sp.	0.24	0.03	12.90	23.70	ND	1.27	ND	7.44	ND
	<i>Dicrotendipes tritomus</i>	0.00	ND	ND	24.50	ND	ND	ND	10.00	ND
II	<i>Corophium sinense</i>	0.62	0.74	22.60	27.90	77.60	22.20	0.64	22.70	0.27
	<i>Chironomus</i> sp.	0.19	0.39	21.10	42.20	197.00	21.30	0.88	21.20	1.38
IV	<i>Neanthes succinea</i>	0.69	0.13	8.28	35.80	74.10	7.83	0.97	8.32	0.05
	Class Bivalvia	0.25	2.27	19.60	17.20	146.00	19.30	1.27	19.40	0.12
	<i>Moerella jedoensis</i>	0.15	0.28	2.84	15.00	12.80	1.43	0.63	1.52	ND
	<i>Philine</i> sp.	0.20	1.11	5.56	54.20	51.20	9.98	0.42	5.56	0.07
	<i>Macrophthalmus japonicus</i>	0.12	0.57	3.43	33.50	27.70	3.35	0.62	3.40	ND

Note: ND means that the concentration was below the limit of detection.

Table 2. Heavy metal concentrations in the dominant macrobenthos species in the experimental areas (I to III) and the reference area (IV) in the autumn of 2014.

Area	Species	Concentration (mg/kg)								
		As	Cd	Cr	Cu	Mn	Ni	Pb	Zn	Hg
I	<i>Einfeldia</i> sp.	0.41	7.16	3.14	33.80	34.20	ND	0.60	14.30	ND
	<i>Cladotanytarsus mancus</i>	ND	2.03	1.85	13.60	15.80	ND	ND	20.90	0.10
	<i>Polypedilum</i> sp.	ND	1.72	ND	22.50	ND	2.35	0.26	15.20	0.09
II	<i>Corophium sinense</i>	0.80	0.10	1.89	23.40	71.60	3.65	0.51	33.80	ND
	<i>Polypedilum</i> sp.	1.91	ND	3.28	12.70	40.30	1.89	0.72	63.10	ND
	<i>Cladotanytarsus mancus</i>	1.53	0.10	4.52	15.30	24.50	1.48	ND	19.40	0.10
	Family Ceratopogonidae	0.42	0.13	5.61	17.10	18.60	10.30	0.37	26.60	0.10
III	<i>Dicrotendipes tritonus</i>	ND	0.55	0.17	15.70	ND	1.90	0.41	16.50	ND
	<i>Corophium sinense</i>	0.01	0.13	4.62	9.53	24.90	9.04	ND	23.60	ND
	<i>Cladotanytarsus</i> sp.	0.71	0.65	2.79	21.90	14.60	2.37	ND	48.30	ND

	<i>Sigara substriata</i>	ND	0.26	1.85	37.10	26.10	2.01	0.71	18.10	ND
	<i>Notomastus latericeus</i>	3.54	ND	3.17	39.00	51.20	1.58	0.90	15.70	ND
IV	Nemertea sp.	1.44	0.12	0.73	43.20	70.90	4.25	0.29	13.00	ND
	<i>Corophium sinense</i>	1.04	ND	1.08	23.40	51.20	5.53	0.37	98.60	ND
	<i>Nereis</i> sp.	3.01	0.81	10.20	16.60	19.10	7.10	1.48	62.70	ND
	<i>Neanthes succinea</i>	1.12	1.76	25.90	29.50	36.80	1.08	0.66	88.80	0.20
	<i>Notomastus latericeus</i>	1.33	0.98	8.35	ND	ND	6.39	ND	30.00	ND
	<i>Macrophthalmus japonicus</i>	0.17	2.24	4.44	57.90	58.90	10.20	1.29	38.50	0.02

Note: ND means that the concentration was below the limit of detection.

Table 3. Values of the single-factor contamination index (*SFCI*) for the dominant species in the experimental areas (I to III) and the reference area (IV) in the spring of 2014. (No macrobenthos sample were collected in the spring in area III, which had dried out completely at that time.)

Area	Species	<i>SFCI</i>						
		As	Cd	Cr	Cu	Pb	Zn	Hg
I	<i>Chironomus</i> sp.	0.24	0.15	25.80	2.37	0.00	0.37	0.00
	<i>Dicrotendipes tritomus</i>	0.00	0.00	0.00	2.45	0.00	0.50	0.00
II	<i>Corophium sinense</i>	0.62	3.72	45.20	2.79	6.40	1.14	5.36
	<i>Chironomus</i> sp.	0.19	1.94	42.20	4.22	8.76	1.06	27.60
IV	<i>Neanthes succinea</i>	0.69	0.67	16.56	3.58	9.72	0.42	0.90
	Class Bivalvia	0.25	11.35	39.20	1.72	12.70	0.97	2.34
	<i>Moerella jodoensis</i>	0.15	1.42	5.68	1.50	6.32	0.08	0.00
	<i>Philine</i> sp.	0.20	5.55	11.12	5.42	4.16	0.28	1.34
	<i>Macrophthalmus japonicus</i>	0.12	2.83	6.86	3.35	6.24	0.17	0.00
	Mean $\pm$ SD	0.97 $\pm$ 0.72	5.21 $\pm$ 4.00	9.29 $\pm$ 0.73	2.40 $\pm$ 1.04	4.76 $\pm$ 0.20	1.80 $\pm$ 0.06	0.68 $\pm$ 0.87

Table 4. Values of the single-factor contamination index (*SFCI*) for the dominant species in the experimental areas (I to III) and the reference area (IV) in the autumn of 2014.

Area	Species	<i>SFCI</i>						
		As	Cd	Cr	Cu	Pb	Zn	Hg
I	<i>Einfeldia</i> sp.	0.41	35.80	6.28	3.38	6.03	0.72	0.00
	<i>Cladotanytarsus mancus</i>	0.00	10.15	3.70	1.36	0.00	1.05	2.06
	<i>Polypedilum</i> sp.	0.00	8.60	0.00	2.25	2.55	0.76	1.74
II	<i>Corophium sinense</i>	0.80	0.52	3.78	2.34	5.12	1.69	0.00
	<i>Polypedilum</i> sp.	1.91	0.00	6.56	1.27	7.16	3.16	0.00
	<i>Cladotanytarsus mancus</i>	1.53	0.49	9.04	1.53	0.00	0.97	1.92
	Family Ceratopogonidae	0.42	0.64	11.22	1.71	3.68	1.33	2.02
III	<i>Dicrotendipes tritomus</i>	0.00	2.73	0.33	1.57	4.06	0.83	0.00
	<i>Corophium sinense</i>	0.01	0.67	9.24	0.95	0.00	1.18	0.00
	<i>Cladotanytarsus</i> sp.	0.71	3.24	5.58	2.19	0.00	2.42	0.00

	<i>Sigara substriata</i>	0.00	1.31	3.70	3.71	7.12	0.91	0.00
	<i>Notomastus latericeus</i>	3.54	0.00	6.34	3.90	9.03	0.79	0.00
IV	Nemertea sp.	1.44	0.60	1.45	4.32	2.94	0.65	0.00
	<i>Corophium sinense</i>	1.04	0.00	2.16	2.34	3.65	4.93	0.00
	<i>Nereis</i> sp.	3.01	4.07	20.40	1.66	14.80	3.14	0.00
	<i>Neanthes succinea</i>	1.12	8.80	51.80	2.95	6.62	4.44	4.02
	<i>Notomastus latericeus</i>	1.33	4.92	16.70	0.00	0.00	1.50	0.00
	<i>Macrophthalmus japonicus</i>	0.17	11.20	8.88	5.79	12.90	1.93	0.42
	Mean $\pm$ SD	0.27 $\pm$ 0.22	3.07 $\pm$ 3.38	21.40 $\pm$ 16.26	3.04 $\pm$ 1.17	6.03 $\pm$ 3.97	0.55 $\pm$ 0.38	4.17 $\pm$ 8.45

Table 5. The biota-sediment accumulation factor (*BSAF*) values for the heavy metals in the macrobenthos in the experimental areas (I to III) and the reference area (IV) during the spring of 2014. (No macrobenthos samples were collected in the spring in area III, which had dried out completely at that time.)

Area	Species	<i>BSAF</i>								
		As	Cd	Cr	Cu	Mn	Ni	Pb	Zn	Hg
I	<i>Chironomus</i> sp.	0.02	0.12	0.21	0.84	0.00	0.04	0.00	0.10	0.00
	<i>Dicrotendipes</i>	0.00	0.00	0.00	0.87	0.00	0.00	0.00	0.13	0.00
	<i>tritonus</i>									
II	<i>Corophium</i>	0.04	2.91	0.36	0.99	0.11	0.69	0.02	0.30	0.00
	<i>sinense</i>									
	<i>Chironomus</i> sp.	0.01	1.51	0.33	1.50	0.27	0.66	0.03	0.28	0.00
IV	<i>Neanthes</i>	0.06	0.75	0.16	1.79	0.13	0.31	0.05	0.14	0.00
	<i>succinea</i>									
	Class Bivalvia	0.02	12.86	0.38	0.86	0.25	0.77	0.07	0.33	0.00

<i>Moerella</i>	0.01	1.61	0.06	0.75	0.02	0.06	0.03	0.03	0.00
<i>jedoensis</i>									
<i>Philine</i> sp.	0.02	6.29	0.11	2.70	0.09	0.40	0.02	0.10	0.00
<i>Macrophthalmus</i>									
<i>japonicus</i>	0.01	3.20	0.07	1.67	0.05	0.13	0.03	0.06	0.00
Mean ± SD	0.02±0.02	3.25±3.86	0.19±0.13	1.33±0.61	0.10±0.09	0.34±0.29	0.03±0.02	0.16±0.11	0.00±0.00

Table 6. The biota-sediment accumulation factor (*BSAF*) values for the heavy metals in the macrobenthos in the experimental areas (I to III) and the reference area (IV) during the autumn of 2014.

Area	Species	<i>BSAF</i>								
		As	Cd	Cr	Cu	Mn	Ni	Pb	Zn	Hg
I	<i>Einfeldia</i> sp.	0.03	42.62	0.05	1.21	0.05	0.00	0.03	0.19	0.00
	<i>Cladotanytarsus mancus</i>	0.00	12.08	0.03	0.49	0.02	0.00	0.00	0.28	0.00
	<i>Polypedilum</i> sp.	0.00	10.24	0.00	0.81	0.00	0.07	0.01	0.20	0.00
II	<i>Corophium sinense</i>	0.04	0.50	0.02	0.78	0.10	0.10	0.02	0.42	0.00
	<i>Polypedilum</i> sp.	0.10	0.00	0.04	0.42	0.06	0.05	0.03	0.78	0.00
	<i>Cladotanytarsus mancus</i>	0.08	0.47	0.06	0.51	0.03	0.04	0.00	0.24	0.00
	Family Ceratopogonidae	0.02	0.61	0.07	0.57	0.03	0.28	0.02	0.33	0.00
III	<i>Dicrotendipes tritonus</i>	0.00	3.88	0.00	0.68	0.00	0.06	0.03	0.25	0.00
	<i>Corophium sinense</i>	0.00	0.95	0.07	0.41	0.04	0.30	0.00	0.36	0.00
	<i>Cladotanytarsus</i> sp.	0.05	4.60	0.04	0.94	0.02	0.08	0.00	0.74	0.00

IV	<i>Sigara substriata</i>	0.00	1.86	0.03	1.60	0.04	0.07	0.04	0.28	0.00
	<i>Notomastus latericeus</i>	0.26	0.00	0.05	1.68	0.09	0.05	0.06	0.24	0.00
	Nemertea sp.	0.11	1.26	0.01	2.27	0.13	0.16	0.02	0.24	0.00
	<i>Corophium sinense</i>	0.08	0.00	0.02	1.23	0.09	0.21	0.03	1.79	0.00
	<i>Nereis</i> sp.	0.24	8.61	0.19	0.87	0.03	0.27	0.10	1.14	0.00
	<i>Neanthes succinea</i>	0.09	18.61	0.48	1.55	0.07	0.04	0.05	1.61	0.00
	<i>Notomastus latericeus</i>	0.10	10.39	0.15	0.00	0.00	0.24	0.00	0.55	0.00
	<i>Macrophthalmus</i>	0.01	23.68	0.08	3.05	0.10	0.38	0.09	0.70	0.00
	<i>japonicus</i>									
	Mean±SD	0.07±0.0	7.80±10.8	0.08±0.1	1.06±0.7	0.05±0.0	0.13±0.1	0.03±0.0	0.57±0.4	0.00±0.0
		7	1	1	3	4	1	3	7	0

Table 7. Dominant species from areas I to IV in the spring and the autumn of 2014. *Y* is the relative dominance of each species. (No macrobenthos samples were collected in the spring in area III, which had dried out completely at that time.)

Area	Season	Species name	Taxon	Number of individuals	<i>Y</i>
I	Spring	<i>Chironomus</i> sp.	Insecta	215	0.72
		<i>Dicrotendipes tritonus</i>	Insecta	22	0.17
	Autumn	<i>Cladotanytarsus mancus</i>	Insecta	18	0.17
		<i>Einfeldia</i> sp.	Insecta	14	0.27
		<i>Polypedilum</i> sp.	Insecta	7	0.07
II	Spring	<i>Corophium sinense</i>	Crustacea	1980	0.93
		<i>Chironomus</i> sp.	Insecta	215	0.06
	Autumn	<i>Corophium sinense</i>	Crustacea	294	0.69
		Family Ceratopogonidae	Insecta	33	0.06
		<i>Cladotanytarsus mancus</i>	Insecta	17	0.03

		<i>Polypedilum</i> sp.	Insecta	41	0.07
III	Autumn	<i>Notomastus latericeus</i>	Polychaeta	37	0.12
		<i>Corophium sinense</i>	Crustacea	23	0.08
		<i>Cladotanytarsus</i> sp.	Insecta	35	0.06
		<i>Dicrotendipes tritonus</i>	Insecta	25	0.04
		<i>Sigara substriata</i>	Insecta	13	0.07
IV	Spring	<i>Neanthes succinea</i>	Polychaeta	33	0.08
		Class Bivalvia	Mollusca	87	0.07
		<i>Moerella jedoensis</i>	Mollusca	42	0.08
		<i>Philine</i> sp.	Mollusca	61	0.12
		<i>Macrophthalmus japonicus</i>	Crustacea	13	0.03
	Autumn	<i>Corophium sinense</i>	Crustacea	1050	0.62

Table 8. A comparison between the heavy metal concentrations in our study area and published results from other big rivers around the world.

	Concentration (mg/kg)					Source
	Cd	Cr	Cu	Pb	Zn	
Yellow River	0.10-0.28	51.25-77.44	18.36-30.14	14.06-25.30	53.73-81.08	Present study
Yellow River	0.08-0.13	51.37-79.75	--	30.9-74.4	--	Zhang et al. (2013b)
Mississippi River	1.00-1.05	8.65-17.30	18.12-23.34	14.30-32.80	51.20-72.30	Santschi et al. (2001); Balogh et al. (2009)
Amazon River	--	31.72-72.80	3.81-38.84	16.56-37.26	81.25-386.10	Marchand et al. (2006)
Yangtze River	0.08-0.30	40.90-61.50	18.90-36.90	19.50-41.50	52.60-124.80	Huang et al. (2006)
Pearl River	7.20-7.80	23.80-59.70	49.30-69.00	167.70-264.20	267.70-426.00	Cheung et al. (2003)

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10