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Bioavailability of trace metals in sediments of a recovering freshwater coastal wetland in China's Yellow River Delta, and risk assessment for the macrobenthic community

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HIGHLIGHTS

- We assessed the instability and bioavailability of sediments.
- Mn and Cd showed moderate and high risks and only Cd exhibited high bioaccumulation.
- As showed a high accumulation risk in despite of its low *ISI* and *BAI* values.
- *BSAF* and $BSAF_m$ values differed greatly for As, Cr, Cd, and Ni.
- Chironomids may serve as a bioindicator in restoration areas.

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ABSTRACT

We investigated the speciation of trace metals and their ecological risks to macrobenthic communities in a recovering coastal wetland of China's Yellow River Delta during the freshwater release project. We established 16 sampling sites in three restoration areas and one intertidal reference area, and collected sediments and macrobenthos four times from 2014 to 2015. The instability index for the trace metals showed a moderate risk for Mn and a high risk for Cd. For both Mn and Cd, the carbonate and Fe–Mn-bound fractions appear to contribute mostly to the instability and bioavailability indexes, but for Cd, the exchangeable fraction also have a much higher contribution. The bioavailability index indicated higher bioavailability of trace metals in freshwater restoration areas than that in the intertidal area. The single-factor contamination index indicated that most trace metal concentrations in the macrobenthos were in excess of the national standard. The biota-sediment accumulation factor suggested that the macrobenthos accumulated most As, Cd, and Cu. Redundancy analysis showed clear relationships between the macrobenthos and sediment metal concentrations. Our results will help wetland managers to assess the bioaccumulation risks based on metal speciation, and to improve management of these recovering freshwater wetland ecosystems.

1. Introduction

Coastal wetlands are transitional areas between land and sea, so their hydrology and ecology are strongly influenced by both freshwater and salt water (Wu and Xu, 2007). However, large areas of coastal wetlands have been lost worldwide because of the decreased freshwater inflows due to damming of rivers, freshwater diversion to other uses, and climate change that has decreased precipitation in headwater areas (Ward et al., 2002; Blankespoor et al., 2012). Freshwater releases into degraded coastal wetlands have become a popular direct restoration activity. This is done by allocating water from other systems (e.g., reservoirs) to provide periodic inundation of wetlands by freshwater (e.g., Merendino and Smith, 1991; Alexander and Dunton, 2002; Cui et al., 2009; Montagna et al., 2015). Most researchs about the effects of restored freshwater inflows to coastal wetlands has focused on the recovery of native vegetation communities (Montagna et al., 2002), changes in salinity and groundwater characteristics (Kalantzi et al., 2014; Li et al., 2016a), increased vegetation cover (Yang et al., 2017), and the delivery of ecosystem services (Montagna et al., 2016).

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Increasing attention has recently been given on the macrobenthic communities (Montagna et al., 2002; Kalantzi et al., 2014; Li et al., 2016a). On the one hand, the macrobenthic community responds to inflows with increased abundance, biomass, and diversity (Montagna et al., 2016; Li et al., 2016b); on the other hand, trace metal contamination induced by freshwater releases represents a potential ecological risk (Cui et al., 2011; Wu et al., 2014; Bai et al., 2016). The macrobenthos can bioaccumulate, biomagnify, or biotransfer certain metals to achieve concentrations high enough to cause harm (Wcislo et al., 2002; Dauvin, 2008). Most previous risk evaluations focused on the total trace metal concentrations. However, researchers have increasingly confirmed that trace metal bioavailability may be more important, and that bioavailability depends on their physicochemical fractions rather than their total concentrations (Li et al., 2007; Burton, 2010; Wu et al., 2016). Roig et al. (2016) demonstrated that monitoring programs based only on the total metal contents in water and sediment were ineffective, as metals present even at undetectable concentrations in the environment can accumulate strongly in benthic organisms.

Quantification of trace metal fractions therefore offers a more realistic estimate of the actual environmental impact (Singh et al., 2005; Cuong and Obbard, 2006; Zhang et al., 2017a). For aquatic sediments, sequential extraction has provided important insights into potentially toxic elements (Tessier et al., 1979; Mossop and Davidson, 2003; Sutherland, 2010). However, the significantly ecological responses of the macrobenthic community to trace metal pollution in these sediments have usually been measured by the variations in organism abundance (De Jonge et al., 2008) or species diversity (Ryu et al., 2011; Sharifinia et al., 2016), or by the changes in qualitative measures such as expert consensus (e.g., AMBI and BENTIX from Rabaoui et al., 2015; IBMWP from Roig et al., 2016). We found no reports about the immediate effects of speciation on bioaccumulation of these metals in macrobenthic communities or on the responses of these processes to freshwater releases into restored coastal wetlands.

In this study, we used field surveys to fill gaps in our knowledge of the effects of trace metal speciation on bioaccumulation in macrobenthos under a freshwater restoration project. We hypothesized that trace metal speciation would create different degrees of bioaccumulation in the macrobenthos and would provide stronger insights into the bioaccumulation than results based only on total trace metal concentrations. Our goals were to: (1) evaluate the instability and bioavailability of trace metals in sediments under freshwater restoration project; (2) explore the accumulation risks of trace metals in macrobenthos; (3) examine the relationships between trace metal fractions and trace metal accumulation in macrobenthos.

2. Methods and materials

2.1. Study area

The study area is located in the Yellow River Delta national nature reserve (37°35′ N to 38°12′ N, 118°33′ E to 119°20′ E) (Fig. 1). In recent years, ecological freshwater release projects have been conducted (Cui et al., 2009). In this study, we selected the northern part of the wetlands (the Yiqianer National Nature Reserve) as a case study, as it has received early-summer freshwater releases that coincided with water and sediment regulation for the upstream Xiaolangdi Reservoir since June of 2010. Each release was a single annual event that lasted for 10–45 days, and involved transfers of 1.3×10^7 to 3.6×10^7 m^3 of water interestoration areas.

The study region includes four subareas (Fig. 1): areas I and II have received freshwater releases since 2010, area III firstly received freshwater releases in 2012, and area IV is the intertidal zone serving as a reference area that did not receive freshwater releases. The reference area and three restoration areas are separated by intertidal barriers that prevent sea water influx into the restoration areas, and the three restoration areas are hydrologically connected by culverts with a 50-cm radius.

2.2. Collection of sediment and macrobenthos samples

Freshwater was released from the Xiaolangdi reservoir from July 4th to July 13th of 2014 and from July 4th to July 23rd of 2015. We conducted field surveys four times in the study areas before the releases (from 20 April to 10 May) and after the releases (from 15 September to 5 October) in both years. We collected four sediment samples in each area during each sampling period. At each sampling site, surface sediment samples were acquired using a core sampler with a 5.0-cm diameter and 5.0-cm depth. We measured five sediment physicochemical parameters: the salinity, median particle size (MPS), total organic carbon (TOC) content, water content, and pH (Supplemental Materials S2, section 2.1). For details of these measurements, see Li et al. (2016a).

Four fixed sampling sites were studied in the reference area and three restoration areas during each survey to sample the macrobenthic communities. At each sampling site, we obtained three sediment samples inside a stainless-steel frame (surface area 0.1 m^2, depth 0.3 m) and then mixed, rinsed, and passed them through a 0.5-mm sieve to separate the macrobenthos. The organisms were preserved in 99.7% ethanol until they could be identified. Each organism was ideally identified to the species level, but to the class or family level if that was not possible. Organisms were examined and counted under a stereomicroscope (Olympus, Center Valley, PA, USA).

2.3. Trace metal determination in sediments and macrobenthos

We powdered the sediment to pass through a 0.15-mm sieve before the trace metal analysis. The sequential trace metal extraction was carried out as described by Tessier et al. (1979), with minor modifications (Supplemental Materials S1). For each trace metal, the total trace metal concentration in sediments was the sum of the concentration of all the fractions.

For the Insecta and Anopla, we combined and homogenized the whole bodies of individuals from the same taxon. For the Polychaeta, Malacostraca, Bivalvia, and Gastropoda, we instead collected and homogenized foot and abdominal muscle tissues. We oven-dried the chosen organisms and tissues at 55 °C to constant weight (Ahmed et al., 2011). The dried samples were then powdered and passed through a 0.15-mm sieve. We digested the benthic samples with HNO_3 at 15 °C for 1 h, followed by 140 °C for 3 h.

All the digested samples were analyzed using coupled plasma-atomic emission spectrometry (ICP-AES) with a JY-Ultima spectrometer (Horiba Jobin Yvon, Longjumeau, France). We measured the concentrations of As, Cd, Cr, Cu, Mn, Ni, Pb, and Zn in the extracted solutions using the wavelengths specified by the manufacturer. Quality assurance was measured using a combination of standard reference materials (GBW07401, from the Chinese Academy of Measurement Sciences), with blank controls; 1 blank and 1 standard were measured after each 10 samples to quantify the accuracy and precision of the metal analysis. The recovery ratios for the standard samples ranged from 95% to 106%.

2.4. Ecological risk analysis

Exchangeable and carbonate-bound metal fractions (F_1 and F_2, respectively) tend to be released under neutral and acidic