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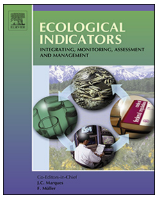

Original Articles

Arsenic and heavy metals pollution along a salinity gradient in drained coastal wetland soils: Depth distributions, sources and toxic risks

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A R T I C L E I N F O

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A B S T R A C T

Salt ions can affect the toxicity, mobility and transfer of metals/metalloids in estuarine wetlands. Soil samples were collected to a depth of 30 cm along a salinity gradient at four sampling sites, including bare land (B), *Tamarix Chinensis* wetlands (T), *Suaeda salsa* wetlands (S) and *Phragmites australis* wetlands (P) in drained coastal wetlands in the Yellow River Estuary, China. Arsenic (As) and heavy metals (i.e., Cd, Cr, Cu, Pb and Zn) were measured to investigate their levels, depth distributions, sources and toxic risks. The results showed that As and heavy metals generally showed a decline with increasing salinities. Arsenic concentrations in all samples exceeded the threshold effects levels (TELs) value and were below the probable effects levels (PELs). The concentrations of Cd, Pb and Zn in all soil samples were below TEL values, while Cr and Cu concentrations were grouped to the range of TELs-PELs in several soil samples. According to the geoaccumulation index (I_{geo}), Cd exhibited unpolluted to moderate pollution at Sites B, S and P, whereas no pollution for other heavy metals were observed in all soils. Generally, higher I_{geo} values for Cd were observed at those sites with lower salinities. The average toxic unit (TU) values of As and heavy metals at Sites B and T followed the order Cr > As > Zn > Cu > Pb > Cd, and the followed order was As > Cr > Zn > Cu > Pb > Cd at Sites S and P. As and Cr showed higher contributions to the TUs than other metals. Correlation analysis showed that As and heavy metals were negatively correlated with electrical conductivity (EC), sand content, Cl^-, Cl^-/SO_4^{2-} ratio and Mg^{2+} ($P < 0.05$), while were positively correlated with soil moisture, clay and silt contents and soil organic carbon (SOC) ($P < 0.05$). The mutivariate analysis indicated that these heavy metals originated from the same source, while As also had another source. The findings of this work can contribute to pollution control and ecosystem health conservation of coastal wetlands.

1. Introduction

Wetland soils can serve as sources, sinks and transformers of nutrients and chemical pollutants and can protect water quality of rivers and lakes adjacent to wetlands (Reddy and DeLaune, 2008; de Andrade Passos et al., 2010; Li et al., 2014a). However, wetland soils around the world are facing metal contamination due to intense human activities (Reddy et al., 2010). Heavy metals are reported to be persistent and could be bioaccumulated in the whole ecosystem (de Andrade Passos et al., 2010). The mobility and toxicity of metals in wetland soils are determined by their chemical properties, concentrations, and availability (Wuana and Okieimen, 2011). Therefore, a better understanding of heavy metal pollution status in wetland soils could contribute to wetland ecosystem health.

Variations in soil characteristics (i.e., organic matter, clay mineral, pH and oxidation/reduction status), structure and development could change the solubility and mobility of toxic metals (Reddy et al., 2010; Li et al., 2014a). Once the changes in the oxidation-reduction status of soil occur, the chemical forms of toxic metals would be transformed, thus affecting their mobility and availability (Reddy et al., 2010). Furthermore, soil oxidation-reduction status could impact soil pH, which affects metal chemistry to a certain degree (Aydinalpi and Marinova, 2003). Aydinalpi and Marinova (2003) showed that more heavy metals could be immobilized with increasing soil pH under mildly alkaline conditions.

Salinity is another key factor affecting toxicity, mobility and transfer of metals in estuarine wetlands (Riba et al., 2003; Fritioff et al., 2005). Gonzalez-Davila et al. (1995) observed that Na^+ ions could

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release Cd from sediment to the overlaying water. A decrease in salinity could enhance the bioavailability and toxicity of heavy metals (e.g., Ca, Zn, Pb) on marine and estuarine isopods (Jones, 1975; Riba et al., 2003). At low salinity ranges, the biological effects of heavy metals were higher than those at high salinity ranges (Riba et al., 2003). Fritioff et al. (2005) reported that heavy metals assimilated by plants at low salinity levels were twice as high as those at higher salinity levels.

With rapid economic development and population growth, most of coastal wetlands worldwide have been drained for construction projects (i.e. port construction, dam building and channel construction) or agricultural lands (Drexler et al., 2009; Brunet and Westbrook, 2012). The drained wetland soils would suffer from major changes in wetland structures and functions (Drexler et al., 2009). Wetland drainage could increase the redox potential and potentially decrease pH values, thereby changing the solubility of metals (Reddy et al., 2010). Meanwhile, soil salinity would increase in the process of wetland drainage (Brunet and Westbrook, 2012). Therefore, metal cycling in wetland soils would be altered due to the changes in soil properties after wetland drainage.

As a result of urbanization and reclamation activities such as agriculture, aquaculture, harbor construction and traffic construction, metal contamination in coastal regions has generated increasing concerns over the past few decades. Moreover, heavy metals and antibiotics from agricultural and aquaculture chemicals could increase the chance of antibiotic resistance (Yang et al., 2017). Dredging or constructing activities might be accompanied by potential environmental issues (i.e. metals and toxic substances), which need to be carefully managed (Erftemeijer et al., 2013). Moreover, saline soils polluted by heavy metals are difficult to remediate, due to the high mobility of heavy metals in soil with higher salinity (Li et al., 2014b). Assessment of metal distribution and toxicity in soils has been listed as a key issue in environmental sciences for a long time (Tessier and Campbell, 1987). Arsenic, Cd, Cr, Cu, Pb and Zn are considered as priority heavy metals that could pose environmental health risks (Wuana and Okieimen, 2011). Most researchers have focused on heavy metal pollution in freshwater or coastal sediments/soils in natural wetlands (Gao et al., 2013; Bai et al., 2014). However, little information is available about profile distributions, sources and risk assessment of heavy metals in wetland soils with changing salinity after wetland drainage. The objectives of this study were: (1) to investigate the profile distributions and risk assessment of As and heavy metals (i.e., Cr, Cd, Cu, Pb and Zn) along a salinity gradient in drained coastal wetlands; (2) to identify the potential sources of As and heavy metals; and (3) to assess the toxic risks of As and heavy metals.

2. Materials and methods

2.1. Site description

This study was conducted in drained coastal wetlands located near Dongying Port, in the Yellow River Delta, Shandong Province of China (Fig. 1). For the construction of Dongying Port, the surrounding coastal wetlands were drained and tidal flows were blocked, resulting in the degradation of coastal wetlands due to drying and salinization. The Yellow River Delta is characterized of a temperate monsoon climate with rain and heat over the same period. It has a clear distinction between the four seasons. The annual average air temperature is 12.4 °C, the annual average precipitation is 551.6 mm and the annual average evaporation is 1928.2 mm (Cui et al., 2008; Gao et al., 2012). Dominant wetland plant species are *Suaeda salsa*, *Tamarix chinensis* and *Phragmites australis* in this region.

2.2. Sample collection and analysis

In the drained coastal wetlands, four sampling sites (i.e., bare land (B), *Suaeda salsa* (S), *Tamarix chinensis* (T) and *Phragmites australis* (P)) with lower soil moisture (< 20%, Table 1) were selected in August 2013. Threre are approximately 200 m away between two adjacent sampling sites. The four sampling sites exhibited different salinity levels, following the order B (8.68 ± 4.25 mS/cm) > T (5.89 ± 3.17 mS/cm) > S (3.19 ± 1.01 mS/cm) > P (2.26 ± 0.39 mS/cm) (Table 1). Three soil cores (5 cm diam.) to a depth of 30 cm were collected at 10 cm intervals at each sampling site, and three replicates were collected within a radius of 1 m and mixed for each sample. In total nine composite samples were obtained at each sampling site. These samples were placed in polyethylene bags. A part of each soil sample was stored in the portable refrigerator for microbial analysis. All the rest of soil samples were brought to the laboratory at once. After air-drying at room temperature for two or three weeks, soil samples were sieved through a 2-mm nylon sieve to move coarse debris and stones. Some air-dried samples were ground with a pestle and mortar until the samples passed through a 0.85 mm nylon sieve for the physical analysis (i.e., pH and EC). The remaining soils were ground until all particles passed through a 0.149-mm nylon sieve for chemical analysis (i.e., As, heavy metals and SOC).

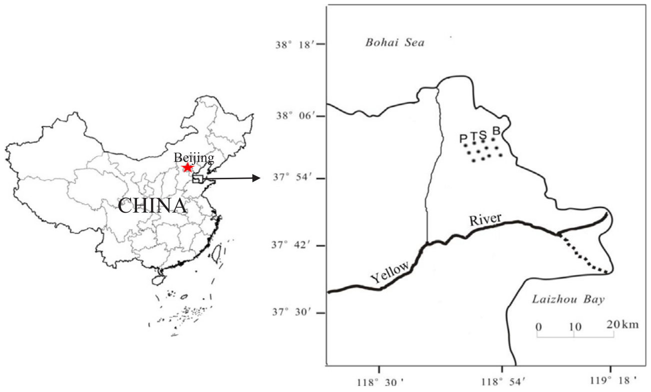

Fig. 1. Location map of sampling sites in the study area.

Table 1
The regional background values and sediment quality guidelines.

mg/kg	As	Cd	Cr	Cu	Pb	Zn
Background values[1]	10.7	0.095	68	21.1	21.6	64.5
TEL[2]	7.24	0.68	52.3	18.7	30.2	124
PEL[2]	41.6	4.21	160	108	112	271

[1] Background values in the Yellow River Delta (CNEMC, 1990).

[2] TEL (threshold effects level), PEL (probable effects level) (Long et al., 1995).

Approximately 0.1 g of each soil sample was weighed and transferred into Teflon tubes to determine the concentrations of As and heavy metals. Each soil sample was digested with 3 ml HNO_3, 1 ml $HClO_4$ and 1 ml HF at 160 °C. After digestion, 1 ml 4 M HCl was added to each tube, and the mixture was diluted to 10 ml with deionized water for the determination of As, Cd, Cr, Cu, Pb and Zn. The concentrations of As and heavy metals in soils were analyzed using inductively coupled plasma atomic emission spectrometry (ICP-AES). For quality assurance and quality control, each batch of samples were analyzed with duplicates, method blanks and standard reference materials (GBW07401) from the Chinese Academy of Measurement Sciences. During the analysis of heavy metals in soils of this study, one blank and one standard were included with every ten samples. The recoveries from the spiked standard samples ranged from 95% to 105%.

The soil pH and electronic conductivity (EC) were measured in the supernatants of 1:5 soil-water mixtures using a Hach pH meter (Hach Company, Loveland, CO, USA) and a conductivity meter (Mettler Toledo, USA), respectively. Salt ions were determined on an ion chromatograph (ICS-2100, USA). Sodium chloride (NaCl) has been proved to be the dominant type of soil salinity in the Yellow River Delta,