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Mobility and potential risk of sediment-associated heavy metal fractions under continuous drought-rewetting cycles

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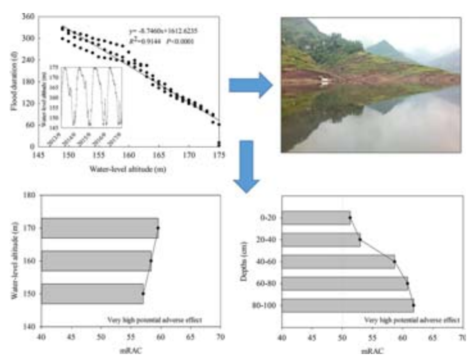
HIGHLIGHTS

- Sediment physical-chemical properties showed an altitude and depth-dependent trend;
- Long-term flooding increased heavy metals in the loosely bounds at the lower portion of WLF zone;
- Organic matter-bound fraction of heavy metal was a key mediator for ecological risk;
- The positive response of LOS of heavy metals to DRW process minifies the risk of heavy metals in the WLF zone sediment

GRAPHICAL ABSTRACT



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ABSTRACT

Ecological decline in the water level fluctuating (WLF) zone of the Three Gorges Reservoir (TGR) has been well established over the past decades. However, the effect of heavy metal fractions present in the sediment and their potential ecological risk under the anti-seasonal hydrological regime are still unclear. The Pengxi River is a tributary of the Yangtze River and it has a typical annual water level fluctuation ranging from 145 to 175 m above sea level. The current study examined heavy metal fractions in sediments containing Cr, Cd, Cu, Pb and Mn collected along the WLF zone using the Tessier sequential extraction scheme. The total organic carbon (TC), total nitrogen (TN), carbon/nitrogen ratio (C/N), pH, particle size composition and content of nitrate (NO_3^--N), and ammonium (NH_4^+-N) differed dramatically among the sampled altitudes and depths and was significantly correlated with the flooding time of the WLF zone. At lower altitudes of the WLF zone, the amounts of the exchangeable (EXC), carbonate-bound (CA) and total heavy metal contents of the surface sediment were much higher compared to those of higher altitudes of the WLF zone. The risk assessment code (RAC) for Cd and Mn showed an opposite trend to that of Cr, Cu, and Pb and mainly depended on the organic matter-bound fraction (OM). The modified RAC (mRAC) indicated a very high potential adverse effect for the whole WLF zone, although the risk value was much lower at the lower altitudes and upper depths of the WLF zone. Our results showed that the positive response of the loosely bound fractions (LOS) of heavy metals to the drought-rewetting (DRW) process minimizes the risk of heavy metals in the WLF zone sediment.

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1. Introduction

The water level fluctuating (WLF) zone of the Three Gorges Reservoir (TGR) is the largest freshwater WLF zone in the world (Bao et al., 2015), comprising an area of 348.92 km^2 across 18 administrative units. The majority of the floodplain sediment exposed due to the artificial water drainage of the TGR conducted from May to September is utilized for farming (Yang et al., 2015). Since the floodplain sediment could serve both as a source of potential noxious metals and as a transformation media (Ye et al., 2013), it may pose a potential ecological risk for Three Gorges tributaries under flood pulse conditions. Over the past 14 years, the WLF zone has been subjected to a continuous interannual flood pulse, and the water table variation was further intensified since 2010 due to full operation of the Three Gorges Dam. The common yearly hydrological variation consists of four stages: (1) water filling leading to a rising water table, (2) a fully filled reservoir with a constant maximum level approximately 175 m, (3) a reservoir drainage period wherein the water table gradually decreases, and (4) natural floods causing highly variable water levels (Tang et al., 2014). With the anti-seasonal hydrological regime (Wang et al., 2016; Xiao et al., 2017), the drought-rewetting (DRW) process was reported to induce a significant decrease in biodiversity (New and Xie, 2008; Zhang et al., 2016) by increasing soil erosion (Qian et al., 2016) and the input of elements into the water (Zhao et al., 2013).

Due to the water table fluctuation in the DRW process, natural absorbents, such as organic matter, iron oxide, and clay and silt minerals, are heterogeneously distributed in the floodplain sediment (Ivanić et al., 2017; Zhang et al., 2014). Thus, the sediment may serve as a sink for heavy metals by gradual physical adsorption or/and chemical transformation, suggesting that heavy metal pollution in the sediment could pose a severe threat to the aquatic ecosystem due to their high toxicity, non-biodegradable and persistent nature and bioaccumulation in the food chain (Alves et al., 2014; Rahman et al., 2015). It remains to be investigated what mechanisms mediate the relationship of heavy metal mobility and ecological risk, as well as whether the relationship shifts under the enhanced water-soil interface process. Elucidation of this relationship will help to better predict the response of sediment heavy metal fractions to the DRW process in the WLF zone.

The DRW process may alter the association mechanisms of heavy metals via sediment solid phase binding processes, causing the incorporation of the metals into the lattice structure of minerals via adsorption, coagulation or flocculation (e.g., Fe-Mn oxides) or via deposition of metals onto sediment surfaces and precipitation, leading to the formation of insoluble fractions as a product of the intensified flooding stress (Zhang et al., 2014). However, the mechanisms of DRW-controlled solid-phase sediment binding with metals are still poorly understood, and factors influencing the mobility of heavy metals in the WLF zone sediment have never been comprehensively clarified in relation to the DRW process (Gao et al., 2016).

We conducted an in situ investigation of the sediment at higher, middle and lower altitudes and five different depths in the WLF zone of the Pengxi River, a tributary of the Yangtze River. Our investigation serves two purposes. First, we attempted to assess the response of heavy metal fractions and sediment particle-size fractions along declining altitudes of the WLF zone to the increased flooding stress caused by the DRW process. Second, we examined the controlling factors of heavy metal mobility and the posed ecological risk using the risk assessment code (RAC) and modified RAC (mRAC). We hypothesized that the DRW process changed the distribution of sediment particle-size fractions along the slope of the WLF zone and affected the release of both nutrients and heavy metal fractions, which mediated the ecological risk of the heavy metals. These effects may be particularly pronounced at lower altitudes of the WLF zone, which suffer much longer artificial flooding periods. A comparison of the effects between higher and lower altitudes of the WLF is thus useful in the prediction of heavy metal responses to the continuous DRW process.

2. Materials and methods

2.1. Study area

The Pengxi River is a typical tributary of the Yangtze River (N30°49′30″ - 31°41′30″, E107°55′48″ - 108°54′48″) located between Kai and Yunyang counties (Fig. 2) (Lin et al., 2016). Areas of up to 27.96 km^2 across the WLF zone of the Pengxi River are exposed to the air during the drainage period of the TGR. The region experiences northern subtropical humid monsoonal climatic conditions with an average annual precipitation of 1053.15 mm, with 80% falling during the rainy season from May to September. The average annual temperature is 18.2 °C. The water level starts to rise in September and drops down to 160 m in April of the following year. It is notable that the period from April to September is suitable for sediment sampling.

2.2. Sampling

When the water level was at the base level of 145 m in June of 2016, we collected sediment samples along three representative transects, named Qukou, Gaoyang, and Shuangjiang (Fig. 1). Along each transect, surface samples (0–20 cm) were collected from three sampling plots of 1 × 1 m selected randomly at the water-level altitudes of 150, 160 and 170 m. The five depths samples (0–20 cm, 20–40 cm, 40–60 cm, 60–80 cm and 80–100 cm) were taken from another individual profile at each transect with a plastic shovel. The GPS coordinates of the sampling points are shown in Table S1.

2.3. Laboratory analysis

All samples were sealed in polyethylene bags and stored at 4 °C for <2 d, then dried at 105 °C in a forced air oven. They were subsequently ground in an agate mortar and then homogenized and sieved through a 100-μm mesh. One gram of the sediment sample was used to initially extract and fractionate the heavy metals into five fractions—exchangeable (EXC), carbonate-bound (CA), Fe-Mn oxide-bound (Fe-Mn), organic matter-bound (OM) and residual (RES)—with a five-step

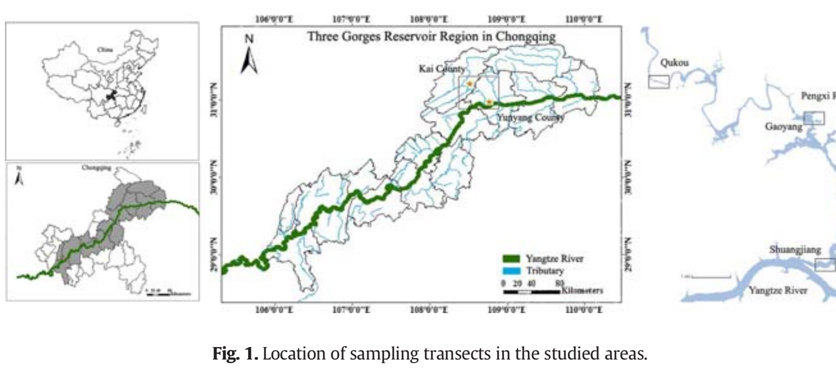

Fig. 1. Location of sampling transects in the studied areas.