



## Spatial variation of heavy metal contamination in the riparian sediments after two-year flow regulation in the Three Gorges Reservoir, China

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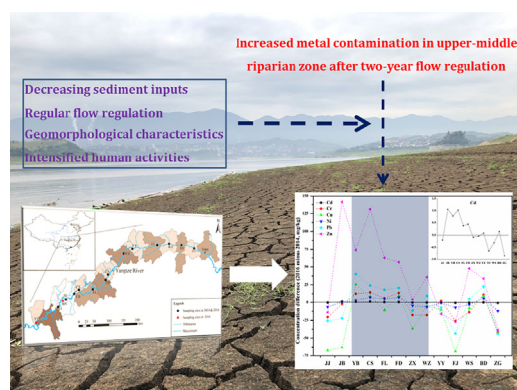
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### HIGHLIGHTS

- Spatial variation of metal contamination in the riparian sediments could occur after the TGR flow regulation.
- Higher contamination and eco-risk of metals existed in the downstream TGR in 2014 but in the upper-middle regions in 2016.
- Cd was the concerned metal with high contamination and potential eco-risk in both years.
- Human activities increasingly contributed to metal contamination in the upper-middle TGR region.
- Decreasing sediment inputs, flow regulation, geomorphological and anthropogenic factors determined the metal redistribution.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Regular impoundment of the Three Gorges Reservoir (TGR) with intensified human activities in the watershed imparts a significant effect on the environmental changes in the riparian zone. In this study, six heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) in the riparian sediments of the entire TGR mainstream were investigated in 2014 and 2016 to identify their contamination and risk characteristics and decipher the main factors for the variation of the metal contamination. The results showed that the concentrations of the heavy metals in the sediments did not vary significantly between 2014 and 2016, and their contamination degrees decreased in the order of  $Cd > Cu \approx Zn > Pb > Cr \approx Ni$  in 2014 and  $Cd > Zn > Cu \approx Pb > Cr \approx Ni$  in 2016. The potential eco-risk of Cd was extremely high in the two years, while the eco-risk of other metals was very low. The sediments showed a moderate to high contamination level, a high potential eco-risk but a low toxic risk to aquatic biota in the two years. Spatially, the contamination and risk levels of heavy metals were relatively higher in the downstream TGR region in 2014 except for the sites close to the urban areas but in the upper-middle TGR region in 2016. Increasing anthropogenic influence contributed to the high contamination and risk levels of Cd, Cu, Pb and Zn in the upper-middle region in 2016. The results indicated that the Cd contamination in the riparian sediments of the TGR was still a vital environmental issue, and the decreased sediment inputs from the upstream major tributaries, the periodic and anti-seasonal flow regulation, local geomorphological characteristics and anthropogenic activities determined the contamination distribution of heavy metals in the riparian sediments.

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## 1. Introduction

Heavy metal contamination as a serious threat to various ecosystems has become a global issue due to their toxicity, non-biodegradable nature and biological enrichment by food chain (Bai et al., 2016; Duan et al., 2018; El Nemr et al., 2016; Rosado et al., 2016; Singh et al., 2005). Except for a small portion of heavy metals existing in water column in the dissolved form, over 90% of the metals in aquatic systems occur in the large reservoir of suspended particles and sediments (Zahra et al., 2013; Zheng et al., 2008). Due to physical and chemical disturbances, heavy metals in the sediments may be released into the water column. This means that the sediments act as a crucial sink and a long term discharge agent of heavy metals into the environment even long after the initial input, which is of great significance for the aquatic safety. Meanwhile, in contrast to overlying water column that is frequently dynamic, the fluvial sediments provide a better archive to assess the contamination and potential ecological risk of heavy metals in aquatic environment and to obtain the information of anthropogenic influence.

Over the past decades, many pristine fluvial systems around the world have been fragmented and impacted by the construction of dams and the irrigation projects for the requirements of the services such as flood control and power generation (Lv et al., 2015; Nilsson et al., 2005). The dammed rivers favor sediment accumulation through decreasing water flow velocity and increasing sediment residence time (Friedl and Wüest, 2002). Consequently, the areas with the contamination of heavy metals in the fluvial sediments may be redistributed by the changes in hydrodynamic conditions, sediment deposition and physico-chemical properties (Feng et al., 2014; Fremion et al., 2016; Zhang et al., 2014). Meanwhile, with growing populations and rapid urbanization and industrialization at the watershed scale, the contamination of heavy metals in the sediments is expected to be more complicated with time.

The Three Gorges Reservoir (TGR) at the upper reach of the Yangtze River is the largest hydroelectric project in the world (Fu et al., 2010; Nilsson et al., 2005), and it covers the regions between Chongqing and Yichang with a distance of ~660 km (Fig. S1). The TGR began to impound water since the late 2003, the water level reached 175 m above sea level (a.s.l.) first in the late 2010, and since then the water level has been fluctuating between 145 m a.s.l. in summer and 175 m a.s.l. in winter. After the impoundment in 2003, the TGR intercepted substantial sediment loads (Li et al., 2011; Yang et al., 2014); however, the sediment discharge into the TGR has been decreasing in recent years due to the natural and anthropogenic influences such as the construction of large upstream Cascade Reservoirs, decreasing precipitation in the watershed, and water and soil conservation projects (Dai and Lu, 2014; Yang et al., 2015, 2018; Zhao et al., 2017). Meanwhile, the flow regulation causes the particle size sorting along the TGR mainstream, and a fining trend of the sediment particle size towards the Three Gorges Dam (TGD) has been observed (Wu et al., 2016; Tang et al., 2014; Yang et al., 2014). On the other hand, the periodic and anti-seasonal regulation of the water level in the TGR and the intensive disturbance in its surrounding areas formed a unique geomorphological unit of the riparian zone (Bao et al., 2015). The riparian zone serves as an important ecotone with various environmental changes, which provides multiple ecological services such as biodiversity conservation, abatement of diffuse pollutants, overland runoff regulation, and bank stabilization (Tang et al., 2018a). In each summer flooding season, substantial sediments are left in this riparian zone with the water withdrawal, especially in the channels with large widths and gentle slope gradients (Tang et al., 2018b; Wang et al., 2016c; Zhao et al., 2017). In the context of the unstable variability in the sediment inputs from the upstream tributaries, hydrodynamic regime, catchment erosion and runoff, and human activities in the upper region of the Yangtze River, increasing concerns have thus been raised over the new challenges for the impacts of heavy metal contamination in the riparian sediments of the TGR on the ecological environmental development.

Given the significance of the aquatic environmental quality, the contamination, ecological and toxic risk of heavy metals in the riparian sediments after the full operation of the TGR have been examined by many researchers (e.g., Gao et al., 2018; Tang et al., 2014; Wang et al., 2016a; Wei et al., 2016; Ye et al., 2011). However, most of these studies were conducted either in tributaries or in certain reaches of the TGR mainstream. Compared with the tributaries, the riparian zones in the mainstream are much easier to be disturbed by stronger hydrodynamic conditions, direct sediment inputs, and intensified anthropogenic activities along the reservoir. This will result in much more variable spatial distribution of heavy metals in the riparian sediments of the TGR. In addition, the spatial and temporal variation of heavy metal contamination in the water of the entire TGR from 2008 to 2013 was reported to be intricate, although the water quality was generally acceptable (Gao et al., 2016). Whether the spatial and temporal distribution of heavy metal contamination in the riparian sediments of the TGR mainstream varies with the flow regulation and what key factors or processes determine this variation still need to be comprehensively explored.

In this study, we selected six heavy metals including Cd, Cr, Cu, Ni, Pb and Zn in the riparian sediments of the entire TGR mainstream to investigate their concentrations in 2014 and 2016, respectively. The objectives of our study are (1) to assess the contamination and risk characteristics of the metals as well as their spatial variation in the two years; (2) to identify the source variation of these metals between the two years using statistical methods and Pb isotopic ratios; (3) to decipher the key factors or processes regulating the spatial variation of the heavy metal contamination in the riparian sediments. This research could be helpful to understand the sediment quality variation under the effects of the regular TGR operation and anthropogenic activities, and thus support the strategic management of heavy metal contamination in the TGR.

## 2. Materials and methods

### 2.1. Sample collection

The sampling sites in 2014 and 2016 spanned the entire mainstream of the TGR region (Fig. S1). The detailed information in the study area including climate, regional landform and geology, the attributes of the water-level fluctuation zone of the TGR and land use could be found in elsewhere (Bao et al., 2015; Tang et al., 2016). Most of the elevation ranges selected to collect the sediment samples in the riparian zone varied between 145 m and 160 m a.s.l. except for several sites in the upper backwater section, since the sediments were not subjected to settle down in the high elevation after the water withdrawal. Meanwhile, the two-year interval of sampling time allowed obtaining the new surface sediments in 2016 compared with in 2014 due to the high sedimentation rate and flow regulation (Tang et al., 2016).

In July 2014, thirteen sites in the riparian zones including Jiangjin (JJ), Jiangbei (JB), Yubei (YB), Changshou (CS), Fuling (FL), Fengdu (FD), Zhongxian (ZX), Wanzhou (WZ), Yunyang (YY), Fengjie (FJ), Wushan (WS), Badong (BD) and Zigui (ZG) were selected to collect the surface sediment samples (0–20 cm) with a plastic shovel. In July 2016, besides the thirteen sites in 2014, another two sites of Yongchuan (YC) and Banan (BN) were selected to collect the sediments (Fig. S1). The site of YC was selected in 2016 in order to identify the quality of the sediments from the upper reach of the Yangtze River. The surface sediments of 0–10 cm were collected at the sites of YC, WS, BD and ZG due to the small sediment loads. At each site, at least three replicates of sediment samples were collected. Since the riparian zones and sediment loads are mainly located at the central TGR regions (from FL to FJ), much more sub-sites were selected to increase the sample collection. In total, 81 and 63 sediment samples were collected in 2014 and 2016, respectively.

All sediment samples were kept at 4 °C in the field. After returning to the laboratory, the samples were freeze-dried and sieved <2 mm to

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