



# Current state, sources, and potential risk of heavy metals in sediments of Three Gorges Reservoir, China<sup>☆</sup>



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## ABSTRACT

Heavy metal (HM) contamination in sediments of Three Gorges Reservoir (TGR) is a particularly important issue for the safety of water quality due to the potential threats of metal toxicity to local and downstream human health. Surface sediments from riparian and submerged areas in the entire TGR mainstream were collected in 2014 to investigate the spatial distribution of HMs (Cd, Cu, Pb, and Zn), identify their possible sources, and assess their potential risk by multiple indices and metal fraction. Results showed that the concentrations of HMs in the sediments increased after the TGR operation, but were lower than those in other Chinese rivers of developed areas. The acid-soluble Cd accounted for more than 50% of total Cd in the sediments, whereas that of other HMs was very low. The Cd concentrations in the riparian sediments increased towards the dam; however, other metals in the riparian sediments and all HMs in the submerged sediments did not show any regular variation trend spatially. The stocks of HMs were significantly higher in the submerged sediments than in the riparian sediments. The high accumulation of HMs in the riparian sediments emerged between Fuling and Fengjie, and those in the submerged sediments existed in the near dam areas. Grain size and Fe/Mn oxides controlled the mobility and transfer of HMs in the sediments. Human activity in the catchment including industrial and agricultural production, shipping industry, mining, etc., increased inputs of HMs in the sediments, and altered their spatial distribution patterns. The sediments were moderately to highly contaminated by Cd, and slightly contaminated by other HMs. The results indicate the current priority of Cd contamination in the TGR, and will conduce to ecological protection in the TGR region.

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## 1. Introduction

Heavy metals (HMs) create permanent pressure on the environment due to their abundance, persistence, and toxicity. Sediments act as a major sink of HMs in aquatic ecosystems, and have been recognized as an effective indicator of metal contamination (Viers et al., 2009; Baborowski et al., 2012). Heavy metals in river sediments may be released to water column via hydrodynamic disturbance, chemical and biological processes under variable sedimentary conditions, causing potential threat to aquatic biota and human health (Singh et al., 2005). Therefore, it is necessary to understand comprehensively the mechanisms of geochemical

distribution, source signatures, and risk situations of HMs in sediments in order to develop pollution control strategies and approaches to water quality management in riverine areas.

Human activity has increased discharge of HMs into aquatic systems. Different methods have been proposed to distinguish the anthropogenic sources of HMs in sediments from natural sources including statistical methods, geochemical indices, and isotope tracing techniques (Townsend and Seen, 2012; Han et al., 2015a; Bing et al., 2016; Chen et al., 2016). Moreover, sedimentary conditions play a vital role in the distribution and transfer of HMs in sediments. For example, fine-grained sediments not only adsorb more HMs, but also contain metal iron/manganese oxide surface coatings, which will limit the mobility and bioavailability of HMs in sediments (Ip et al., 2007; Bing et al., 2013). As demands on water resources increase, many river systems have been altered through impoundments and diversions. There are more than 45,000 dams above 15 m high globally, with over 300 dams defined as giant

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dams (above 150 m high) (Nilsson et al., 2005). In China, nearly 100,000 reservoirs have been constructed until 2011, and the giant reservoirs account for 0.8% (Lv et al., 2015a). In the catchment of Yangtze River, the large and medium sized reservoirs reach 174 and 1122 respectively in 2010 according to Changjiang and Southwest Rivers Water Resources Bulletin, and some other reservoirs have been building up recently. Changes in hydrological conditions (e.g., reduced water flow velocity) have the potential to affect the distribution and transfer of contaminants in sediments (Feng et al., 2014).

The Three Gorges Reservoir (TGR) in China is the world largest hydroelectric project, which has a large storage and flood control capacity with a water area of 1080 km<sup>2</sup> (Fu et al., 2010). The backwater of the TGR is about 660 km long and forms a typical river-type reservoir from the Three Gorges Dam (TGD) up to Jiangjin City in Chongqing Municipality. The TGR was impounded through three stages to elevations of 135 m, 156 m, and 175 m above sea level in 2003, 2006, and 2010, respectively (Bao et al., 2015). After the completion of the project, the water level varies between 145 m and 175 m, which breaks the natural transport balance of sediments from the upstream of the TGD. Meanwhile, the TGR operation features the characteristic of anti-season water level variation; that is, the high water level emerges in winter (dry season), and the low level occurs in summer (wet season). This will alter original inputs or outputs of contaminants and their distribution in the sediments. In last few decades, the contamination issues of HMs in the sediments of TGR have been increasingly highlighted, whereas the researches were conducted either in tributaries (Wang et al., 2012a; Han et al., 2015b) or in several mainstream sites (Wang et al., 2012b; Tang et al., 2014). In addition, the research mainly focused on the spatiotemporal distribution and eco-risk assessment of HMs in the sediments, and scant information is available on their fractions and bioavailability (Lv et al., 2015b).

Therefore, the main objectives of this study are (1) to analyze the influence of the complete operation of TGR on the distribution of HMs (Cd, Cu, Pb, and Zn) concentrations and stocks in the riparian and submerged sediments along the entire TGR mainstream, (2) to identify the possible sources of HMs in the sediments, and (3) to assess their potential eco-risk by multiple assessment indices and HMs fractions. This work is expected to provide new insights into the contamination state of HMs and thereby the ecological remediation in the TGR region.

## 2. Materials and methods

### 2.1. Study area

The region of TGR (28°28′–31°44′N, 105°49′–110°12′E) is located at the upper reach of the Yangtze River, mainly in Chongqing Municipality and Hubei Province (Fig. 1). The weather in the TGR is mainly controlled by the humid subtropical monsoon climate. The mean annual temperature and precipitation are 16–19 °C and 1000–1200 mm, respectively. The main soil types in the region of TGR include purple soil, yellow soil, paddy soil, calcareous soil, and so on, and the purple soil is the most widely distributed soil. The geomorphological characteristics of the TGR can be found in details elsewhere (Bao et al., 2015).

### 2.2. Sample collection

The sampling sites spanned the entire TGR mainstream from the upstream Jiangjin to the dam area Zigui, covering more than 600 km-long distance (Fig. 1, Table S1). In July–August 2014, twenty-third sites in the riparian zones and eight in the submerged

areas were selected respectively to collect the surface sediment samples (0–30 cm). However, based on the field investigation, the sediment loads were small in Wushan (S20), Badong (S21), and Zigui (S22–23), and thus the sediments in the surface of 0–10 cm were collected. At each riparian site, three repeated samples were collected with a plastic shovel, and the bulk density ring was used to collect samples for sediment density. At each submerged site, three sediment cores were collected using a gravity sediment core sampler (100 cm in length and 6 cm in diameter). Each core was directly extruded and sliced into 30 cm in field with a plastic sheet. The water depth at all submerged sites was more than 30 m. All samples were kept at 4 °C upon returning to the laboratory.

### 2.3. Chemical analysis

Original sediment samples (5 g) were placed in a beaker which was rinse beforehand. HCl (5% in volume) and H<sub>2</sub>O<sub>2</sub> were added to remove carbonates and organic matters respectively, and then the sample was washed to neutral by deionized water. The residues were dispersed with an ultrasonic oscillator for 1 min after mixing with a dispersant solution ((Na<sub>2</sub>PO<sub>3</sub>)<sub>6</sub>). Grain size was measured using a Mastersizer 2000 Laser Grain-size Meter (Clay: <4 μm; Silt: 4–64 μm; Sand: >64 μm).

The rest of sediment samples were air-dried at room temperature for several weeks. Then, the sediments were pulverized by an agate mortar to pass through a 200-mesh Nylon screen. The content of sediment organic matter was determined by the method of loss on ignition (LOI), consisting of burning of a known amount of samples for 4 h at 550 °C in a muffle and recording the percentage of weight loss. Sediment pH was measured by a pH meter after mixing the sediments and deionized water with a ratio of 1:2.5 for 30 min.

The acid-soluble fraction (exchangeable and carbonate-bound metals) of Cd, Cu, Pb, and Zn in the sediments was extracted followed by the BCR approach (Rauret et al., 1999). The solid samples were digested with HCl–HF–HNO<sub>3</sub>–HClO<sub>4</sub>. The concentrations of major elements (Al, Ca, Fe, K, Mg, Mn, Na, P, and Ti) were measured by an American Leeman Labs Profile inductively coupled plasma atomic emission spectrometer (ICP-AES), and those of Cd, Cu, Pb, Zn, and their fractions were detected by inductively coupled plasma mass spectroscopy (ICP-MS). Standard solution SPEX™ from the United States was used as the standard. Quality control was assured by the analysis of duplicate samples, blanks, and reference materials (GSD-9 and GSD-11). According to the measurement of the repeated samples and reference materials, the relative standard deviation was below 3% for ICP-AES and below 5% for ICP-MS, respectively. The recovery of the reference materials was 92–108% for ICP-AES and 90–110% for ICP-MS, respectively.

Lead isotopes (<sup>206</sup>Pb, <sup>207</sup>Pb, and <sup>208</sup>Pb) of the sediments after the corresponding digestion above were determined by ICP-MS (Agilent 7700x). An international standard reference material (SRM981-NIST, United States) was used for instrument calibration and standard material (GBW04426, China) for the analytical control. The maximum deviations of <sup>207</sup>Pb/<sup>206</sup>Pb and <sup>208</sup>Pb/<sup>206</sup>Pb related to the repeated measurements of the GBW04426 Pb standard were less than 0.002, and the RSD of isotopic ratios were 0.18 and 0.09%, respectively.

### 2.4. Calculation of contamination (and eco-risk) indices and HMs stocks

Enrichment factors (EFs), geoaccumulation index (*I*<sub>geo</sub>), contamination index (*P*<sub>i</sub>), potential eco-risk index (*E*<sub>r</sub><sup>i</sup>), and risk assessment code (RAC) were employed to evaluate the contamination intensity of HMs in the sediments. The detailed calculations

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