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Multiple assessments of trace metals in sediments and their response to the water level fluctuation in the Three Gorges Reservoir, China

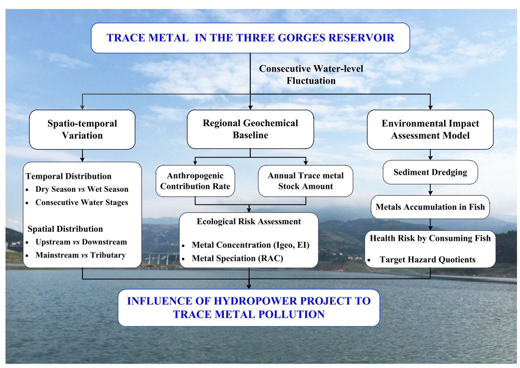
Li Gao [a,b], Bo Gao [a,b,*], Dongyu Xu [b], Wenqi Peng [a,b], Jin Lu [b]

[a] State Key Laboratory of Simulation and Regulation of Water Cycle in River Basin, China Institute of Water Resources and Hydropower Research, Beijing 100038, China
[b] Department of Water Environment, China Institute of Water Resources and Hydropower Research, Beijing 100038, China

HIGHLIGHTS

- Anthropogenic contribution and metal stock are calculated via geochemical baseline.
- Trace metal contents stabilized after TGR operated normally for more than five years.
- Anthropogenic contribution rate for trace metals range from 8.51% to 24.86%.
- Sediment load is the main factor influencing trace metal stock in TGR sediment.
- Fish consumption was safe, but health risk caused by Hg is a potential issue.

GRAPHICAL ABSTRACT



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ABSTRACT

Response of trace metals to the consecutive water level fluctuation in the Three Gorges Reservoir (TGR) sediments remains unclear. Here, we evaluated the influence of consecutive stages of water level fluctuation on trace metal pollution using multiple analytical approaches. The spatio-temporal distributions of trace metals in TGR sediments were investigated for five consecutive water impoundment stages from 2015 to 2017. Anthropogenic contributions and trace metal stocks in the TGR were quantitatively estimated using a combination of a regional geochemical baseline (RGB) and annual sediment load. Results showed that trace metals were accumulated after the construction and impoundment of the TGR. However, after the TGR operated normally for more than five years, trace metals concentrations stabilized in sediments. Trace metal concentrations in the mainstream were slightly higher than those in the tributaries. In the mainstream, metal concentrations in the upstream were lower than those in the midstream and downstream except for Cd. Anthropogenic contributions of trace metals ranged from 8.51 to 24.86% and were highest for Hg and Cd. The sediment load was the main factor influencing trace metal stock in TGR sediments. Although the total Cd stock amount was relatively low, its potential ecological effects are of great concern due to its high mobile fraction percentages and toxicity. The RGB-based geo-accumulation index and potential ecological risk index showed that TGR sediments were uncontaminated, and were subject to low ecological risk from trace metals. This result differs from traditional assessment results, indicating that previous assessments may overestimate the ecological risks of the trace metals in the TGR. The health risks posed by trace metals bio-accumulated in fish, stemming from sediment resuspension, were assessed using an environmental impact assessment model. Results suggested that residents should not experience significant health risks from the intake of individual metals through fish consumption.

* Corresponding author at: State Key Laboratory of Simulation and Regulation of Water Cycle in River Basin, China Institute of Water Resources and Hydropower Research, Beijing 100038, China.
E-mail address: gaobo@iwhr.com (B. Gao).

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1. Introduction

Hydroelectric dam construction has become one of most intense human impacts on river systems (Li et al., 2011). Over 50,000 large dams have been constructed globally for water sustenance, energy generation, flood control, infrastructure, and economic benefits (Li et al., 2013; Holbach et al., 2014). China hosts more than half of all globally commissioned large dams (Nilsson et al., 2005). The construction of dams disrupts river continuities, alters water regimes, and unavoidably impacts the environmental behavior of pollutants (Gao et al., 2016). Presently, the Three Gorges Reservoir (TGR) on the Yangtze River in China is the largest hydropower project in the world, and is a typical example of hydropower engineering. The potential environmental risks and impacts of the TGR have been main focus during the process of its design, construction and operation phases (Shen and Xie, 2004; Stone, 2008, 2010). Multiple studies have investigated environmental threats, including geohazards (Seeber et al., 2010; Yang and Lu, 2013), ecological changes (Wu et al., 2003; Zhang and Lou, 2011), eutrophication (Holbach et al., 2014; Tang et al., 2018) and trace metal levels (Bing et al., 2016).

Among all contaminants, trace metals pose a permanent risk to the environment due to their persistence, bioaccumulation, and toxicity. Sediments serve as the main sink for contaminants in aquatic ecosystem, with over 90% of inorganic pollutants being trapped in sediments during the hydrological cycle (Viers et al., 2009; Fremion et al., 2016; Chen et al., 2018). The construction of the Three Gorges Dam (TGD) limited the natural transfer of water, leading to a decrease in the water flow velocity and an increase in the sediment residence time, favoring trace metal deposition (Bing et al., 2016; Friedl and Wuest, 2002). In fact, after the TGR's impoundment, 60% of the sediments that entered the TGR were trapped, primarily during high-discharge months (June–September) in 2003–2006 (Xu and Milliman, 2009). Moreover, after the TGR commenced normal operation, water level periodically varies between 145 and 175 m, which interrupted the natural transport balance of sediments in the TGR and created an artificial riparian zone, with a vertical height of 30 m (Bing et al., 2016). Additionally, anti-season water operation features alter the original distribution and transfer behavior of contaminants in the sediments. The unique operational mode of the TGR has led to the release of metals from the sediments to the water column through sulfide oxidization and/or sediment organic fraction degradation (Fremion et al., 2016; Caetano et al., 2003). Most studies on trace metals in TGR sediments have focused on their distribution and pollution risk assessment in tributaries (Wei et al., 2016; Gao et al., 2014a) and several mainstream sites (Wang et al., 2012). Some studies have assessed the entire TGR region (Bing et al., 2016; Wang et al., 2017; Gao et al., 2014b, 2015a), without accounting for consecutive water impoundment periods. In fact, sediment background values in the Yangtze have often been used as the references in traditional pollution assessments of metals in the TGR (Bing et al., 2016; Wei et al., 2016; Tang et al., 2014). It is an established fact that the environmental risk level is directly related to the choice of the background value. Methods employed by previous studies are inadequate to scientifically evaluate the pollutant level of metals in the TGR due to the following two reasons: (1) due to anthropogenic activities of many years, especially construction of the TGD, a "natural background" without human interference does not exist (Karim et al., 2015), therefore, pollution risks may be overestimated; (2) the Yangtze River basin is large with a variable geologic background. The background of trace metals in the entire basin may be lower or higher than the actual background in the TGR, and it may not reflect the regional background of trace metals in the TGR. In comparison, a geochemical baseline represents the natural level of trace metals in soils and sediments (Tian et al., 2017; Lin et al., 2012). Moreover, a geochemical baseline can distinguish between natural and anthropogenic concentrations (Tian et al., 2017; Zhang et al., 2014), and allows for the calculation of the rate of anthropogenic contributions of metals at each sampling site accordingly. Furthermore, the amount of metal input from human activities in a region can be quantitatively estimated.

In this study, we investigated the metals in the TGR during five consecutive water phases, from 2015 to 2017. The main objectives were to (1) analyze the spatio-temporal distributions of trace metals through five consecutive water periods in TGR sediments; (2) establish a regional geochemical baseline (RGB), calculate the anthropogenic contribution rate of metals, and estimate the annual stock of metals in TGR sediments; (3) assess pollution risks of trace metals using multiple assessment indices and metal chemical speciation; and (4) investigate the accumulation of metals in fish during sediment dredging and assess the health threat from consuming fish.

2. Materials and methods

2.1. Sample collection

Sediment samples were collected in the wet season (June) and dry season (December) during five consecutive water impoundment stages from 2015 to 2017. At each sampling time, a total of forty-seven sediment samples (thirty-seven samples in the mainstream and ten samples in the tributaries) were collected. The sampling locations are described in the Fig. 1. Sampling sites S1–S6, S7–S21, and S22–S37 were located in the upstream, midstream and downstream areas of the mainstream, respectively, and sites T1–T10 were located in the tributaries. The collected sediment samples were stored in clean polyethylene bags and treated immediately upon returning to the laboratory. The sediment samples were then frozen, lyophilized at −80 °C, and ground in an agate mortar to ensure homogeneity.

2.2. Trace metal analysis

Sediment samples were digested using concentrated HNO_3, H_2O_2, and HF; the detailed procedure is described in the supplementary material. The Cr, Ni, Cu, Zn, As, Cd, and Pb concentrations in the digested sediment samples were analyzed using an Elan DRC-e inductively coupled plasma-mass spectrometry (ICP-MS, Perkin Elmer, USA). Concentrations of Hg were measured using a Direct Mercury Analyzer (Milestone DMA-80). Quality control was tested using certified reference material of stream sediment (GSD-10, GBW07310), produced by the Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences. Analytical reagent blanks were prepared with each batch of digestions and then analyzed for the same elements. The average recoveries of the different metals were in the range of 80.8–108.9% (Table S1).

The chemical speciations of the metals were determined using a three-stage European Community Bureau of Reference (BCR) sequential extraction (Rauret et al., 2001). During the extraction, metals (Cr, Ni, Cu, Zn, As, Cd and Pb) were classified into four fractions: acid-soluble/exchangeable fraction (F1), reducible fraction (F2), oxidizable fraction (F3) and residual fraction (F4). The F4 concentration was calculated using the equation: total concentration - F1 - F2 - F3. The accuracy of the analytical procedures employed for the analysis of metals in sediments was evaluated using the BCR-701 reference material. The recovery values were 88.8–118.1% for F1, 82.7–84.4% for F2 and 95.9–107.81% for F3, respectively.

2.3. Establishment of regional geochemical baseline values and risk assessment of trace metals in the TGR sediments

The RGB values of trace metals in sediments were calculated by normalization. The inert element Li was selected as the reference element in this study. The linear regression equations between the metals and Li were established as:

$$C_m = a \times C_{Li} + b \quad (1)$$