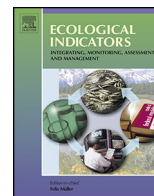




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Distribution, bioavailability, and potential risk assessment of the metals in tributary sediments of Three Gorges Reservoir: The impact of water impoundment

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ABSTRACT

To investigate the impact of water impoundment on the metal contamination in sediments cores from the three tributaries of Three Gorges Reservoir (TGR), the concentrations, distribution, bioavailability, and potential risk of eight trace metals between summer and winter were analyzed using sequential analysis. The mean contents of all studied metals were higher than the geochemical background value, and were higher in summer than in winter. The results of the partitioning study indicated that Cr and Ni prevailed in the residual fraction, while a small proportion was found in the easily soluble fractions. Cu and Zn were distributed mainly in the residual and reducible fraction, while Cd and Pb were predominantly associated with non-residual fractions. These observations suggested that the most easily mobilized metals in the study area were Cd and Pb. The mean enrichment factors (EF) of Cu, Zn, Cd and Hg were higher than 1.5, revealing the potential anthropogenic inputs, whilst the EF of other metals remained within the range of natural variability. The positive correlation between non-residual Cu, Zn and Cd and their EF values further indicated that anthropogenic inputs were the potentially major contributor for the enrichment of Cu, Zn and Cd in TGR sediments. The results evaluated by both potential ecological risk index and modified risk assessment code (mRAC) of all sampling sites demonstrated the relatively high potential risk of sediment contamination effect in TGR.

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1. Introduction

Trace metal contamination has elicited attention due to its inherent toxicity, vast sources, persistence and non-degradability (Audry et al., 2004; Cobbina et al., 2015; Pejman et al., 2015). With rapid urbanization and industrialization, large amount of metals from anthropogenic sources are discharged into the aquatic environment where they are possibly bio-accumulated by aquatic organisms and even bio-magnified through the food chain, thereby posing potential harmful effects on human health and even the whole ecosystem (Mountouris et al., 2002; Rainbow and Luoma, 2011). Therefore, a better understanding of the contamination status of trace metals in aquatic environment is important to the food

security, public health concerns and the sustainable development of the ecosystems (Banerjee, 2003; Godwill et al., 2015).

Sediments are important deposits of metal accumulation in aquatic environment (Forstner, 1989). It has been estimated that more than 90% of trace metals in aquatic systems are related to suspended particles and sediments (Calmano et al., 1993). The high capability of sediments to accumulate metal compounds makes them a major repository of natural and anthropogenic metals and one of the most important media to assess the contamination level in aquatic ecosystem (Silva and Rezende, 2002). Due to their stability within the sedimentary column, most of the contaminants can leave their fingerprint in sediments (Duan et al., 2010). On the other hand, the non-residual fractions of metals (i.e. exchangeable, bound to Fe–Mn oxides and bound to organic matter fractions) may be conditionally released to water column through any disturbance, such as hydrological, physical, chemical and biological activities (Arain et al., 2008; Cuong and Obbard, 2006; Tessier et al., 1979). Thus, sediments are a potential source of metal contamination for surrounding water column.

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The Three Gorges Reservoir (TGR) area plays a key role in economic development, national drinking water security and sustainability of ecosystem (Plateau, 2006). However, the alteration of hydrologic conditions of TGR by impoundment has caused a number of environmental changes (Zhou et al., 2014). The metals contamination before impoundment of 2008 in the TGR sediments has been in focus for decades, while few investigations have been conducted after water impoundment (Zhao et al., 2008; Wang et al., 2011; Yang et al., 2009). Additionally, limited data is available for comparisons between different impoundment periods. To fill these knowledge gaps, it is necessary to find out the detailed information, such as the spatial variation, bioavailability and health risk assessment of trace metals in sediments collected from TGR during different impoundment periods.

The primary objectives of this study were therefore to determine the concentrations and spatial distributions of metals in sediment cores collected from Fengjie area of TGR at summer and winter after impoundment. Additionally, the bioavailability of trace metals in sediment cores was assessed by differentiating their chemical forms. And enrichment factors (EF), risk index (RI) and modified risk assessment code (mRAC) were performed to evaluate the contamination level and ecological risk.

2. Material and methods

2.1. Samples collection

The Three Gorges Dam (TGD) on the Yangtze River is the largest hydroelectric project ever built in the world. The reservoir waters and their fringe areas cover an overall area of 58,000 km² and including totally 20 districts and counties (cities) (Plateau, 2006). The TGR is impounded to the maximum level of 175 m in winter for energy generation and subsequently emptied to the base level of 145 m in summer for flood control (Tang et al., 2014). In this study, twelve sediment cores in July 2014 (summer) and six sediment cores in December 2014 (winter) were collected in a typical area (Fengjie) of TGR. The sampling sites were described within the zone (Fig. 1). One sampling site was selected from the main river channel (CJ) and eleven sampling sites were on the upstream, midstream and downstream of three tributaries (Zhuyi River (ZY), Meixi River (MX) and Caotang River (CT)). At each sampling site, surface samples were taken using core sampler (K-B type, Wildco, USA) near the middle of the flow of the stream. The sediment cores were sliced at 5 cm thick intervals with slice equipment whose surface is coated with a Polytetrafluoroethylene. The sediment samples were placed into plastic bags, sealed into clean polyethylene bags and treated immediately after returning to the laboratory. The sediment samples were wet sieved through an acid-cleaned 63 μm mesh nylon sieve to obtain the chemically active materials (Gao et al., 2015), dried at 40 °C to constant weight, and ground in an agate mortar to ensure homogeneity.

2.2. Analysis of metal concentration

Total metal concentrations in sediments were measured using an established method (Liu et al., 1996). Briefly, a mass of 40 mg of dry sample was weighed and dissolved into 10 mL Teflon bombs. Approximately 4 mL concentrated HNO₃ + 0.4 mL concentrated H₂O₂ were added to the samples and these were put on a hot plate for 24 h. This step was to remove organic materials from sediment samples. The samples were then dried at 120 °C. The residue was dissolved in 1.5 mL HNO₃ + 1.5 mL HF of sample. After 20 min ultrasonic treatment, these samples were taken into a sealed bomb and placed in an oven at 190 °C for 48 h. This procedure resulted in clear solutions for sediments. After evaporation at 120 °C, samples

were dissolved in 1% HNO₃ (v/v). Concentrations of Cr, Ni, Cu, Zn, As, Cd, Pb were measured by inductively coupled plasma-mass spectrometry (ICP-MS, Perkin Elmer Elan DRC-e). Hg was measured using a Direct Mercury Analyzer (Milestone DMA-80). The quality controls for the strong acid digestion method included reagent blanks, duplicate samples, and standard reference materials. The QA/QC results show no sign of contamination in all the analyses. The accuracy of the analytical procedures employed for the analysis of the trace metals in sediments was checked using the certified reference material of China stream sediment (GSD-10, GBW-07310), obtaining recovery of 109.1% for Cr, 105.7% for Ni, 104.3% for Cu, 101.1% for Zn, 106.7% for As, 105.4% for Cd, 99.00% for Pb and 96.43% for Hg (Table S1).

2.3. Sequential extraction procedure of metals

Sequential extraction was performed using a three-stage modified procedure recommended by Community Bureau of Reference (BCR) plus the residual fraction (Pueyo et al., 2008). During the extraction, metals (Cr, Ni, Cu, Zn, Cd and Pb) were classified into four fractions: acid-soluble/exchangeable fraction (F1), reducible fraction (F2), oxidizable fraction (F3) and residual fraction (F4). The accuracy of the analytical procedures employed for the analysis of metals in sediments was checked using the BCR Reference Material (BCR 701). The recovery values were 139.23% for F1, 106.16% for F2 and 81.48% for F3.

2.4. Estimation of sediment contamination

2.4.1. Sediment quality assessment guidelines (SQGs)

Sediment quality assessment guidelines (SQGs) are very useful in terms of revealing sediment contamination by comparing the sediment concentration of metals with the corresponding quality guideline (Long et al., 1995; MacDonald et al., 2000; Smith et al., 1996). According to MacDonald et al. (2000), the reliability of the probable effect level (PEL), lowest effect level (LEL), threshold effect level (TEL), minimal effect threshold (MET), effect range low (ERL), effect range median (ERM), threshold effect concentration (TEC) and probable effect concentration (PEC) for assessing sediment quality conditions is determined based on their predictive ability (Table 1).

2.4.2. Enrichment factors (EF)

Enrichment factors (EF) can be utilized as a reference to predict the extent of metal contamination (Selvaraj et al., 2004). The EF values for all the metals were calculated according to the following formula:

$$EF = \frac{(X/Y)_{\text{sample}}}{(X/Y)_{\text{crust}}} \quad (1)$$

where $(X/Y)_{\text{sample}}$ is the ratio of element (X) to normalizer element (Y) for the studied samples and $(X/Y)_{\text{crust}}$ is the ratio of element (X) to normalizer element (Y) for earth crust. In this study, Co displayed significant relationships with almost all metals (Table 2). Moreover, the mean concentration (18.04 mg/kg) of Co in the studied area was almost equal to local background value (17.00 mg/kg). Matthai and Birch (2001) have suggested that Co is a suitable normalizing element in the area for which there is a demonstrated absence of substantial anthropogenic origin of this trace metal. Hence, Co was employed as the reference element. If an EF value is between 0.5 and 1.5, it suggests that metals may be entirely from crustal materials or natural weathering processes (Zhang and Liu, 2002). Samples having an enrichment factor of >1.5 are considered indicative of human influence and an EF between 1.5–3, 3–5, 5–10 and >10 is considered evidence of minor, moderate, severe, and very severe enrichment, respectively (Birch and Olmos, 2008).

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