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Research papers

Thallium pollution in sediments response to consecutive water seasons in Three Gorges Reservoir using geochemical baseline concentrations

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ABSTRACT

The Three Gorges Reservoir (TGR) is the world's largest hydrological project. However, the response of thallium (Tl) in TGR sediments to reservoir operation remains highly uncertain. Here, we investigated the pollution characteristics of Tl in TGR sediments during five consecutive water seasons. Geochemical baseline concentrations (GBCs) were used to distinguish the anthropogenic input of Tl and to quantitatively estimate the Tl anthropogenic contribution in TGR sediments. The results showed that the average Tl concentration in TGR sediments was 0.72 ± 0.14 mg/kg (ranging from 0.26 to 1.07 mg/kg) during five consecutive water seasons. The average Tl content in tributary sediment was higher than that in mainstream sediment. In the mainstream, Tl in TGR sediments showed an accumulation tendency from the upstream to downstream. Additionally, the temporal variation revealed that the mean value of Tl in TGR sediments in the dry season was higher than that in the wet season during 2015–2016. Furthermore, the average GBCs of Tl (0.71 mg/kg) was higher than background values (BVs) in the Yangtze River sediments. Using the GBCs of different water seasons to calculate the anthropogenic input of Tl in the TGR, the average anthropogenic contribution was found to be approximately 6.43% (2.81%–9.12%). The assessment results by two different methods (geo-accumulation index and potential ecological risk index) used GBCs as the BVs, indicating that TGR sediments were at "uncontaminated" level and close to the "low ecological risk", respectively. Comparing to the GBC of Tl, the assessment of Tl pollution in TGR using BVs in the Yangtze River sediments may be over-evaluated. This study is beneficial for assessing Tl pollution in TGR sediments response to consecutive water seasons and estimating the anthropogenic contribution of Tl.

1. Introduction

Thallium (Tl) is a non-essential element found in trace amounts in the earth's crust with average concentrations of 0.1–1.7 mg/kg (Peter and Viraraghavan, 2005). Due to its acute and chronic toxicity, Tl is considered a priority toxic pollutant (Kazantzis, 2000; Peter and Viraraghavan, 2005). Emissions from anthropogenic industrial activities (e.g., coal combustion, and ferrous and non-ferrous mining/smelting) lead to the increase of Tl concentrations in the environment (Karbowska, 2016; Kazantzis, 2000; Peter and Viraraghavan, 2005). Therefore, in order to control environmental contamination by Tl, several governments have established water standards for Tl. The maximum contaminant level in drinking water is 0.1 μg/L in China (CNS, 2006), 0.8 μg/L in Canada (CCME, 2003), and 2 μg/L in the USA (USEPA, 1992). Compared with Tl in water, limited research has been conducted on Tl in sediments, and the relevant sediments standards are lacking. Sediments in water environments can act as sources and sinks of trace metals, and metals in the sediment can be released into the water body. Hence, a better understanding of the anthropogenic impacts and geochemical behavior of Tl in sediments would be worthwhile.

The Three Gorges Reservoir (TGR) is the world's largest hydrological project. It sustains the environment in the middle and lower reaches of the Yangtze River and is important to national drinking water security. After the completion of the project, the water level fluctuates between 145 and 175 m. The TGR operation follows characteristics of anti-seasonal water level variations, with high water levels in winter (dry season) and low levels in summer (wet season) (Bing et al., 2016). In recent years, water contamination issues regarding trace metals in TGR sediments have been increasingly highlighted. In

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comparison with the common monitored trace metals (e.g., Cu, Zn, Pb and Cd) (Han et al., 2015; Tang et al., 2014; Wang et al., 2012), limited research has been focused on the pollution status of Tl in TGR sediments. Han et al. (2015) and Jia et al. (2014) reported on the concentrations and distributions of Tl in sediments of several tributaries of the TGR. Another study investigated Tl concentrations in sediments of the entire TGR and found that the mean Tl concentration was 0.58 mg/kg in the dry season (Gao et al., 2014). However, the response of Tl environmental behavior in TGR sediments to consecutive water seasons remains uncertain. In fact, reservoir operation could affect the migration and transformation of trace metals in TGR sediments (Bing et al., 2016; Holbach et al., 2014).

The geo-accumulation index (I_{geo}), enrichment factor (EF), and potential ecological risk parameter (E_r) have often been used to quantitatively assess the trace metal contamination in sediments (Gao et al., 2015; Han et al., 2015). The selection of background values (BVs) is important for the pollution assessment by these methods. Most previous studies selected BVs in soils and sediments in China or the Yangtze River as background reference values in the TGR. However, the BVs of trace metals for soils and sediments mainly depend on local geological properties, such as the crustal composition (Jiang et al., 2013). Multiple BVs are required across a large area, as the use of a single regional BV has the disadvantage of disregarding the variability of trace metal contents due to geological conditions (Covelli and Fontolan, 1997; Fukue et al., 2006). Consequently, previous studies may not have provided accurate assessments of the pollution status in TGR sediments. Furthermore, it is necessary to select suitable BVs for assessing trace metal pollution in the TGR. Geochemical baseline concentrations (GBCs) are defined as the natural levels of trace metals in soils (Bech et al., 2005; Galán et al., 2008; Tian et al., 2017) and sediments (Liu et al., 2013). Studies have been conducted to determine the GBCs of trace metals in different regions, and these have been successfully applied to the assessment of trace metal pollution (Martin et al., 2016; Tian et al., 2017). Therefore, GBCs should be established for TGR; in particular, this could lead to more suitable assessments for Tl in TGR sediments.

This study was the attempt to use the GBCs to assess Tl contamination in the entire TGR area during the consecutive water seasons. The objectives of this study were (1) to clarify the spatial and temporal variation of Tl in TGR sediments for five consecutive water seasons, (2) to establish the GBCs for Tl in TGR sediments, and (3) to evaluate the pollution status of Tl in TGR sediments using GBCs.

2. Materials and methods

2.1. Study area and sample collection

The TGR is located in the main stem of the Yangtze River in China, and it covers an area of 58,000 km^2, including a water area of 1080 km^2 (Stone, 2008; Zhao et al., 2017). The backwater of the TGR is approximately 660 km long and forms a typical river-type reservoir from the Three Gorges Dam (TGD) to Jiangjin City in Chongqing Municipality (Bing et al., 2016). The TGR area is located in western Hubei Province (Yichang City) to the east of Chongqing City (28°32′–31°44′ N and 105°44′–111°39′ E) (Fig. 1). The climate in the TGR area is primarily of the humid subtropical monsoonal type. The mean annual temperature ranges from 16.5 to 19.0 °C, and the annual mean precipitation is approximately 1000–1200 mm, with 80% falling during April-October period (Bao et al., 2015; Ye et al., 2013). The water operation of this project is of the anti-season type, characterized by a high water level (175 m) in winter (dry season) and a low water level (145 m) in summer (wet season).

A total of 240 sediment samples were collected during five consecutive water seasons (including 2015-wet season, 2015-dry season, 2016-wet season, 2016-dry season and 2017-wet season, respectively). For each water season, 48 sampling sites were located in the mainstream from upstream to downstream, and tributaries of TGR. Thirty-seven sediment samples (S1-S37) were collected from the mainstream, namely from the upstream to the front of the TGR dam. Eleven sediment samples (T1-T11) were collected from major tributaries (Fig. 1). In this study, the upstream area of the mainstream was defined ranging from Jiangbei District to Fengdu Town (S37-S32) (Guo et al., 2016). The mid-stream area was defined ranging from Fengdu Town to Fengjie Town (S32-S17), and the downstream area was demarcated as that from Fengjie Town to Yichang City (S17-S1) (Guo et al., 2016). The tributaries of the study area included the Wujiang, Qutang, Pengxi, Modao, Meixi, Daning, Yandu, Qinggan, and Xiangxi Rivers (Fig. 1). Sediment samples were taken from the middle of the stream at each sampling site. Approximately 1 kg of sediment was collected in clean polyethylene bags and immediately transported to the laboratory. The sediment samples were then freeze dried and gently crushed and ground in an agate mortar. The crushed samples were then passed through an acid-cleaned 0.25 mm nylon sieve for further analysis.

2.2. Analysis of metal concentrations

A strong acid digestion method was used to analyze the total Tl and Li concentrations in the TGR sediments. The details of this method have been provided in a previous report (Xu et al., 2016). Tl concentrations were analyzed using an inductively coupled plasma mass spectrometer (PerkinElmer Elan DRC-e, USA). The accuracy of the analytical procedures was controlled by certified reference materials, namely GSD-10 (GBW07310) and GSD-1a (GBW07301a), which were purchased from the Chinese Institute of Geophysical and Geochemical Exploration. These two sediment standard materials were analyzed using the same procedure as those for the TGR sediments, and analysis results agreed with the certified values (Table S1); the recovery of the sediment standard materials ranged from 95.2 to 97.0%. The limit of analysis detection in this study was 0.05 mg/kg for Tl.

2.3. Assessment of sediment pollution

In this study, the GBCs, I_{geo}, and E_r were employed to evaluate the Tl contamination in the TGR sediments.

2.3.1. Geochemical baseline concentration (GBC) of Tl in TGR sediments

The calculation of the GBC requires a normalization method based on a linear regression equation obtained from the correlation of concentrations between the element in question (in this case, Tl), and a conservative reference element (Teng and Ni, 2007). This study used lithium (Li) as the conservative reference/normalizing element because it is the best representative to fine particle size of the sediments (Teng and Ni, 2007). The equation defining the geochemical baseline model is as follows:

$$C_m = aC_N + b \tag{1}$$

where C_m is the concentration of Tl (mg/kg), C_N is the concentration of Li (mg/kg), and a and b are the regression constants.

In the x-y scatterplot described by Eq. (1), data within the 95% confidence limit were characterized as naturally sourced. Data outside the 95% confidence limit were characterized as having anthropogenic sources (Teng and Ni, 2007). Data from the anthropogenic sources were then removed, and a linear regression was fitted to the remaining data from natural sources with new regression constants (c and d). We subsequently arrived at the following equation:

$$B_m = c\bar{C}_N + d \tag{2}$$

where B_m is the GBC of Tl (mg/kg) in the study area, $\bar{C}_N$ is the normalized average of Li (mg/kg), and c and d are the new regression constants. Using the normalized average Li concentration, the naturally sourced Tl concentration was obtained. This Tl value was defined as the