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Assessment of Cr pollution in tributary sediment cores in the Three Gorges Reservoir combining geochemical baseline and in situ DGT

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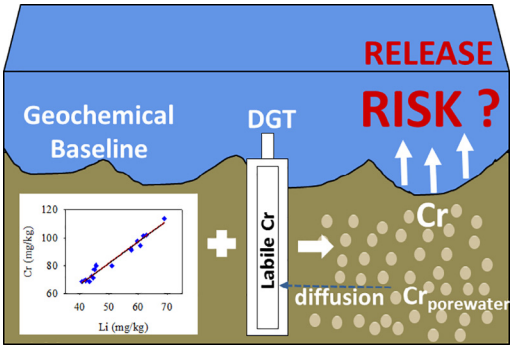
HIGHLIGHTS

- This study assessed Cr pollution by combining geochemical baseline and DGT.
- The carcinogenic risk from Cr in the TGR water did not exceed US EPA recommendations.
- The average anthropogenic contribution rate for Cr in TGR sediment was 6.03%.
- Cr was only being released in the upstream of Meixi River sediments.

GRAPHICAL ABSTRACT



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ABSTRACT

The mobility and transfer of trace metals in sediments are vital to understanding trace metals environmental behavior in water environment. However, as a predominant aquatic carcinogen, an effective method for assessing the release and deposition for Cr at the sediment-water interface (SWI) is still not clearly understood. Here we established a comprehensive methodology to evaluate the release risk of Cr at the SWI combining regional geochemical baseline (RGB) and diffusive gradients in thin films (DGT). Sediment cores and water samples were collected in the two tributaries and mainstream of the Three Gorges Reservoir, which is the world's largest man-made hydroelectric station. Results showed that the calculated Cr carcinogenic risks in surface water did not exceed US EPA maximum recommended level. The RGB of Cr (85.53 ± 14.44 mg/kg) were calculated and the differentials between Cr concentration and RGB in surface sediments showed the average anthropogenic contribution rate was 6.03% and the upstream of Meixi River (MX-S) and mainstream were influenced by anthropogenic activities. The net diffusive flux using DGT showed that Cr in the midstream of Caotang River and MX had the potential to move upwards into the overlying water. Furthermore, combining the results of differential (Cr concentration vs. RGB) and the net flux, MX-S was the only site with a risk of Cr release. To the best of our knowledge, this study is the first attempt to combine RGB and DGT to scientifically assess metal release at SWI and provided a new perspective to comprehensively assess metal pollution in water environment.

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1. Introduction

Chromium (Cr) is a common element in the Earth's crust (Aboul Dahab and Al-Madfa, 1997), but its presence in the environment can also result from its wide use in various industries such as leather tanning, metal finishing and smelting (Burbridge et al., 2012; Lee et al., 2008). The Cr pollution state and mobility in sediment is imperative to understand its geochemical behavior in aquatic environments, especially the release at the sediment-water interface (SWI). Traditional assessment methods in metal pollution assessments mainly consist of pollution indexes such as the contamination factor, geoaccumulation index (I_{geo}) and enrichment factors (Bing et al., 2016; Cheng et al., 2017; Wei et al., 2016); however, assessment results vary according to reference background levels. Using the natural background has the following limitations: (1) it does not consider the natural variation of metals in the environment (Covelli and Fontolan, 1997; Daskalakis and O'Connor, 1995); and a "natural background" that strictly refers to the pristine geochemical composition does not exist at present (Karim et al., 2015). (2) lack of background values in studied areas due to the extensity of the region. The geochemical baseline is defined as the level of natural variation in metals in the surficial environment (Galán et al., 2008; Tian et al., 2017), and it includes both the geogenic natural concentrations and anthropogenic contribution. The geochemical baseline can distinguish the natural concentration and anthropogenically influenced concentration for each sample (Karim et al., 2015; Matschullat et al., 2000; Teng et al., 2009; Zhang et al., 2014b). The geochemical baseline has been applied as the reference background when determining the pollution status of metals in the sediment (Lin et al., 2012; Tian et al., 2017). However, these assessment methods have all been based on the total metal concentrations in the sediment. In reality, metal toxicity in the sediment depends on mobility and bioavailability. Moreover, most studies on metal mobilization are based on ex situ sequential extraction, whereas in situ methods are not widely used. In recent years, the in situ diffusive gradients in thin films (DGT) technique has been used to investigate the remobilization of metals in sediment (Amato et al., 2014; Gao et al., 2017; Wang et al., 2017b). DGT can reflect the dynamic process during deployment rather than the equilibrium concentration (Sun et al., 2014; Zhang et al., 2014a). Using Fick's first law model, the apparent net flux at the SWI can be quantitatively calculated, which can judge the diffusive trend of the pollutants (Ding et al., 2015; Han et al., 2015). Compared with other classical methods, this method can in situ calculate bilateral diffusive flux at the SWI (Ni et al., 2017; Mustajärvi et al., 2017).

Water quality directly affects the socioeconomic development, national drinking water safety, and human health. The Three Gorges Reservoir (TGR) is the world's largest man-made hydroelectric station (Shen and Xie, 2004) and the largest drinking water reservoir in China. After impoundment of the TGR, hydrological and environmental conditions change, accumulated contaminants in sediments from agriculture and waste water discharge can be released back into the watershed (Cheng et al., 2017). As cumulative and non-degradable contaminants, metal pollution poses significant ecological risks and has provoked particular concern in the TGR (Yang et al., 2012; Bing et al., 2016; Cheng et al., 2017). Cr is regularly monitored, and previous studies of Cr in the TGR sediment were mainly based on traditional assessment methods (Wang et al., 2017a; Wei et al., 2016; Zhao et al., 2017a). However, Cr mobility and transfer at the SWI has not been reported.

In this study, the geochemical baseline and DGT technique were employed to investigate the mobility and release of Cr in TGR sediment. The primary objectives are to (1) assess the carcinogenic risk of Cr in the surface water of the TGR; (2) establish the regional geochemical baseline (RGB) and evaluate the contamination level based on the RGB and background values (BV) in the Yangtze and Cr concentration in the crust; and (3) establish a method to investigate the release of Cr at the SWI combining the RGB and DGT.

2. Materials and methods

2.1. Sample preparation

Seven undisturbed sediment cores were collected using a core sampler (K-B type, Wildco, USA) near the middle of the stream in different reaches of two tributaries (the Meixi River (MX) and Caotang River (CT)) and the Yangtze River (CJ) in the TGR in July 2015. Two surface water samples were collected concurrently at each site. Sampling site locations are shown in Fig. 1. Water samples were collected in polymer polyethylene bottles, labeled, and transported to the laboratory. The samples were filtered through 0.45-μm Millipore filter membranes and acidified to pH < 2 by adding ultrapure concentrated nitric acid. All samples were stored at 4 °C until analysis. The sediment cores were protected with nitrogen gas and sealed to prevent interference by oxygen in the air. Then, the sediment cores were immediately transported to the laboratory and stabilized for at least 24 h at 25 °C. During transportation, cores were maintained upright to minimize disturbance and covered with two layers of black bags to block light.

2.2. Human carcinogenic risk assessment methodology

Cr is regarded as a chemical carcinogen in the health risk assessment. The population was divided into four parts, infants (3–6 years old), teenagers (7–19 years old), adults (20–59 years old) and the elderly (60–69 years old). Human carcinogenic risk was assessed using US EPA (1989) methodology. The carcinogenic risk is calculated using the following equations:

$$R = [1 - \exp(-D \times q)]/Y \quad (1)$$

$$D = Q_i \times C/Wi \quad (2)$$

where R is the carcinogenic risk to an individual over their lifetime posed by Cr in drinking water (a^{-1}); D is the daily intake of Cr from drinking water (mg/(kg·day); q is the carcinogenic factor of Cr (41 (kg·d)/mg) (Sun et al., 2009); Y is the average lifespan (75.76 years) (NBS, 2012); C is the concentration of Cr in water (mg/L); Q_i is the ingestion rate of water (1.0, 2.0, 3.2, and 2.7 L/d for infants, teenagers, adults and elderly, respectively) (Duan et al., 2010); and W_i refers to body weight (18.9, 44.4, 63.1, and 62.2 kg for infants, teenagers, adults and elderly, respectively) (GAS, 2011).

Sensitivity analysis based on a Monte-Carlo simulation was introduced to estimate the relative contribution of related random variables to the risk result. Four characteristic parameters were selected: Cr concentration (C), body weight (W), average lifespan (Y), and water ingestion rate (Q). Given that infants are the most susceptible to adverse health risks, the R_{Cr} for infants were selected for the simulation. A positive value for sensitivity indicates positive correlations between the variable and health risk, and the larger the value is, the greater its influence is.

2.3. DGT deployment and measurement

The DGT probes (LSPM) were purchased from DGT Research Ltd., Lancaster, UK. For each sediment core, a DGT probe was deployed for 24 h in the dark at 25 ± 0.5 °C. Before deployment, the overlying water was extracted and retained at a height of approximately 3 cm. A mark was made on the side at 3 cm below the top of the window in each DGT probe. Then, the deoxygenated DGT probes were gently and smoothly pushed into the sediment core until the mark was in line with the SWI. After retrieval, the probes were rinsed with Milli-Q water. Then, each resin gel was segmented into 5 mm (vertical) × 6 mm (horizontal) slices. Each gel was eluted with 0.3 mL of 1 M HNO_3 for 24 h. The eluted solutions were then diluted tenfold with Milli-Q water and measured using inductively coupled plasma-mass