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Deposition behavior, risk assessment and source identification of heavy metals in reservoir sediments of Northeast China

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A B S T R A C T

Sediment cores from five reservoirs, located in the Liaoning and Jilin Provinces in Northeast China, were collected to investigate the accumulation and potential toxicity of heavy metals (Fe, Mn, Cu, Cd, Pb, Zn, and Cr) during a sampling campaign in February, 2015. The results showed that all the detected metals accumulated significantly, especially Cd, compared to their respective background values. Among these reservoirs, Biliuhe Reservoir had markedly increasing trends for organic matter and all the metals, among which Mn was elevated by 280% to 3411 mg/kg in a core of only 18 cm in depth. Xinlicheng Reservoir was characterized by heavy siltation and varying metal distribution due to its regular geometric features and pulsed flood events. The Enrichment factor (EF) and geo-accumulation index (I_{geo}) indicated Cd was strongly enriched by anthropogenic inputs, with the values of EF and I_{geo} greater than 8 and 3, respectively. The toxicity assessment calculated using consensus-based sediment quality guidelines (SQGs) implied the whole cores of Tanghe and Dahuofang and the upper cores of Biliuhe, Xinlicheng and Fengman exhibited toxicity to sediment-dwelling organisms. Cr contributed more to $Q_{m\text{-}PEC}$ than the other heavy metals, because only Cr exceeded the probable effect concentration (PEC) despite its low enrichment. According to the results of correlation analysis (CA) and principal components analysis (PCA), mining industries and agricultural activities within the basin were the main anthropogenic pollution sources for these heavy metals.

1. Introduction

Heavy metals in the ecosystem resulting from natural and anthropogenic inputs may pose considerable threats to the aquatic environment. These metals harm human health through various pathways, due to their bioaccumulation, acute and chronic toxicity and persistence (Mclaughlin et al., 1999; MacDonald et al., 2000; Salmanighabeshi et al., 2015). In aquatic systems, most of the metals are accumulated and sequestered in the particulate phase in sediments; as long as the physiochemical parameters of the sediments or overlying water stay stable (Martínez-Santos et al., 2015; Zhang et al., 2016). Once the conditions change under certain circumstances, such as during remedial dredging, flood events and benthic bioturbation, sediments tend to release these metals into the aquatic media (Wang et al., 2012; Peraza-castro et al., 2016). Reservoirs play an important role worldwide in preventing floods and retaining water for industrial and agricultural activities, generating hydroelectric power, drinking and other activities. In addition to these benefits, the intensified accumulation of sediments and pollutants, especially in the vicinity of the dam, caused by discontinuities in water and sediment transport puts pressure on water quality and aquatic organisms (Morris and Fan, 1998; Frémion et al., 2016). Accumulated sediments physically reduce the water storage capacity, cause ecological and environmental problems and gradually eliminate the benefits that rely on reservoir water (Devi et al., 2008; Palazón and Navas, 2014). In addition to the disturbances mentioned above, reservoir management and even temperature stratification can also influence physiochemical conditions (e.g., pH, dissolved oxygen and redox potential) and lead to heavy metal releases at the sediment water interface (Graham et al., 2012; Frémion et al., 2016). For drinking water reservoirs, released metals in the soluble phase not only increase the amounts of their bioavailable forms but also pose problems to water treatment plants (Bryant et al., 2011).

Northeast China, primarily the three provinces of Heilongjiang, Jilin and Liaoning, was once the cradle of heavy industry during the initial stages of China's modernization due to its plentiful resources (Lin et al., 2012). It has also been a major area of grain production owing to its fertile black soil with an estimated 7.4 million hectares of cropland in 1982 (Zhang et al., 2007). Decades of traditional industrial and

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agricultural activities have led to the pollution of various heavy metals due to fertilizers, manures and chemicals (including fungicides, pesticides and herbicides) being applied to the agriculture. These are also regarded as general anthropogenic inputs, as well as the irrigation (He et al., 2005; Wu et al., 2012). Reports have shown that soil erosion in Northeast China has become remarkable as land reclamation has been active since the 1950s, and inevitably, agrochemicals including heavy metals have entered surface water systems and accumulated in sediments (Yang et al., 2003; Xu et al., 2010). Moreover, atmospheric deposition as a consequence of fine particles enriched with anthropogenic heavy metals indirectly serves as an additional input to the natural environment (Clark et al., 2014). Apart from vehicle exhaust and industrial emissions, coal combustion alone makes northern China more prone to such deposition contamination, particularly from Pb, because coal combustion dominates the energy generation in this region. Additionally, the prevailing wind, together with contaminated particles, is from Mainland China to the north, which aggregates pollution from atmospheric deposition (Yao et al., 2009; Cheng and Hu, 2010). Previous studies in this region have focused on heavy metals in urban and agricultural soils, street dust, river sediments and other aspects (Zheng et al., 2008, 2010; Chai et al., 2015); however, heavy metal investigation in reservoir sediments is rarely studied.

In the present work, we aim to i) reveal the vertical profiles of trace metals in five sediment cores near each reservoir dam in Northeast China; ii) assess the degree of the metal enrichment and potential sediment toxicity; iii) identify the sources of heavy metals through multi-statistical analyses.

2. Materials and methods

2.1. Study area and sampling

The five reservoirs of interest, three in Liaoning Province and the other two in Jilin Province, are listed as nationally important water sources for the peripheral cities. Geographically, Biliuhe (BLH), Tanghe (TH) and Dahuofang (DHF) Reservoirs are almost evenly distributed from south to north in the eastern part of Liaoning Province (Fig. 1). Xinlincheng (XLC) and Fengman (FM) Reservoirs (also known as Songhua Lake) are close to the cities of Changchun and Jilin, respectively. All of these reservoirs have been in operation for decades, from 30 years for the BLH Reservoir to 74 years for the FM Reservoir (Table 1). In addition, there are many agricultural and mining activities in the upstream drainage basins of these reservoirs. The Hongtoushan Group is the largest copper mining enterprise in northeast China and is responsible for the accumulation of heavy metals in the Hun River, which is the main inflow of the DHF Reservoir (Zheng et al., 2016). Various mining enterprises along the Second Songhua River and Huifa River, which are main inflows of the FM Reservoir, produce Au, Cu, Ni and other metals. A report has shown that the gold field in this area has led to the contamination of the upstream sediments and aquatic organisms (Zhu et al., 2016). With regard to the BLH Reservoir, several gold mining companies were distributed along the upper reach and mining tailings reaching up to ten million tons were piled only 200 m from the Biliu River, the main river feeding this reservoir. Locations of the reservoirs, sampling sites and mining areas are shown in Fig. 1.

Given the scarce and unevenly distributed water resources in Northeast China, inter-basin water diversion projects between reservoirs and rivers are performed to balance water supply and demand. Through such projects, the DHF Reservoir alone has provided drinking water for nine cities and benefits more than five million people so far (Zheng et al., 2016). The TH Reservoir also diverts water from Xi and Lan Rivers, which belong to another reservoir basin. Soil erosion, cross contamination and ecological impacts are urgent issues facing the whole process of water diversion.

The sampling campaign was carried out in February 2015 when the five reservoirs were all covered by ice. Ten sediment cores were collected from the vicinity of the reservoir dams using a gravity corer made of stainless steel (6 cm in diameter of the inner PVC tube). The ten collected cores included one in the BLH Reservoir, three in the XLC Reservoir, and two in the TH, DHF and FM Reservoirs. The cores were transported to the laboratory and stored below 0 °C. Before freeze-drying, all the sediment cores were sliced at 1 cm intervals for the upper

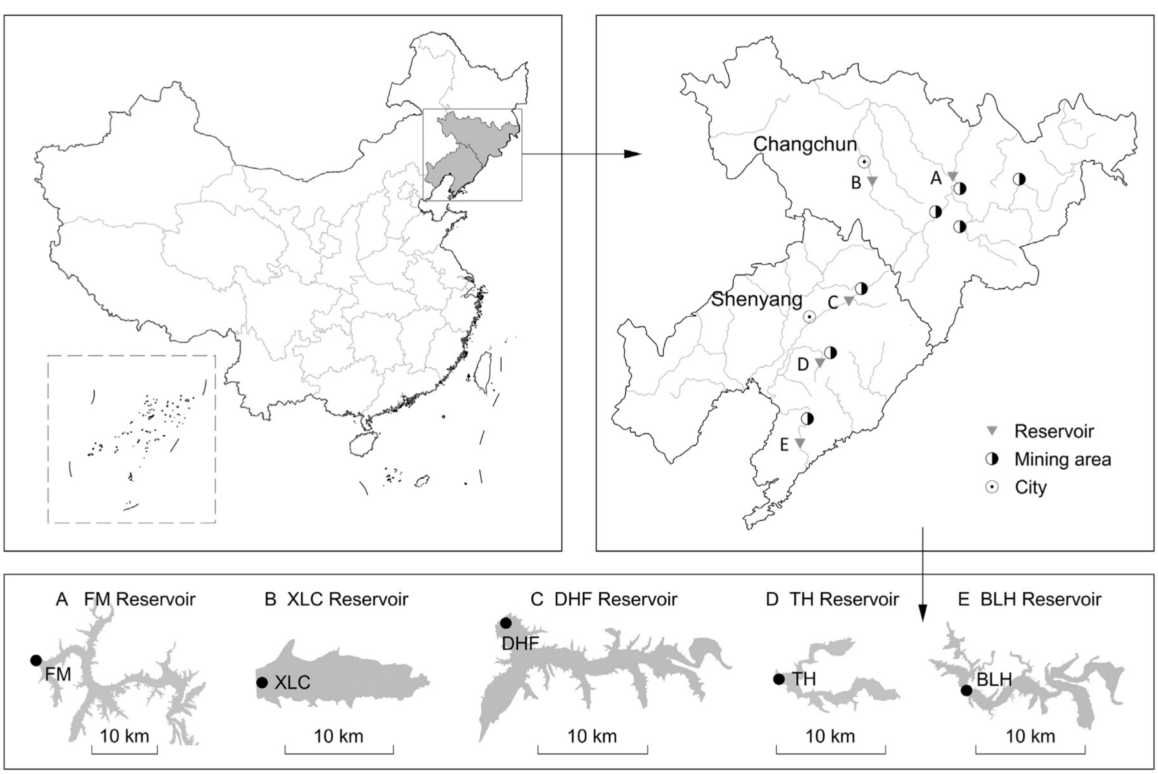

Fig. 1. Location of the reservoirs with sampling sites (orientation is rotated for the five reservoirs and the FM Reservoir is clipped).