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Pollution characteristics and source identification of trace metals in riparian soils of Miyun Reservoir, China

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ABSTRACT

The South-to-North Water Diversion Project, one of China's largest water diversion projects, has aroused widespread concerns about its potential ecological impacts, especially the potential release of trace metals from shoreline soils into Miyun Reservoir (MYR). Here, riparian soil samples from three elevations and four types of land use were collected. Soil particle size distributions, contents and chemical fractionations of trace metals and lead (Pb) isotopic compositions were analyzed. Results showed that soil texture was basically similar in four types of land use, being mainly composed of sand, with minor portions of clay and silt, while recreational land contained more abundant chromium (Cr), copper (Cu), zinc (Zn) and cadmium (Cd), suggesting a possible anthropogenic source for this soil pollution. The potential ecological risk assessment revealed considerable contamination of recreational land, with Cd being the predominant contaminant. Chemical fractionations showed that Cu, arsenic (As), Pb and Cd had potential release risks. Additionally, the $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ values of soils were similar to those of coal combustion. By combining principal component analysis (PCA) with Pb isotopic results, coal combustion was identified as the major anthropogenic source of Zn, Cr, Cu, Cd and Pb. Moreover, isotope ratios of Pb fell in the scope of aerosols, indicating that atmospheric deposition may be the primary input pathway of anthropogenic Zn, Cr, Cu, Cd and Pb. Therefore, controlling coal combustion should be a priority to reduce effectively the introduction of additional Zn, Cu, Cd, and Pb to the area in the future.

1. Introduction

With rapid urbanization and industrialization, large amounts of trace metals (e.g., As, Cd, Cr, Cu and Pb) from anthropogenic sources are discharged into the soil environment through various pathways, including vehicle emissions, chemical production, coal combustion, municipal solid waste, and sedimentation of dust and suspended atmospheric pollutants (Rainbow and Luoma, 2011). Furthermore, many trace metals could be bio-accumulated by soil organisms and even biomagnified through the food chains, thereby posing potential harmful effects to human health and ecosystem (Jiang et al., 2017; Li and Zhang, 2010). Reports have indicated that chronic exposure to Cd could cause diseases such as lung cancer, prostatic proliferative lesions, bone fracturing, kidney dysfunction and hypertension (Waalkes, 2000). Exposure to Pb could cause plumbism, anemia, nephropathy and gastrointestinal colic, and could affect the central nervous system (Żukowska and Biziuk, 2008). It is clear that an improved understanding of trace metal contamination in soils is crucial to food security, public health, and the sustainable development of the ecosystem.

Beijing, the capital of China, is the political, economic, and cultural center of the country. Because the Miyun Reservoir (MYR) is the unique source of drinking water for Beijing city, the water quality of this reservoir has a profound effect on the safety of water supply to the city. To relieve the pressure on the water supply to the arid northern cities of China, the government launched the South-to-North Water Diversion Project (SNWDP) at an estimated cost of 500 billion yuan (US$62.5 billion). This project is designed to divert annually 44.8 km^3 of water from the Yangtze River to northern China (Stone and Jia, 2006). After water impoundment via central route of SNWDP, the operational water level of the MYR is estimated to rise from 130 m to approximately 150 m, leading to the inundation of the surrounding riparian soils at elevations between 130 and 150 m. Upon inundation, soils will be subject to physical, biological and chemical changes, as the transition from a terrestrial to an aquatic ecosystem takes place. Changes in the soil Eh and pH could alter the binding capacity of metal ions to organic matter, reactive iron (Fe) oxides/oxyhydroxides and clay minerals (Dmytriw et al., 1995). Accordingly, the metals associated with these phases may be remobilized and transported into the MYR water, where

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the released metals would probably become pollutants. This impact of water impoundment on the release risk of metals would be directly influenced by the concentration and speciation of metals in soils located at elevations between 130 and 150 m. However, few studies have been published on the metal contamination levels in MYR soils (Chen et al., 2016a, 2016b; Han et al., 2016). A search on Web of Science using "metals/elements; Miyun Reservoir" as search phrases only returns 29 records since 1900. Therefore, the systematical investigation on the contamination status and speciation of metals in these soils is a matter of urgency.

In view of this analysis, the aims of the present study were 1) to study the concentrations and spatial distributions of Cd, Cr, Cu, Pb, As, Hg, and Zn at different elevations (130–145 m) in the riparian soils around the MYR, relevant to four types of land use (i.e., forestland, grassland, wasteland, and recreational land); 2) to determine the speciation of these metals using the modified Community Bureau of Reference (BCR) sequential extraction procedure; 3) to assess the pollution status by employing comprehensive potential ecological risk methods (contamination factor (CF), modified degree of contamination (mCd, and geoaccumulation factor (I_{geo})); 4) to identify the potential sources of metals by using Pearson correlation analysis, principal component analysis (PCA), and Pb isotopic analytical techniques.

2. Materials and methods

2.1. Study area

The maximum water volume of the MYR is 4.38 billion m^3. The maximum depth is 63.5 m (Gu et al., 2006), with the northern part of the reservoir being shallow and the southern part deeper. The upper basin of the reservoir, covering an area of 1,535,400 ha, includes parts of the Miyun and Huairou districts, and the Yanqing counties of Beijing and Guyuan, Chicheng, Chongli, Luanping, Huailai, Fengning, Xinglong, and Chengde, as well as the Zhangjiakou counties or cities in the Hebei Province. The study area has a continental monsoon climate, with semi-humid or semi-arid climatic characteristics. The annual mean temperature is approximately 8–10 °C. The mean annual rainfall is 400–600 mm, with highly variable monthly and annual rainfall.

2.2. Sampling collections

Prior to the water level rise of the reservoir (July 14, 2015), 42 topsoil samples (0–20 cm) were collected with a stainless steel shovel. The coordinates of the sample location were recorded with a GPS, and the sampling location are shown in Fig. 1. Four major types of land use and three elevations (130, 140 and 145 m) (based on the elevation system of Huanghai) were considered in this study and the sample number of each type of land use was determined according to its area, namely, (1) forestland, from which 18 samples were collected from soils located at elevations of 130, 140, and 145 m at the S1, S2, S3, S10, S11, and S14 sampling sites; (2) grassland, from which three samples were collected from the soils located at elevations of 130, 140, and 145 m at the S8 sampling site; (3) wasteland, from which 15 soil samples were collected from soils located at elevations of 130, 140, and 145 m at the S4–S7 and S9 sampling sites; and (4) recreational land, from which six soil samples were collected from the soils located at elevations of 130, 140, and 145 m on the left and right sides at the resort areas (sites S12 and S13). All the collected samples were kept in sealed Kraft bags to avoid contamination and were transported immediately to the laboratory. The soil samples were dried indoors at room temperature, and impurities, such as stones and tree leaves, were removed by sieving and hands. Subsequently, the samples were ground to pass through a 0.15 mm nylon sieve for the analysis.

2.3. Analytical methods

The total metal concentrations in the soils were measured by using an established method (Ying et al., 1996). Each sample was prepared by weighing 40 mg of dry soil into 10 mL Teflon bombs, after which approximately 4 mL 68% HNO_3 and 0.4 mL 40% H_2O_2 were added, and the samples were heated on a hot plate for 24 h. The samples were subsequently dried at 120 °C and the residue was dissolved in 1.5 mL HNO_3 and 1.5 mL hydrofluoric acid (HF) of sample. After 20 min of ultrasonic treatment, the samples were placed into a sealed bomb, which was placed in an oven at 190 °C for 48 h. This procedure resulted in clear solutions of the soils. After evaporation at 120 °C, the samples were dissolved in 1% HNO_3 (v:v). The concentrations of Cd, Cr, Cu, Pb, As and Zn were measured by inductively coupled plasma mass spectrometry (ICP-MS) with the Elan DRC-e (PerkinElmer, USA). A DMA-80 direct mercury analyzer (Milestone, Italy) was used to measure the mercury (Hg) content. Quality controls for the strong acid digestion method included reagent blanks, duplicate samples, and standard reference materials. The quality assurance and quality control (QA/QC) results showed no sign of contamination in any of the analyses. The accuracy of the analytical procedures was checked by using the certified reference material (CRMs) for soils (GSS-9, GBW07423). We obtained good agreement with the certified values (Table S1). The total organic carbon (TOC) in the decarbonated soils was analyzed using a Vario MACRO Cube CHNS analyzer (Elementar Analysensysteme, Germany). The granularity of each sample was analyzed using a particle size analyzer (Microtrac S3500, USA) with the ability to analyze a particle size range of 0.02–2000 µm. The particle size ranges used were < 2 µm

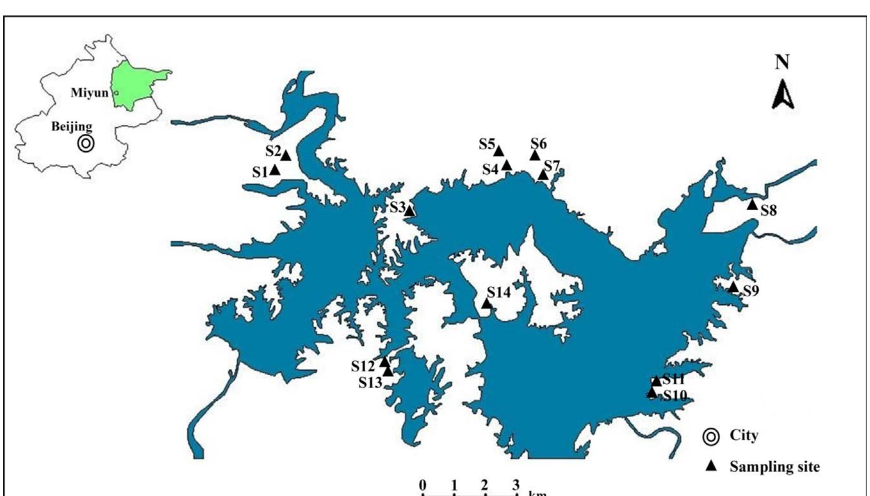

Fig. 1. Map of the sampling sites in the Miyun Reservoir (MYR).