Distribution of heavy metals and metalloids in bulk and particle size fractions of soils from coal-mine brownfield and implications on human health

Hongxia Li [a], Hongbing Ji [a, *], Chunjing Shi [a], Yang Gao [a], Yan Zhang [a], Xiangyu Xu [a], Huaijian Ding [a], Lei Tang [a, b], Yuxin Xing [b]

[a] Beijing Key Laboratory of Resource-oriented Treatment of Industrial Pollution, School of Energy and Environmental Engineering, University of Science and Technology Beijing, Beijing, 100083, China
[b] Beijing Geo-engineering Design and Research Institute, Beijing, 101500, China

HIGHLIGHTS

- Distribution and sources of heavy metals in coal mine brownfield were determined.
- Distribution characteristics of heavy metals in different soil particles were evaluated.
- Sources of heavy metals in various particle sizes were evaluated.
- Human health risk of coal mine brownfield was assessed.

GRAPHICAL ABSTRACT
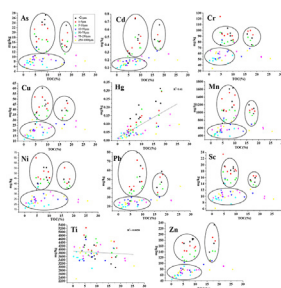
ARTICLE INFO

ABSTRACT

Heavy metals (HMs) and metalloids migrate into their surroundings, thus increasing environmental risks and threatening human health. Current studies on coal-mine brownfields, however, have not thoroughly investigated soil-associated HMs and metalloids produced by coal mining. Therefore, this study explored the spatial and particle fraction distribution and human health implications of HMs and metalloids. The soil-associated HMs and metalloids are Arsenic (As), Cadmium (Cd), Chromium (Cr), Copper (Cu), Mercury (Hg), Manganese (Mn), Nickel (Ni), Lead (Pb), Scandium (Sc), Titanium (Ti) and Zinc (Zn). Results showed that Cd, Cu, Pb, and Ni were enriched in bulk soils. Cadmium, Cu and Pb from anthropogenic source were mainly found at entrance roadsides and in sites closest to coal mines. HMs and metalloids primarily accumulated in fine fractions (<1, 1–5, and 5–10 μm). Moreover, HM and metalloid loadings substantially accumulated in the 75–250 μm and 250–1000 μm fractions. Most fine soil fractions showed moderate to strong potential ecological risks, whereas all the coarse particle fractions (50–75, 75–250, and 250–1000 μm) presented slight potential ecological risk. Exposure to soil-associated HMs and metalloids mainly occurred via ingestion. The total non-carcinogenic risks to children and adults fell below the safe level of 1, whereas the total carcinogenic risks to these individuals were higher than that of the maximum acceptable level set by the United States Environmental Protection Agency (USEPA, 1×10^{-4}). The total carcinogenic risk was mainly contributed by Cd and Ni through ingestion and dermal access. Therefore, hygiene and food security in areas should be emphasized.

© 2017 Elsevier Ltd. All rights reserved.

* Corresponding author. University of Science and Technology Beijing, Xueyuan Road No. 30, Haidian District, Beijing, 100083, China.
  E-mail address: ji.hongbing@hotmail.com (H. Ji).

1. Introduction

China is simultaneously the largest producer and consumer of coal worldwide (Li et al., 2015; Wang et al., 2014). Coal mining produces waste rocks that contain acid-forming metal sulfides, such as pyrite and marcasite (FeS_2) (Bulusu et al., 2007; Schippers et al., 2007). Moreover, the opencast and underground exploitation processes of coal mining produce mine dust that contains HMs and metalloids. Acid mine drainage (AMD) is a significant environmental problem associated with abandoned coal mines. During or after mining activities, sulfides are oxidized upon exposure to atmospheric O_2, thus generating acidic waters that contain high levels of dissolved $SO_4{}^{2-}$, Fe, HMs, and metalloids (Bhuiyan et al., 2010; Zhou et al., 2014; Qi et al., 2008; Yu et al., 2011). The release of indestructible and toxic HMs and metalloids to the surrounding environment exposes local residents to health hazards (Jiang et al., 2017; Khan et al., 2017).

Most research efforts have focused on the geochemical characteristics of HMs and metalloids in the ecological system of coal mining area or coal combustion environments (Ge et al., 2015; Pandey et al., 2015; Liu et al., 2016). Human health problems relevant to coal combustion, such as endemic arsenosis, fluorosis, selenosis and lung cancer, have also been reported (Dai et al., 2004, 2012). However, limited health risk assessment studies have been conducted for soil HM and metalloid pollution from coal-mine brownfield. Furthermore, most previous studies have concentrated on one or few coal mines. Hence, pollution assessment for an entire region of a coal mining area is urgently needed.

The Zhaitang Reservoir is located in Qingshui River, Beijing. The water quality of the reservoir directly affects the safety of drinking water of Beijing residents. Large-scale coal mines are located around the Qingshui River Basin. Although the majority of these mines are closed, their presence has noticeable effects, including changes in aquatic and terrestrial ecosystem health. However, studies on HMs and metalloids in this region are scarce. More information is required to clarify the pollution characteristics of soils in sensitive areas and the human health implications of soil-associated HMs and metalloids. Therefore, the objectives of this work are to (1) explore the spatial distribution and sources of HMs and metalloids in coal-mine brownfield soils; (2) determine the distribution and loadings of HMs and metalloids in different particle size fractions; and (3) assess the potential risk of HMs and metalloids to ecology and human health. This information can then help increase awareness of local stakeholders and authorities and urge them to take immediate measures.

2. Materials and methods

2.1. Study area

The study area (39°51′0″–40°6′0″N, 115°30′0″–116°0′30″E), Qingshui River Basin, is located west of Mentougou district, Beijing, China (Fig. 1). Additional samples for comparison were obtained from the banks of Yongding River after its junction with Qingshui River. Qingshui River is a tributary at the right bank of Yongding River. The water body is approximately 51.4-km-long with a watershed area of approximately 556.6 km^2. The terrain of the Qingshui Rive slopes downward from higher southwest to northeast with a relative topography height difference of approximately 1900 m. The Zhaitang Reservoir built at the middle reach of Qingshui River is the biggest water reservoir in Mentougou District and holds a capacity of 54.2×10^6 m^3 with a watershed area of approximately 354 km^2. The climate in the region is temperate semi-humid to semi-arid continental monsoon. The region experiences an annual precipitation of 571.6 mm.

2.2. Soil sampling and soil particle size fractionation

A total of 87 surface soil samples (0–20 cm in depth) were collected around coal mines in the study area (Fig. 1) during October 2015. Sample sets were obtained from Qingshui (3–84) and Yongding (85–89) River Basin. At each sampling site, a composite soil sample (approximately 1.0–1.5 kg) was obtained by thoroughly mixing 4 subsamples with a stainless steel blade and a plastic scoop. Sample locations were identified via GPS (Garmin GPS 72H). The environmental condition for each sample was recorded. Soil samples were then stored in polyethylene bags and transported to the laboratory. The samples were air-dried and sieved through a 1 mm mesh sieve to remove coarse debris for pH measurement. For total content analyses of HMs or metalloids and TOC (total organic carbon) in bulk soil, portions of each soil sample was further ground (<75 μm) with an agate mortar.

To obtain fractions with different particle sizes, 9 samples were selected from Qingshui River Basin and 1 sample was selected from Yongding River Basin. All of the 10 samples were selected from different areas. According to the classification standard for soil particles, clay (<1 μm), coarse clay(1–5 μm), fine silt (5–10 μm), coarse silt (10–50 μm), fine sand (50–250 μm), and coarse sand (250–1000 μm) were separated into seven size fractions (<1, 1–5, 5–10, 10–50, 50–75, 75–250, and 250–1000 μm) with sedimentation method based on Stokes' Law (Qin et al., 2014).

Air-dried 1000-μm sieved soil samples were passed through 250 μm and 75 μm nylon sieves in sequence. Then, the 75-μm sieved soil samples (approximately 150 g) were dispersed with 900 mL of deionized water via ultrasonic treatment at 100 w for 1 h. The soil solution was then transferred to a 1000-mL measuring cylinder. After an appropriate settling time, which was previously calculated with Stokes' Law, the supernatant (<1 μm) was siphoned off at a fixed depth below the surface and collected. To ensure that each particle fraction was completely separated, the remaining solution was subjected to repeated sedimentation and siphon until the supernatant was clear. After separation, all solutions of different size fractions were dried at 40 °C, weighed, and stored for further analyses.

2.3. Analytical method

Soil pH was measured in a soil–$CaCl_2$ (0.01 M) suspension (soil/solution 1:2.5, ISO10390) with a pH meter (Leici PHS-3C, China). TOC content was determined with a TOC analyzer (Metalar Analysensysteme GmbH, Germany). Cadmium, Cr, Cu, Mn, Ni, Pb, Sc, Ti, and Zn were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) (PerkinElmer NexION 300Q, USA) as previously described in (Ding et al., 2016). As and Hg were analyzed by atomic fluorescence spectroscopy (AFS) (Jitian AFS-960, China) in accordance with a previous method (Chen et al., 2016).

2.4. Quality control and quality assurance

The reagents and chemicals were of the analytical grades. Reagent blanks, the standard reference soil samples (GBW07401, GBW07402, GBW07403, GBW07404, GBW07405, GBW07406, GBW07407, GBW07408, Beijing, China; the Center of National Standard Reference Material of China) and duplicate samples were measured to carry out quality assurance and quality control. The logarithm deviation ($\triangle$lgC) of measured values and standard values were within ±0.046. The relative percentage differences between sample duplicates were within ±7.50%. Hence, analytical accuracy and precision of each metal conformed to requirements.