



Characterization of heavy metals in coal gangue-reclaimed soils from a coal mining area

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ABSTRACT

Coal gangue is practically used as a filling material for soil reclamation in the coal mining subsidence areas. Due to the toxic elements enriched in coal gangue, the reclaimed soils potentially pose great environmental concerns. In this study, we investigated the distribution and mobility of six heavy metals (As, Cr, Cu, Mn, Pb and Zn) in coal gangue-reclaimed soils ($n = 26$) from a typical coal mining area of Huainan Coalfield, China. The concentrations of Cu, Pb and Zn in the reclaimed soils were enriched by a factor of 1.51, 1.48 and 2.05 compared to the original agricultural soils, indicating potential migration of these heavy metals from coal gangue to soils. The soils of the finest particle grain size fraction ($< 74 \mu\text{m}$) had the highest concentrations of these heavy metals. The sequential extraction procedure results demonstrated that major portions of As, Cr and Pb existed in the residual fractions, whereas Mn was dominated in the reducible fraction and Cu, Zn in the oxidizable fractions. The results of risk assessment code and individual contamination factor revealed that Mn had a medium risk to the local ecology and the other heavy metals had lower potential risks. This study has a great implication to understand the behavior of heavy metals in soils and to implement various remedial options for soil pollution.

1. Introduction

The large-scale exploitation of coal resources in China has brought severe damage to coal mining areas (Yin and Zhang, 2011). In recent decades, coal mining subsidence areas have increased due to over-exploitation, accelerating the deterioration of ecological environment (Sun et al., 2010). In China, coal mining land reclamation began in the late 1970s and speeded up since the promulgation of the Regulations of Land Relation in 1988 (Li et al., 2011). It has been reported that the land reclamation rate of coal mining subsidence areas is rising rapidly from $< 1\%$ initially to 25% in 2013 (Dou, 2013). At present, coal gangue-reclamation is the main technology for mining land reclamation (Yang et al., 2011). The coal gangues were backfilled in the subsidence area and covered by soils. This technology not only fills the mining subsidence land, but also decreases the solid waste accumulation (Yang et al., 2011). However, the toxic substances in coal gangue might transfer to the reclaimed soils, which might cause severe soil pollution (Liu et al., 2013; Sun et al., 2014; Yao et al., 2010; Zhou et al., 2014). In contrast to most pollutants, heavy metals are not biodegradable and have long lasting effects in soil due to the strong adsorption of many metals on humid and clay colloids in soils (Bakircioglu et al., 2011; Liu

et al., 2013; Sun et al., 2014). Leaching and weathering of coal gangue-reclamation are the primarily pathways for heavy elements to enter into the ecosystem environment (Sun et al., 2010; Zhou et al., 2014).

Chromium (Cr), copper (Cu), manganese (Mn), lead (Pb) and zinc (Zn) are the most common, toxic heavy metals (Tang et al., 2013b). Metalloid such as arsenic (As) often falls into the heavy metal category due to their similarities in chemical properties and environmental behavior (Li et al., 2014). Heavy metal pollution is persistent, covert and irreversible (Tang et al., 2013b). Heavy metals in the reclaimed soils might have a significant impact on agricultural and fishery production, posing a great threat to human health via the food chain (Gong et al., 2014). For example, chronic exposure to As can cause adverse effects such as dermal lesions, peripheral neuropathy, skin cancer and squamous cell carcinoma (Żukowska and Biziuk, 2008). The excessive intake of Pb can damage the nervous, skeletal, circulatory, enzymatic, endocrine, and immune systems of human beings (Zhang et al., 2012). Several studies have suggested that the toxicity and mobility of heavy metals depend not only on their total amounts but also on their chemical species: exchangeable fraction is bioavailable; carbonate fraction, Fe–Mn oxide fraction and organic matter fraction are potentially bioavailable; residual fraction is un-bioavailable (Mao et al., 2014;

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Yang et al., 2016). The bioavailability of heavy metals to plants and fishes is potentially influenced by the physical and chemical properties of the reclaimed soils, such as soil particle-size distribution, organic matter and pH (Qian et al., 1996). Hence, it is crucial to investigate the characteristics of heavy metals in coal gangue-reclaimed soils, in order to adopt appropriate soil remediation measures.

Numerous studies have been conducted to determine heavy metals in bulk and size-resolved soils and in soils of different physicochemical fractions. However, most previous researches focused on urban soils, garden soils, dusts and sediments (Acosta et al., 2009; Beamer et al., 2012; Sun et al., 2014; Zheng et al., 2015), and only few data (Chen et al., 2017; Liu et al., 2017) are available for the reclaimed soils in coal mining subsidence areas. To better assess the environmental risks and implement environmentally-friendly remedial measures in the future, it is important to properly assess the toxic effect of heavy metals in coal gangue-reclamation soils. The main objectives of this research are to: (1) determine contamination levels of several important toxic heavy metals (As, Cr, Cu, Mn, Pb and Zn) in coal gangue-reclamation soils; (2) investigate the distribution patterns of heavy metals in soils of different particle size fractions; and (3) evaluate the mobility of heavy metals using the sequential extraction methods.

2. Materials and methods

2.1. Study area

Huainan City is located in the mid-north part of Anhui Province and encompasses a total area of 2600 km² and a population of 2 million. Locating in the temperate, semi-humid monsoon climate zone, its mean annual temperature is 15.2 °C and mean annual precipitation is 922 mm. According to the recent report, coal mining has resulted in about 275 km² of land subsidence as of 2015, accounting for 10.6% of the total area of Huainan City (Yao et al., 2010). It is estimated that the land subsidence area will extend to > 1000 km² if all the Huainan coals are mined. This has a significant influence on the sustainable development of mining area and the ecological environment to Huainan city (Yao et al., 2010). The subsided land mostly distributes in Datong, Xiejiaji, Bagongshan, Panji and Fengtai County. Xinzhuangzi Mine (116°49'38"E, 32°35'41"N) is an old mine, which belongs to Bagongshan county (Fig. 1A).

2.2. Sampling

As shown in Fig. 1B, 26 sampling sites were selected across the coal gangue-filled reclamation area at Xinzhuangzi mine, Huainan city, Anhui province, China. Surface (0–20 cm depth) soil samples (each of approximately 1 kg) were systematically collected from agricultural land, orchard, forest land and culture pond of coal gangue-reclamation areas. At each sampling point, three sub-samples, each with a 20 cm × 20 cm surface, were taken and then mixed to obtain a bulk sample, and then sampled using a soil stainless steel hand trowel. Stones and visible plant roots were removed from soil samples before transferring to cleaned polyethylene bags. Precautions were taken to avoid contamination during sampling, drying, grinding and storage.

2.3. Experiments

Soil moisture was determined gravimetrically by drying 30 g of received 26 soil samples at 105 °C for 24 h (Hu et al., 2016). The pH (1:2.5 soil:water, m:v) of soil samples was analyzed using an electrode pH meter (Lisbeth et al., 2009). Soil organic matter was measured using dichromate oxidation (Du and Gao, 2006). Soil total phosphorus (P) content was measured using colorimetric after digestion with HClO₄-H₂SO₄ (Hu et al., 2016) and available phosphorus was established following the Olsen method (Du and Gao, 2006). Soil total nitrogen (N) was determined by the Kjeldahl method and the nitrate nitrogen (NO₃-

N) contents were determined using an ultraviolet spectrophotometer (Du and Gao, 2006).

The 26 dried samples were homogenized, and then pulverized in an agate mortar to pass through a 120 mesh plastic sieve for chemical analysis. About 0.2 g soil sample was weighed into a pre-cleaned Teflon vessel fitted within a microwave digestion system. Then, soil samples were digested by adding 10 mL high purity acids mixture, i.e. HNO₃:HCl:HF = 3:1:1. After cooling, the digestion solution was diluted to 10 mL with 5% HCl. The sample solutions were analyzed using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS) for As, Cr, Cu, Mn, Pb and Zn.

Soil samples from the same region were mixed together for the heavy metal concentration analysis of different particle size and the sequential chemical extraction experiments, respectively. Four soil samples were then divided by particle diameter into five physical size fractions, < 74 μm, 74–420 μm, 420–841 μm, 841–1880 μm, > 1880 μm, respectively. The weights of each size fraction were recorded and their percent distributions were calculated. Analysis procedure of heavy metal contents in soils of different particle sizes was the same as above.

In order to evaluate the different binding fractions of heavy metals, 4 soils were subjected to the four-step sequential extraction procedure (SEP) in the following order (Nemati et al., 2011):

- (i) Exchangeable fraction (F1): 1 g of sample was introduced into a centrifuge tube. 40 mL of 0.11 M acetic acid was added to the tube and shaken for 16 h at room temperature. The extract was subsequently separated from the solid phase by centrifugation at 8000 rpm for 10 min.
- (ii) Reducible fraction (F2): 40 mL of 0.1 M hydroxyl ammonium chloride was added to the solid residue remaining from the first extraction stage. After shaking the extract for 16 h at room temperature, it was centrifuged as described in step 1.
- (iii) Oxidizable fraction (F3): 10 mL of 8.8 M hydrogen peroxide was added to the solid residue in the centrifuge tube. It was occasionally shaken by hand at room temperature for 1 h, followed by digestion at 85 ± 2 °C for another 1 h in a water bath. Heating was continued as before until the volume was reduced to 2–3 mL, after which 50 mL of 1 M hydrogen peroxide was introduced into the residue and shaken at room temperature for 16 h. It was also centrifuged as described above and then decanted slowly into a clean container.
- (iv) Residual fraction (F4): The residue from step 3 was digested using a mixture of aqua regia and hydrofluoric acid (15 mL HNO₃ + 5 mL HCl + 2 mL HF).

The four fractions acquired during each step of the analytical procedure were analyzed for As, Cr, Cu, Mn, Pb and Zn as above.

The method blank, standard material and sample replicates were used to for quality control and assurance (Liang et al., 2015, 2017). Analytical reagent blanks were prepared with each batch of digestion set, and were found to contain negligible levels of the heavy metals. The sediment Standard Reference Materials [GBW-07403 (GSS-3)] provided by the Center of National Reference Materials of China showed that the recoveries of the studied elements ranged from 93% to 106%. The relative difference of heavy metals was < 5% for the sample replicates. Detection limits were 0.6, 0.2, 0.2, 0.07, 0.04 and 0.3 mg L⁻¹ for As, Cr, Cu, Mn, Pb and Zn, respectively.

2.4. Assessment methods

2.4.1. Enrichment factor

Enrichment factor (EF) index was useful in assessing the anthropogenically introduced heavy metal, and it was calculated by Eq. (A.1) (Chan et al., 2001):

$$EF_x = C_x / C_{ref} \quad (\text{A.1})$$

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