



## Partitioning and geochemical fractions of heavy metals from geogenic and anthropogenic sources in various soil particle size fractions



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### ABSTRACT

The size of soil particles can significantly affect the enrichment of heavy metals in soil. However, the size effect of soil particles in colloidal dimension on the partitioning and geochemical fractions of heavy metals has been rarely studied. In this study, four soils, polluted by several heavy metals, were collected from Jiangxi (JX) and Hebei (HB) Provinces and Guangxi Zhuang Autonomous Region (GX and GX') in China, respectively. Five size fractions ( $> 10$ ,  $10-1$ ,  $1-0.45$ ,  $0.45-0.2$  and  $< 0.2 \mu\text{m}$ ) were separated from the studied soils to investigate the partitioning and geochemical fractions of cadmium (Cd), cadmium (Cr), copper (Cu), manganese (Mn), nickel (Ni), lead (Pb) and zinc (Zn). For JX, GX and GX' soils, Cr and Ni in JX and GX' soils and Cr, Ni and Mn in GX soil, as geogenic elements, were comparable with their background values; whereas other heavy metals, as anthropogenic elements, were higher than their background values. For the HB soil that was only polluted by Pb, Cd, Cr, Cu, Mn, Ni and Zn were the geogenic heavy metals. Generally, the contents of most heavy metals in the soil colloids were higher than that in coarse soil particles, and the partitioning tendencies with particle size were not significantly different between geogenic heavy metals and anthropogenic heavy metals. The partitioning of heavy metals in various soil particle size fractions was closely related to the soil particle compositions, such as OM, Fe/Al oxides and clay minerals. For geogenic heavy metals (Cr, Ni), the proportion of the residual fraction, which is imbedded in the silicate crystalline structures of soil particles, was very high, and the geochemical fraction patterns of the various soil particle size fractions were comparable, whereas the bioavailable fraction of anthropogenic heavy metals, such as Cu, Mn, Pb and Zn, decreased with decreasing soil particle size and the proportion of other fractions increased with decreasing size. This result indicates that more anthropogenic heavy metals exist in their stable form in fine soil particles. Correlations between Fe, Al and TOC and the bioavailable fraction of anthropogenic heavy metals were all negative, and most were significant or very significant indicating that high contents of OM and Fe/Al oxides stabilize anthropogenic heavy metals in fine soil particles.

### 1. Introduction

Soil pollution by heavy metals has become of great concern worldwide in the past few decades (Frohne et al., 2014; Hernandez et al., 2003; Kahapanagiotis et al., 1991; Miller and Mc Fee, 1983; Rodríguez et al., 2009). Excess heavy metals in the soil are mainly derived from human activities, such as mining, fertilization, traffic and industry (Chen et al., 2010; Liu et al., 2013; Liu et al., 2014; Rodríguez et al., 2009; Zhai et al., 2003). When anthropogenic heavy metals are discharged into the soil, they can be stabilized by the soil through

adsorption, precipitation, complexation, etc. (Bradl, 2004), which is time dependent and controlled by the soil composition, soil type, environmental condition and source of the heavy metals (Liu et al., 2016; Ma et al., 2013; McLaughlin, 2001; Zhang et al., 2013). Despite this, once the concentration of soil heavy metals exceeds the maximum capacity of the soil environment, the soil heavy metals can migrate downward easily to pollute the ground water and can be taken up easily by crops (Chen et al., 2012; Hou et al., 2014; Huang et al., 2008; Neidhardt et al., 2012; Zhao et al., 2012). Eventually, adverse effects on plant growth and human health can become apparent.

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Soil particles play a crucial role in the mobility and bioavailability of soil heavy metals due to the high contents of metal oxides, OM and clay minerals (Rashad et al., 2013; Rinklebe et al., 2016; Shaheen and Rinklebe, 2014; Wang et al., 2014). Fe/Mn oxides in soil particles can strongly bond with heavy metals via coprecipitation, adsorption, surface complex formation, ion exchange and penetration of the crystal lattice (Chao and Theobald, 1976). OM, an important part of soil particles, possesses high affinity for heavy metals, resulting from complexation of various functional groups (-OH, -COOH, etc.) on the OM surface with heavy metal ions (Bradl, 2004). The affinity of soil particles for heavy metals can also be greatly enhanced by some clay minerals in soil particles (Bhattacharyya and Gupta, 2008). For various soil particle size fractions, the contents of these effective compositions (i.e., metal oxides, OM and clay minerals) change greatly with soil particle size, leading to differences in the heavy metal partitioning in various soil particle size fractions. Qian et al. (1996) characterized the partitioning of heavy metals in different particle size fractions and found that clay and one of the sand fractions contained more heavy metals. It was found by Gong et al. (2014) that the heavy metal contents in various soil particle size fractions increased with decreasing particle size. Generally, more anthropogenic heavy metals are accumulated in fine soil particles due to the increase of specific surface area and effective compositions with decreasing soil particle size (Gong et al., 2014; Zhang et al., 2013), while the contents of geogenic heavy metals seem to show no obvious differences among the various soil particle size fractions (Ajmone-Marsan et al., 2008).

Soil colloids, defined as soil particles with a diameter of less than 1  $\mu\text{m}$ , have often been studied in the past. In the presence of preferential flow, macropores or fractures, soil colloids can be transported faster than the solute because of the size exclusion effect (de Jonge et al., 2004; Kanti Sen and Khilar, 2006). Moreover, soil colloids can strongly bond with heavy metals. Therefore, the transport of heavy metals in soil can be greatly affected by soil colloids (Hu et al., 2008; Wang et al., 2014; Yin et al., 2010; Zhou et al., 2011). Zhou et al. (2011) found that Cu bound to soil colloids played a major part in the leaching solution. A similar result was found by Yin et al. (2010) when they studied Pb transport in two shooting-range soils. The particle size of soil colloids can greatly affect the co-transport of heavy metals with soil colloids because of the difference in mobility and heavy metals' partitioning for various soil colloid size fractions. However, the size effect of soil particles on the partitioning of heavy metals in colloidal dimension is seldom studied. Furthermore, the geochemical fraction patterns of heavy metals from geogenic and anthropogenic sources may be different in various soil particle size fractions. Therefore, the partitioning and geochemical fractions of heavy metals in various soil colloid size fractions must be examined for heavy metal transport and environmental risk assessment. The objectives of this study are to separate four soils polluted by heavy metals into five size fractions based on their mobility, i.e., > 10  $\mu\text{m}$  (immobile soil matrix), 10–1  $\mu\text{m}$  (mobile soil particles), 1–0.45  $\mu\text{m}$  (coarse soil colloids), 0.45–0.2  $\mu\text{m}$  (medium soil colloids) and < 0.2  $\mu\text{m}$  (fine soil colloids), and to characterize the effect of soil particle size on the partitioning and geochemical fractions of heavy metals from geogenic and anthropogenic sources, which could contribute to the understanding of the co-transport of heavy metals by soil colloids.

## 2. Material and methods

### 2.1. Soils and separation of soil particle size fractions

Four soil samples polluted by heavy metals were collected from different regions in China. HB soil is a landscaping soil (calcareous soil) in Hebei province, which was polluted by dust from a lead-acid battery factory. GX and GX' soils (red soil), which were polluted by waste water from mining activities of a polymetallic mine, were collected from dry land and a paddy field, respectively, in Guangxi Zhuang Autonomous

Region. In addition, GX and GX' soils were sampled at adjacent arable lands to ensure the same soil heavy metal source and pollution history. JX soil was collected from dry land (red soil) in Jiangxi Province, which had also been polluted by waste water from mining activities and had been used in our previous study (Liu et al., 2017). The soil samples were air-dried, sieved through a 2-mm polyethylene sieve, and then stored in polyethylene Ziploc bags before further analysis and particle size separation. Natural sedimentation and fractional centrifugation were performed to obtain soil particles with different sizes of > 10, 10–1, 1–0.45, 0.45–0.2 and < 0.2  $\mu\text{m}$  (Gimbert et al., 2005). The separation procedure is described in detail in the supplementary materials (File S1).

### 2.2. Physical and chemical analysis

Particle size distributions were analysed using a laser particle size analyser (Microtrac S3500, USA). Mineral compositions were identified using X-ray diffraction (XRD; PANalytical, X'Pert PRO MPD, Netherland) at 40 mA and 40 kV using Cu K $\alpha$  radiation at 25 °C; the instrument was operated in the range of 5–80°. The OM content was measured by an element analyser (Eurovector, EA3000, Italy) after acidification with 0.1 mol/L HCl and dilution to neutral pH with deionized water. The concentrations of Al, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn in the obtained soil particles and bulk soils were measured by inductively coupled plasma atomic emission spectrometry (ICP-AES; Jarrel-ASH ICAP-9000, American) after the samples had been digested with HNO<sub>3</sub>-HClO<sub>4</sub>-HF (3:1:1, v/v/v). To ensure analytical quality, geochemical standard soils (GSS-1 and GSS-2), provided by the National Research Center for Geoanalysis of China, were used to validate the analytical method. The recoveries of the standard samples ranged from 90%–110%.

The sequential extraction of heavy metals in soils was carried out following the modified European Community Bureau of Reference (BCR) sequential extraction procedure (Rauret et al., 1999). This procedure separates the heavy metals in soils into three fractions, i.e., exchangeable and carbonate-associated (F1), reducible (bound to Fe/Mn oxides, F2) and oxidizable (bound to OM, F3) fractions, which are extracted by 0.11 mol/L acetic acid solution, 0.1 mol/L hydroxylamine hydrochloride solution adjusted to pH 2.0 with HNO<sub>3</sub> and 1 mol/L ammonium acetate solution after digestion with H<sub>2</sub>O<sub>2</sub>, respectively. To obtain the mass balance of heavy metals in the soils, the residual fraction (F4) was measured after acid digestion using the procedure described above for total metal determination. The recoveries which were defined as the sum of F1, F2, F3 and F4 divided by the total content, ranged from 98%–113%. To ensure adequate extraction efficiency, the reference sediment material BCR-701 was used. The measured heavy metal concentrations in the F1, F2, and F3 fractions were not significantly different from the certified values, as shown in Table S1 (supplementary materials). The properties, total Cd and Cd geochemical fractions of the JX soil particles, which were shown in our previous study (Liu et al., 2017), were also used in the present study for comparison with other soil particles.

### 2.3. Statistical analysis

Statistical analyses were performed with the statistical software package SPSS version 20.0 for Windows.

## 3. Results and discussion

### 3.1. Characteristics of the bulk soils and various soil particle size fractions

The physicochemical properties of the bulk soils and various soil particle size fractions are shown in Table 1. The proportion of clay in the HB soil was 2.64%, which is lower than that in the other soils. For the JX, GX and GX' soils, the proportions of clay were comparable with

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