



Arsenic, chromium, molybdenum, and selenium: Geochemical fractions and potential mobilization in riverine soil profiles originating from Germany and Egypt



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H I G H L I G H T S

- Selenium showed the highest potential mobilization followed by Cr, Mo, and As.
- Potential mobilization of As, Cr, and Se was high in the Wupper soils.
- Potential mobilization of Mo was high in the Nile soils.
- Residual fraction of As, Cr, Mo, and Se was dominant in the Nile soils.
- Oxidizable fraction of Cr and Se was dominant in the Wupper soils.

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The fractionation and potential mobilization of As, Cr, Mo, and Se in four floodplain soil profiles collected along the Nile (Egypt) and Wupper (Germany) Rivers were assessed using the BCR sequential extraction procedure. The concentrations of total and the geochemical fractions (acid soluble (F1), reducible (F2), oxidizable (F3), and residual (F4) fraction) of the elements were determined. The Wupper soils had the highest total concentrations (mg kg^{-1}) of As (378) and Cr (2,797) while the Nile soils contained the highest total Mo (12) and Se (42). The residual fraction of As, Cr, Mo, and Se was dominant in the Nile soils suggesting the geogenic source of the elements in these soils. The residual fraction of As and Mo and the oxidizable fraction of Cr and Se were dominant in the Wupper soils. Among the non-residual fractions (potential mobile fractions; $\text{PMF} = \sum \text{F1} - \text{F3}$), the oxidizable fraction was dominant for Cr, Mo, and Se in the Nile soils and for Mo in the Wupper soils, while the reducible fraction was dominant for As in both soils. The PMF of As, Cr, and Se was higher in the Wupper than in the Nile soils which might reflect the anthropogenic sources of these elements in the Wupper soils, while the opposite was the case for the PMF of Mo. The high PMF of Se (87%), Cr (87%), and As (21%) in the Wupper soils suggested that a release of these toxic elements may happen which increase the potential environmental risks in the anthropogenically polluted soils.

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1. Introduction

High concentrations of potentially toxic elements (PTEs) such as arsenic (As), chromium (Cr), molybdenum (Mo), and selenium (Se) in agricultural ecosystems may create important public and environmental health problems (Sun et al., 2012; Antoniadis et al., 2017a,b; Rinklebe et al., 2017). The binding between soil and PTEs can affect both the potential mobilization and phytoavailability of these elements (Shaheen and Rinklebe, 2015). It is generally agreed that total element concentration alone is a poor indicator of soil contamination (Rinklebe and Shaheen, 2014; Rivera et al., 2016), because it does not provide predictive insights on mobilization, bioavailability, and fate of the contaminants (D'Amore et al., 2005; Degryse et al., 2009). Therefore, more attention has been focused on the analysis of geochemical fractionation of PTEs rather than solely the total element content. Knowledge of PTEs partitioning is importance to determine geochemically reactive pools as a better indication of potential risk of PTEs (Shaheen and Rinklebe, 2014; Matong et al., 2016). Therefore, fractionation of PTEs in soil using sequential extraction procedures together with total element content has been generally conducted to specify the chemical fractions in which element are present in different environmental matrices (Shaheen et al., 2015; Ma et al., 2016). Sequential extraction is an important and widely applied tool for gaining information on potential mobility of PTEs in soils. Sequential extraction procedures are applied for a large number of PTEs in a wide range of sample types (Bacon and Davidson, 2008).

Among many sequential extraction protocols (Tessier et al., 1979; Zeien and Brummer, 1989), the sequential extraction procedure of the Community Bureau of Reference (BCR) has the advantage that it is well standardized (Ure et al., 1993; Quevauviller, 1998; Rauret et al., 1999; Bacon and Davidson, 2008; Sutherland, 2010). It is a widely used technique and the results can be interpreted as 'operational fractionations' according to the definition of Ure (1991) and Ure et al. (1993). For example, PTEs recovered in Step 1 of the BCR procedure should be described as "acid soluble fraction"; in step 2 described as associated with the "reducible fraction", rather than as being 'bound to iron/manganese oxyhydroxides'; in step 3 as "oxidizable fraction" rather than as being 'bound to organic matter or sulfate' (Bacon and Davidson, 2008). This technique was applied by many scientists (e.g., Arain et al., 2008; Gadepalle et al., 2008; Baig et al., 2009; Bakircioglu et al., 2011; Sungur et al., 2014; Rosas-Castor et al., 2015; Matong et al., 2016; Wang et al., 2016) to extract sequentially several PTEs including As, Cr, Mo, and Se.

Although soil pollution with anthropogenic PTEs and its remediation has been the focus of increasing research (e.g., Choppala et al., 2016; Rizwan et al., 2016; Niazi et al., 2017) in the last few decades due to their toxic impacts on our ecosystems, soils that contain elevated levels of PTEs derived from geological sources have received scant attention, even though they provide some fascinating topics to study (e.g. Fernández-Caliani, 2012; Li et al., 2014; Rivera et al., 2016). Naturally occurring or geogenic PTEs are relatively immobile and rarely toxic (Adriano, 2001); however, they may pose a potential risk to the environment (Rapant et al., 2008). Therefore, investigation of the geochemical fractions of PTEs in floodplain soils varying in origin, characterization, and source of elements is worth to be studied.

Previous studies have determined the geochemical fractions of trace element cations in floodplain soils in Germany, Egypt and Greece (e.g., Shaheen and Rinklebe, 2014; Shaheen et al., 2015). However, considerable knowledge gaps exist regarding the fractionation and depth distribution of element anions such as As, Cr, Mo, and Se in soils which are still not well studied especially under different anthropogenic and geogenic sources. Moreover, the

geochemical fractions of As, Cr, Mo, and Se in various floodplain soil profiles originating from different regions under large variation of the soil forming factors, process and climates is unclear. Furthermore, the relationships between soil properties and the vertical distribution of As, Cr, Mo, and Se fractions in floodplain soils of two different regions have not been studied to our knowledge to date. Therefore, our objectives were 1) to quantify and compare the total and the geochemical fractions of As, Cr, Mo, and Se in four entire soil profiles representing floodplain soils originating from the Nile River (Egypt) and from the Wupper River (Germany), and 2) to assess the vertical profile distribution and potential mobilization of As, Cr, Mo, and Se as affected by relevant soil properties.

2. Materials and methods

2.1. Study sites

2.1.1. German soils

Two profiles representing floodplain soils along the Wupper River were sampled. The respective study area is located close to the confluence of the Wupper River into the Rhine River near Cologne in North Rhine-Westphalia, Germany. We selected two study sites within the study area and one soil profile was dug at each study site (Profile 1: 51°4'0.48"N, 6°4'0.48"E; Profile 2: 51°7'49.86"N, 7°1'35.10" E). More details about the first study site are given by Frohne et al. (2011). The two study sites are used as grassland and are flooded seasonally by the Wupper River, usually in springtime. Details about the second study site are given by Mihajlovic et al. (2014).

2.1.2. Egyptian soils

Two soil profiles representing floodplain soils in the north of the Nile River delta were sampled. Both profiles were collected from Kafr El-Sheikh governorate, Egypt (31° 14' 60" N and 31° 14' 60" E) (Appleby et al., 2001). The first profile (P3) was very close to the Nile River and collected from Desouk at the western part of Kafrelsheikh Governorate (31° 07' 42" N and 30° 38' 46" E). The second profile (P4) was collected from the south of Kafr El-Sheikh. The two study sites are used as agricultural soils and cultivated by wheat/clover/vegetables in the winter and by corn/rice/vegetables in the summer. The studied soils are under an arid climate with an annual precipitation of <100 mm (Appleby et al., 2001; Shaheen et al., 2014a).

2.2. Soil preparation and characterization

The soils were described in detail and classified according to "Bodenkundliche Kartieranleitung" (Ad-hoc Arbeitsgemeinschaft Boden, 2005), and "World reference base for soil resources 2006" (IUSS Working Group WRB, 2014) (Table 1). Soil samples have been collected in accordance to genetic horizons in the Wupper soils and according to depth in the Nile soils because the latter are not developed yet to show genetic horizons. Sampling from each horizon or depth was performed in four replicates of about 1 kg each, which were pooled to one composite sample. The collected samples were transported to the laboratory and all visible roots, macro fauna, and fresh litter were removed from the samples. The composite soil sample was homogenized, air dried for several days, and ground to pass through a 2-mm sieved and stored in the laboratory for the physico-chemical analyses (Shaheen and Rinklebe, 2017). Soil properties were analyzed according to standard soil methods as provided elsewhere (Blume et al., 2011; Sparks et al., 1996) as described in details in Shaheen and Rinklebe (2017). Dithionite-extractable iron (Fe) and manganese (Mn) in both soils were extracted with 3 M sodium citrate + 1 M sodium

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