



Effect of aging on the bioavailability and fractionation of arsenic in soils derived from five parent materials in a red soil region of Southern China



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ABSTRACT

The effects of aging time and soil parent materials on the bioavailability and fractionations of arsenic (As) in five red soils were studied. The results indicated that As bioavailability in all soils decreased during aging, especially with a sharp decline occurring in the first 30 days. After aging for 360 days, the highest available As concentration, which accounted for 12.3% of the total, was observed in soils derived from purple sandy shale. While 2.67% was the lowest proportion of the available As in soils derived from quaternary red clay. Furthermore, the best fit of the available As changing with aging time was obtained using the pseudo-second-order model ($R^2 = 0.939\text{--}0.998$, $P < 0.05$). Notably, Al oxides played a more crucial role ($R^2 = 0.89$, $P < 0.05$) than did Fe oxides in controlling the rate of As aging. The non-specially and specially absorbed As constituted the primary forms of available As.

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1. Introduction

Arsenic (As) is a naturally ubiquitous toxic element that exists in a wide range of minerals and soils. In the environment, arsenic can originate from both natural and anthropogenic sources (Gonzaga et al., 2006); the scale of problematic As contamination can exist in a range from local to regional. China is regarded as one of the most seriously As-contaminated countries in the world. Owing to mining, smelting, and geogenic origins, the extent of As-contaminated agricultural soils is increasing rapidly in China (Zhao et al., 2014). Further, the average As concentrations in Southern China are much higher than those in other regions of China, and the As concentrations are particularly high in red soil regions (Chen et al., 2015). For example, the As concentration in

soils of arable land were 284.9 and 114 mg kg⁻¹ in Shimen and Liangjianzhen in Hunan and Guangxi Province, respectively (Wei and Chen, 2002; Zhao et al., 2014).

It is widely recognized that measurement of the total concentration of metal(loid)s in soil is frequently a poor predictor of the potential biological availability of these materials, and is thus a poor proxy as a parameter for assessing toxicity (Fendorf et al., 2004; Ma et al., 2006). In the environment, the magnitude of the bioavailability of contaminants can be reduced with increasing time (Pignatello and Xing, 1995). The changes in As bioavailability during aging depend on the As partitioning process in soils (Tang et al., 2007; Liang et al., 2014). With time, the As distribution in soils changes from more available forms to less available forms (Fendorf et al., 2004). It is known that many processes can alter the bioavailability of metal(loid)s during aging, including surface precipitation/surface oxidation, Ostwald ripening, cavity entrapment, diffusion into micropores, or incorporation into crystal lattices (Ma et al., 2006; Wendling et al., 2009). The specific aging mechanisms of As in soils are not well understood.

Soil properties are key factors in controlling the bioavailability of As. These properties thus influence potential risks in contaminated soils. Amorphous oxides of Al, Fe and Mn occur as a discrete phase

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or as coatings on other mineral surfaces. These oxides with large surface areas, microporous structures, and an abundance of binding sites show a significant impact on contaminant mobility and bioavailability (Axe and Trivedi, 2002). Yang et al. (2002) and Tang et al. (2007) found that Fe, Al oxides content and soil pH were the most important soil properties controlling the initial degree of adsorption and the magnitude of the bioavailability of As, which involves the partition of As among different soil components. Soil clay minerals are also crucial to the natural attenuation of As; these minerals offer important sites where the adsorption/desorption or oxidation/reduction of As can take place. Lin and Plus (2002) reported that halloysite and chlorite have much greater As adsorption capacity than do other clay minerals. In addition, the variable-charge components of soil, phosphate, organic matter (OM), and CaCO_3 in soils have been implicated as factors that impact As mobility and bioavailability in soils, although there have been controversial conclusions in this area of study (Bolan et al., 2013; Redman et al., 2002). For instance, Wang and Mulligan (2009) indicated that sorption onto organic matter might reduce the mobility of As in mine tailings under acidic pH conditions ($\text{pH} < 3$), whereas the mobility of As might be increased under alkaline conditions ($\text{pH} > 7$). Soil parent material is a foundational component in soil formation and can impact soil particulate sizes, mineral content, and soil properties. Soil parent materials also influence As concentrations and toxicity. Chen et al. (2002) and Yamasaki et al. (2013) indicated that parent material was the main source of As in non-polluted red soil, with red soils derived from limestone containing high amounts of As and Cd. Although many studies have illustrated the effects of soil properties and soil parent materials on the bioavailability and adsorption of As, such studies have generally discussed either individual soil minerals or have compared soils from different regions. Soils in the same red soil area can be derived from different parent materials. To date, there is little information available about how different soil parent materials influence the bioavailability and redistribution of As in red soil regions during the aging process.

In this case, five soils derived from different parent materials were spiked with exogenous As and then incubated for 360 days; and soil sampling was performed at predetermined time points. Conventional NaHCO_3 extraction methods were used for the analysis of bioavailable As. Wenzel's (2001) sequential extractions were carried out to determine whether changes in the redistribution of As in different soil fractions could account for changes in the bioavailability of As. The observations will be helpful for understanding the exogenous As aging process in soils as affected by parent materials and for establishing an empirical basis for the rational utilization and management of soils in As-contaminated red soil regions.

2. Materials and methods

2.1. Soil samples and treatments

Five soils derived from different parent materials including quaternary red clay (RS1), purple sandy shale (RS2), limestone (RS3), plate shale (RS4), and granite (RS5) were collected from a cultivated red soil region (0–20 cm) in Hunan Province, China ($26^\circ 75' \sim 26^\circ 97' \text{N}$, $111^\circ 85' \sim 111^\circ 95' \text{E}$). Soil samples were air-dried, homogenized, and ground until they were able to pass through 2 mm nylon fiber sieve. Samples (1000 g equivalent dry mass) of each soil were spiked with appropriate volumes (RS1, 72.02 ml; RS2, 89.29 ml; RS3, 76.34 ml; RS4, 67.43 ml; RS5, 79.20 ml) of $\text{Na}_3\text{AsO}_4 \cdot 12\text{H}_2\text{O}$ (1000 mg l^{-1}) solution to attain the same total As concentration of 100 mg kg^{-1} in soils. This concentration was almost the average As concentration in arable land impacted by

mining and smelting activities in Hunan Province (Li et al., 2010, 2014; Wu and Chen, 2010). Meanwhile, the same spiked concentration was also adopted by Yang et al. (2003) and the results demonstrated that there was no significant difference in As bioavailability over the concentration range of $10\text{--}100 \text{ mg kg}^{-1}$. The soil samples were then mixed thoroughly and double-distilled water was added to adjust the soil moisture to 70% of the water holding capacity. 300 g subsamples were weighed into glass cups with filter membranes on the top to allow air influx and avoid the evaporation of moisture. All soils were stored in the dark at $25 \pm 1^\circ \text{C}$, and double distilled water was added to maintain soil moisture every 2 or 3 days. Soils were sampled at different aging periods (1, 9, 15, 30, 60, 90, 200, and 360 days) after As addition, then analyzed for available As and the fractionation of As. All treatments were repeated in triplicate.

2.2. Soil physicochemical properties

The evaluation of soil physicochemical properties was performed in duplicate. Particle size composition of the soils was analyzed with a laser particle size analyzer (Helos-Rodos, Sympatec GmbH, Germany) after dispersion by adding 0.5 M NaOH to acidic soils and 0.5 M sodium oxalate to soils with pH values above 6.0 (Lu, 2000).

Soil pH was measured potentiometrically at a 1:2.5 ratio of soil to 0.01 M CaCl_2 after 1 h of end-to-end shaking. Cation exchange capacity (CEC) was measured with methods presented by Sparks (1996). Soil organic carbon (SOC) was measured using the acid dichromate oxidation method described by Lu (2000). Available P was measured using the method of Olsen et al. (1954). Total P was measured after digestion of soil with $\text{HClO}_4\text{--H}_2\text{SO}_4$ (Lu, 2000).

Amorphous Fe, Al, and Mn oxides were extracted by shaking 1 g of soil in 30 ml of acidified ammonium oxalate buffer for 2 h (Loeppert and Inskeep, 1996). Dithionite-citrate-bicarbonate (DCB) extractable Fe, Al, and Mn were measured with the procedure described by Manning and Goldberg (1997), which involved shaking duplicate 250 ml centrifuge tubes containing 4 g soil in 120 ml of 0.57 M sodium citrate ($\text{Na}_3\text{C}_6\text{H}_5\text{O}_7 \cdot 5\text{H}_2\text{O}$) and 0.1 M sodium dithionite ($\text{Na}_2\text{S}_2\text{O}_4$) for 16 h (140 rpm). After shaking, the suspensions were centrifuged (4100g, 10min), and 15 ml aliquots were removed and filtered, the extracts correspond to free Fe, Al, and Mn oxides. Concentrations of Fe, Al, Mn, and P were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES, Optima2000DV, Perkin Elmer, USA). Total arsenic in soil was determined by hydride generation atomic fluorescence spectrometry (HG-AFS, AFS-9120, Two-channel atomic fluorescence spectrophotometer, Beijing Titan Instrumental Company, China) after digestion of soils with aqua-regia.

2.3. Available As and the sequential extraction of soil As

Soil available As was determined using a 0.5 M NaHCO_3 chemical extraction method. This method was selected because 0.5 M NaHCO_3 extraction of As were known to show good correlation with As concentrations in plants, no matter the soil pH (Peryea, 2002; Wang et al., 2014; Su et al., 2015). Soil samples (5.00 g) were suspended in 50 ml of 0.5 M NaHCO_3 and shaken for 2 h at room temperature, and then filtered through 0.45 μm filter paper before As measurement by HG-AFS.

The sequential extraction procedure of Wenzel et al. (2001) was used to evaluate the fractionation of As in spiked, aged soil. Soil (1 g) was extracted sequentially with 25 ml of 0.05 M $(\text{NH}_4)_2\text{SO}_4$, corresponding to non-specifically adsorbed As (F1); 25 ml of 0.05 M $\text{NH}_4\text{H}_2\text{PO}_4$, corresponding to specifically adsorbed As (F2); 37.5 ml of 0.2 M NH_4 -oxalate, corresponding to As associated with amorphous and poorly-crystalline hydrous oxides of Fe and Al (F3);

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