



# Changes of arsenic fractionation and bioaccessibility in wastewater-irrigated soils as a function of aging: Influence of redox condition and arsenic load



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## ABSTRACT

Redox conditions and arsenic (As) loads may affect the aging mechanisms of As in soils. A batch experiment of one wastewater-irrigated soil under four different conditions was performed to investigate the impact of redox conditions and As loads on the aging mechanisms of As in soils in this study. The results showed that the aerobic condition substantially decreased the more labile fractions and increased the less mobilizable or immobilizable fractions in soils over time, which was likely due to the oxidation of As(III) to As(V) and the decomposition of organic matter. The redistribution of the strongly adsorbed and organically bound fraction (F4) occurred in all soils during the aging process, and both low As load and aerobic conditions accelerated this redistribution. The aging processes of As were characterized by three stages in various soils except the low As load soil with two stages within 180 days: The transformations of As fractions were from three former fractions to three later ones in all soils in the first stage (0 day–10 days), while the main transformations after 10 days were from F4 to bound to Fe and Mn/Al oxides fraction (F5) and from water soluble fraction (F1) to F5 in soils with low and high As loads, respectively; In contrast, the main transformations in the second stage were from F1 to F4 and from F1 to F5 in soils at low oxygen and aerobic conditions, respectively, and were from F4 to F5 in the third stage in both redox conditions. The bioaccessibility of As in the low As load soil decreased markedly from approximately 40% down to <10% due to the aging and more remarkably from >70% down to <20% in three other soils. Both high As load and low oxygen conditions preserved higher bioaccessibility of As in soils after long aging process. In terms of As bioaccessibility, high As load and aerobic condition prolonged the aging process of arsenic in soils.

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## 1. Introduction

Arsenic (As)-rich groundwater is widely distributed in a number of countries including China, Bangladesh, India, Vietnam, Thailand, Mongolia, Pakistan, Japan, Korea, Philippines, Indonesia, Turkey, New Zealand, Spain, Greece, USA, Canada, Mexico, Argentina, Chile, South Africa and Australia (Smedley and Kinniburgh, 2002; Guo et al., 2014; Huang et al., 2014). Unfortunately, in many areas of the above countries, As-rich groundwater is commonly used for irrigation, which results in a transfer of water soluble As from groundwater into soils, and ultimately increases As levels in soils (Xie et al., 2012; Polizzotto et al., 2013; Tong et al., 2014). It is known that mobile and bioavailable fractions of As in soils will move into crops via soil–plant transfer, and finally cause harm to human health through food chains (Williams et al., 2005;

Karak et al., 2011; Wang et al., 2013). Hence, it is necessary to understand the fractionation and bioaccessibility of As in soils irrigated by As-rich groundwater when assessing the ecological risk of these soils. However, as we know, after water soluble As is added into soils, the fractionation and bioaccessibility of As in soils are significantly influenced by the aging; that is, more mobilizable fractions will transform into less mobilizable fractions and the bioaccessibility decreases due to the aging effect (Tang et al., 2007). Thus, the aging effect should be taken into account when assessing the ecological risk of the soils irrigated by As-rich groundwater. If not, the assumption would be that all As fractions in soils are bioavailable, which would overestimate the risks (Juhász et al., 2008).

To date, some factors controlling aging mechanisms of As in soils such as soil type, organic matter content and As species have been discussed (e.g., Yang et al., 2002; Yang et al., 2005; Quazi et al., 2010; Quazi et al., 2011). It was reported that aging greatly weakened the bioaccessibility of As in some soils such as isohumosol, argosol, and ferrosol, owing to the transformation of more labile fractions to the

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less mobilizable fractions over time, but aging did not have a significant effect on the chromosol and ferralsol (Tang et al., 2007; Juhasz et al., 2008). The bioaccessibilities of both inorganic As and organic As added to soils were significantly decreased during the aging process, and a portion of arsenite added to soils under aerobic conditions was oxidized to arsenate over time (Yang et al., 2005; Quazi et al., 2010; Quazi et al., 2011). In contrast, the addition of organic matter increases the bioaccessibility of As in soils, indicating that abundant organic matter weakens or even inhibits the influence of aging on the bioaccessibility of As in soils (Meunier et al., 2011), while it may be changed after the mineralization or stabilization of organic matter such as some low molecular weight organic substances in soils (Dippold et al., 2014; Gunina et al., 2014). In addition to the factors mentioned above, As loads and redox conditions such as aerobic or low oxygen conditions may also be important factors for the aging mechanisms of As in soils. Because the transformation of As species and soil components such as iron hydroxides and organic matter are generally affected by the redox conditions (Zhu et al., 2008; Frohne et al., 2011; Parsons et al., 2013; Ma et al., 2014; Han et al., 2015), and the fixation of As on soil minerals is significantly influenced by As loads due to the higher adsorption amount of As on soils with increasing As loads (Goh and Lim, 2004; Huang et al., 2013). As far as we know, however, the impact of redox conditions and As loads on the aging process of As in Chinese irrigated soils has received little attention.

It is known that the variation of redox conditions and the increase of As loads in Chinese irrigated soils occur under the intermittent irrigation (alternating wetting and drying) by As-rich groundwater for crops (Neidhardt et al., 2012). Therefore, it is very important to understand the influences of redox conditions and As loads on the aging of As in soils, which will be beneficial to explain the specific mechanisms of As aging in soils and then to propose more reasonable remediation strategies for As-contaminated soils. Additionally, it will be helpful to assess the ecological risk of As-contaminated soils irrigated by As-rich groundwater. The objectives of this study are to investigate (1) the impact of As loads and redox conditions on the fractionation and bioaccessibility of As in soils over time; and (2) how the aging mechanisms of As in soils are affected by the redox conditions and As loads.

## 2. Materials and methods

### 2.1. Soil sampling and characterization

Fourteen top soils (0–10 cm) were sampled from a wastewater-irrigated area of the Pearl River Delta region in South China, which is located at the longitude of 113°08'E and latitude of 23°04'N (Huang et al., 2011). Soil samples were air-dried, ground, then passed through a 0.9-mm sieve, and finally pooled into one composite sample and mixed thoroughly to ensure homogeneity. The distribution of soil particle size was determined by the sedimentation method, and the soil pH was measured with a glass electrode using a 1:5 (w:w) suspension of soil to water (Lu, 2000). The total iron and aluminum were extracted by the HCl-HNO<sub>3</sub>-HClO<sub>4</sub>-HF method (Lu, 2000), and the free and amorphous iron oxides were extracted by the dithionite-citrate-bicarbonate method and the NH<sub>4</sub><sup>+</sup>-oxalate method, respectively (Pansu and Gautheyrou, 2006). The soil organic matter was measured using the K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> method, and the cation exchange capacity of the soil was determined by the NH<sub>4</sub>OAc method (Pansu and Gautheyrou, 2006). One modified Tessier's sequential extraction procedure (SEP) (details in Section 2.3) was used to measure the background values of As fractions in soils (Tessier et al., 1979). The properties of the experimental soil are shown in Table 1.

### 2.2. Soil preparation and handling

The effect of As loads was investigated by taking two samples with 300 g each, and spiking them with 100 mL of either 0.0008 mol/L or 0.008 mol/L NaAsO<sub>2</sub> solution to artificially increase the total As load in

**Table 1**

Physical and chemical properties of the experimental soil.

pH	7.67
Organic matter (g/kg)	28.7
Total Fe (g/kg)	43.1
Free iron oxides (g/kg)	17.0
Amorphous iron oxides (g/kg)	6.7
Total Al (g/kg)	93.7
Total Mn (g/kg)	0.627
CEC (cmol/kg)	12.7
Particle size (%)	
Sand (>0.05 mm)	31.8
Silt (0.002–0.05 mm)	55.6
Clay (<0.002 mm)	12.6
As fractions (mg/kg)	
F1	0.045
F2	0.082
F3	0.199
F4	2.59
F5	1.77
F6	16.5

the samples by approximately 20 mg/kg and 200 mg/kg, respectively. Correspondingly, the samples are denoted as S<sub>LL</sub> and S<sub>HL</sub>, which represent low load of As and high load of As, respectively. These two samples were mixed thoroughly by an artificial stir with two glass rods and maintained the soil moisture at close to field capacity (32.9%) to mimic the field conditions and were then sealed at room temperature. The effect of redox conditions was also investigated by taking two samples with 300 g each, spiked with 100 mL of 0.004 mol/L or 210 mL of 0.0019 mol/L NaAsO<sub>2</sub> solution to artificially increase the total As load in the samples by approximately 100 mg/kg, and made the soil moisture close to field capacity (32.9%) and supersaturation, respectively. These two samples are denoted as S<sub>L</sub> and S<sub>A</sub>, which represent the low oxygen condition and aerobic condition (rich oxygen), respectively. S<sub>L</sub> was sealed at room temperature and the soil moisture maintained at supersaturation, while S<sub>A</sub> was opened at room temperature and the soil moisture maintained at field capacity by intermittent filling with water. It is worth mentioning that we had performed a pre-experiment before this experiment, and a significant difference of redox values between supersaturation and field capacity at the fourth day after treatment was observed. At different times (0 d, 3 d, 10 d, 30 d, 60 d, 120 d and 180 d) after spiking, soil samples were collected and freeze-dried.

By the way, the reason these dosages and source of As were selected in this study is as follows: the highest concentrations of As in groundwater in the Pearl River Delta (include the irrigated area where the experimental soil was collected) and near the irrigated area were 0.581 mg/L and 0.303 mg/L, respectively (Huang et al., 2014), these concentrations are approximately 0.008 mol/L and 0.004 mol/L of As, respectively; As(III) is the main form of As in the groundwater of the area where the experimental soil was collected (Huang et al., 2014).

### 2.3. Sequential extraction procedure

The dried samples were ground and mixed thoroughly, and 1.00 g dried samples were weighed into acid-washed 50 mL centrifuge tubes. The fractionation of As in soils is generally determined by the SEP such as some Tessier's modifications (Li et al., 2009; Basu and Schreiber, 2013). This study also adopted one modified Tessier's SEP, the extractants of F1 and F6 are different from that of Tessier's SEP, and the shaking time of F1–F5 are also different from that of Tessier's SEP. The details are shown in Table 2.

### 2.4. Analysis and quality control

All reagents in this study were analytical grade or better. Before the analysis of As, extracts after the SEP were filtered through 0.45- $\mu$ m filter

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