

## The ageing effect on the bioaccessibility and fractionation of arsenic in soils from China

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### Abstract

Ingestion of contaminated soil has been recognized as an important exposure pathway of arsenic for humans, especially for children through outdoor hand-to-mouth activities. An improved sequential extraction procedure was employed in an attempt to reveal the relationship between bioaccessibility and fractionation of As in five soils from China. Arsenic bioaccessibility in acidic ( $\approx$ pH 4.5) soils reached approximately stable levels after a sharp decline within one week of ageing. In contrast, As bioaccessibility in higher pH ( $>6.0$ ) soils was found to be significantly higher and took two weeks of ageing to reach stable levels. The artificially added As was more labile than indigenous As. The main proportions of added As were found in the specifically sorbed and amorphous and poorly-crystalline hydrous Fe/Al oxide-bound fractions. Correlation analysis shows that the non-specifically and specifically sorbed As are likely to constitute the main proportion of bioaccessible soil As. The soil content of amorphous and crystalline Fe/Al oxides and soil pH appear to be the key factors controlling, not only the time needed to reach a steady state, but also the magnitude of the bioaccessibility of As added to the soils.

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### 1. Introduction

Soil ingestion is regarded as an important human exposure pathway of contaminants, especially for children through outdoor hand-to-mouth activities. The daily amount of soil ingestion has been estimated to be in the range of 50–200 mg d<sup>-1</sup> (Van Wijnen et al., 1990). Moreover, children have the ability to absorb higher proportions of metals through the digestive tract to reach systemic circulation than adults, which may make them subject to higher adverse health effects (Miller and Schriker, 1981;

Calabrese and Stanek, 1995). Arsenic contaminated soil has long been recognized as a threat to human health.

It has been recognized that soil contaminants are usually less than 100% bioavailable (Rodriguez et al., 1999). Given the expense and time constraints associated with animal testing, much recent attention has been directed to the development of *in vitro* extraction tests that mimic the human digestion process. Such tests estimate bioaccessibility on the basis of the maximum amount of soil contaminant that is soluble in the digestive fluid, and is therefore available for intestinal absorption (Ruby et al., 1999).

The decreasing bioaccessibility of a metal to a given biological organism with increasing contact time with the soil is often termed “ageing”, although the specific retention mechanisms responsible are not well understood.

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However, it is supposed that adsorption of cations and/or anions (e.g. lead,  $\text{Pb}^{2+}$ ; selenite,  $\text{SeO}_3^{2-}$ ; selenate,  $\text{SeO}_4^{2-}$ ) on soil surfaces initially form outer-sphere complexes, followed by the formation of inner-sphere complexes (Hayes and Leckie, 1986; Zhang and Sparks, 1990). Upon ageing, intraparticle surface diffusion of metal contaminants within micropores may result in reduced lability, and may also result in nucleation and surface precipitation (Ford and Sparks, 2000; Axe and Trivedi, 2002). These mechanisms appear to be reasonable explanations for the decreasing potential for contaminant release from polluted soil during the ageing process. However, such a general explanation may not be sufficient to account for variations in the bioaccessibility of soil As with ageing, which involves the partition of As among different soil components.

To the authors' knowledge, only a limited number of reports have focused on the effects of ageing on the bioaccessibility of As in soil and the underlying mechanisms involved. Yang et al. (2002) found that the bioaccessibility of arsenate added to soils was significantly reduced in some soils over time, and that the iron oxide content along with pH appeared to be the main soil factors controlling the steady-state bioaccessibility of arsenate. Fendorf et al. (2004) also reported that exchangeable As in two soil samples (as extracted with 1 M  $\text{MgSO}_4$ , pH 7), decreased with incubation time and was highly correlated with an exponential decline in the bioaccessibility of As. Another recent paper by Yang et al. (2005) showed that a significant portion of arsenite added to soils was oxidized to arsenate over six month's aerobic ageing, decreasing remarkably the bioaccessibility of As. However, in all these three studies, As bioaccessibility was determined using an artificial gastric phase only, without further assessment using an artificial small intestinal phase.

The objectives of this study were to investigate (i) the ageing effect on the bioaccessibility of As in five soils from China using a modified physiologically based *in vitro* test, and (ii) the availability of soil As fractions for intestinal absorption using a sequential extraction procedure As.

Such investigations could be crucial for assessing temporal variations in health risks at either new or historically As-contaminated sites.

## 2. Materials and methods

### 2.1. Soil preparation

Five typical uncontaminated, non-cultivated soils were collected from sites throughout China in Changchun, Beijing, Nanjing, Yujiang, and Hainan. The soils were classified as a Cryi-Ustic Isohumosol (CC), a Hapli-Ustic Argosol (BJ), a Ferri-Udic Argosol (NJ), a Haplic-Udic Ferrisol (YJ), and a Rhodi-Udic Ferrallosol (HN) (Cooperative Research Group on Chinese Soil Taxonomy, 2001). The soils were air-dried, ground, and passed through a 2 mm mesh sieve. Samples (400 g) of each soil were spiked with 60 ml of  $\text{Na}_3\text{AsO}_4$  (600 mg  $\text{l}^{-1}$  of As) solution to artificially increase the total As concentration in the soils by about 120 mg  $\text{kg}^{-1}$ . The samples were then mixed thoroughly, and after being air-dried and homogenized again by sieving, 40 g subsamples were weighed into plastic cups. Double distilled water was added to the cups to adjust the soil moisture to 15% on a weekly basis to simulate field-like drying-wetting cycles. All soils were stored in the dark at 25 °C. At different times (1, 3, 7, 14, 30, 60, and 120 d) after the As addition, the samples were air dried, and then ground gently with a wooden spatula. Since it is usually assumed that only soil particles less than 0.25 mm in diameter adhere to the hands of children (Duggan et al., 1985), this soil fraction only (<0.25 mm) was used for both the *in vitro* extraction test and the sequential fractionation. Some basic characteristics of the soils (<0.25 fraction) including the background As concentrations are shown in Table 1.

### 2.2. Characterization of soils

Particle size composition of the <0.25 mm soil fraction was analyzed with a laser particle size analyser (Mastersizer

Table 1  
Selected physical and chemical properties of the experimental soils

Soil	CC	BJ	NJ	YJ	HN	
pH ( $\text{CaCl}_2$ )	6.09	7.43	6.09	4.50	4.56	
$\text{CaCO}_3$ ( $\text{g kg}^{-1}$ )	8.75	36.88	7.88	4.38	2.38	
OC ( $\text{g kg}^{-1}$ )	15.40	41.02	26.46	3.68	4.86	
CEC ( $\text{cmol kg}^{-1}$ )	22.50	15.75	23.25	10.88	6.75	
Fe ( $\text{g kg}^{-1}$ ) <sup>a</sup>	2.47	1.14	2.91	2.79	2.70	
Fe ( $\text{g kg}^{-1}$ ) <sup>b</sup>	1.75	2.90	3.60	19.77	46.15	
Al ( $\text{g kg}^{-1}$ ) <sup>a</sup>	0.87	1.06	0.69	1.36	1.70	
Al ( $\text{g kg}^{-1}$ ) <sup>b</sup>	0.00	0.00	0.06	1.74	4.54	
Mn ( $\text{g kg}^{-1}$ ) <sup>a</sup>	0.27	0.14	0.46	0.03	0.19	
Mn ( $\text{g kg}^{-1}$ ) <sup>b</sup>	0.00	0.01	0.00	0.06	0.32	
As ( $\text{mg kg}^{-1}$ )	11.41	11.00	9.37	20.84	17.05	
Particle size	Sand (>0.05 mm)	5.1	5.1	4.0	0.9	3.0
composition (%)	Silt (0.002–0.05 mm)	82.2	77.3	88.1	58.8	64.3
	Clay (<0.002 mm)	12.7	17.6	7.9	40.3	32.7

<sup>a</sup> and <sup>b</sup> represent amorphous and crystallised oxides, respectively.

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