



Arsenic in agricultural soils across China: Distribution pattern, accumulation trend, influencing factors, and risk assessment



Yuting Zhou ^a, Lili Niu ^a, Kai Liu ^{b,*}, Shanshan Yin ^a, Weiping Liu ^{a,*}

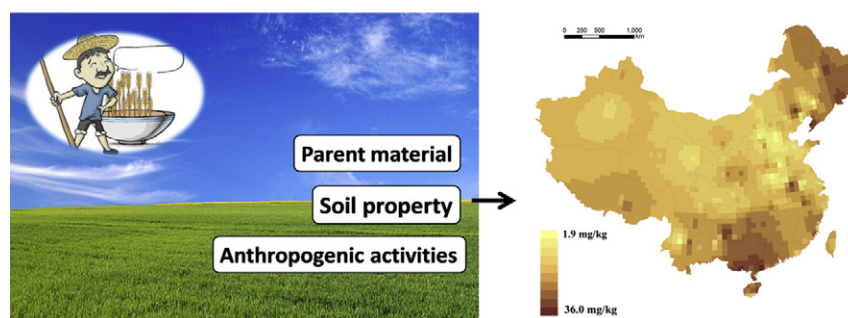
^a MOE Key Laboratory of Environmental Remediation and Ecosystem Health, College of Environmental and Resource Sciences, Zhejiang University, Hangzhou 310058, China

^b Division of Engineering and Applied Science, California Institute of Technology, Pasadena, CA 91125, USA

HIGHLIGHTS

- Nation-wide distribution, and potential ecological and human health risks of arsenic in agricultural soils were determined.
- Comprehensive analyses were performed to estimate the correlations between arsenic and soil properties.
- Temporal accumulation trend of soil arsenic were identified and the influencing factors were further discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As) in the environment is of concern due to its strong toxicity and high risks to the ecosystems and humans. In this study, soil samples across China collected in 2011 and 2016 were used to determine the concentrations of arsenic in arable soils. The median concentration of arsenic in surface soils was 9.7 mg/kg. The inventory of arsenic in the Chinese agricultural surface soils was estimated to be 3.7×10^6 tons. In general, arsenic contamination was found higher in South and Northeast China than in other regions, with means of 18.7 and 15.8 mg/kg, respectively. Vertically, arsenic concentrations were higher in top layer (0–15 cm) soils (median of 9.8 mg/kg) and decreased with soil depth (medians of 8.9 mg/kg at 15–30 cm and 8.0 mg/kg at 30–45 cm). By comparing with published data, an increasing accumulation trend over the past decades was found and this enhancement was positively related with the long-term application of fertilizers in agricultural practice, especially phosphate fertilizers. Soil pH was found to affect the movement of arsenic in soil, and high-pH conditions enhanced the pool of arsenic. The ecological risk assessment revealed that arsenic in Chinese agricultural soil posed a low risk to the ecosystem. Regarding human health, the mean hazard indices (HIs) of arsenic were below 1, suggesting an absence of non-carcinogenic risks. In addition, the cancer risks of arsenic in all soil samples were within the acceptable range (below 1×10^{-4}), indicating low to very low risks to the exposed population. Findings from this study are valuable to provide effective management options for risk avoidance and to control the persistent accumulation of arsenic in the agriculture sector across the world.

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1. Introduction

Arsenic (As) is a metalloid element which is extensively present in the natural environment and all human populations are exposed to it through various pathways (Abernathy et al., 1999). Arsenic is

* Corresponding authors.

E-mail addresses: kliu7@caltech.edu (K. Liu), wliu@zju.edu (W. Liu).

bioaccumulative and non-biodegradable (Wei and Yang, 2010). A variety of adverse health effects, such as cancers, cardiovascular and neurological diseases, and diabetes, have been attributed to arsenic exposure in human. Compared to organic forms, inorganic arsenic is much more toxic and considered to be a Class 1 non-threshold carcinogen (Tchounwou et al., 2004). The World Health Organization (WHO) and International Agency for Research on Cancer (IARC) have recognized arsenic as a carcinogen. Numerous epidemic-like health accidents linked to arsenic contamination have happened in recent years (Bundschuh et al., 2010; Mandal and Suzuki, 2002; Kabay et al., 2010), and hundreds of thousands of people in the world have already been affected by the arsenic contamination (Net, 2001; Wang, 2008).

Agricultural soil affects public health via the quantity and quality of human food supply. Excessive arsenic in agricultural soil can reduce soil productivity, lower the quality of crops, and pose a threat to human health via food consumption (Chen et al., 1999; Raghunath et al., 1999). Arsenic in soils can be derived from not only natural sources such as weathering and erosion of rocks, but also anthropogenic activities including industrial emissions, application of fertilizers and pesticides, sewage irrigation, and atmospheric deposition (Chary et al., 2008; Kumaresan and Riyazuddin, 2001; Lu et al., 2010; Norra et al., 2005; Zhang et al., 2014). These intensive human activities lead to a general increase in arsenic on local, regional, and global scales (Hagan et al., 2015; Seigneur et al., 2004). It was estimated that atmospheric deposition related to industrial processes accounted for 58.2% of total annual input of arsenic in agricultural soil, and the application of various fertilizers was identified as another big contributor to the arsenic burden in soil (Luo et al., 2009).

China has a long history of being an agricultural country. Farmland quality directly links to the health and life of billions of people. Previous studies on arsenic in China focused on the contamination in the industrial areas or were just small regional scale studies (Huang et al., 2007; Pan et al., 2016; Wong et al., 2002). Nationwide spatial and vertical distributions of arsenic in soils, especially the agricultural soils and the associated risks to humans and ecosystems have not been systematically investigated before. Such knowledge is crucial for maintaining sustainable agriculture in China (PRC, 2005).

In this study, we measured the total arsenic content in agricultural soil samples collected across China in 2011 and 2016. The objectives were to (a) comprehensively characterize the concentrations, spatial and vertical distributions, and calculate the inventory of arsenic in agricultural soil, (b) identify the main source of arsenic using a geo-accumulation index and principle component analysis (PCA), (c) better understand the temporal accumulation trend of arsenic during the past ten years, (d) explore the influence of soil properties and the impact of human activities, and (e) evaluate the potential ecological risk and human health risk associated with exposure to arsenic for adults and children.

2. Materials and methods

2.1. Sample collection

A total of 242 surface agricultural soil samples were collected from 31 provinces, autonomous regions, and municipalities in China (Fig. S1-a), including 121 samples in 2011 and 121 samples in May in 2016. In addition, 25 soil core samples were collected from 21 provinces across China (Fig. S1-b) in May in 2016. The soils collected in the South China are paddy soils, while the other areas are mainly vegetable and wheat fields. Each core was sectioned into the surface (0–15 cm), middle (15–30 cm) and bottom (30–45 cm) segments. The sampling sites in 2016 were chosen to match the sites used in 2011 (Niu et al., 2013) and located by GPS. At each site, more than five discrete subsamples were collected by a plastic scoop and mixed to form a composite sample. All samples were placed in sealed plastic bags and stored at -20°C before analysis.

2.2. Extraction and analysis

The soil samples were air-dried, grounded, and sieved through a 2 mm sieve. Then, a portion of each sample was added to CO_2 -free, de-ionized water (soil to water ratio of 1:2.5) to measure soil pH using a pH electrode. Another portion of the soil was used to measure soil organic matter by potassium dichromate oxidation (Lu, 2000).

Arsenic was extracted from collected soil samples according to the method established by Huang and Chen (2007). Briefly, approximately 0.5 g of each sample was placed in a 50 mL flask. A mixture liquor of 10 mL aqua regia (3:1 HCl/ HNO_3 , v/v) was added. Then, the soils were digested at about 100°C with a water bath. Additional details of the digestion procedure were described by Zhou et al. (2017). After digestion, de-ionized water was added to 50 mL and then filtered through a $4\ \mu\text{m}$ filter. The concentrations of arsenic were measured with an inductively coupled plasma-mass spectrometer (ICP-MS, Agilent 7500a, Agilent, Santa Clara, California). All the chemical analysis was carried out in duplicate.

2.3. Quality control and quality assurance

All analytical procedures were carried out with strict quality control measures. The glassware and Teflon crucibles used in the experiment were soaked in 10% HNO_3 overnight before use. Standard reference materials (GBW07424, GSS-10 and GBW07429, GSS-15), provided by the Center of National Standard Reference Materials of China, were also detected during the determination process to validate the accuracy of laboratory analyses. The measured arsenic concentrations were 85.8–109% ($n = 32$) of the reference values. Procedural blank samples ($n = 32$) were used with each 10 samples to validate the accuracy and examine contamination of metal analysis. Duplicate samples were also analyzed and the relative deviations of the duplicated analyses were below 5% for all the samples.

2.4. Calculation of inventories

The total inventory of arsenic in agricultural surface soil in China was calculated by the following equation (Jia et al., 2009).

$$R = \sum \text{ALDC}_{\text{soil}} \times 10 \quad (1)$$

where R is in kg, summed over 30 provinces; A is the area (km^2) and D is the soil density (g/cm^3) of cultivated lands in each province (National Bureau of Statistics of China, 2016), L is the depth (20 cm) of the soil sampled in this study, and C_{soil} is the mean concentration (mg/kg) of arsenic in all samples collected from each province (except Taiwan). The soil densities in China ranged from 0.53 to $1.79\ \text{g}/\text{cm}^3$ (Jia et al., 2009). The average of the maximum and minimum ($1.16\ \text{g}/\text{cm}^3$) was used in Eq. (1) to provide an order of magnitude estimate of the arsenic inventory in Chinese soils. Factor 10 is conversion factor (unitless).

2.5. Risk assessment

Geo-accumulation index (I_{geo}) is an indicator that quantifies the degree of contamination by heavy metals in soils. The value is calculated as follows (Müller, 1979):

$$I_{\text{geo}} = \log_2 \frac{C_i}{1.5C_0} \quad (2)$$

where C_i is the concentration of arsenic in soil samples; C_0 is the provincewise average background value of the arsenic in soils of China (China National Environmental Monitoring Center, 1990). The contamination of soils could be categorized into seven levels based on the calculated I_{geo} values, including practically uncontaminated ($I_{\text{geo}} < 0$), uncontaminated to moderately contaminated ($0 < I_{\text{geo}} \leq 1$),

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