ANNALS OF THE NEW YORK ACADEMY OF SCIENCES

Issue: *Ecological Complexity and Sustainability*

Effect of long-term fertilization on total soil arsenic in China

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To assess the effects of long-term fertilization on arsenic (As) accumulation in soils and crops with different agricultural practices, five experimental stations (Changshu, Taoyuan, Hailun, Fengqiu, and Qiyang) with long-term fertilization practices, representing five typical soils of China, were selected to investigate the soil As concentrations. Results indicated that the geological source, that is, parent materials, played a dominant role in determining the soil As concentrations. Long-term application of manure and phosphorus fertilizers led to a decrease of As concentration in the surface paddy soil at Taoyuan, while the effects of fertilization on As concentration in other samples were minimal. In addition, other agricultural practices, such as the removal of crop biomass, reduced the As concentrations in the surface soils with low levels of soil As (Fengqiu, Changshu, and Taoyuan). In the upland soils with higher As concentrations, wheat may have risk to human health through food-chain and maize can be considered as a favorable crop.

Keywords: arsenic; long-term fertilization; crop; risk assessment; Chinese soil

Introduction

Arsenic (As) can cause abnormality, mutation, and carcinogenesis to humans (Bates *et al.* 1992). Millions of people are being threatened by As contaminated drinking water, most notably in Bangladesh and West Bengal (Smith *et al.* 2000). In China, As poisoning was also found in Taiwan, Xinjiang, Inner Mongolia, and Shanxi, and 3.7 million people were affected (Shen *et al.* 2005).

Arsenic is widely spread in soil and mostly at low levels ranging from 2.5 to 33.5 mg kg⁻¹ in surface soils of China (Weng *et al.* 2000). Except for the parent materials of the soil, agricultural activities, such as the applications of As-containing pesticides, herbicides, phosphorous fertilizers, and livestock manures, are considered to be the major contributors of As in the agricultural soils (Peters *et al.* 1996; Chirenje *et al.* 2003; Camm *et al.* 2004). Li and Chen (2005) found that the application of pig manure increased soil As accumulation rate ranging from 11.8 to 78.9 µg kg⁻¹ yr⁻¹ in Beijing, China. Once the

As enters into the soil ecosystem, it will experience complicated geochemical processes. The plants and microbes can play important roles in the processes, such as As species transformation or leaching from soil to groundwater (Bentley and Chasteen 2002; Mukhopadhyay *et al.* 2002; Oremland and Stolz 2003, 2005; Islam *et al.* 2004).

There are diverse soil ecosystems, fertilization treatments, plants, and cropping systems in China. Although some researches have focused on the investigation of As-polluted agricultural soils, the influence of As on growth of crops and its critical level, and the As risk to human health through food-chain (Xie *et al.* 1991; Chen and Liu 1993; Xia 1994; Cheng *et al.* 2005), there are still no reports about the effect of long-term fertilization on As concentrations in agricultural soils, crops, and the corresponding risk to human health, even less in the comparison between different soil ecosystems. Thus, it is essential to understand the relationships among the long-term fertilization, the soil As concentrations, and the As accumulation in crops for further

human risk assessment. In this study, five soils under different long-term fertilization treatments were investigated to reveal the effects of long-term fertilization on soil As and the human health risk through food-chain.

Materials and methods

Site descriptions

Five long-term experimental stations, that is, Changshu, Taoyuan, Hailun, Fengqiu, and Qiyang, were selected to represent the typical soils and agricultural farming systems in China (Table 1). The Changshu and Taoyuan stations are paddy soils, while the Hailun, Fengqiu, and Qiyang stations are upland soils. Totally, 12 fertilization treatments, that is, CK0, CK, M, N, N + M, NP, NP + M, NK, PK,

NPK, NPK + M, and NPK + 1/2M (CK0 represents fallow [without growing crop and without fertilizer application]; CK represents unfertilized (control); N, P, and K represent chemical nitrogen, phosphorus, and potassium fertilizers, respectively; M represents manure.), were investigated in the five stations (Table 2). There were three and four replicates for each treatment of paddy soils and upland soils, respectively.

Sampling and analysis

Soil samples were collected in 2006 from experimental plots at two horizons, that is, horizon A (surface soil, 0–20 cm in depth) and horizon B (sub-surface soil, 20–40 cm in depth). The soils were air dried, ground, and passed through a 0.149 mm

Table 1. Description and soil characteristics of the long-term experimental stations

Station ^a	Location	Soil type	Initiation year	pH (H ₂ O)	OM (g kg ⁻¹)	Annual average temperature (°C)	Annual average precipitation (mm)
Changshu station (CAS)	31°32'N, 120°41'E	Paddy soil (inceptisol, derived from silt loam deposit)	1989	5.50	37.0	15.5	1316
Taoyuan station (CAS)	28°55'N, 111°26'E	Paddy soil (ultisols, derived from quaternary red clay)	1990	5.74	23.2	16.5	1448
Hailun station (CAS)	47°26'N, 126°38'E	Black soil (mollisol, upland)	1985	6.20	54.0	1.5	570
Fengqiu station (CAS)	35°04'N, 113°10'E	Fluvo-aquic soil (inceptisol, derived from alluvial sediments of the Yellow River, upland)	1989	8.65	10.1	13.9	605
Qiyang station (CAAS)	26°45'N, 111°52'E	Red soil (ultisols, derived from quaternary red clay, upland)	1991	5.70	11.5	18.2	1200

^aCAS and CAAS represent Chinese Academy of Sciences and Chinese Academy of Agricultural Sciences, respectively.

Table 2. Fertilization treatments and the applied fertilizers in the experimental stations

Station ^a	Fertilization treatment ^b	Recommended chemical fertilizers	Recommended manure	Cropping system
Changshu station (CAS)	CK, NPK, NPK + 1/2M, NPK + M	N: 300.0 kg hm ⁻² for wheat and rice; P: 32.7 kg hm ⁻² for wheat and rice; K: 124.5 kg hm ⁻² for wheat and rice.	1/2M represents 1500 kg hm ⁻² of rice straw; M represents 3000 kg hm ⁻² of rice straw.	Wheat—rice
Taoyuan station (CAS)	CK, N, NK, NP, PK, NPK, M, N + M, NP + M, NPK+M	N: 72.9 kg hm ⁻² for early rice, 109.4 kg hm ⁻² for late rice; P: 39.3 kg hm ⁻² for early rice; K: 137.0 kg hm ⁻² for late rice.	The manure was composed by the rice straw and the pig manure which was fed by 50% of the harvested rice.	Early rice—late rice
Hailun station (CAS)	CK, NK, NP, PK, NPK	N: 112.5 kg hm ⁻² for maize, 13.5 kg hm ⁻² for soybean; P: 45.0 kg hm ⁻² for maize, 34.5 kg hm ⁻² for soybean; K: 60.0 kg hm ⁻² for maize, 60.0 kg hm ⁻² for soybean.	The manure was composed by 80% of the rice straw and the pig manure, which was fed by 80% of the harvested rice.	Maize—soybean
Fengqiu station (CAS)	CK, NK, NP, PK, NPK, M, NPK + M	N: 150 kg hm ⁻² for wheat, 150 kg hm ⁻² for maize; P: 75.0 kg hm ⁻² for maize, 75.0 kg hm ⁻² for wheat; K: 150.0 kg hm ⁻² for wheat, 150.0 kg hm ⁻² for maize.	The manure was composed by wheat straw, bean cake and cotton kernel cake. The amount of manure was established by the N which was equivalent to the N in chemical fertilizers, and the deficient PK was supplemented by chemical PK fertilizers.	Wheat—maize
Qiyang station (CAAS)	CK, CK0, N, NK, NP, PK, NPK, M, NPK + M	N: 90.0 kg hm ⁻² for wheat, 210.0 kg hm ⁻² for maize; P: 36.0 kg hm ⁻² for wheat, 84.0 kg hm ⁻² for maize; K: 36.0 kg hm ⁻² for wheat, 84.0 kg hm ⁻² for maize.	The manure of the pig. In the M treatment, the amount of manure was established by the N which was equivalent to the N in chemical fertilizers; While in NPK + M treatment, 70% of the total N was applied as manure and the left 30% of N was applied as chemical fertilizers.	Wheat—maize

^aCAS and CAAS represent Chinese Academy of Sciences and Chinese Academy of Agricultural Sciences, respectively.
^bCK0 and CK represent fallow (without growing crop and without fertilizer application) and unfertilized (control) treatments, respectively. N, P, and K represent chemical nitrogen, phosphorus, and potassium fertilizers, respectively; M represents manure.

sieve. Crop samples were collected in two stations, that is, rice in the Taoyuan station, and maize and wheat in the Qiyang station. The crops were dried at 60° C, gently ground, and kept in the desiccator for analysis.

Soils were digested in digestion tubes with HCl-HNO₃-HClO₄ using 0.15 g sample (Lu 2000). The soil sample was first digested with 5.0 mL aqua regia (HCl:HNO₃ = 3:1 in v/v) at 120° C for 12 h. Then 4.0 mL HClO₄ was added and the sample was digested at 120° C continuously until it became white or gray white. The digested solution was transferred to a volumetric flask and diluted to 50.0 mL with distilled water. Crop samples were digested in digestion tubes with HNO₃ using 0.3 g sample (Lu 2000). 5.0 mL of concentrated HNO₃ was added and the sample was digested at 120° C until the mixture became white or clear. The digested solution was transferred to a volumetric flask and diluted to 25.0 mL with distilled water. All the glassware used for the experiment was previously soaked in 15% HNO₃ (v/v) and rinsed with distilled water. All reagents used in this study were of reagent grade.

The concentrations of As in the digested solutions were determined using hydride generation atomic fluorescence spectrometer (AFS-2202E, Haiguang Company, Beijing, China). Quality assurance was conducted by spiking the digested solutions with certified element standards.

Statistical analyses were performed using ANOVA procedure in STATISTICA (Version 6.0, Statsoft Inc., USA).

Results

Soil total arsenic concentrations under different fertilization treatments and horizons

In the paddy soils of the two stations (Changshu, Taoyuan), there were no significant differences in soil As concentrations among the treatments of the horizon A as well as the horizon B of the Changshu station (Table 3). There were also no significant differences among the treatments of the horizon B of the Taoyuan station. However, for the horizon A of the Taoyuan station, soil As concentrations of the N and NK treatments (14.29 mg kg⁻¹ and 13.93 mg kg⁻¹, respectively) were significantly higher than those of the NP + M treatment (10.27 mg kg⁻¹) ($P < 0.05$) (Table 3). The appli-

cations of manure and phosphorus (P) fertilizers decreased the As concentrations of the surface soils in the Taoyuan station through the comparisons between the treatments with and without manure or P fertilizers ($P < 0.05$). In the two stations, As concentrations were significantly lower in the surface soils than in the subsurface soils ($P < 0.05$) (Fig. 1).

In the upland soils of the three stations (Hailun, Fengqiu, Qiyang), there were no significant differences in the soil As concentrations among the treatments in horizon A of the Hailun station, horizons A and B of the Fengqiu station, and horizon B of the Qiyang station (Table 3). While in horizon A of the Qiyang station, As concentration of the CK0 (58.78 mg kg⁻¹) was significantly higher than any other treatments ($P < 0.05$). As concentrations in the surface soils were significantly lower than those in the subsurface soils in the Fengqiu station ($P < 0.05$) (Fig. 1).

Soil total As concentrations in different stations

The highest soil As concentrations in horizon A were found in the Qiyang station ranging from 43.37 mg kg⁻¹ to 58.78 mg kg⁻¹, and the lowest ones were in the Changshu station (7.22–7.78 mg kg⁻¹) (Table 3). The order of the stations in the total As concentrations were Qiyang > Taoyuan > Hailun > Fengqiu > Changshu. There were significant differences between any two stations except Taoyuan and Hailun, Hailun and Fengqiu, Fengqiu and Changshu ($P < 0.05$) (Fig. 1).

Soil As concentrations of the horizon B in the Taoyuan station were comparable with those in the Fengqiu station, and there were significant differences between any other two stations ($P < 0.05$). The highest As concentrations were also found in Qiyang station (43.60–65.89 mg kg⁻¹), which were at least three times higher than those in the other stations (Table 3, Fig. 1).

As concentrations in crops

In the Taoyuan station, rice As concentration in the CK treatment was the lowest one (0.20 mg kg⁻¹), which was significantly lower than those in the NP (0.31 mg kg⁻¹) and NPK (0.29 mg kg⁻¹) treatments ($P < 0.05$). While there were no significant differences in the rice As concentrations among the other treatments (Table 4).

Table 3. Total As concentrations under different fertilization treatments in the experimental stations (mg kg⁻¹)

Fertilization treatment	Station ^a				
	Changshu	Taoyuan	Hailun	Fengqiu	Qiyang
Horizon A					
CK0	NA	NA	NA	NA	58.78 ± 14.14a
CK	7.22 ± 0.32a	12.58 ± 1.12ab	10.78 ± 0.62a	9.41 ± 0.81a	52.64 ± 8.57ab
M	NA	11.96 ± 1.23ab	NA	9.25 ± 0.37a	51.61 ± 2.59ab
N	NA	14.29 ± 1.16a	NA	NA	46.98 ± 8.32ab
N + M	NA	11.66 ± 0.48ab	NA	NA	NA
NP	NA	12.87 ± 3.22b	10.22 ± 0.57a	9.16 ± 0.24a	43.37 ± 6.20b
NP + M	NA	10.27 ± 0.71b	NA	NA	NA
NK	NA	13.93 ± 2.36a	10.46 ± 0.47a	9.10 ± 0.74a	44.09 ± 7.45b
PK	NA	NA	10.65 ± 0.26a	9.12 ± 0.55a	48.72 ± 6.93ab
NPK	7.78 ± 0.78a	12.94 ± 1.49b	9.98 ± 0.50a	8.76 ± 0.26a	48.43 ± 5.70ab
NPK + M	7.25 ± 0.26a	11.13 ± 1.16ab	NA	8.67 ± 1.07a	47.97 ± 0.75ab
NPK + 1/2M	7.51 ± 0.27a	NA	NA	NA	NA
Horizon B					
CK0	NA	NA	NA	NA	NA
CK	8.00 ± 0.22a	14.67 ± 0.52a	NA	12.92 ± 1.11a	65.89 ± 15.70a
M	NA	NA	NA	12.89 ± 1.43a	56.28 ± 0.90a
N	NA	14.84 ± 0.71a	NA	NA	NA
N + M	NA	NA	NA	NA	NA
NP	NA	16.75 ± 2.06a	NA	12.68 ± 0.93a	43.60 ± 10.83a
NP + M	NA	NA	NA	NA	NA
NK	NA	15.32 ± 2.69a	NA	12.34 ± 1.21a	46.48 ± 9.77a
PK	NA	NA	NA	12.51 ± 1.41a	49.38 ± 7.14a
NPK	8.01 ± 0.66a	15.36 ± 1.11a	NA	12.00 ± 0.67a	48.83 ± 6.51a
NPK + M	8.07 ± 0.98a	15.82 ± 0.61a	NA	12.58 ± 0.78a	49.63 ± 7.04a
NPK + 1/2M	8.02 ± 0.59a	NA	NA	NA	NA

^aThe results are expressed as mean ± SD of the three or four replicates; NA, no such sample; different letters within each column of each horizon indicate significant differences among the treatments (*P* < 0.05, *n* = 3, *n* = 4).

In the Qiyang station, the accumulations of As in the tissues of maize were different with an order of root >> leaf >> stem > grain (“>>” indicates significant difference [*P* < 0.05]) (Table 5). There was no correlation among the As concentrations in the four maize tissues. The highest As concentration (0.14 mg kg⁻¹) in the maize grain was found in the NPK treatment, while the CK and PK treatments had the lowest one (0.04 mg kg⁻¹). For the wheat grain, the CK treatment had the highest As concentration (0.78 mg kg⁻¹), which was as twice higher as the lowest one (PK, 0.30 mg kg⁻¹). The As concentrations in wheat grains were much higher (about 5 to 20 times higher) than those in the maize grains (*P* < 0.05) (Table 5).

Discussion

Effect of long-term fertilization on soil total As concentrations

Long-term fertilization had minor influences on soil As in both horizon A and horizon B of the most stations, which indicated that the environmental As risk from fertilization was minimal.

However, in the Taoyuan paddy soil, As concentrations in horizon A decreased in the treatments applied with manure or P fertilizers. Several approaches, such as anion competitive adsorption (Brouwere *et al.* 2004; Caille *et al.* 2004; Signes-Pastor *et al.* 2007), arsenate reducing to arsenite, As-bearing iron oxides (Fe^{III}) reducing to Fe^{II}

Figure 1. Average soil As concentrations in different experimental stations. Different letters within each horizon (small letters on columns of Horizon A, capital letters on columns of Horizon B) indicate significant differences among the stations ($P < 0.05$, $n = 3$, $n = 4$). Asterisks (*) above columns of horizon B indicate significant differences between horizon B and horizon A in the same station ($P < 0.05$).

(Cummings *et al.* 1999; Tadanier *et al.* 2005), could make As release from soil adsorbing sites owing to the use of P fertilizers and manure. The released As might be removed with biomass or leached to the deeper horizon, and thus decrease the soil As concentration.

On the other hand, nitrogen and phosphorus deficiency could restrict the crop growth in the soils, and thus the application of manure or P fertilizers

significantly increased the crop biomass (Qin *et al.* 1998; Shen *et al.* 1998; Yu *et al.* 2003). The increased crop biomass could remove more As from the soil and caused the decline of surface soil As.

In the CK0 (fallow) treatment of the Qiyang station, no As would be removed by biomass, and its As concentration was the highest one in horizon A of all the treatments. This also confirmed that biomass harvest was an important reason of As removal from the surface soil. Chen *et al.* (2007) developed a generalized trace element mass balance model, which showed that the plant uptake factor was a primary factor affecting the fate and transport of As in cropland soils.

Table 4. As concentrations of rice grain in the Taoyuan station (mg kg⁻¹)

Fertilization treatment	Rice grain ^a (mg kg ⁻¹)
CK	0.20 ± 0.01 c
M	0.28 ± 0.02 abc
N	0.21 ± 0.01 bc
N + M	0.24 ± 0.02 abc
NP	0.31 ± 0.09 a
NP + M	0.21 ± 0.05 bc
NK	0.25 ± 0.02 abc
NPK	0.29 ± 0.06 ab
NPK + M	0.24 ± 0.03 abc

^aDifferent letters within the column “rice grain” indicate significant ($P < 0.05$, $n = 3$) differences among the treatments. The toxic limit was 0.15 mg kg⁻¹ of inorganic As in rice grain (GB2715–2005, China).

Effect of soil type on soil total As concentrations

Parent material is an important factor determining the concentration of trace metals in the soil (Kauranne 1992). In this study, the five stations were all long-term experimental stations and there were no notable As pollution sources. The effect of external pollutants on soil As was minor. In general, the concentrations of trace metals in subsurface soils were considered to be more close to the parent materials. The differences of soil As concentrations among the treatments of horizon B in each station were little, while the differences of the

Table 5. As concentrations of maize and wheat in the Qiyang station (mg kg^{-1})

Fertilization treatment	Maize				Wheat Grain
	Root	Stem	Leaf	Grain	
CK	1.49	0.25	0.75	0.04	0.78
M	1.44	0.14	1.30	0.06	0.73
NP	1.95	0.18	0.69	0.06	0.69
PK	1.37	0.20	0.64	0.04	0.30
NPK	1.54	0.18	0.50	0.14	0.77
NPK + M	2.35	0.16	0.87	0.06	0.70
Average ^a	1.69 ± 0.38 a	0.19 ± 0.04 c	0.79 ± 0.28 b	0.07 ± 0.04 c	0.66 ± 0.18

^aDifferent letters within the row “average” indicate significant ($P < 0.05$, $n = 6$) differences among the maize tissues. The toxic limits of inorganic As in maize grain and wheat grain were 0.20 mg kg^{-1} and 0.10 mg kg^{-1} , respectively (GB2715–2005, China).

soil As concentrations in horizon B among the stations were great. Thus, geological source played a dominant role in determining the As concentrations of agricultural soils. In the various parent materials, quartzite, silicate, and aluminosilicate might be the main contributors to soil As (Zhang *et al.* 1996). Further investigation is needed to clarify the contribution of different minerals to the soil As concentrations.

Soil As concentrations in the Qiyang station were higher than those in other stations. Two mixed soil samples of the paddy fields in this station were also analyzed and high As concentrations were found in horizon A (41.19 mg kg^{-1}) and B (48.00 mg kg^{-1}). This confirmed that the high soil As concentration in this station was mostly related with the parent materials.

On the basis of the parent materials of the soils, agricultural practices have limited influences on the soil As concentrations. For example, only in the soils with low As levels (Changshu, Fengqiu, Taoyuan), soil As in horizon B was significantly higher than that in horizon A, due to the As removal by crop biomass. In the soils with a high As level (Qiyang), the differences between the two horizons were not significant.

At the same time, our investigation of drinking groundwater in the Qiyang station showed that the As concentration ($3.07 \mu\text{g L}^{-1}$) was lower than the safe limit for drinking water ($10 \mu\text{g L}^{-1}$) as recommended by the World Health Organization (WHO). There was no relationship between the high As concentration in the upland red soil and the low As con-

centration in the groundwater. Therefore, the high As level in the soil had not led to the groundwater contamination in the Qiyang station.

Risk assessment of crop As on human health
In the five stations, Qiyang (upland soil) and Taoyuan stations (paddy soil) had higher As concentrations. Thus, the crops of the two stations were investigated to assess their As risk to human health.

In the Taoyuan station, the highest concentration of rice As was 0.31 mg kg^{-1} . A few recent studies had reported that 85–95% of the total As in rice was inorganic (Hossain 2006). The highest inorganic As in the rice grain of the Taoyuan station ranged from 0.26 to 0.29 mg kg^{-1} according to the reported ratio. These values were higher than the toxic threshold for As (0.15 mg kg^{-1}) according to the Hygienic Standard for Grains of China (GB2715–2005, China), but lower than the permissible limit of 1.0 mg kg^{-1} dry matter according to the WHO recommendation. This indicated that the rice of the Taoyuan station was relatively safe to human. Although the fertilization could slightly increase the rice As concentrations, the total As of the rice grain was relatively low and the environmental risk to human health through food-chain should be minimal.

In the Qiyang station, root had the highest As concentration among the maize tissues while grain was the lowest one. As was mainly accumulated in the maize root and its transfer ability from root to grain was minor. According to the Hygienic Standard for Grains of China (GB2715–2005, China), the limits

of inorganic As in maize grain and wheat grain were 0.20 mg kg^{-1} and 0.10 mg kg^{-1} , respectively. The As concentrations in the maize grain were all lower than the limit and was relatively safe to human. However, the As concentrations in the wheat grain were all much higher (about six times higher on average As concentrations) than the limit. In the upland agricultural practice, wheat would accumulate As in the grain more easily than maize (Liu *et al.* 2005). When the soil had high As concentrations, maize should be considered as the favorable crop instead of wheat. It is a concern that the NPK treatment resulted in higher As concentrations in both maize grain and wheat grain but it is a popular fertilization practice in this region.

Conclusions

Geological source (i.e., parent material) played a dominant role in determining the soil As concentrations in the long-term (15–21 years) fertilization experiments. The effects of long-term fertilization on soil As concentrations were not significant in the upland soils. However, in the paddy soils of the Taoyuan station, the application of manure and P fertilizers decreased the As concentrations of the surface soils. Meanwhile, the crop harvest could remove As from the surface soils at low levels of soil As (Fengqiu, Changshu, and Taoyuan stations) and decrease its concentration in the surface soils.

In the upland soils with high As concentrations, wheat may bring risk to human health through food-chain, while maize should be considered as a favorable crop.

Acknowledgments

This research was sponsored by the National Basic Research Program (Grant No. 2005CB121104) and the Natural Science Foundation of China (50621804, 40671172). We are grateful to D. J. Wang at the Changshu station, X. L. Xie at the Taoyuan Station, X. Z. Han at the Hailun station, S. W. Qin at the Fengqiu Station, and B. R. Wang and D. Z. Qin at the Qiyang Station for their assistance in soil sampling.

Conflicts of interest

The authors declare no conflicts of interest.

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