Seasonal effect on the load in soil and subsequent transfer of arsenic to rice

S.M. Imamul Huq, J.C. Joardar and A.F.M. Manzurul Hoque

Abstract

The BR 29 rice variety was cultivated in potted soil (Aeric Endoaquept) for four consecutive seasons, using As-contaminated water in the dry (boro) season and fresh water in the monsoon (aman), in order to observe the effect of alternate irrigation on the As loading in the soil and on its transfer to rice. Soil arsenic content increased significantly ($p = 0.003$ for the first boro season and 0.013 for the second boro season) after each boro season, whereas the soil As was found to have decreased significantly ($p < 0.001$) at the end of both of the aman seasons. The soil As was found to be significantly reduced ($p < 0.001$) at the end of the fourth cropping season (boro–aman–boro–aman). At the end of the fourth cycle, the arsenic loading was found to be almost unchanged. Arsenic in rice grains was detected only when As-contaminated water was used during the boro season. Moreover, the As accumulation in rice grains was lower in the second boro season of the four cropping season cycles.

Key words: arsenic, BR 29 rice, cropping season, irrigation, soil loading

INTRODUCTION

Lowland rice cultivation requires submerged and wet soils: the land is flooded by irrigation water in the dry season when boro rice is grown, and by rainwater in the monsoon when the aman rice is grown (Kyuma 2004). In Bangladesh, about 75% of the total cropped area is used for rice culture, of which approximately 78% is irrigated with groundwater in the dry season. Shallow aquifers are heavily used as the major source of irrigation water for rice cultivation, over about 80% of the total irrigated area (Ministry of Agriculture 2004). Since most of the shallow aquifers of Bangladesh are contaminated with arsenic (Burgess and Ahmed 2006), this irrigation with As-contaminated groundwater carries a risk of soil accumulation of this toxic element, eventually entering the food chain through plant uptake and consumption of these plants by animals (Imamul Huq and Naidu 2005). Irrigation with arsenic-contaminated groundwater is leading to elevated levels of arsenic in paddy soils (Alam and Sattar 2000), which may lead to increased concentrations of arsenic in rice (Duxbury *et al.* 2003; Meharg and Rahman 2003; Imamul Huq and Naidu 2005; Imamul Huq *et al.* 2006; Williams *et al.* 2006); wheat (Imamul Huq and Naidu 2005; Imamul Huq *et al.* 2006); vegetables (Imamul Huq and Naidu 2005; Imamul Huq *et al.* 2006); and other agricultural products (Abedin *et al.* 2002). According to Imamul Huq *et al.* (2006), the total As loading in irrigated soils for a boro rice requiring 1000 mm of irrigation water per season ranges from 1.36 to 5.5 kg/ha/yr. Similarly, for winter wheat that requires 150 mm of irrigation water per season, the As loading from irrigation ranges from 0.12 to 0.82 kg/ha/yr.

Arsenic that enters the soil via groundwater irrigation subsequently accumulates in different parts of the rice plants (Duxbury *et al.* 2003; Meharg and Rahman 2003; Imamul Huq and Naidu 2005; Imamul Huq *et al.* 2006; Williams *et al.* 2006) and in different soil horizons (Imamul Huq *et al.* 2007*a*; Imamul Huq *et al.* 2008) to varying extents. It has been observed, how-

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ever, that arsenic in the topsoil of rice fields increases significantly after the dry-season irrigation with As-contaminated water (Imamul Huq *et al.* 2008). Arsenic thus accumulated in the topsoil would be bioavailable to the next crop of rice, even if the crop is cultivated with arsenic-free irrigation water or with rainwater (Imamul Huq *et al.* 2007*b*). A positive significant relationship between soil As and rain-fed rice-grain As (Imamul Huq *et al.* 2007*b*) observed in sites where As-contaminated groundwater is used during the boro season, indicates the carry-over effect of irrigation-water As to rice cultivated under the alternate (As-free rainwater) irrigation system. No information is so far available on the long-term effects of As accumulation in soils and its subsequent transfer to rice grains of the same variety cultivated under the alternate irrigation system (dry-season irrigation with arsenic-contaminated groundwater followed by rice cultivation with rainwater irrigation). From this standpoint, a single variety (BR 29) of rice was grown in potted soil with alternate watering in order to obtain a clear understanding of the soil As load and of the long-term As transfer to rice (BR 29) in such circumstances.

MATERIALS AND METHODS

The experiment was conducted in the net house of the Department of Soil, Water and Environment, University of Dhaka, Bangladesh.

Soil sample collection and sample preparation

The floodplain of the river Ganges was selected for soil sampling. At the field selected as the sampling site, As-contaminated groundwater has been in use for irrigation for the last five years, and the field is used for both irrigated and rain-fed rice. The site thus selected was Doyarampur village in the Gerda union (local administrative unit) of the Faridpur Sadar upazila (sub-district). The soil is a Grey Calcareous Floodplain Soil in the Ishwardi soil series (SRDI 2005) and is taxonomically referred to as an Aeric Endoaquept (Rahman 2005). The location of the sampling site is 23°33.289' N and 89°54.638' E (Figure 1). The soil is poorly drained, flooded up to 0.61–0.91 m for three to four months, and remains unsaturated for about seven to eight months in the dry season (SRDI 2005).

The soil samples representing 0–15 cm depth from the surface were collected using the composite soil-sampling method as suggested by the Soil Survey Staff of the United States Department of Agriculture (USDA 1951), and the samples were collected before the start of irrigation. The procedures and relevant precautions needed for proper sampling and the preservation of samples were followed, as described in Imamul Huq and Alam (2005).

The collected soil samples were air dried, and the visible roots and debris were removed manually. Some of the larger and massive aggregates were broken by gentle crushing with a wooden hammer, after which the ground samples were passed through a 0.5-mm stainless-steel sieve. The sieved samples were then mixed thoroughly to make the composite sample, and were preserved for laboratory analysis. The remaining soil samples were crushed to smaller clods and passed through a 5-mm sieve. These soils were used for the pot experiment.

Experimental set-up

The experiment was set up under net house conditions. Clay pots of 20-L capacity, with no drainage holes at the bottom, were used for the experiment. Each of the pots was filled with 20 kg of soil, and the pots were arranged randomly in the net house. Rice was grown with As-contaminated irrigation water in the dry (boro) season, and with As-free rainwater in the monsoon season (aman). There was only one treatment for As-contaminated irrigation water (0.5 mgAs/L). The same variety of rice was grown for four consecutive cropping seasons (boro–aman–boro–aman). To simulate the irrigation water, normal tap water, artificially spiked with arsenic salt (sodium meta-arsenite) at the rate of 0.5 mg As/L water, was used, and the same tap water was used for irrigation for rain-fed conditions as well as for the control. Irrigation water was added every second day in the dry season, whereas water was used only when required during a relatively longer dry spell in the wet season. Care was taken not to allow the water to spill over during the wet season. However, in the second aman season (i.e. the fourth growing season) there was a spill-over on one occasion, due to incessant rainfall for two days. A total of twelve pots were used in the experiment: six as control and six for treatment. All the pots were used consecutively for each season. The background As level of the soil was 4.01 mg/kg. This was taken as the control. For each cropping season, the required amount of P, K, and one-third of the required N fertilizer (BARC 2005) were mixed with the soil. Of the remaining two-thirds of the N fertilizer, one-third was applied 35 days after seed sowing, and the final one-third was applied during the panicle initiation stage of the rice plants. The sources of N, P and K were urea, triple superphosphate and muriate of potash, respectively. Operations such as weeding and pest control were practised during the experimental period.

Sample collection from pots

The plants were harvested when the rice was ripening. The roots, straw and grains were collected separately. After harvest, soil samples were also collected from all the pots, and these samples were processed for laboratory analysis by following the same procedure employed earlier to prepare the field soil samples. The plant roots were washed several times with deionized distilled water, in order to remove adhering soil particles from the root surface as quickly as possible after

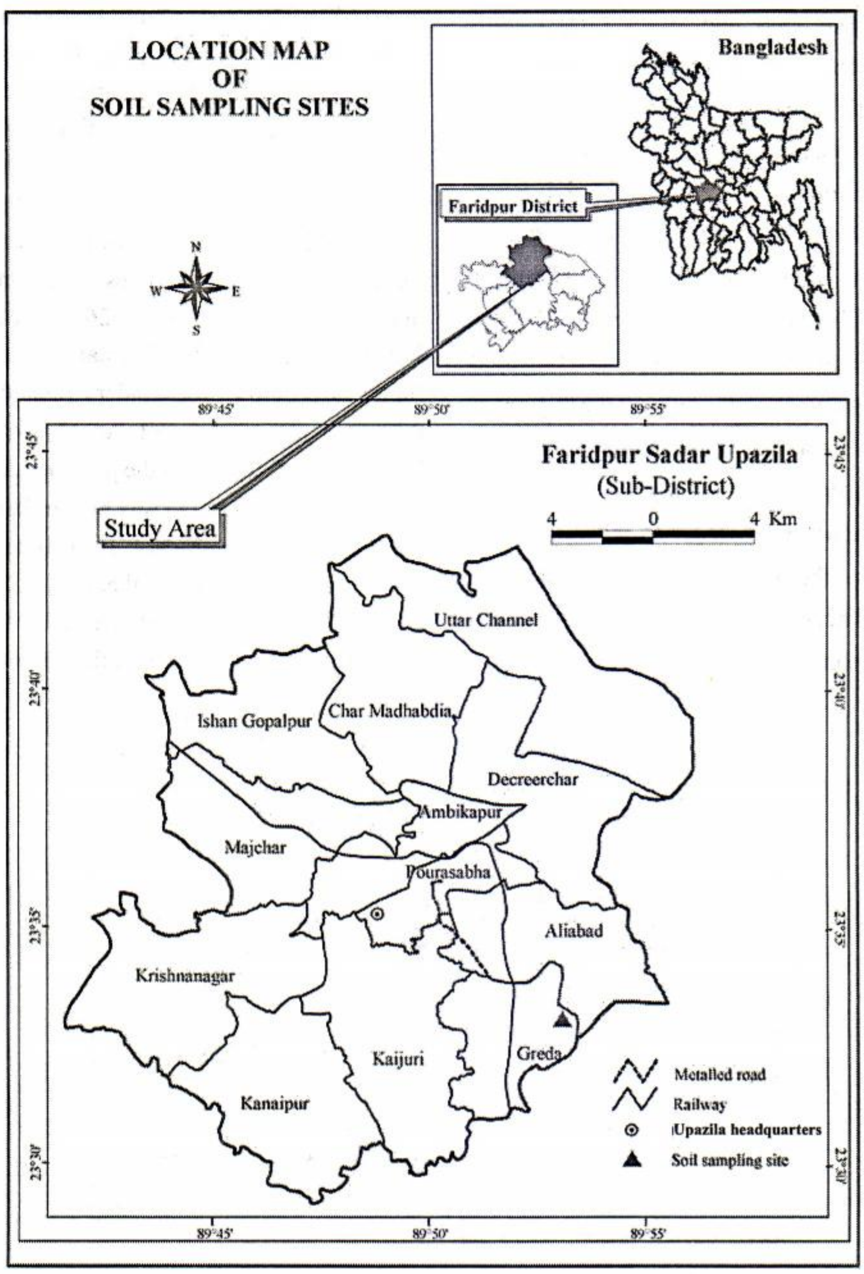

Figure 1. Location map of the sampling site

sample collection. The upper parts of plants were also washed. The fresh weights of the plant samples were recorded. These samples were first air-dried, and then oven-dried at 70 ± 5°C for 48 hours, and the dry weights were recorded. The dried plant samples were ground, sieved through a 0.2-mm sieve, and preserved for analysis.

Laboratory analysis

Some routine analysis of the soil samples was carried out in the laboratory, following prescribed methods as described in Imamul Huq and Alam (2005). Soil arsenic (both pre- and post-experiment) was extracted by digestion with aqua regia ($HCl:HNO_3$, 3:1), whereas the plant arsenic was extracted by digestion with concentrated nitric acid (Portman and Riley 1964). The arsenic in the extract was estimated by hydride generation atomic absorption spectrometer (HG–AAS), with the help of potassium iodide and urea, following calibration of the equipment. Certified reference materials were used throughout the digestion, and were analysed as a part of the quality assurance/quality control protocol. Reagent blanks and internal standards were used where appropriate, to ensure accuracy and precision in the analysis of arsenic. Each batch of ten samples was accompanied by reference standard samples, in order to ensure strict QA/QC procedures.

Data analysis

The experimental data were statistically analysed using the well-known statistical software Minitab 13.0.

RESULTS AND DISCUSSION

Arsenic in soil

The soil As increased significantly ($p = 0.003$) from its initial level after the harvest of the boro season rice (irrigated rice). However, in the control soils where As-free tap water was used, the soil As content was reduced, and this decrease was statistically significant ($p < 0.001$). The arsenic build-up in soil was calculated on the basis of the initial level of As in soil and the As that accumulated in the soil after the crop is removed (Figure 2). The build-up was estimated to be 1.26 kg/ha for the boro season. After the aman rice, the soil As, both in the control and in the treated pots, decreased significantly ($p < 0.001$ for the treatment and $p = 0.005$ for the control) and the values were significantly lower than the initial values ($p = 0.026$ for the treatment and $p < 0.001$ for the control). Thereafter, the pots were used for the next boro season (under irrigated conditions). In this case again, the soil As showed a significant increase ($p = 0.013$) over the previous aman season values. Although this value was higher than the initial soil value, it was not significant. The estimated build-up for this season was lower than that for the first season (1.03 kg/ha). On the other hand, the As in the control soils decreased progressively, and the decrease was statisti-

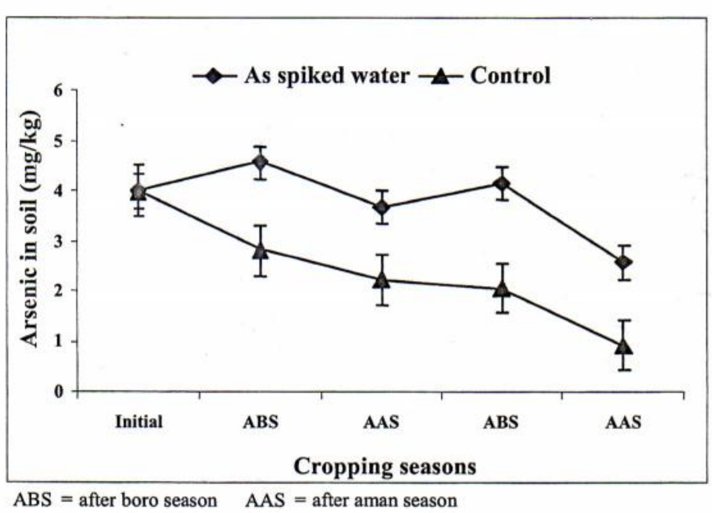

Figure 2. Changes in soil As after the harvest of four rice cropping season under the alternate irrigation system

cally significant ($p = 0.024$). After the fourth cropping season (second aman season), the soil As was reduced drastically and significantly ($p < 0.001$ for the control and $p < 0.001$ for the treatment). This drastic reduction could also be related to excessive rainfall during the fourth season. The reduction in soil As content from its initial value in the four cropping cycles was also significant ($p < 0.001$ for both the control and the treatment).

Arsenic in rice

The experiment involved the cultivation of BR 29 rice in four consecutive cropping seasons on the same soil (boro–aman–boro–aman) with, alternately, As-contaminated irrigation water in the dry (boro) season and As-free rainwater in the monsoon (aman).

It is clear from Table 1 that a greater amount of As accumulated in the different parts of the rice plants when they were grown with As-contaminated water. This trend of higher As accumulation is evident for both of the boro seasons. A very small fraction of As was found in rice grains throughout the four cropping seasons, particularly when it received only As-contaminated water (boro season). Arsenic in the grains of aman rice was below the detection limit (0.02 µg/kg) of the AAS (Varian Spectra 220) that was used. Moreover, a very small fraction of As was detected in rice grains of the control plants in the first boro season. This could be due to the bioavailable fraction of soil As (0.27 mg/kg).

In the boro season, the application of As-contaminated irrigation water provided water-soluble As which may have supplemented the inherent bioavailability of As in the soil to rice, allowing more As to be transferred from soil to rice plants in this case. It needs to be further mentioned that pot experiments tend to increase the uptake of trace elements. This phenomenon could have played an additive role in As accumulation. The As biotransformation, particularly the microbial reduction of the As content in paddy soil (Xie and Naidu 2006), and, among others, the sorption–desorption processes influenced predominantly by the clay content in soil as well as the soil pH, could govern As bioavailability to and consequent accumulation in rice plants (Horswell and Speir 2006). In the rice grown in the aman season, the As transfer from the soil to roots and shoots of the BR 29 was lower than that which occurred in the preceding boro season. This could be related to the use of As-free water. It appears that such water does not provide water-soluble As to supplement the bioavailable fraction of soil As.

Arsenic could be retained in soil by various adsorption processes, which are by no means totally irreversible (McLaren *et al.* 2006). It is therefore likely that an increase of As in the soil, resulting from the application of As-contaminated irrigation water, could be due to the preponderance of As adsorption over the total As loss from the soil via uptake by the rice plant, biomethylation and leaching processes in the dry season. On the other hand, a decrease in As in the soil following application of As-free tap water in the dry season signifies that the desorption of As into solution could be the predominant process making the As more bioavailable to rice, as well as more liable to be volatilized to the atmosphere or leached out of the surface soil. Similarly, the decrease of As in soil after the monsoon would occur as a consequence of As desorption in the soil because As-free water was used.

Table 1. Mean As concentration in different parts (roots, shoots and grains) of BR 29 rice cultivated using alternate irrigation system

Rice cultivation season	Type of irrigation system	Arsenic (mean ± SD) concentration (mg/kg) in BR 29 rice						As load in soil (kg/ha)
		Roots		Shoots		Grains		
		Treatment	Control	Treatment	Control	Treatment	Control	
Boro	As-spiked water	3.79 ± 0.23	2.48 ± 0.12	0.43 ± 0.06	0.26 ± 0.07	0.25 ± 0.04	0.02 ± 0.01	1.26
Aman	Rainwater	2.88 ± 0.13	1.98 ± 0.14	0.26 ± 0.06	0.15 ± 0.04	bdl	bdl	–
Boro	As-spiked water	3.72 ± 0.18	2.73 ± 0.11	1.63 ± 0.11	0.36 ± 0.05	0.07 ± 0.02	bdl	1.03
Aman	Rainwater	2.53 ± 0.10	1.34 ± 0.12	1.25 ± 0.09	0.25 ± 0.05	bdl	bdl	–

bdl = below detection limit (0.02 µg/kg)

Although arsenic sequestration varied in different parts of the same variety of rice in different seasons, arsenic accumulation has always been higher from As-contaminated soils as compared to uncontaminated ones, irrespective of the season. Previous observations by the authors (Imamul Huq *et al*. 2006) that the total soil content has no bearing on arsenic accumulation in plants, have been further substantiated by the present study. The overall tendency for As accumulation to decrease in rice plants cultivated in both contaminated and uncontaminated soils under an alternate irrigation system rather indicates that submergence for a period of time might accentuate the bioavailability of the As and increase the subsequent mobility of the element in the soil–plant system. Yang *et al.* (2002) found a significant reduction in As bioavailability in soils over time. Our study suggests that application of As-free irrigation water to rice fields could substantially decrease the bioavailability of As in soil, thus reducing As uptake by rice. Moreover, our understanding of the process of slow build-up of As in irrigated fields (with As-contaminated water) needs further research. Further field studies should be carried out with more of the boro rice varieties currently cultivated in Bangladesh.

REFERENCES

Abedin, M.J., Cresser, M.S., Meharg, A.A., Feldmann, J. and Cotter-Howells, J. (2002) Arsenic accumulation and metabolism in rice (*Oryza sativa* L.). *Environ. Sci. Technol.*, **36**, 962–968

Alam, M.B. and Sattar, M.A. (2000) Assessment of arsenic contamination in soils and waters in some areas of Bangladesh. *Water Science and Technology*, **42**, 185–193

BARC (Bangladesh Agricultural Research Council) (2005) *Fertilizer Recommendation Guide – 2005* (Miah, M.M.U., Farid, A.T.M., Miah, M.A.M., Jahiruddin, M., Rahman, S.M.K., Quayyum, M.A., Sattar, M.A., Motalib, M.A., Islam, M.F., Ahsan, M. and Razia, S. eds), BARC Soils Publication No. 45. Bangladesh Agricultural Research Council, Dhaka, Bangladesh. 260 pp.

Burgess, W. and Ahmed, K.M. (2006) Arsenic in aquifers of the Bengal Basin: from sediment source to tube-wells used for domestic water supply and irrigation. In: *Managing Arsenic in the Environment: From Soil to Human Health.* (Naidu, R., Smith, E., Owens, G., Bhattacharya, P. and Nadebaum, P. eds), pp. 31–56. CSIRO Publishing, Melbourne, Australia

Duxbury, J.M., Mayer, A.B., Lauren, J.G. and Hassan, N. (2003) Food chain aspects of arsenic contamination in Bangladesh: effects on quality and productivity of rice. *J. Environ. Sci. and Health, Part A: Toxic Hazardous Substances and Environmental Engineering*, **38**, 61–69

Horswell, J. and Speir, T. (2006) Arsenic phytotoxicity: effect on crop yield and crop quality. In: *Managing Arsenic in the Environment: From Soil to Human Health* (Naidu, R., Smith, E., Owens, G., Bhattacharya, P. and Nadebaum, P. eds), pp. 183–207. CSIRO Publishing, Melbourne, Australia

Imamul Huq, S.M. and Alam, M.D. (eds) (2005) *A Handbook on Analyses of Soil, Plant, and Water.* BACER-DU, University of Dhaka, Bangladesh. 246 pp.

Imamul Huq, S.M. and Naidu, R. (2005) Arsenic in groundwater and contamination of the food chain: Bangladesh scenario. In: *Natural Arsenic in Groundwater: Occurrence, Remediation and Management* (Bundschuh, J., Bhattacharya, P. and Chandrasekharam, D. eds), pp. 95–101. Balkema, Leiden, The Netherlands

Imamul Huq, S.M., Joardar, J.C., Parvin, S., Correll, R. and Naidu, R. (2006) Arsenic contamination in food chain: arsenic transfer into food materials through groundwater irrigation. *J. Health Popul. Nutr.*, **24 (3)**, 305–316

Imamul Huq, S.M., Manzurul Hoque, A.F.M., Biswas, A. and Joardar, J.C. (2007*a*) Fate of irrigation water arsenic in soil profile. In: *Proceedings of International Workshop on Arsenic Sourcing and Mobilisation in Holocene Deltas* (Basu, B. ed.), pp. 95–98. School of Fundamental Research, Kolkata, India

Imamul Huq, S.M., Haque, H.A., Joardar, J.C. and Hossain, M.S.A. (2007*b*) Arsenic accumulation in rice grown in aman and boro seasons. *Dhaka Univ. J. Biol. Sci.*, **16** (2), 91–97

Imamul Huq, S.M., Manzurul Hoque, A.F.M., Joardar, J.C. and Shoaib, J.U. (2008) Arsenic movement in the profiles of some Bangladesh soils. *Canadian J. Pure and Applied Sci.*, **2** (1), 251–259

Kyuma, K. (2004) *Paddy Soil Science*. pp. 7–59. Kyoto University Press, Japan and Trans Pacific Press, Australia

McLaren, R.G., Megharaj, M. and Naidu, R. (2006) Fate of arsenic in the soil environment. In: *Managing Arsenic in the Environment: From Soil to Human Health* (Naidu, R., Smith, E., Owens, G., Bhattacharya, P. and Nadebaum, P. eds), pp. 157–182. CSIRO Publishing, Melbourne, Australia

Meharg, A.A. and Rahman, M.M. (2003) Arsenic contamination of Bangladesh paddy field soils: implications for rice contribution to arsenic consumption. *Environ. Sci. Technol.*, **37**, 229–234

Ministry of Agriculture (Sector Monitoring Unit) (2004) *Handbook of Agricultural Statistics*. Ministry of Agriculture, Government of Bangladesh. 81 pp.

Portman, J.E. and Riley, J.P. (1964) Determination of arsenic in seawater, marine plants and silicate and carbonate sediments. *Anal. Chem. Acta,* **31,** 509–519

Rahman, M.R. (2005) *Soils of Bangladesh*. Darpon Publications, Dhaka, Bangladesh. 264 pp.

SRDI (Soil Resource Development Institute) (2005) *Soil and Land Resource Utilization Guide for Faridpur Sadar Upazila Under Faridpur District*. SRDI, Dhaka, Ministry of Agriculture, Government of the People's Republic of Bangladesh. 91 pp.

USDA (1951) *Soil Survey Manual*. Soil Survey Staff, Bureau of Plant Industry, Soils, and Agricultural Engineering, United States Department of Agriculture, Washington. 503 pp.

Williams, P.N., Islam, M.R., Adomako, E.E., Raab, A., Hossain, S.A., Zhu, Y.G., Feldmann, J. and Meharg, A.A. (2006) Increase in rice grain arsenic for regions of Bangladesh irrigating paddies with elevated arsenic in ground waters. *Environ. Sci. Technol.*, **40** (16), 4903–4908

Xie, Z.M. and Naidu, R. (2006) Factors influencing bioavailability of arsenic to crops. In: *Managing Arsenic in the Environment: From Soil to Human Health* (Naidu, R., Smith, E., Owens, G., Bhattacharya, P. and Nadebaum, P. eds), pp. 223–234. CSIRO Publishing, Melbourne, Australia

Yang, J.K., Barnett, M.O., Jardine, P.M., Basta, N.T. and Casteel, S.W. (2002) Adsorption, sequestration and bioavailability of As^{V} in soils. *Environ. Sci. Technol.*, **36**, 4526–4529