3 Arsenic estimation in foodstuffs of arsenic exposed areas in Bangladesh

M. Abdur Rahman Khan
Directorate General of Health Services
Ministry of Health and Family Welfare
Government of Bangladesh
Mohakhali, Dhaka.

Mir Misbahuddin
Division of Arsenic Research
Department of Pharmacology
Bangabandhu Sheikh Mujib Medical University
Shahbag, Dhaka.

M. Siddiqur Rahman
Arsenic Cell
Directorate General of Health Services
Ministry of Health and Family Welfare
Government of Bangladesh
Mohakhali, Dhaka.

Salamat Khandker, Ifthaker-Al-Mahmud
WHO Arsenic Cell
DPHE Bahaban
Kakrail, Dhaka.

Abstract
One thousand samples of different foodstuffs were collected from two Upazillas of Bangladesh for the estimation of arsenic using atomic absorption spectrometer with hydride generator. The mean (± SD) concentration of arsenic in raw rice was 878.5 ± 379.3 µg/kg. Among the foodstuffs studied, lowest concentration of arsenic (61.3 ± 63.7 µg/kg) was found in the leaves of *Convolvus arvensis* (local name- kalmi leaf). Highest concentration of arsenic was found in arum leaf and root (1181.1 ± 658.9 µg/kg and 1153.9 ± 479.7 µg/kg). Although the mean amounts of arsenic in tube well water and soil were several fold higher in Shahrasti Upazilla (Chadpur District) in comparison to Bashail Upazilla (Tangail District), the amounts of arsenic in different foodstuffs were not increased significantly except in arum root. This study suggests that rice as well as vegetables were contaminated with high concentration of arsenic and their concentration was not related with increased concentration of arsenic in soil.

Introduction

Bangladesh is now facing the world's most threatening environmental disaster, the ground water arsenic contamination (IPCS, 2001). It is estimated that about 97% of rural population uses tube well water for drinking and cooking purposes. This increase use of tube well water resulted in an unintended consequences of chronic arsenic poisoning observed since 1993. Arsenic is present in the tube well water in Bangladesh in concentration much higher than the WHO guideline of 10 μg/l and even higher than the standard of 50 μg/l set by the Government of Bangladesh. About half of the total populations are drinking high concentration of arsenic (Mudur, 2000).

Recent reports show that some of our foodstuffs are also contaminated with arsenic (Huq et al., 2003; Meharg and Rahman, 2003; Das et al., 2004). It might be due to high concentration of arsenic in soil where foodstuffs are grown. Only a few studies were done. The numbers of samples were not sufficient as well. So, the extent of contamination is not clear.

The presence of arsenic in foodstuffs is an important factor of contamination as because if we know the arsenic contaminated tube well water we can marked the tube well as red color. That is, one should not drink that water. If the alternate source is available or the people are aware about the hazard of arsenic intake through drinking water and cooking, then the extent of arsenic intake may be reduced. On the other hand, intake of low arsenic containing foodstuffs cannot be ensured in an arsenic endemic area until extensive studies are done. In addition, the total daily amount of arsenic through food is not known.

Foodstuffs grown in arsenic contaminated soil not only consumed by the people of arsenic exposed areas but also by the people of arsenic non-exposed areas in Bangladesh, even some of the vegetables are exported to other countries. A study shows that arsenic content of the vegetables from the United Kingdom was approximately 2 to 3 fold lower than those observed for the vegetables imported from Bangladesh (Al Rmalli et al., 2005). That is, people drinking arsenic safe drinking water have the chance to expose of arsenic contaminated foodstuffs.

The present study detects the extent of arsenic contamination of different foodstuffs of two arsenic exposed areas (Upazillas) in Bangladesh.

Materials and Methods

Study area: Through retrospective study of Bangladesh Arsenic Mitigation and Water Supply Project's tube well screening report, we selected Bashail and Shahrasti Upazilla of Tangail and Chadpur Districts respectively. There was no reported arsenic contaminated tube well in Bashail Upazilla. On the other hand, above 95% tube wells of Shahrasti Upazilla were contaminated with high concentration of arsenic.

Duration of study: The samples were collected during June and July, 2005.

Sample collectors: We selected 5 health workers from each Upazilla for collection of different foodstuffs. The health workers received one day intensive orientation training regarding sample collection procedure. Local Civil Surgeon, Upazilla Health & Family Planning Officer and four Health Inspectors received orientation training too. They monitored the process of sample collection.

Collection of samples: After a discussion with the local key persons, we selected a village purposively. From middle point of the village we selected the direction by coin throwing method. Individual house was rarely found in the villages. There were cluster of houses. From each cluster we selected one house randomly. Every 10^{th} cluster of house from the starting point was selected for sample collection. The number of samples from each house was at least 10. Thus, 500 hundred samples were collected from arsenic exposed area (Shahrasti Upazilla) and another 500 samples from arsenic non-exposed area (Bashail Upazilla). In this connection, it may be mentioned that water, soil and rice samples were the common items. Each sample was collected in propylene bag and identified as code number. All the samples were then transported to the laboratory on the same day of collection. The health workers collected 100 ml of tube well water in a plastic bottle containing 0.1% nitric acid and kept in a refrigerator at 0~4°C until analysis. In total 85 samples of water were collected. About 100 g of surface soil (0~15 cm depth) was collected in propylene bag and kept at room temperature. The total number of soil samples was 73. They were dried and filters before estimation. Ten to 50 gram of raw rice, leafy and non-leafy vegetables, roots and tubers, and fruits were collected in separate propylene bags. These were preserved at -20°C at the Division of Arsenic Research, Department of Pharmacology, Bangabandhu Sheikh Mujib Medical University, Dhaka, Bangladesh where estimations of arsenic were done.

Extraction of arsenic from foodstuffs: At first, each sample was weighed and homogenized by an electric blender and the extraction of arsenic was done with a mixture of water-methanol (50:50, v/v) for 10 hours (D'Amato et al., 2004).

Extraction of arsenic from soil sample: Extractable arsenic of the soil was assessed following aqua-regia digestion of soils. Soil (500 mg) was digested with 10 ml of aqua-regia solution. The procedure was repeated. Following digestion, the extracts were diluted to 50 ml using aqua-regia.

Estimation of arsenic (in brief) (Wang et al., 1994)*):* Each sample was introduced into the hydride generator by continuous flow of 10% hydrochloric acid/3% sulfuric acid and 1% sodium borohydride into a gas-liquid separator. The arsine vapor produced by arsenic and the hydrogen gas (produced by sodium borohydride and acid) was carried by flowing argon gas into quartz T-tube. The tube was heated in an air-acetylene flame, served as atomization cell.

Reagents: Hydrochloric acid, sulfuric acid, nitric acid, perchloric acid, sodium borohydride, sodium hydroxide and potassium iodide were reagent grade and were obtained from E. Merck (Germany). Atomic absorption arsenic (As^{3+}) standard solution (1 mg/ml) was supplied from the Buck Scientific Co., CT, USA. Acetylene gas and argon were from Bangladesh Oxygen Limited (BOL), Dhaka.

Quality control: The limit of detection and limit of quantification were estimated using 3 and 10 standard deviation of low concentration of standard estimated for 6 times. Using our method, the limit of quantification of total arsenic was 0.88 μg/l.

Analyses of data: Statistical analyses were carried out using Statistical Package for Social Science (SPSS), version 9.0, USA. Student's t-test was also used where necessary. Statistical significance was determined by p value less than 0.05. The values are expressed as mean ± SD. One way ANOVA was done for comparison.

Results

Foodstuffs collected from the two Upazillas were shown in Table 1. The collected foodstuffs were raw rice, green leafy and non-leafy vegetables, roots and tubers, and fruits. Varieties of vegetables and fruits are available in Bangladesh but the availability depends on the seasons. Availability of foodstuffs, in other sense, means the consumption by the rural people. Among the leafy vegetables available during the months June and July, 32.2% were indian spinach. Other commonly used leafy vegetables were arum leaf (21.5%), amaranth leaf (15.0%), pumpkin leaf (12.0%) and halancha leaf (10.3%). Commonly used non-leafy vegetables were lady's finger (17.9%), egg plant (16.8%), arum stem (15.8%), green papaya (15.8%), and amaranth (14.8%). Arum is commonly consumed by the rural people of Bangladesh. Its root, stem and leaf are cooked by different methods. The wide use of arum may be due to easy to cultivate and low cost. Only guava, jumbura and papaya were available fruits during the study period. Among them guava was mainly consumed (89.0%).

We collected 42 samples of tube well water from 50 houses of Bashail Upazilla and 43 samples from Shahrasti Upazilla. The mean concentration of arsenic in the tube well water of Bashail Upazilla was 53.8 ± 73.0 μg/l whereas it was 195.6 ± 255.3 μg/l in Shahrasti Upazilla. 36.6% tube well of Bashail Upazilla had the level of arsenic more than 50 μg/l whereas 76.1% tube well of Shahrasti Upazilla had the level of arsenic more than 50 μg/l.

Table 1: Foodstuffs collected from the two Upazillas of Bangladesh

Samples	*Local name*	*Number*	*Percentage*
Raw rice	Chaal	84	84.0
Leafy vegetables			
Amaranth leaf	Data shak	35	15.0
Arum leaf	Kachu shak	50	21.5
Halancha leaf	Halancha shak	24	10.3
Indian spinach	Pui shak	74	32.2
Jute leaf	Pat shak	10	4.3
Kalmi leaf	Kalmi shak	4	1.7
Potato leaf	Alu shak	7	3.0
Pumpkin leaf	Kumra pata	28	12.0
Non-leafy vegetables			
Amaranth	Data	43	14.8
Arum stem	Kachu data	46	15.8
Banana (green)	Kacha kala	12	4.1
Egg plant	Begun	49	16.8
Snake gourd	Chichinga	21	7.2
Dhundal	Dhundal	11	3.8
Ridge gourd	Jingha	11	3.8
Lady's finger	Dharash	52	17.9
Papaya (green)	Pepe	46	15.8
Pumpkin	Kumra	2	0.7
Roots and tubers			
Arum lati	Kachur lati	38	63.3
Arum root	Mun kachu	22	36.7
Fruits			
Guava	Peyara	81	89.0
Jambura	Jambura	5	5.5
Papaya	Pepe	5	5.5

Table 2: Amount of arsenic in different foodstuffs

Foodstuffs	*Botanical name*	*Number of samples*	*Amount of arsenic (μg/kg)*
Amaranth	*Amaranthus esculentus*	43	619.3 ± 453.6
Amaranth leaf	*Amaranthus esculentus*	35	588.7 ± 451.8
Arum lati		38	1,140.6 ± 403.0
Arum leaf	*Colocasia esculenta*	50	1,181.1 ± 658.9
Arum root	*Colocasia esculenta*	22	1,153.9 ± 479.7
Arum stem	*Colocasia esculenta*	46	967.5 ± 445.8
Banana	*Musa sapientum*	12	452.7 ± 345.3
Egg plant	*Solanum xanthocarpum*	49	894.7 ± 488.3
Chili		35	699.1 ± 296.3
Dhundal		11	638.7 ± 206.6
Guava	*Psidium guajava*	82	772.2 ± 427.0
Halancha leaf		24	674.4 ± 398.7
Indian spinach	*Basella alba*	74	771.6 ± 368.1
Jute leaf	*Corchorus olitortus*	10	632.4 ± 448.7
Kalmi leaf	*Convolvus arvensis*	4	61.3 ± 63.7
Lady's finger	*Hibiscus esculentus*	52	729.2 ± 463.0
Lemon	*Citrus limon*	31	619.0 ± 309.3
Onion		2	504.9 ± 185.8
Papaya (green)	*Carica papaya*	47	683.2 ± 413.3
Potato	*Solanum tuberosum*	2	448.5 ± 314.7
Potato leaf	*Solanum tuberosum*	7	671.4 ± 316.1
Pumpkin	*Cucurbita moschata*	2	264.9 ± 43.6
Pumpkin leaf	*Cucurbita moschata*	28	748.0 ± 486.0
Raw rice	*Oryza sativa*	75	878.5 ± 379.5
Ridge gourd		11	716.7 ± 439.3
Snake gourd	*Trichosanthes anguina*	22	686.3 ± 403.6

Highest concentration of arsenic in the tube well water in Shahrasti Upazilla was 1580.0 μg/l. The mean concentration of arsenic in the tube well of Shahrasti Upazilla was about 4-fold higher than that of Bashail Upazilla.

Forty three samples of soils were collected from the 43 houses of Bashail Upazilla and 30 samples from Shahrasti Upazilla. The mean concentration of arsenic in the soil of Bashail Upazilla and Shahrasti Upazilla were 517.0 ± 539.6 and 883.5 ± 621.7 μg/kg respectively. That is, the mean concentration of arsenic in the soil of Shahrasti Upazilla was more than 1.5 fold higher than that of the soil of Bashail Upazilla.

Table 2 shows the amount of arsenic in different foodstuffs collected from the two Upazillas. Among the foodstuffs studied, lowest concentration of arsenic (61.3 ± 63.7 μg/kg) was found in the leaves of *Convolvus arvensis* (local name- kalmi leaf). Highest concentration of arsenic was found in arum leaf and root (1181.1 ± 658.9 μg/kg and 1153.9 ± 479.7 μg/kg). The amount of arsenic in raw rice was 878.5 ± 379.5 μg/kg.

Table 3: Comparison of the amount of arsenic in different foodstuffs collected from two Upazillas

Foodstuffs	*Bashail Upazilla*		*Shahrasti Upazilla*	
	Number of samples	*Amount of arsenic (μg/kg)*	*Number of samples*	*Amount of arsenic (μg/kg)*
Amaranth	12	521.7 ± 236.7	23	642.2 ± 526.1
Amaranth leaf	7	495.9 ± 503.2	28	610 ± 446.9
Arum lati	10	1,222.9 ± 389.1	24	1,106.3 ± 411.8
Arum leaf	13	1,191.9 ± 720.7	37	1,177.2 ± 646.9
Arum root	6	830.9 ± 379.8	16	1,300.8 ± 460.1
Arum stem	29	969.6 ± 480.1	17	963.7 ± 390.9
Egg plant	27	887.1 ± 428.7	22	904.9 ± 571.2
Chili	26	685.0 ± 312.5	9	739.7 ± 256.0
Guava	43	747.7 ± 411.2	39	803.2 ± 450.6
Indian spinach	41	826.8 ± 378.9	33	704.8 ± 354.1
Lady's finger	22	756.4 ± 495.9	30	704.4 ± 441.1
Lemon	20	570.3 ± 279.2	11	707.4 ± 354.3
Onion	0	-	2	504.9 ± 185.8
Papaya (green)	29	704.4 ± 406.0	18	648.4 ± 435.4
Potato	0	-	2	448.5 ± 314.7
Potato leaf	6	708.5 ± 337.7	1	440.9
Pumpkin leaf	25	766.1 ± 515.1	3	608.8 ± 37.2
Raw rice	48	864.0 ± 321.9	36	901.5 ± 461.6
Snake gourd	22	686.3 ± 403.6	0	-

We compared the amount of arsenic in foodstuffs of two Upazillas (Table 3). Data of the samples having sufficient numbers were presented for comparison. The difference in the amount of arsenic in the selected foodstuffs was not statistically significant except in arum roots. The amount of arsenic in the arum root of Shahrasti Upazilla (1300.8 ± 460.1 μg/kg) was significantly higher than the arum root of Bashail Upazilla (830.9 ± 379.8 μg/kg).

The amounts of arsenic in foodstuffs were compared with the amount of arsenic in soil (Table 4). The ranges of arsenic in soil were divided into three groups- 0-500 μg/kg, 501-1000 μg/kg and >1000 μg/kg. High amount of arsenic in soil did not increase the amount of arsenic in foodstuff significantly.

Table 4: Amount of arsenic in foodstuffs grown in soil containing different range of arsenic

Foodstuffs	*Amount of arsenic in soil (μg/kg)*		*Number of Samples*	*Amount of arsenic in foodstuffs (μg/kg)*	
	range	*mean*		*mean*	*SD*
Raw rice	0-500	188.6	26	806.1	266.6
	501-1000	688.1	12	885.8	351.8
	>1000	1455.1	16	866.8	429.6
Amaranth	0-500	234.5	12	638.7	558.5
	501-1000	675.0	5	513.4	138.8
	>1000	1482.6	9	402.5	257.3
Amaranth leaf	0-500	211.3	10	643.4	427.0
	501-1000	619.3	2	848.3	270.1
	>1000	1452.8	11	573.4	519.3
Arum leaf	0-500	243.5	16	1150.7	584.3
	501-1000	698.9	4	1226.9	311.4
	>1000	1507.4	13	1191.2	638.5
Arum lati	0-500	255.7	13	1317.2	399.7
	501-1000	698.9	4	944.8	420.9
	>1000	1522.4	10	1092.5	382.5
Arum root	0-500	231.8	6	1186.4	473.4
	501-1000	738.8	2	1529.0	18.9
	>1000	1387.9	3	1018.3	547.6
Arum stem	0-500	186.2	20	955.1	447.2
	501-1000	694.4	6	894.4	405.4
	>1000	1563.9	6	1025.9	466.3
Egg plant	0-500	205.5	24	925.9	529.7
	501-1000	635.8	4	806.2	242.9
	>1000	1570.7	6	720.1	425.3
Halancha leaf	0-500	203.0	14	736.4	403.9

	501-1000	585.2	2	638.1	36.1
	>1000	1439.7	2	1065.3	259.0
Indian spinach	0-500	213.2	32	815.7	348.3
	501-1000	724.8	9	896.9	493.1
	>1000	1463.7	17	817.7	349.8
Lady's finger	0-500	223.2	17	654.0	450.0
	501-1000	739.1	4	1057.0	370.9
	>1000	1475.6	10	799.3	637.1
Papaya (green)	0-500	222.9	16	670.4	360.8
	501-1000	750.0	7	547.1	261.7
	>1000	1512.1	8	561.0	291.1
Pumpkin leaf	0-500	151.2	11	728.0	317.6
	501-1000	684.2	7	528.0	575.1
	>1000	1602.0	3	1337.7	418.5
Snake gourd	0-500	115.1	8	700.1	425.0
	501-1000	698.1	4	409.0	251.1
	>1000	1463.3	5	693.5	255.7

Discussion

Bangladesh is typically a rice growing country. About 80% of the total agricultural land (61% of the land area of the country) is under rice cultivation. Bangladeshi people are mainly consuming rice and vegetables (Jahan and Hossain, 1998). Fish, meat, egg and milk product are less than 15% of the total intake of foodstuffs. Rice comprises 73% of a Bangladeshi's calorific intake (del Ninno and Dorosh, 2001) whereas in USA it is only 5% of mean food intake (Tao and Bolger, 1999). The mean concentration of arsenic in raw rice was 878.5 μg/kg. This concentration is actually 10~30% higher due to processing of raw rice by arsenic contaminated water (Misbahuddin, 2003). The mean concentrations of arsenic in the rice of different districts of Bangladesh were reviewed (Williams et al., 2005). The range of arsenic was 100 to 950 μg/kg. Our data is within this range.

A wide variety of vegetables are available in Bangladesh that depends on the seasons. Most of the vegetables that we collected during June and July contained high amount of arsenic. More than 1 mg of arsenic/kg was found in arum which is the common, easy and widely cultivated vegetable in rural Bangladesh. All the parts of this vegetable (leaf, stem and root) are edible. Huq et al., (2003) also showed the presence of large amount arsenic in arum (15,970 μg/kg) which is about 15 fold higher than our result. In another study, Alam et al., (2002) showed the amount of arsenic in arum to 1240 μg/kg which is similar to our data. Among the vegetables in UK imported from Bangladesh, arum contained highest concentration of arsenic (540 μg/kg)(Al Rmalli et al., 2005).

We collected the samples from two Upazillas- one is arsenic-affected and another is not based on the survey conducted by Bangladesh Arsenic Mitigation and Water Supply Project. Our findings showed that the water of 36.6% tube well of arsenic unaffected area had the level of arsenic more than 50 μg/l. This discrepancy may be due to the method of estimation of arsenic. The mitigation project used the kit method whereas we estimated by atomic absorption spectrometer. Kit method is less sensitive than the atomic absorption spectrometer.

The average arsenic contents in the soils of Bashail and Shahrasti Upazillas were 517.0 ± 539.6 μg/kg and 883.5 ± 621.7 μg/kg respectively. Huq et al (2003) also estimated the amount of arsenic in different districts of Bangladesh and showed that it was less than 10 mg/kg. The amount of arsenic in soil of Shahrasti Upazilla was almost double the value in Bashail Upazilla. But the average amount of arsenic in foodstuffs of Shahrasti Upazilla was not significantly differed from the foodstuffs of Bashail Upazilla. The reason why the increased amount of arsenic was not accumulated within the foodstuffs is not clear. It may be due to the presence of some protective mechanism.

High concentration of lead and copper were detected in the vegetables available the Dhaka city, which is not a arsenic affected area (Hossain and Bodruzzaman, 2002). Studies are required to examine the amount of these heavy metals in the foodstuffs in the arsenic-affected areas as well as their relationship with the extent of arsenic absorbed within the body.

Acknowledgement

This work was supported by a grant from World Health Organization, Bangladesh (under ICP-budget). We thank Dr. Deoraj Caussy (epidemiologist, SEARO) and Dr. Andrew Trevett (WHO Advisor for Environmental Health) for their critical comments of the study.

References

Al Rmalli SW, Haris PI, Harrington CF, Ayub M. A survey of arsenic in foodstuffs on sale in the United Kingdom and imported from Bangladesh. Sci Total Environ. 2005; 337: 23-30.

Alam AMS, Islam MA, Quader MA, Hossain MG, Siddique MNEA. Level of arsenic in water, soil and food samples in an arsenic affected area. In: Bangladesh environment 2002. Ahmed MF, Tanveer SA, Badruzzaman ABM. ed. Dhaka, ITN-Bangladesh, 2002, pp 161-66.

D'Amato M, Forte G, Caroli S. Identification and quantification of major species of arsenic in rice. J AOAC Int. 2004; 87: 238-43.

Das HK, Mitra AK, Sengupta PK, Hossain A, Islam F, Rabbani GH. Arsenic concentrations in rice, vegetables, and fish in Bangladesh: a preliminary study. Environ Int. 2004; 30: 383-87.

Del Ninno C, Dorosh PA. Averting a food crisis: private imports and public targeted distribution in Bangladesh after the 1998 flood. Agric Econ. 2001; 25: 337-46.

Hossain MA, Badruzzaman ABM. Analysis of heavy metals in vegetable samples in some local Bazaars. In: Bangladesh environment 2002. Ahmed MF, Tanveer SA, Badruzzaman ABM. ed. Dhaka, ITN-Bangladesh, 2002, pp 558-70.

Huq SMI, Naidu R. Arsenic in groundwater of Bangladesh: contamination in the foodchain. In: arsenic contamination: Bangladesh perspective. Ahmed MF. Ed. Dhaka, ITN-Bangladesh, 2003, pp 203-26.

IPCS. Arsenic and arsenic compounds. Environmental health criteria 224. 2nd ed. Geneva, World Health Organization, 2001: pp 7-8.

Jahan K, Hossain M. Nature and extent of malnutrition in Bangladesh. Bangladesh National Nutrition Survey 1995-96. Dhaka, University of Dhaka, 1998. pp 13-18.

Meharg AA, Rahman M. Arsenic contamination of Bangladesh paddy field soils: implications for rice contribution to arsenic consumption. Environ Sci Technol. 2003; 37: 229-34.

Misbahuddin M. Consumption of arsenic through cooked rice. Lancet. 2003; 361: 435-36.

Mudur G. Half of Bangladeshi population at risk of arsenic poisoning. Br Med J. 2000; 359:1127.

Tao SSH, Bolger PM. Dietary arsenic intakes in the United States: FDA total diet study, September 1991-December 1996. Food Addit Contam. 1999; 16: 465-72.

Wang CT, Chang WT, Huang CW, Chou SS, Lin CT, Wang RT. Studies on the concentrations of arsenic, selenium, copper, zinc and iron in the hair of Blackfoot disease patients in different clinical stages. Eur J Clin Chem Clin Biochem. 1994; 32:107–11.

Williams PN, Price AH, Raab A, Hossain SA, Feldmann J, Meharg AA. Variation in arsenic speciation and concentration in paddy rice related to dietary exposure. Environ Sci Technol. 2005; 39: 5531-40.