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Arsenic (As) Contamination in Different Food and Dietary Samples from Several Districts of Bangladesh and Arsenic (As) Detection, Mitigation and Toxicity Measurement and impact of Dietary Arsenic Exposure on Human Health

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Abstract

Objective: To determine the level of arsenic concentration in vegetables and other food categories in three selected areas of Pabna district and to estimate quantitatively the dietary arsenic exposure in one of the arsenic contaminated areas of Bangladesh.

Materials and Methods: The study was conducted in Char Ruppur, Char mirkamari and Lakshmikunda village of Ishwardi Upzila in Pabna district. Ishwardi (Town) consists of 12 wards and 37 mahallas. Arsenic was detected in the ADM Lab, Department of Pharmacology, Bangladesh Agricultural University, Mymensingh with Hydride Generation Atomic Absorption Spectrophotometer (HG-AAS; PG-990, PG Instruments Ltd. UK). Arsenic was detected by forming AsH_3 at below pH 1.0 after the reaction of As with a solution of sodium borohydride ($NaBH_4$), sodium hydroxide (NaOH, M=40,000 g/mol,) and 10% HCl. In this test, standard was maintained as AsV ranging from 0 to 12.5 µg/L.

Results: A total of 120 vegetable samples, 15 rice samples and 15 fish samples were collected from five different markets of three different villages of Pabna district and were tested for arsenic concentration. Findings demonstrated that the mean concentration of As in leafy vegetables (0.52 µg g^{-1}) was significantly higher compared to those found in fruity (0.422 µg g^{-1}) and root & tuber vegetables (0.486 µg g^{-1}).

Conclusion: Underground Contaminated water was the major source for the As contamination of various products in Pabna. The arsenic levels were found higher among the leafy vegetables samples in comparison to fruit and root & tuber vegetables. Further studies will be conducted to search the genetic risk factors of arsenic toxicity in the population of the mostly affected people.

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Introduction:

Arsenic is recognized as one of the world's greatest environmental hazards, threatening the lives of several hundred million people. However, in terms of population exposure the most severe case of such contamination is observed in Bangladesh where it is considered to be the largest mass poisoning in history [21]. Out of 465 upazillas (sub-districts) in Bangladesh, 270 are affected with high levels (>50 ppb or µg L-1) of arsenic [3] and it was estimated that over 35 million Bangladeshis are at augmented risk of arsenic poisoning [16], [19]. Furthermore, evidences of high soil and plant arsenic levels have been found due to the introduction of arsenic-contaminated water in crop fields. Again, there is a possibility of using the contaminated water for cooking. As a result, even more people will become exposed to toxic level of arsenic through food chain.

The problem of arsenic in ground water is more severe in the south and south-eastern division of Bangladesh and occurs naturally [9]. Till now, there are two popular hypotheses regarding the origin of arsenic in Bangladesh ground water. According to the first hypothesis, arsenic is derived from 'erosion of lithified sediments and crystalline rocks of the Himalayan range and adjacent regions' [5], [17] and was deposited on floodplains across the lower delta where it was subjected to intense chemical weathering. According to Bhattacharya and McArthur, the mechanism of the presence of high arsenic in ground water is due to the reduction of Fe (III) oxides and release of associated as to solution [4], [11].

So far, many studies have been performed to assess the scale of the problem, its possible consequence and remediation or minimization measures. To date the focus of arsenic poisoning has been mainly on exposure through drinking water sources. However, due to the widespread use of ground water in drinking, cooking, agriculture, etc. arsenic can reach the human body from many possible routes. And the dietary arsenic concentration often exceeds the WHO recommended provisional maximum tolerable daily intake of ingested inorganic arsenic which is 2.14 μg/kg of the body mass [20].

As part of this study, vegetables, rice and fish samples were collected from Char Ruppur, Char mirkamari and Lakshmikunda; previously identified arsenic affected villages of Ishwardi upzilla of Pabna district (Table 1). Arsenic concentration was determined by hydride generation atomic absorption spectrophotometer. Those villages of Ishwardi upzilla are highly affected with As contamination in groundwater and the contaminated groundwater is used for irrigation purposes. The study was carried out based on the information provided by Upzilla Shastho Complex, Ishwardi. The objectives of the study was to determine the level of arsenic concentration in vegetables and other food categories in three selected areas of Ishwardi upazilla, Pabna district and to estimate quantitatively the dietary arsenic exposure in one of the arsenic contaminated areas of Bangladesh.

Materials and Methods:

The study was conducted in Char Ruppur, Char mirkamari and Lakshmikunda village of Ishwardi Upzila in Pabna district. Ishwardi (Town) consists of 12 wards and 37 mahallas. The area of the town is 23.73 sq km. The town has a population of 63106; male 51.64%, female 48.36%. The density of population is 2659 per sq km. Literacy rate among the town people is 51.8%. Bangladesh Sugarcane Research Institute (BSRI), Regional Agricultural Research Station (RARS, BARI), Pulse Research Centre, Silk Seed Store and many industrial establishments are located in the town. Main crops are paddy, wheat, sugarcane, betel leaf, potato, onion and brinjal.

Sampling and pretreatment

Fresh samples of some common vegetables were collected in January 2009. Eight different vegetables of the following food groups were studied: leafy vegetables (Kachu, cabbage,), fruity vegetables (tomato, bean, chilli), root and tuber vegetables (potato, radish, onion).Rice from local market was collected and two types of fish was also collected. Vegetables from local market were selected using a random sampling procedure. They were collected by hand using vinyl gloves, packed into zipper bags and brought to the laboratory. The cleaning of samples was performed by shaking and in the case of potatoes, taro, and cabbage by means of a dry brush. The samples were washed with distilled water to remove dusts adhering on the surface, cut into small pieces, dried in an oven at 65°C for 48 h, and then ground to powder and used for arsenic analysis. The leaves from onion, cabbage, taro leaf and chilli were taken and the non-edible parts of potato and bean were removed using a knife. The edible portions of vegetables were washed three times with distilled water and finally rinsed with deionised water and dried in an oven at 65°C, ground using a grinder and used for analysis.

The rice samples were collected by random sampling procedure. After collection they were cleaned with distilled water and finally rinsed with deionized water before oven drying and also dried for 48h at 65°C and then ground to powder, used for analysis. Fish samples were cleaned with distilled water and flesh part was cut into small pieces before digestion.

Digestion of samples

The digestion tubes were cleaned with detergent and then immersed in 1% hydrochloric acid (HCl) overnight at room temperature. In the following day it was first washed with tap water and finally washed with deionized water and dried in a digital electric oven (Memmert,) at 150-200 0C until drying. Each sample was marked with sample identification number. 0.5g of each sample was taken in separate digestion tube, then 5 ml of digesting agent- 69% nitric acid (HNO3) was added to each digestion tube containing sample and left overnight at room temperature. Following the similar procedure, 5 ml of same acid mixture was taken in another empty digestion tube (without sample) for blank determination (as control).

On the next day, the content in the digestion tubes was heated by placing these tubes in a block digester (M-24 plazas/samples, J.P. Selecta, Spain) at 118°C for 4 hours until nitrous oxide fume emission (brown coloured fume) stopped. During the heating period every digestion tubes were gently shaken several times to make the contents homogeneous and to enhance fume release. Then the samples were cooled to room temperature. Following cooling, 2 ml of 30% hydrogen peroxide (H_2O_2) was added slowly to each digestion tube and shaken gently then heated again at the same temperature (118°C) for 1 hour and left for cooling to room temperature. Then again 2 ml of 30% H_2O_2 was added slowly to each digestion tube and heated further at 118°C until the colorless watery solution appeared and left for cooling to room temperature. After completion of the digestion process the samples were then ready for dilution.

Dilution of digested samples

After digestion, the samples were diluted individually by adding deionized water separately in 25 ml calibrated volumetric flask. After dilution, each sample was filtered individually with filter paper (Whatman 42) into correspondingly marked sterile 30 ml screw capped sterile plastic vials and preserved at 4°C in refrigerator until tested for arsenic.

Measurement of arsenic

Arsenic was detected in the ADM Lab, Department of Pharmacology, Bangladesh Agricultural University, Mymensingh with Hydride Generation Atomic Absorption Spectrophotometer (HG-AAS; PG-990. Arsenic was detected by forming AsH_3 at below pH 1.0 after the reaction of As with a solution of sodium borohydride ($NaBH_4$), sodium hydroxide (NaOH, M=40,000 g/mol,) and 10% HCl. In this test, standard was maintained as AsV ranging from 0 to 12.5 μg/L.

Table 1. **Information about arsenic contaminated area of Ishwardi upzilla**

Area/ Village/Union	Population	Number of tube-wells	Number of contaminated tube-wells	Percentage of contamination	Persons affected
Solimpur Union in Ishwardi upzilla					
Boroichara	3527	15	4	26.66	5
Shekherdair	1775	7	2	28.57	12
Kolerkandi	539	8	3	37.55	45
Charmirkamari	4611	39	5	12.82	-
Charsilimpur	1935	9	3	33.33	-
Kathalbaria	893	8	3	37.55	-
Jagannathpur	2937	19	6	31.57	-
Dashuria Union in Ishwardi upzilla					
Nawdapara	2321	26	4	15.38	-
Dikshaildawan	2115	32	2	6.25	-
Dadpur	1198	4	1	25.00	-
Lakshmikunda Union in Ishwardi upzilla					
Bilkada	22536	30	10	33.33	40
Sahapur Union in Ishwardi upzilla					
Gorgori	1368	17	8	47.05	-
Rahimpur	1596	20	9	45.00	2
Sahapur	1442	58	15	25.86	13
Pakshi					
Charruppur	16360	161	45	27.95	150
Baghail	10850	114	25	21.92	20

Results:

A total of 120 vegetable samples, 15 rice samples and 15 fish samples were collected from five different markets of three different villages of Pabna district and were tested for arsenic concentration. Experimental results showed that there was a wide variation in As concentrations of different type of vegetables (Table 2). Arsenic concentration of leafy vegetables ranged from 0.366 μg g^{-1} in cabbage to 0.674 μg g^{-1} in taro. Among the fruity vegetables the concentration varied between 0.298 μg g^{-1} in tomato to 0.552 μg g^{-1} in bean and in case of root and tuber vegetables it was μg g^{-1} in radish to 0.56 μg g^{-1} in onion. The vegetables followed the order: taro (0.674 μg g^{-1}) > onion (0.56 μg g^{-1}) > bean (0.552 μg g^{-1}) > potato (0.46 μg g^{-1}) > radish (0.438 μg g^{-1}) > chilli (0.418 μg g^{-1}) > cabbage (0.366 μg g^{-1}) > tomato (0.298 μg g^{-1}). The mean concentration of As in leafy vegetables (0.52 μg g^{-1}) was significantly higher compared to those found in fruity (0.422 μg g^{-1}) and root & tuber vegetables (0.486 μg g^{-1}).

Table 2. Arsenic concentration in different types of vegetables, rice and fish samples

Sample	Arsenic concentration in μg/g (Mean. ± S.E)
Leafy vegetables	
Taro	0.673±0.071
Cabbage	0.367±0.038
Fruity vegetables	
Tomato	0.299±0.032
Bean	0.552±0.063
Chilli	0.418±0.086
Root and tuber vegetables	
Potato	0.460±0.068
Radish	0.438±0.077
Onion	0.566±0.052
Rice	0.454±0021.
Fish	0.292±0.015

Discussion:

Leafy vegetables like taro leaf, bottle gourd and pumpkin leaves are very abundant in Bangladesh and constitute a significant portion of low income diet. But high arsenic levels have been reported in such vegetables from many studies [13], [14], [15], when contaminated water is used for irrigation for these vegetables, water on leaves dries up leaving an arsenic residue.

Hence, high arsenic concentration builds up on leaf surfaces. Fruity vegetables like brinjal, tomato, various gourds, cucumber, plantain etc. are considerably costly and also contain less arsenic. But high arsenic levels have been reported in such vegetables from many studies (Roychowdhury et al. 2002; Shah et al, 2004; Heikens, 2006). According to Roychowdhury et al. (2003), when contaminated water is used for irrigation for these vegetables, water on leaves dries up leaving an arsenic residue.

The high AS concentration in vegetables might be due to high levels of As (415.34 μg/L) in water irrigating the vegetable crops. The ground water in Bangladesh is important considering its use for drinking and irrigation. It has been estimated that 95% of the rural people depend on tube well waters for drinking and cooking purposes. Based on population data and use of contaminated water, Lokuge et al. (2004) estimated that about 24% of people of Bangladesh use drinking water having arsenic above the Bangladesh permissible limit. Yu et al. (2003) surveyed 34 Geological-Geographic regions of Bangladesh for ground water arsenic as well as for the incidence of toxicity symptoms. They concluded that about 68.3% of the population is exposed to arsenic levels above 50 μg L^{-1} and about 1.8 million people suffer from arsenic related diseases (arsenicosis), while another 130 thousand suffer from other arsenic-related cancers.

Average AS concentration in rice was found to be 0.454 μg g^{-1} which is higher to those reported by Bae et al. (2002), Duxbury (2003), Williams *et al.* (2005), Smith *et al.* (2006 who documented As concentration in raw rice between 0.043 and 1.835 μg g^{-1}.The variations were observed between varieties, locations of cultivation and seasons of cultivation [2], [6], [16], [18].

Table 3. Ground water Arsenic concentration in three villages of Ishwardi upzilli, Pabna

Well owner Well type: Shallow tube well		Well depth(Feet)	Arsenic concentration in μg/L
Village: Char Ruppur			
	Well A	180	1111.6
	Well B	110	1053.9
	Well C	150	721.7
	Well D	60	714.4
	Well E	90	637.0
	Well F	165	613.9
	Well G	150	613.9
Village: Char Mirkamari			
	Well A	55	513.9
	Well B	100	332.8
	Well C	105	344.6
	Well D	180	121.5
	Well E	90	121.5
	Well F	80	113.8
Village: Lakshmikunda			
	Well A	75	106.1
	Well B	80	98.9
	Well C	120	90.7
	Well D	65	90.7
	Well E	90	75.3

Average AS content of fish samples was 0.292 μg g^{-1} within the range of other documented reports. Only two studies dealing with arsenic content of fresh water fish of Bangladesh have so far been found. The average arsenic content was 0.19 μg g-1 (0.04 – 0.58 μg g^{-1} As) the highest being reported in Bombay duck (Harpodon nehereus).

An adult male consumes 591 g rice, 160 g vegetables, 2.7 L water, and 28 g fish per day [7]. The potential maximum tolerable daily intake (PMTDI) for an adult (70 kg) is 140μg d^{-1}. From available data it was found that only by consuming rice, water, vegetables and fish a person consumes an amount of AS exceeding the PMTDI value. Therefore, dietary ingestion of AS by the people in these areas is alarming.

Conclusion

Underground Contaminated water was the major source for the contamination of various food products in Pabna and upazila. The arsenic levels were found higher among the leafy vegetables samples in comparison to fruit and root & tuber vegetables. These finding highlight the need of water treatment plant for the irrigational purposes in order to limit this major problem from further progression.

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