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Effect of arsenic-contaminated water on food chain in Bangladesh: Analysis of arsenic in soil, water and plants

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Abstract

Arsenic contamination of groundwater is a serious environmental hazard and national health problem in Bangladesh. Arsenic has contaminated the groundwater in about 85% of the total area of Bangladesh. It is accumulating in human bodies through the intake of contaminated drinking water and its presence in the food. This paper presents a brief overview of an analysis of soil, water and plant samples in affected areas. The arsenic levels in soil varied substantially in different locations and also in rice growing seasons. The accumulation of arsenic was higher in irrigated boro (December-June) season than rainfed aman (July-November) season. The level of arsenic ranged from 7.37-10.97 mg kg^{-1} in study areas. The highest concentration was found in Monirampur upazilla in Jessore district in boro season followed by Chapainawabgonj sadar in Chapainawabgonj district in the same season. The successive irrigations with arsenic contaminated water increased the level of arsenic accumulation of soil in boro season. The arsenic concentration in water also varied greatly in different locations of Bangladesh. Out of 8 locations, the tubewell water of 6 locations was found to be higher than the safe limit (0.05 mg L^{-1}). The tubewell water from two locations of Chapainawabgonj was highly contaminated having concentrations of 0.48 mg L^{-1} and 0.46 mg L^{-1}, respectively. The accumulation of arsenic in rice grain was lower than the rice straw in all cases. The contaminated rice straw might increase the arsenic in cattle raised in areas where straw is used as feed. The leaf vegetables amaranth had higher arsenic accumulation than the fruit vegetables tomato, brinjal. Therefore, the use of leaf vegetables in arsenic contaminated areas would greatly increase health concern.

Key words: Arsenic, food chain, soil, water, rice, vegetables, contamination, Bangladesh.

Introduction

Groundwater is the main source of drinking water for the people of Bangladesh. About 97% of the population of the country use tubewells to obtain water for drinking and cooking purposes [7]. Tubewells have been used in Bangladesh since the 1940s. Previously drinking water came from rivers, lakes and shallow wells, but epidemics of cholera and diarrhea were traced to these sources. Tubewell water drastically reduced the rate of water–borne diseases but the current arsenic crisis overshadows that success. Recent measurements show that in many parts of the Ganges and Brahmaputra basin, more than 60% of shallow and deep tubewells contain arsenic above the WHO guideline of 0.01 mg L^{-1} [20]; in addition, more than 30% of the tubewells contain arsenic above the Bangladesh standard of 0.05 mg L^{-1} [6, 7]. Chakraborti [6] showed that the groundwater of hill tracts and tablelands (Barind Tract, Madhupur Garh and Lalmai Hills) was almost free from arsenic contamination but flood plain and deltaic region with coastal belt was highly contaminated. Nowadays, the arsenic contamination of groundwater in Bangladesh is suspected to be the biggest calamity of arsenic in the world [8]. People are getting contaminated by drinking underground water and consuming food with high levels of arsenic. Huq et al. [12] have shown that arsenic may accumulate in the edible parts of some leafy vegetables. During the past few decades, hundreds of newspapers have reported contamination of water in different areas of Bangladesh, but limited studies have been conducted to assess the presence of arsenic in food chain [5, 12, 14]. However, more studies are needed to develop a reliable database on the presence of arsenic in irrigation water, irrigated soil and food samples, and to assess their interrelationships. The objectives of our present study are to: 1) determine the amount of arsenic in soil of some selected upazillas in Bangladesh, 2) examine water from tube wells in those areas and 3) determine the levels of arsenic in rice and vegetables.

Materials and Methods

Collection of samples: Soil samples were collected from fields in aman rice, October 2000, and boro rice, June 2001. A similar time schedule was followed for rice and water sample collection. The samples were taken during the aman and boro seasons from the same plots. Samples were collected from five points of the selected plots: two on both diagonals (4), one meter from the corners, and one on the intersection points of the diagonals. Samples were collected at a 0-30 cm depth using a soil auger from the selected locations of all the plots. Selected sites were in Gopalgonj sadar, Muksudpur in Gopalgonj, Jessore sadar, Monirampur in Jessore, Pirgacha in Rangpur, Rajarhat in Kurigram, Charghat in Rajshahi and Chapainawabgonj sadar upazillas in the Chapainawabgonj district. Samples were packed in polythene bags and labelled; 5 g of each sample was brought back to the laboratory for chemical analysis at the Department of Environmental Planning in Rural Areas in Tottori University, Japan. The tubewell water was collected in 100 ml plastic bottles from 10 locations at 8 selected upazillas. Bottles were rinsed 2-3 times with the same tubewell water before sampling. Water samples were collected 10-15 minutes after starting the tubewells. Then the bottles were properly labelled, tightly sealed and brought back to the laboratory for arsenic detection. Samples of roots, straw and grains of rice were collected from five locations in Gopalgonj district in an area of 1 m^2 during harvesting from selected plots. Samples were oven-dried and ground and 5 g of each sample was placed in a properly labeled paper bag for analysis in Japan. For vegetables, the selected crops eggplant, tomatoes and amaranth were grown in six plots; three plots were irrigated with arsenic contaminated water and the other three plots with good quality water. Grain, straw and root samples of these crops were also analyzed for arsenic.

Analysis of soil, water and plants: The study was conducted at the Department of Environmental Planning in Rural Areas, Tottori University. X-ray Fluorescence Spectroscopy was used for the analysis of samples DBDTC (dibenzyldithiocarbamate) method (precipitation of heavy metals with DBDTC) was carried out for analyzing the samples. In this method, 1 g of soil/plant was placed in a 200-mL beaker and digested with 10 mL of concentrated nitric acid and 20 mL of hydrochloric acid, and evaporated to 15 mL. Twenty mL of HNO_3 was added again to the sample and evaporated to 20 mL. The beaker side was rinsed with little water and 50 mL of distilled water was added before samples were filtrated and evaporated to 1–2 mL. After adding 10 mL of 10% HCL, 30 mL of 50% HCL and 40 mL of 4-methyl,2-pentanon, the lower portion of the samples was collected and volume increased to 100 mL with distilled water. Twenty mL was taken from the samples and diluted 5 times for the determination of arsenic concentration in soil and plants. Three mL of cobalt solution and 5 mL of buffer solution were added to the sample and the pH was adjusted to 4–5. The samples were incubated for 15 minutes at 30°C. Then, 10 mL of DBDTC was added and solution was filtered with 47 mm filter paper. Further treatment was needed for pentavalent arsenic (total volume of filtered solution was used). Both of the filter papers (tri and penta) were dried in an oven and samples were subjected to X–ray fluorescent analysis. To determine arsenic levels in water, 100-mL water samples (50 mL of water sample + 50 mL of distilled water) were placed in 200-mL beaker and digested with HNO_3 and evaporated to 15 mL. Twenty mL nitric acid was added again into the sample and evaporated to 20 mL. The sample was increased to 100 mL with distilled water and was followed by the same procedure as that for the soil and plant samples. A calibration curve was prepared with 0, 0.02, 0.05, 0.1 and 0.5 mg L^{-1} standard solutions of arsenic; values from the analysis were applied to the calibration curve to calculate the concentration of arsenic in water, soil and plant samples.

Results and Discussion

Arsenic levels in soil: The levels of arsenic in soil at 8 locations from 8 upazillas are shown in Fig. 1. The arsenic levels in Chapainawabgonj varied from 7.91 to 11.38 mg kg^{-1} during aman and from 7.82 to 11.19 mg kg^{-1} during boro season. Soil in Chapainawabgonj in both the aman and boro fields was judged to be slightly contaminated as described by Huq et al. [12]. According to his finding, less than 10 mg kg^{-1} of arsenic in soil is below the level of contamination. The arsenic levels in soil in Ghonapara at Gopalgonj sadar during aman and boro were 8.52 and 9.74 mg kg^{-1} respectively, which were below the level of contamination. In Nayakandi at Muksudpur upazilla, the level of arsenic during aman was slightly contaminated (9.69 mg kg^{-1}). During boro the levels of arsenic was 10.14 mg kg^{-1}, which was also slightly contaminated. The arsenic levels in Sajiali at Jessore sadar during aman and boro were 9.33 mg kg^{-1} and 10.45 mg kg^{-1}, respectively. Therefore, we concluded that the levels of arsenic contamination increased gradually in boro season. In Dolaria at Monirampur upazilla, the arsenic levels during aman and boro were 9.54 mg kg^{-1} and 10.97 mg kg^{-1}, respectively. Here the contamination of arsenic was much higher in boro season than aman season. The arsenic levels in Miapur at Charghat upazilla during aman were 9.37 mg kg^{-1} and during boro season they slightly increased to 9.93 mg kg^{-1}. The arsenic levels in Taluksubol at Rajarhat upazilla soil during aman were 7.97 mg kg^{-1} and it was 9.93 mg kg^{-1} during boro. Thus, we concluded that the soil of Rajarhat during aman was below the level of contamination and during boro it was slightly contaminated. The arsenic levels in Agarghoria at Pirgacha upazilla during aman and boro were 7.57 mg kg^{-1} and 7.83 mg kg^{-1}, respectively. Thus the soil was considered not contaminated during both seasons. In general, the surface soils contained arsenic at normal levels although a few soil samples showed contamination. Alam and Sattar [1] conducted an experiment on soil and water from 25 locations representing five upazilas in four districts of Bangladesh. The soils were collected from the depth of 0–15, 15–30 and 30–45 cm and water samples from same locations. The arsenic content in soils ranged 1.27–56.68, 18–54.77, 1.2–50.95, 1.27–39.48 and 18–35.66 mg kg^{-1} in Chapainawabgonj sadar, Kushtia sadar, Bera, Ishardi and Sarishabari upazila, respectively. Out of a total 25 samples arsenic was detectable for 18 samples at 0–15 cm, 17 samples at 15–30 cm and 15 samples at 30–45 cm depth. One sample at 0–15, 7 samples at 15–30 cm and 4 samples at 30–45 cm depth were found to be slightly contaminated. Huq et al. [12] observed that whereas the maximum permissible limit of arsenic in soil is 10 mg kg^{-1}, 80 mg kg^{-1} of arsenic has been detected in Bangladesh soil. It is apparent

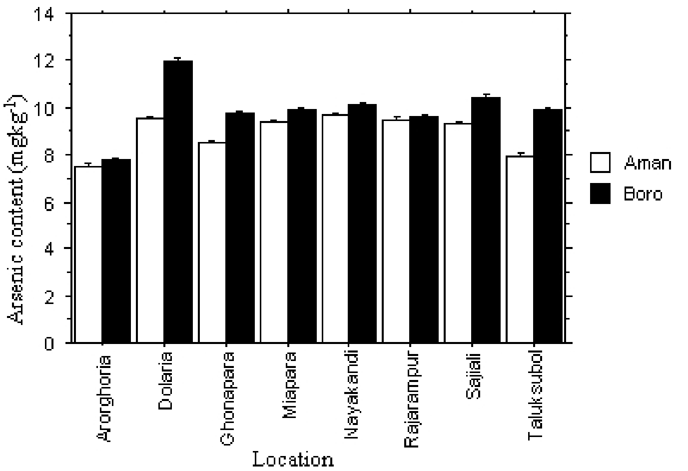

Figure 1. Relative arsenic content of soils during aman and boro seasons.

from the findings of Huq et al. [12], that the average arsenic contents in Bangladesh soils are less than 10 mg kg^{-1}. The surface 15 cm soils contain more arsenic than the subsurface 30 cm soil. There is a positive relationship between arsenic adsorption in soils and clay content. pH of soils seems to play some roles in this regard. Ali et al.[4] showed that the presence of arsenic in irrigation water results in significant increase in arsenic concentration in the irrigated soil, particularly in the top layer (up to about 15 cm) [4]. These findings support the current trend of higher As content in boro season as the soil was irrigated several times with As contaminated water. Therefore, the As contaminated water plays an important role in accumulating higher As content in soil. The successive irrigation with contaminated water would lead to hazards and accumulation of As in plants. Ashraf Ali et al. [4], showed that, in general, arsenic concentrations in soil were significantly higher at the end of the irrigation season, compared to concentrations at the beginning of the season. However, after the flood, arsenic concentration in soil came down to levels comparable to those at the beginning of the irrigation season. The reduction in arsenic concentration in soil after the flood may result from bio–geochemical processes occurring in the rice fields. They also found that, compared to the rice field, arsenic concentrations in the soil samples collected from the vegetable fields were significantly lower. For example, mean arsenic concentration in the topsoil layer (7.5 cm) at the Srinagar site was 7.8 mg kg^{-1} compared to 14.5 mg kg^{-1} for rice field samples. For the Sonargoan site, the mean for the top layer (7.5 cm) of vegetable field samples was 3.5 mg kg^{-1} compared to 8.9 mg kg^{-1} for the rice field samples. They found that arsenic concentration in soil decreased with depth, and that arsenic accumulation in the canal samples were relatively higher compared to the field samples. Among the field samples at the Srinagar sites, arsenic concentration in the topsoil layer varied from about 7 to 27.5 mg kg^{-1}. According to Das et al. [9], the concentrations in soils varied from 7.21 to 27.58 mg kg^{-1}. In their study, three samples in Daudkandi upazila showed arsenic content (ranged 25.14 to 27.58 mg kg^{-1}) above the critical level set by Japan.

Arsenic levels in water: The arsenic concentrations in water at different locations are shown in Fig. 2. Arsenic concentrations in tubewells from the 8 upazillas varied greatly from 0.03 to 0.48 mg L^{-1}. The highest arsenic concentration in water in Rajarampur and Miapara in Chapainawabgonj was 0.48 and 0.46 mg L^{-1}, respectively. The arsenic concentrations in Ghonapara and Nayakandi in Gopalgonj and Muksudpur upazilla were the second highest (0.11 and 0.19 mg L^{-1}). The lowest concentration of arsenic (0.03 mg L^{-1}) was detected at Taluksubol in Rajarhat upazilla, which was uncontaminated according to the Bangladesh arsenic standards (0.05 mg L^{-1}). At Dolaria (Monirampur) and Sajiali (Jessore), the concentrations of arsenic were 0.06 and 0.05 mg L^{-1}, respectively. The arsenic concentrations at Miapur and Vatpara of Charghat were 0.04 and 0.06 mg L^{-1}, respectively. The arsenic concentration in Agorghoria in Pirgacha was 0.08 mg L^{-1}. Out of 10 samples, 2 (Taluksubol and Miapur) were free from arsenic contamination and the water was determined suitable for drinking; 7 samples were highly contaminated and unsuitable for drinking. In addition, 9.5% of the arsenic was in a trivalent form and 90.5% in pentavalent form. Note that trivalent arsenic is more toxic than pentavalent arsenic. Nickson et al.[16] reported that the arsenic derives from reductive dissolution of arsenic-rich iron oxyhydroxides which in turn are derived from weathering of base-metal sulphides. This finding means that it should be possible, by sedimentological study of the Ganges alluvial sediments, to guide the placement of new water wells so that they will be less polluted with arsenic. From the results of study by Alam and Sattar [1], the arsenic content of the tubewell water measured from Chapainawabgonj sadar, Kushtia sadar, Bera, Ishardi and Sarishabari thana ranged 0.010-0.056, 0.01-0.071, 0.01-0.056, 0.01-0.056 and 0.025-0.071 mg L^{-1}, respectively. Out of 25 water samples 17 contained variable amounts of arsenic where 6 sample sites contained arsenic levels above 0.05 mg L^{-1}, and these sites were Rajarampur of Chapainawabgonj upazila, Jordaha of Bera, Courtpara of Kushtia sadar upazila, Nalgari of Ishardi and Ijarapara of Sarishabari upazila. The acceptable limit of arsenic in water as recommended by WHO is 0.01 mg L^{-1}, and the maximum permissible limits are 0.05, 0.04 and 0.058 mg L^{-1} in public water, fresh water and saline water, respectively[19]. Das [9] analyzed 77 samples in two studied upazilas of Daudkandi and Begumgonj. He showed elevated levels of arsenic contents 0.29-0.71 mgL^{-1} in water, 5.64-29.47 $mgkg^{-1}$ in soil and 0.02-3.40 $mgkg^{-1}$ in rice. He suspected that extensive withdrawal of contaminated groundwater contaminates surface soils and plants thus affecting the food chain. Ali et al. [4] conducted an experiment on arsenic in plant–soil environment in Bangladesh. According to the test, arsenic concentration in groundwater samples from 10 irrigation wells at Srinagar site varied from about 0.17 to about 0.6 mg L^{-1}. The arsenic concentrations varied from 0.22 to 0.53 and 0.050 to 0.40 mg L^{-1} in well water and rice–field irrigation water, respectively.

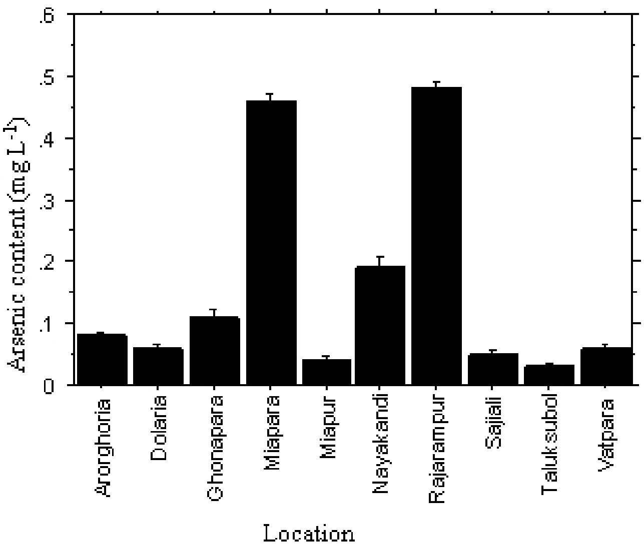

Figure 2. Arsenic content of water at locations across Bangladesh.

Arsenic levels in rice: The arsenic concentrations in rice grains and straw at different locations in the Gopalgonj district are shown in Figs 3 and 4. The results varied from 0.01-0.02 to 1.79-2.52 mg kg^{-1} in rice grain and rice straw, respectively. Out of 15 samples, most straw samples showed arsenic levels above the permissible limit (0.05 mg kg^{-1}) whereas, the grain samples were free of contamination. Arsenic enters rice, Bangladesh's staple crop, through irrigation water pumped from contaminated soil [17]. Meharg and Rahman [15] reported that the accumulation of arsenic in rice-grain was above 1.7 mg kg^{-1} in three samples. Rice comprises

73% of a Bangladeshi's caloric intake and arsenic is in much of the country's groundwater[17]. In polluted regions, arsenic intake could be much higher than estimated. In some regions rice could be the dominant source of arsenic [14]. A study by Ali et al. [4] revealed that high arsenic in irrigation water and soil appears to result in higher concentration of arsenic in root, stem and leaf of rice plants. These trends were similar with those reported by Abedin et al. [5] based on a greenhouse study. This suggests that arsenic can be translocated to paddy shoot. Since rice straw is widely used as cattle feed in Bangladesh and India, high arsenic in rice stem and leaf may result in adverse health impacts on cattle and increase human arsenic exposure via the plant-animal-human pathway. Das et al.[9] reported that concentration in root; shoot and rice-grain of studied paddy were different. Roots of paddy accumulated the highest arsenic followed by shoot and rice-grain. The mean arsenic in rice grain (0.23 mg kg^{-1}) is 6 times less than in shoot and 10 times less than in the root. The arsenic concentration of rice produced in a polluted area of Bangladesh was about 0.3 mg kg^{-1}, which was 2 to 3 times higher than the arsenic content of rice produced in a non-polluted district [13]. Alam et al. [3] found that arsenic accumulation in rice varies from variety to variety and the quantity of accumulated arsenic decreases gradually from root to shoot. From their result the arsenic concentration was 0.0, 0.052, 0.29 and 15.8 mg kg^{-1} in rice grain, husk, leaf and stem, respectively. They also observed that variations in arsenic could depend on many factors such as rice variety, use of irrigation water, soil quality, fertilizer applied, etc. However, it is obvious that arsenic from contaminated irrigation water accumulates in root, stem and leaf of rice plant that may cause risks for cattle and human health. They suggested that rice variety 'Purbachi' was more susceptible to arsenic accumulation.

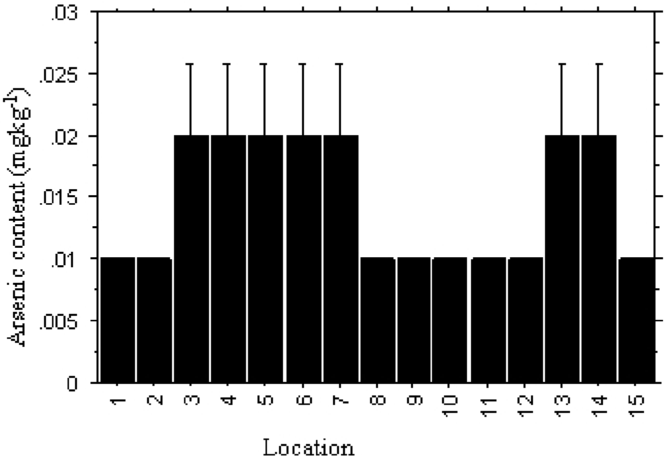

Figure 3. Relative arsenic content of rice grain in some selected locations at Gopalgonj district in Bangladesh.

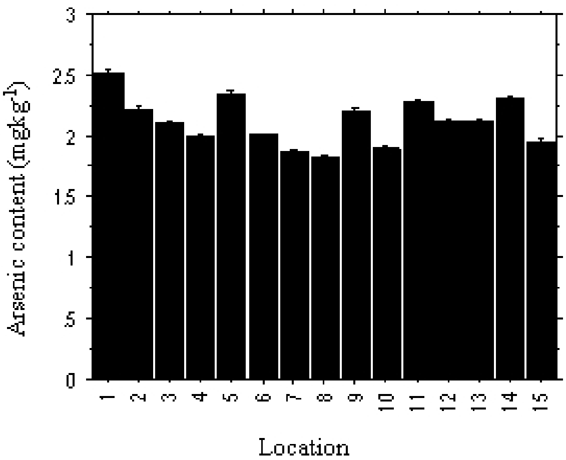

Figure 4. Relative arsenic content of rice straw in some selected locations at Gopalgonj district in Bangladesh.

Arsenic levels in vegetables: The arsenic concentrations in different vegetables are shown in Fig. 5. It is found that, plant parts like stem accumulated more As than the fruits when the plants were allowed to grow in As contaminated water. Among the three vegetables, brinjal plants accumulated the highest amount of As (1.73 mg kg^{-1}) followed by tomato stem (1.04 mg kg^{-1}). The accumulation of As in tomato and brinjal was not significantly higher than the level in uncontaminated water. Therefore, we concluded that the translocation of As from plants to fruits is very limited. Ali et al. [4] reported that the content of arsenic, in general, was higher in the vegetables grown with arsenic contaminated irrigation water than in those grown with arsenic free water. Therefore, fruit vegetables (like amaranth) are much safer than the leafy or stem vegetables as As accumulation is higher in stem or body parts. The trend of arsenic accumulation was higher in leaf vegetables and lower in fruit vegetables. Arsenic uptake by different crops was physiologically variable. Out of 30 samples, the average value of arsenic in tomato stem, brinjal stem and fruit and amaranth exceeded the permissible level (0.05 mg kg^{-1}) in Bangladesh. Brinjal fruit and amaranth plant were safe in uncontaminated water. Tomato fruit was safe in both contaminated and uncontaminated water. Huq et al. [12] conducted an experiment on vegetables in Bangladesh. According to his data the arsenic levels mg kg^{-1} were 2.43–2.79 (gourd), 1.77–2.66 (amaranth), 0.33–1.81 (cucumber), 0.7–0.84 (potato), 1.28–1.86 (cabbage), 0.83–1.128 (papaya), 15.97–19.78 (taro) and 0.58 (bean) [12]. Dhaka University in Bangladesh has reported 7–115, 0.8–1, 0.64–1.42, 1.14–2.12 mg kg^{-1} arsenic in taro, spinach, pulses and green papaya,

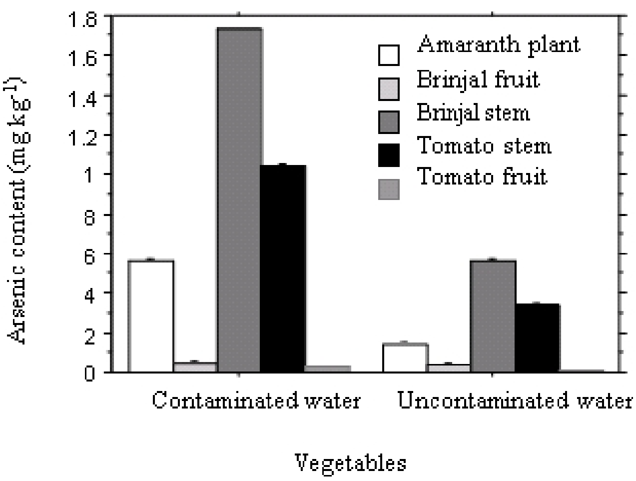

Figure 5. Relative arsenic content of different vegetables using arsenic-free and contaminated water growing at Chapainawabgonj, Bangladesh.

respectively. On the other hand, these values are 0.08–0.39, 0.1–0.15, 0.09 and 0.21–0.46 mg kg^{-1}, respectively in arsenic free areas. Therefore, it is assumed that, a large portion of arsenic is accumulating in the food which will definitely accumulate in humans via the food chain. Roychowdhury[18] found that the individual food composite and food groups containing the highest mean arsenic concentrations (μg kg^{-1}) are potato skin (292.62 and 104), leaf of vegetables (212.34 and 294.67), arum leaf (331 and 341) and papaya (196.50 and 373). Arsenic is absorbed by the skin of most of the vegetables. Higher levels of arsenic were also observed in cooked items compared with raw foods.

Conclusions

We concluded that there was significant and profound level of arsenic contamination in the topsoil, water and plant samples collected from different locations in Bangladesh. Majority of the plants exhibited the arsenic levels above the safe limit. The arsenic problem in Bangladesh is increasing very rapidly. Therefore, there is an urgent need for extensive and detailed study of heavy metal pollution across Bangladesh. Moreover, cheap and economically viable preventive measure should be identified to control this menace for the poor susceptible people of Bangladesh.

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