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Arsenic Contamination in Soil, Vegetable, Groundwater, and Riverine Fish Species in Joypurhat, Bangladesh

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A B S T R A C T

Arsenic contamination is one of the major concerns today in Bangladesh. Various potential and threatening health risks for humankind could be occurred due to the consumption of Arsenic which has mixed with the food chain like fish and vegetables. Therefore, in this study, we analyzed 100 samples of deep tube well water, sediment, soil, vegetable, and fish samples, collected from five different Upazila of Joypurhat district in Bangladesh. The farming process, unplanned groundwater consumption, etc. could be marked as the main culprit for arsenic contamination in a region. However, Arsenic concentrations were determined by atomic absorption spectroscopy followed by the wet digestion method. From this analysis, a shocking result has been found that almost every sample contains arsenic and its level crossed the permissible limit set by WHO guidelines for water and food samples (vegetable and rice) which is an alarming issue for southeast Asian countries.

INTRODUCTION

Consumption of trace elements in the environment (water, sediment, soil, etc.) can cause a potential health risk because of the spreading of such toxic elements in water-based media, which leads to a toxic food chain as the plants absorb them. Hence, monitoring of the human exposure to such toxic food chains which are polluted with trace elements is highly required. Already numerous studies have found the presence of toxic elements in consumable foods in various quantities (Dabeka et al., 1993; Roychowdhury et al., 2002, 2003; Sapunar-Postružnik et al., 1996; Tsuda et al., 1995).

Arsenic, one of the toxic elements providing potential human health risk, is abundant in nature, existing in air, water, soil, and food whereas concentrations might get varied through the anthropogenic status (Villa-Lojo et al., 2002). Various inorganic, as well as organic forms of Arsenic often, could be found in the environment, such as As (III), As (V), monomethylarsonic acid (MMA III, V), dimethylarsinic acid (DMA V), arsenobetaine, arsenocholine, etc. (Jaishankar et al., 2014). However, Monomethylarsonic acid (MMA III) is basically an intermediate compound that is found to have several times higher toxicity compared to the other arsenic compounds and may be one of the potential causes of cancer (Jaishankar et al., 2014).

Humans are generally affected by various organic and inorganic species of arsenic that are present in water, food, and other media of the environment whereas the paths of penetration for dust and fumes could be the respiratory process, and oral digestion for water, beverages, and foods. The most vital or crucial mode of this arsenic toxicity in the human body is that it hampers the process of necessary enzymatic system through forming bonds with various biological ligands (Nriagu, 1994). Humankind is bound to get in touch with arsenic-rich environments occupationally. Chronic exposure to arsenic may result in various health problems related to the skin, liver, cardiovascular system, gastrointestinal tract, hematopoietic system, nervous system, respiratory tract, etc. (Jaishankar et al., 2014). Nowadays, some of these problematic issues with human health are noticeable among people from the southeastern part of Asia, specifically in countries like Bangladesh, Taiwan, and India. According to the WHO and Environmental Protection Agency (EPA), arsenic is classified as a harmful element and also marked as a human carcinogen to skin, lung, bladder, liver, and kidney due to its toxicity. (Afzal et al., 2018; World Health Organization, 2017). A relevant study, by Londesborough and co-researchers (Londesborough et al., 1999) found that arsenic toxicity reduces with the increase of methylation although trivalent forms of MMA and DMA were found more carcinogenic in another study by Vega et al., (2001).

Bangladesh, a South Asian country have about 150 million of population, is considered one of the most vulnerable countries that have been contaminated by groundwater arsenic resulting from the underlying mineralogy of the area. A study by Samanta et al., (1999) found arsenic contamination in 41 among 64 districts of Bangladesh. Approximately, 61% of tube-wells water was found to contain arsenic of more than 0.05 mg per liter. whereas 13% have had arsenic content above 0.01 mg/l (Ali & Tarafdar, 2004). The arsenic concentration in contaminated water, on average, was 0.26 mg/l having an upper level of 0.83 mg/l which is unfortunately way higher than the maximum permissible limit (0.05 mg/l) and upper limit (0.01 mg/l) declared by the WHO (World Health Organization, 2017) and Environment Protection Agency (Flanagan, 1990), respectively. The aspects of food chain contamination by arsenic in Bangladesh came in limelight as the main source of potable water is groundwater which is mostly used for agriculture purposes (Duxbury et al., 2003). A considerable amount (1.7mg/l) of arsenic in staple foods was reported in a recent study by Meharg& Rahman, (2003) whereas a large concentration of arsenic has also been found in vegetables from the arsenic polluted areas in Bangladesh (Alam et al., 2003; Das et al., 2004).

Similar studies regarding this topic, have also been conducted to determine the content of arsenic inside foods from various regions of various countries. For instance, a study by Sapunar-Postružnik et al., (1996) found very lower levels of arsenic inside the fruits (0.002 mg/l) and vegetables (0.004 mg/l) from Croatia whereas foods from various regions in Canada were found to have in the range between <0.0001-4.83 $mgkg^{-1}$ (Dabeka et al., 1993). In Japan, a total diet study showed the daily intake of arsenic leveled between 0.16 mg and 0.28 mg while the value got decreased to 0.088 mg and 0.065 mg in the USA and United Kingdom, respectively. The arsenic content was quite higher in seafood samples (2.36 mgkg-1) compared to rice grains (0.074 mgkg-1) (Schoof et al., 1999). Recently, a study by Alam et al., (2003) reported the average and range of arsenic in affected areas as 0.225 mg per kg and 0.019-0.489 mg per kg, respectively while the arsenic content in vegetables was between 0.07 and 0.3990 mg per kg(Das et al., 2004).

This study focuses on finding out the current arsenic exposure scenario of Bangladeshi people. This study suggests the implementation of proper control and monitoring systems by the government on regular basis, especially in this area.

MATERIALS AND METHODS

Sample Collection Area

All samples were collected randomly from five different locations of Joypurhat district, Bangladesh which are pointed out in the following figure. Most of the deep tube well samples in this area were known to be adversely contaminated with arsenic. In a sterilized polyethylene bottle 25 water samples were collected from 5 different locations from each Upazila. All of the samples from waterbodies were then acidified to near pH < 2 using hydrochloric acid and then kept under 4°C temp until analysis. 5 rice-sample and 20 vegetables-samples were collected from the field before being stocked in bags of polyethylene and preserved under 4°C temp. Soil samples were resourced from the depth of 15-30cm below the surface level of the ground. Five fish samples of each species from the same locations, a total of 25 samples were collected from the river passing through those Upazilas.

Digestion and Arsenic Determination Method

For arsenic determination from water samples, a similar method reported by Kundu et al., (2018) was used. According to this journal article 50mL volumetric flask was used to take 1mL of sample; added 4 mL concentrated HCl, 5 mL of ascorbic acid. The digestion process was continued for 45 min then the solution was prepared with 50 ml of distilled water (Kundu et al., 2018). The concentrations of arsenic were then measured by a flame atomic absorption spectroscopy at 197 nm (Farzana Akter et al., 2005; Mihucz et al., 2017).

Plant, soil, and fish samples were digested with a similar method, reported by Ma et al., (2016) and Sadee et al., (2016) were employed. In short, this method involves important steps such as approximately 0.5 gm of plant/soil samples being weighed into separate vessels that are Teflon reaction. Then 8 mL of concentrated HNO_3 (70%) as well as 2 ml of H_2O_2 (30%) were added to the vessel. Samples were then digested for one hour at 1600 W with two steps. In the first step, the temp raised up to 160°C within 15 mins and remained at this temp for an additional 5 min. Then in the next step, the temp elevated from 160 to 200°C within 8 mins and kept for a further 15 min. After completing the digestion, Teflon reacted vessels were cooled down at room temp. The digested sample solution was then migrated to a 25 mL volumetric flask and made volume up to the mark with Milli-Q water. The amount of arsenic was then determined by using flame atomic absorption spectroscopy described above.

RESULTS AND DISCUSSION

Concentrations of Arsenic in Vegetables and Soil

In Bangladesh, 270 sub-districts (Upazilla) among 465 are highly affected by Arsenic pollution (Ashraf Ali, 2006). The remarked reason for which soil and vegetables are contaminated by heavy metals is due to a massive amount of the withdrawal of groundwater for irrigating the crops field (Meharg & Rahman, 2003). Soil Arsenic levels could raise up to 83 mgkg-1 in Bangladesh because of the irrigation carried out with Arsenic contaminated groundwater (Ullah, 1998). In the present study, mean concentration of arsenic in soil were 24.1 mgkg-1 to 40.1 mgkg-1 (dry weight) with an average of 32.62 mgkg-1, 27.2 mgkg-1 to 40.1 mgkg-1 with an average of 33.8 mgkg-1, 25.5 mgkg-1 to 37.4 mgkg-1 with an average of 33.26 mgkg-1, 28.7 mgkg-1 to 34.1 mgkg-1 with an average of 30.34 mgkg-1 and 30.5 mgkg-1 to 38.7 mgkg-1 with an average of 34.7 mgkg-1 at arsenic affected Joypurhat, Akkelpur, Kalai, Khetlal and Pachbibiupazila of Joypurhat district, respectively. However, a similar study was performed before by Kabir (2017) on soil Arsenic in B.Baria, Faridpur, Tala, and Sonargong regions in Bangladesh respectively with the findings of mean Arsenic 9.42, 13.15, 20.40, and 8.82 ppm. In comparison with the findings of Kabir's article, the findings of this current article are higher in value.

To evaluate the extent of contamination by arsenic in affected regions of Joypurhat district in Bangladesh, various fish and vegetable samples were investigated. Irrigation with Arsenic contaminated water increases the Arsenic pollution also in vegetables. (Abedin, 2002; Alam& Rahman, 2003; Duxbury et al., 2003; Mehrag& Rahman, 2003). As shown in Table 1. The concentration of arsenic in rice (Oryza sativa)grains ranged from 0.31to 0.91 mgkg-1 with a mean value of 0.58 mgkg-1 whereas the values ranged from 0.24 mgkg-1 to 0.61mgkg-1 with a mean value of 0.46 mgkg-1, 0.49 mgkg-1 to 0.88 mgkg-1 with a mean value of 0.65 mgkg-1, 0.30 mgkg-1 to 0.69 mgkg-1 with a mean value of 0.46 mgkg-1, 0.40 mgkg-1 to 0.93 mgkg-1 with a mean value of 0.63 mgkg-1 in tomatoes (Solanum lycopersicum), potatoes (Solanum tuberosum), arum leaves (Colocasiaantiquorum) and radishes (Raphanusraphanistrum), respectively. The maximum level of arsenic was found in rice (Oryza sativa) sample from Kalaiupazila of Joypurhat district contributing to the excessive use of fertilizers and pesticides along with irrigation of land by arsenic-contaminated water from local water bodies. As mentioned in M.F. Hossain. (2006), in soil, the value of Arsenic concentration varies from 0.009 to 1.5 mg Kg and the leafy vegetables contain the higher concentration while the fruits contain a lower concentration. This mentioned range has a similarity with the findings of this article.

In Bangladesh, excessive removal of contaminated underground water for farming purposes triggered the elevating concentrations of arsenic in surface soil layers (Meharg & Rahman, 2003; Nickson et al., 1998; Panaullah et al., 2003). In the present study, the concentration of arsenic in soil ranged from 24.1 mg/ kg (dry weight) to 40.1 mgkg-1 (dry weight) with an average of 32.62 mgkg-1, 27.2 mgkg-1 to 40.1 mgkg-1 with an average of 33.8 mgkg-1, 25.5 mgkg-1 to 37.4 mgkg-1 with an average of 33.26 mgkg-1, 28.7 mgkg-1 to 34.1 mgkg-1 with an average of 30.34 mgkg-1 and 30.5 mgkg-1 to 38.7 mgkg-1 with an average of 34.7 mgkg-1 at arsenic affected Joypurhat, Akkelpur, Kalai, Khetlal and Pachbibiupazila of Joypurhat district, respectively (Table 2). All the values found were significantly several times higher than that of the standard permissible concentration (10 mgkg-1) throughout the world (Das et al., 2004). Such a high concentration of arsenic in the soil of the affected areas can be attributed to the use of fertilizers and pesticides, irrigation of crops by underground water, sediment deposition, types of sediment, and the diagenesis process as well (Das et al., 2004).

Table 1. Concentrations of arsenic in vegetable and soil samples from five affected areas

Sample Name	Scientific Name	Location	Vegetable samples: Range of Arsenic content (mgkg-1)	Vegetable samples: Mean of Arsenic content (mgkg-1)	Soil samples: Range of Arsenic content (mgkg-1)	Soil samples: Mean of Arsenic content (mgkg-1)
Rice	*Oryza sativa*	Joypurhat	0.16-0.71	0.44	16.5-41.22	30.4
		Akkelpur	0.18-0.55	0.31	22.5-55.2	40.1
		Kalai	0.4-1.44	0.91	13.7-39.8	25.5
		Khetlal	0.21-1.85	0.83	17.2-36.7	28.7
		Pachbibi	0.16-0.66	0.41	23.5-46.4	35.4
Tomato	*Solanumlycopersicum*	Joypurhat	0.08-0.43	0.24	27.96-61.11	40.1
		Akkelpur	0.16-0.62	0.39	22.5-50.2	38.3
		Kalai	0.21-0.84	0.52	25.3-46.6	34.1
		Khetlal	0.15-0.77	0.51	17.2-42.7	30.4
		Pachbibi	0.23-0.91	0.61	22.3-46.8	34.8
Potato	*Solanum tuberosum*	Joypurhat	0.32-0.87	0.66	22.2-51.23	35.1
		Akkelpur	0.15-0.82	0.53	15.4-36.0	27.2
		Kalai	0.15-0.89	0.49	20.6-41.9	31.9
		Khetlal	0.43-1.4	0.88	19.0-42.3	29.1
		Pachbibi	0.43-1.12	0.71	30.0-48.4	38.7
Arum leaves	*Colocasiaantiquorum*	Joypurhat	0.06-0.70	0.30	23.4-42.09	33.4
		Akkelpur	0.12-1.34	0.46	18.6-41.4	31.3
		Kalai	0.16-0.67	0.39	27.8-48.1	37.4
		Khetlal	0.22-0.87	0.47	22.3-37.2	29.4
		Pachbibi	0.41-1.20	0.69	26.8-44.1	34.1
Radish	*Raphanusraphanistrum*	Joypurhat	0.21-0.62	0.48	15.79-31.88	24.1
		Akkelpur	0.31-0.75	0.51	22.8-42.8	32.1
		Kalai	0.21-0.82	0.40	27.8-49.8	37.8
		Khetlal	0.32-1.32	0.81	26.0-42.7	34.1
		Pachbibi	0.45-1.76	0.93	23.3-38.0	30.5

Concentrations of Arsenic in water and fish

Arsenic occurs in water by four main species which are: arsenite (H_2AsO_3), arsenate (H_2AsO_4), Methyl-arsenic-acid [$Ch_3AsO(OH)_2$], and dimethyl-arsenic-acid [$(CH_3)_2As(OH)$]. However, the two inorganic species i.e., arsenite (H_2AsO_3) and arsenate (H_2AsO_4) are mainly found in Bangladeshi groundwater (Fazal et al., 2001) and create Arsenic pollution in river water. In Bangladesh, nearly 35 million people are in a vulnerable situation to Arsenic contamination due to the drinking water that contains more than 50 micro g/L of arsenic (Kabir, 2017).

The concentration of arsenic in water could create an impact on the water-based ecosystem. This study found that in fish samples the concentration of Arsenic ranged from 0.04 mgkg-1 to 0.5 mgkg-1 with an average of 0.224 mgkg-1 (Table 3). As the river water fish were concerned, the highest concentration of arsenic was found in catfish (popular in certain parts of Bangladesh) from the river Tulshiganga (0.5 mgkg-1) which is several times higher than the of permissible limit (0.05 mgkg-1) Nevertheless, the maximum percentages are present as nontoxic arsenobetaine.

Table 2. Concentrations of arsenic in fish samples from five different rivers

Sample Name	**Scientific name**	**Location**	**Fish samples**	
			Range of Arsenic content (mgkg-1)	**Mean of Arsenic content (mgkg-1)**
Cat fish	*Heteropneustesfos silis*	Jamuna (Joypurhat-Sadar)	0.04-0.21	0.112
		Tulshiganga (Akkelpur)	0.25-0.75	0.524
		Nandail (Kalai)	0.03-0.06	0.042
		Tulshiganga (Khetlal)	0.14-0.47	0.29
		Jamuna (Pachbibi)	0.04-0.31	0.126
Spotted Head fish	*Channa punctatus*	Jamuna (Joypurhat-Sadar)	0.15-0.42	0.30
		Tulshiganga (Akkelpur)	0.08-0.25	0.15
		Nandail (Kalai)	0.04-0.18	0.10
		Tulshiganga (Khetlal)	0.09-0.40	0.212
		Jamuna (Pachbibi)	0.14-0.29	0.21
Lata fish	*Channastriata*	Jamuna (Joypurhat-Sadar)	0.14-0.41	0.232
		Tulshiganga (Akkelpur)	0.18-0.47	0.316
		Nandail (Kalai)	0.07-0.41	0.20
		Tulshiganga (Khetlal)	0.08-0.31	0.21
		Jamuna (Pachbibi)	0.04-0.51	0.34
Koi fish	*Anabas cobojius*	Jamuna (Joypurhat-Sadar)	0.14-0.56	0.412
		Tulshiganga (Akkelpur)	0.14-0.28	0.208
		Nandail (Kalai)	0.01-0.09	0.05
		Tulshiganga (Khetlal)	0.18-0.40	0.298
		Jamuna (Pachbibi)	0.19-0.34	0.252
Tyangra fish	*Macronesvittalus*	Jamuna (Joypurhat-Sadar)	0.09-0.31	0.214
		Tulshiganga (Akkelpur)	0.12-0.31	0.23
		Nandail (Kalai)	0.07-0.28	0.14
		Tulshiganga (Khetlal)	0.24-0.64	0.40
		Jamuna (Pachbibi)	0.09-0.25	0.18

As the speciation of arsenic was not the concern of this work, several studies found that about 70% of arsenic content in the samples of fish tissue is present as nontoxic arsenobetaine (Ebisuda et al., 2002).

Table 3. The concentration of arsenic in deep tube well water and sediments from affected areas

Sample	**Jamuna (Joypurhat-Sadar) (mgkg-1)**	**Tulshiganga (Akkelpur) (mgkg-1)**	**Nandail (Kalai) (mgkg-1)**	**Tulshiganga (Khetlal) (mgkg-1)**	**Jamuna (Pachbibi) (mgkg-1)**
Deep tube well water (mg/l)	0.21	0.43	0.10	0.72	0.52
Sediment (mgkg-1)	0.44	0.21	0.13	0.23	0.45

Deep tube well water and sediments collected from those areas were found to contain arsenic ranging from 0.1 mg/l to 0.7 mg/l with a mean of 0.38 mg/l and 0.1 mgkg-1 to 0.45 mgkg-1 with an average of 0.28 mgkg-1, respectively exceeding the limit promisingly (Table 3). The concentration of Arsenic was found 0.52mg/kg in deep tube well water at Pachbibi near Jamuna river while the lowest amount was observed at 0.10 mg/kg at Kalai area near Nandail. Similarly, the amount of arsenic in sediment samples exceeds the permissible limit also. The highest and lowest concentration was noted as 0.45 mg/kg and 0.13 mg/kg at the bank of Jamuna and Nandail rivers respectively. This finding denotes pollution of drinking water and other water bodies in the area. Government should take necessary steps to mark the tube wells to restrict the people from drinking the arsenic-contaminated water. Most importantly, arsenic-contaminated river bank areas should be marked to inhibit local people from cultivating crops, vegetables, and other food items.

Conclusion

Arsenic contamination has been an alarming issue in the Southeast Asian regions. Several investigations have found the contamination level to be detrimental to human health. The present study has investigated the severity of As pollution in soil, vegetable, water, and fish of Joypurhat district, Bangladesh. The results showed that the pollution has reached the level across multiple trophic levels. All the values of As concentration found for soil and vegetable samples were significantly considerably higher than the international permissible limit (10 mgkg-1) (Das et al., 2004). The values were alarmingly higher in the case of fish and water samples as well. This points to the need for immediate action to be taken to reduce this As contamination of soil, vegetables, fish, and water which is becoming the reason for different health issues throughout the country.

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