University of Tehran Press

Pollution

Print ISSN: 2383-451X
Online ISSN: 2383-4501

https://jpoll.ut.ac.ir/

Hazard Estimations Result from Arsenic Contamination in Common Foodstuffs, Soil, Sediment, and Water of Joypurhat District, Bangladesh

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Article Info

Article type:
Research Article

Article history:
Received: 18 Aug 2022
Revised: 27 Oct 2022
Accepted: 11 Jan 2023

Keywords:
Heavy metals
Pollution
Health risk
etc

ABSTRACT

We analyzed 125 samples collected from Joypurhat district, Bangladesh, in this study. Average inorganic arsenic (IAs) content obtained from collected polished rice, tomato, potato, radish, and arum leaves 0.31 - 0.91, 0.24 - 0.61, 0.49 - 0.88, 0.40 - 0.93, and 0.30 - 0.69 mg/kg, respectively. This report summarized that almost every agronomic sample contains arsenic; the As contents remain within the permissible limit set by FAO/WHO's guideline (1.00 mg/kg) except for the rice sample. The As concentration for the rice sample was significantly higher (0.31 - 0.91) than the prescribed limit (0.20 mg/kg). But, the As level for water (mean range, 0.10 - 0.72 mg/l), sediment (0.13 - 0.53 mg/kg), and soil samples (24.1 - 43.1 mg/kg) also significantly surpassed the permissible level. The present study is alarming for water samples, where the highest IAs concentration (0.72 mg/l) is 72 times [14 times] higher than WHO/FAO's [Bangladesh's] allowable limit (0.01mg/l) [0.05 mg/l]. All agronomic fields contain higher IAs (25.50 - 43.10 mg/kg) than the world standard limit (10 mg/kg). Statistical Igeo confirmed the moderate pollution of the entire agronomic field of Joypurhat except for the river's sediment. Again, EF values ensured the anthropogenic pollution by the moderately severe enrichment of As for the 65% agronomic field and significant enrichment of As for the 35% agronomic field. Hazard estimation results revealed the privileged possibility of non-carcinogenic [carcinogenic] health hazards to regular polished rice [water] consumers. So, present study suggests that authorities should take necessary steps to prevent contamination/upcoming health risks.

Cite this article: Author, A. A., Author, B. B., & Author, C. C. (2023). Hazard Estimations Result from Arsenic Contamination in Common Foodstuffs, Soil, Sediment, and Water of Joypurhat District, Bangladesh. *Pollution*, 9 (2), 531-544. http://doi.org/10.22059/POLL.2022.347351.1578

DOI: http://doi.org/10.22059/POLL.2022.347351.1578

INTRODUCTION

Recently, 12 countries in Asia have reported excess Arsenic (As) pollution in groundwater resources for some locations in East Ganges Basin (EGBs) region. Among these 12 countries, Bangladesh possesses the worst situation. Here 61 districts (out of 64) are extremely polluted by As. Around 20 percent of shallow aquifer/shallow tube wells (STWs) are contaminated (higher than the permissible limit of 0.05 mg/l for Bangladesh). And approximately 30 million people

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depend mainly on this contaminated water for their domestic activities in Bangladesh. The most alarming is that 10 – 30 thousand people have been affected by severe arsenicosis (Heikens, 2006; Chakraborti et al., 2010). As contamination in groundwater becomes a potential health risk in Bangladesh, especially for direct drinking water from the STWs. In the last couple of years, inexpensive STWs have increased drastically due to the green revolution, which facilitates plenty of crop production according to necessity, even in drought. Initial investigation and further scientific studies conducted in the last decade suggest significant As pollution due to easy interruption of the food chain through irrigation. Potentially, a more hazardous impact is the continuous accumulation of As in the plow land through the irrigation water; contaminated by As; which ultimately destroys the country's nutritional status and rural economy (Chakraborti et al., 2010; Suzuki et al., 2022).

Joypurhat is known as a storehouse of food in Bangladesh for producing different types of agricultural commodities. But, the prime concern is that this region lacks quality irrigation and drinking water. Sometimes, contaminating elements, especially trace elements, makes the situation more challenging. Groundwater is the prime source of irrigation in Joypurhat, where around 65% of cultivable lands are irrigated by groundwater (Chakraborti et al., 2010). The water samples collected from the deep tube wells contain arsenic in this area. Contamination of trace elements in the environment (water, sediment, soil, etc.) can cause a potential health risk due to the transfer of such toxic aspects in aquatic media and their consequent uptake and bio-accumulate by plants leading to entrance into the food chain. Hence, it is necessary to monitor human exposure to various poisonous trace elements present (toxins) in the food cycle. Numerous studies have already reported the presence of toxic elements within foodstuffs with different concentrations (Suzuki et al., 2022; Wong et al., 2022; Lyu et al., 2022; Roychowdhury et al., 2002; Raju, 2022). The above-stated facts motivated us to perform a survey-type study of As contamination in water, sediment, soil, and foodstuff collected from different parts of Joypurhat.

Arsenic (As) is a toxic trace element. It is abundant in nature and is present in the soil, sediment, water, food items, etc. In a previous study, Mehrdadi et al. (2009) claimed excess withdrawal of groundwater/continual droughts results in a drop-down of water tables, which facilitates the oxidation of natural arsenic ores to turn into the liquid phase as As ions and enhances As pollution. As, is usually founded in various organic and inorganic chemical forms such as As (III), As (V), monomethylarsonic acid (MMA III, V), dimethylarsinic acid (DMA V), arsenobetaine, arsenocholine, etc. ((Mehrdadi et al., 2009); Rahman et al., 2022). An intermediate product of As, [i.e., Monomethylarsonic acid (MMA III)] is known as inorganic Arsenic (IAs); which is more toxic compared to other arsenic compounds and may be one of the potential causes of cancer (Langasco et al., 2022). Humans are generally affected by various organic and inorganic forms of As present in water, foodstuffs, and other environmental/atmospheric media where the paths of penetrations include respiration/inhalation via dust and fumes; and orally via water, beverages, different types of food items, etc. Mainly prominent mode of action for IAs toxicity in humans is the inactivation of a living enzyme through binding with other organic ligands (Heikens, 2006). Humans who are occupationally bound to get exposed to IAs-rich environments are consistently monitored for IAs exposure. Chronic exposure to IAs may result in various health hazards to the respiratory tract, gastrointestinal tract, liver, skin, hematopoietic system, cardiovascular system, nervous system, circulatory system, etc. (Langasco et al., 2022; Khatun et al., 2021). Recently, many of these have been shared among inhabitants in the southeastern subcontinents, specifically in Bangladesh, Myanmar, India, etc. According to the WHO, FAO, and Environmental Protection Agency (EPA), IAs has been categorized as a toxic trace metal and grouped as the number one (1) carcinogen to the skin/membrane, liver, lung, bladder, and kidney (Langasco et al., 2022; Khatun et al., 2021; WHO, 2017; Ali and Tarafdar, 2003; EPA 2001).

A significant amount (1.7 mg/l) of As in staple foods was reported by Meharg and Rahman (2003). A higher amount of As in fruits and vegetables from the As-affected areas of Bangladesh has also been reported (Duxbury et al., 2003; Das et al., 2004). Some researchers studied the As content in foods from various regions in many countries. For instance, a study by Sapunar-Postruznik et al. (1996) found the minute amount of As in horticultural fruits (0.002 mg/kg), leaves, and vegetables (0.004 mg/kg) in Croatia. The As content in the foods collected from the different parts of Canada was in the range of 0.0001-4.83 mg/kg. In Japan, a total diet study showed the daily intake of As ranges between 0.16 - 0.28 mg (Tsuda et al., 1995) while it was decreased to 0.088 mg and 0.065 mg in USA and United Kingdom, respectively. The As content was quite higher in seafood samples (2.36 mg/kg) compared to rice grains (0.074 mg/kg) (Schoof et al., 1999). Recently, Alam et al. (2003) claimed the mean [range] 0.225 mg/kg [0.019-0.489 mg/kg] of IAs in affected areas, while the arsenic content in vegetables was between 0.07 and 0.3990 mg/kg.

The previously reported results indicated that IAs concentration in different agricultural commodities varies extensively due to different levels of contaminated STWs used for irrigating plow land. Nevertheless, IAs contamination in the food chain is characterized by other factors, such as natural availability and easy uptake of IAs by crops that need to be extensively studied. Considering these criteria in the present study, we report the level of IAs contaminations in some familiar sources, like water, soil, sediment, rice, major agronomic crops, and fish from Joypurhat district, Bangladesh.

MATERIALS AND METHOD

Sample collection area

The soil and crops samples were collected randomly from five dissimilar Upazila [i.e., Joypurhat = S_1 (25.0677°N and 88.9946°E), Akkelpur = S_2 (24.9034°N and 89.0327°E), Kalai = S_3 (25.0677°N and 89.2175°E), Khetlal = S_4 (25.0029°N and 89.1447°E), and Pachbibi = S_5 (25.2444°N and 89.0408°E)] of Joypurhat district, Bangladesh which is figured out in the following Fig. 1. Samples of polished rice (N_1 = 5 samples collected from S_1 to S_5 sites), Potato (N_2 = 5 samples

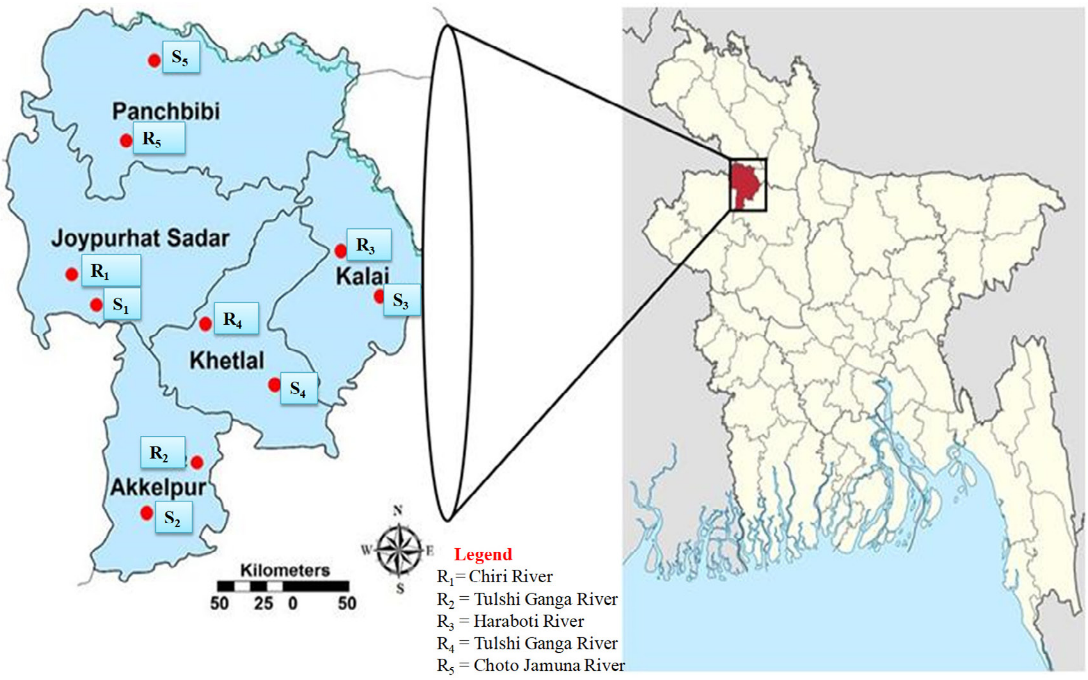

Fig. 1. Sampling site (Joypurhat District in Bangladesh)

collected from S_1 to S_5 sites), Tomato (N_3 = 5 samples collected from S_1 to S_5 sites), Radish (N_4 = 5 samples collected from S_1 to S_5 sites), Arum leaves (N_5 = 5 samples collected from S_1 to S_5 sites) collected from the field were stored in polyethylene bags separately and preserved at 4°C. Soil samples (N_6 = 25 samples) were collected from 15-30 cm depth below the ground surface of the plant collecting location. Fish, surface water, and sediment were collected from the same location and symbolized as R_1 (Chiri River, 25.0890°N and 88.9676°E), R_2 (Tulshi Ganga River, 24.9355°N and 89.0718°E), R_3 (Haraboti River, 25.0986°N and 89.1997°E), R_4 (Tulshi Ganga River, 25.07288°N and 89.0713°E), and R_5 (Choto Jamuna River, 25.1707°N and 89.0191°E) shown in Fig. 1. While each of the fish samples was collected from R_1 to R_5 locations where Walking catfish, Spotted snakehead fish, Lata fish, Koi fish, and Tangra fish were marked as N_7, N_8, N_9, N_{10}, and N_{11}, respectively. N_{12} (total 25 samples) and N_{13} (total 25 samples) represent the water and sediment samples from R_1 to R_5 collected from the same location of each fish collecting area and stored in sterilized plastic bottles before analysis. All analyses were done within one week of the sample collection time.

Digestion and Arsenic determination method

For the determination of the arsenic content in the water samples, an identical analytical technique of Kundu et al. (2018) was used. Accordingly, 1 ml of the sample was taken in a 50 ml volumetric flask in which 4 ml of concentrated HCl and 5 ml of ascorbic acid were added. Digestion was carried out for 45 minutes at 60°C; after then, added the distilled water to make the solution of 50 ml. The flame atomic absorption spectroscopy technique was used to determine the amount of arsenic. Hollow Cathode Lamps (HCL) with 193.7 nm wavelength and 0.5 nm slit were used for IAs determination (Akter et al., 2005; Mihucz et al., 2017). Plant, soil, and fish samples were digested with a similar method reported by Sadee et al. (2016) and Ma et al. (2016). Later 8 ml 70% HNO_3 (concentrated) and 2 ml 30% H_2O_2 were inserted into the Teflon vessel. After then, the samples were digested for 43 minutes at 1600 W with two steps. At first, within 15 minutes, the vessel temperature reached 160°C and held for 5 minutes. Then, within 8 minutes, the vessel temperature changed to 200°C and kept for 15 minutes. After finishing the digestion process, the Teflon vessels were kept at room temperature until cool. The digested sample solution was then taken into a 25 ml volumetric flask, and Milli-Q water was made up to the mark of the flask. The amount of IAs was then determined by using flame atomic absorption spectroscopy (FAAS) described above.

Sample identification: For this study, the five most economical and popular fish species (Table 1) were collected from the river passing through the selected Upazila shown in Fig. 1. Each of the five fish species with three replicates was collected from the same location.

Quality control schedule: All the standards (Fluka solution), reagents, solvent, chemicals (Merck), and equipment were analytical grade. Internal and external qualities were controlled by regular inspection. Authentic data were kept by regular records, statistical analysis, method validation, calibration, etc. Before every use, the machines and instruments were calibrated accurately. External quality was maintained by the skilled analyst, and appropriate handling of the device, glassware, samples, chemicals, reagents, etc. Acid blanks were run periodically to keep the purification of the samples and chemicals. The precision of the experimental method, typical recoveries moreover the reputability are approximately 94 - 96%. Each time calibration curve was maintained by four (04) dissimilar known standard concentrations.

Assessment of soil contamination level

This study calculated widely used Enrichment factor (EF) and geo-accumulation index (I_{geo}) to assess the level of soil contamination. I_{geo} and EF were determined by the following equations (Abdullah et al., 2020):

Table 1. Sample specification of five selected fish species

Serial no.	English name	Local name	Scientific name	Stratum / Inhabit
1	Walking catfish	Magur fish	*Clarias batrachus*	Omnivorous
2	Spotted snakehead fish	Shol fish	*Chilodus punctatus*	Demersal
3	Lata fish	Taki fish	*Channa punctatus*	Carnivorous
4	Koi fish	Koi fish	*Cyprinus rubrofuscus*	Omnivorous demers
5	Tangra fish	Tangra fish	*Bagrus pelusius*	Bottom feeder

Table 2. Soil classification based on statistical Igeo and EF (Abdullah et al., 2020).

Class	I_{geo} range	I_{geo} based Soil/sediment quality	EF range	Level of metal enrichment
0	$I_{geo} \leq 0$	Unpolluted	< 2	Low enrichment
1	$0 \leq I_{geo} \leq 1$	Unpolluted to moderately polluted	2 ≤ EF < 5	Moderate enrichment
2	$1 \leq I_{geo} \leq 2$	Moderately polluted	5 ≤ EF < 20	Significant enrichment
3	$2 \leq I_{geo} \leq 3$	Moderately to strongly polluted	20 ≤ EF < 40	Moderately severe enrichment
4	$3 \leq I_{geo} \leq 4$	Strongly polluted	EF > 40	Severe enrichment
5	$4 \leq I_{geo} \leq 5$	Strongly to extremely polluted	----	----
6	$I_{geo} > 6$	Extremely polluted	----	----

$$I_{geo} = \log_2 \left(\frac{Cn}{1.5Bn}\right) \ldots\ldots\ldots\ldots\ldots\ldots\ldots (i)$$

Here, C_n and B_n are the concentration of obtained IAs in the sampling sites and Earth's crust IAs content (1.8 mg/kg), respectively. 1.5 is a constant value incorporated here to overcome the lithogenic effect (Taylor, 1964).

$$EF_c = \frac{(C_M / C_{Fe})_{Sample}}{(C_M / C_{Fe})_{Earth's\ crust}} \ldots\ldots\ldots\ldots\ldots\ldots\ldots (ii)$$

Here, $(C_M/C_N)_{sample}$ = Obtained IAs content to Fe (i.e. used as a metal of normalization) concentration proportion divided by the Earth's crust IAs content (1.8 mg/kg)/ Earth's crust Fe concentration (56300 mg/kg) (Taylor 1964). Fe was extensively selected as the normalization metal due to its natural occurrence plus strong immobility. Based on I_{geo} and EF, evaluated soil samples were categorized according to Abdullah et al. (2020).

Calculation of human health risk

The dietary intake of IAs has been calculated utilizing obtained IAs concentrations of different agronomic crops and fish species (Ahmed et al., 2016).

$$ADI = \frac{FIR \times C}{ABW} \ \mu g/kg\text{-}BW/day \ldots\ldots\ldots (iii)$$

FIR = Foodstuff Intake Rate (g/person/day) and C= Obtained IAs concentration in different foodstuff (mg/kg, wet weight). Typically, a mature man (average 60 kg) daily consumes 416g polished rice, 73.342 g potato (ARP 2018), 11.31 g tomato (Ahmed et al., 2016), 30g radish, 30g arum leaves (Reimers and Keast, 2016) and 62.58 g fish (DoF 2019) respectively.

Estimation of non-carcinogenic and carcinogenic health risk

Hazard Quotient (HQ) and Carcinogenic risk (CR) estimation: HQ and HI are extensively used to assess non-carcinogenic health risks associated with heavy metals via consuming individual foodstuffs. Besides, CR results from the lifetime disclosure of trace metals like IAs (Khatun et al., 2021). The following equations are used to calculate the HQ and CR (Islam et al., 2020).

$$HQ = \frac{ER \times EI \times FIR \times CF \times C}{WAB \times ATn \times RfD} \times 10^{-3} \qquad \text{(iv)}$$

$$CR = \frac{EF \times ED \times FIR \times CF \times C \times CSFo}{ABW \times AT\mathrm{c}} \times 10^{-3} \qquad \text{(v)}$$

Where, HQ = Hazard Quotient, ER = Exposure Rate (365 days/year), EI = Exposure Interval (30 years for non-carcinogenic risk) (USEPA 2022), FIR = Foodstuffs Ingestion Rate (as explained previously), CF = Conversion Factor/feature (0.208 to change fresh weight to dry weight), C = Obtained Concentration of IAs (mg/kg), ABW = Average Body Weight = 60kg, ATn = Average exposure Time (EF×ED) (i.e. for Non-carcinogens 365 days/year for 30 years = 10950 days), RfD = Reference Oral Dose intake by human (IAs = 3.000E-4), CSFo = carcinogenic slope factor caused by Oral ingestion (mg/kg BW/day)$^{-1}$ (1.500 for IAs), and ATc = Average time for carcinogenic metal exposure (i.e. ATc = EF × ED = 365 days/ years×70years) (USEPA 2022). IAs is a carcinogenic trace metal, and its carcinogenic effect is stated from E-4 to E-6.

RESULTS AND DISCUSSION

Inorganic arsenic concentrations in agronomic crops and corresponding soil samples

In Bangladesh, soil and vegetables are contaminated by different trace metals due to the extensive withdrawal of contaminated groundwater for irrigating crops field (Rahaman et al., 2022). To evaluate the extent of contamination by arsenic in affected regions of Joypurhat district, various agronomic crops [viz. Rice (*Oryza sativa*), potato (*Solanum tuberosum*), tomato (*Solanum lycopersicum*), Arum leaves (*Colocasia esculenta*), and radish (*Raphanus sativus*)] samples were considered as shown in Table 3. The highest IAs concentration is obtained in S_3 sites (mean 0.91 mg/kg) followed by other sites for the polished rice samples. The concentration level for different Upazila based on their mean concentration can be expressed as follows: S_3 (mean 0.91 mg/kg) > S_4 (mean 0.83 mg/kg) > S_1 (mean 0.44 mg/kg) > S_5 (mean 0.41 mg/kg) > S_2 (mean 0.31 mg/kg). However, Table 3 also shows that the concentration for soil samples collected from corresponding rice plow land of different Upazila is as follows: S_2 (mean 40.10 mg/kg) > S_5 (mean 35.40 mg/kg) > S_1 (mean 30.40 mg/kg) > S_4 (mean 28.70 mg/kg) > S_3 (mean 25.50 mg/kg).

The maximum IAs concentration is observed for potatoes collected from the S_5 location (mean 0.61 mg/kg) than those collected from other locations. The order of the mean accumulation of IAs in case of potatoes collected from selected sites is as follows (Table 3): S_5 (mean 0.61 mg/kg) > S_3 (mean 0.52 mg/kg) > S_4 (mean 0.51 mg/kg) > S_2 (mean 0.39 mg/kg) > S_1 (mean 0.24 mg/kg). This has changed as follows for the corresponding soil samples: S_1 (mean 40.10 mg/kg) > S_2 (mean 38.30 mg/kg) > S_5 (mean 34.81 mg/kg) > S_3 (mean 34.11mg/kg) > S_4 (mean 30.41 mg/kg).

However, tomato and radish samples collected from different Upazila follow the same hierarchy though the range and mean concentration vary slightly [Table 3]. Here, the hierarchy for tomato [radish] is as follows: S_4 (mean 0.88 mg/kg) [0.93mg/kg] > S_5 (mean 0.71 mg/kg) [0.81mg/kg] > S_1 (mean 0.66 mg/kg) [0.51mg/kg] > S_2 (mean 0.53 mg/kg) [0.48mg/kg] > S_3

Table 3. Concentrations of IAs and their exposure consequence of selected Agronomic crops plus soil samples from five affected areas

Sample Name	Location	Agronomic samples		Health risk Assessment			Soil samples = N_6		Soil contamination level Assessment			
		Range of As con. (mg/kg)	Mean of As con. (mg/kg)	ADI (µg/kg-BW/day)	HQ	CR	Range of As con. (mg/kg)	Mean of As con. (mg/kg)	I_{geo} range	Soil category based on I_{geo}	EF range	Level of metal enrichment
N_1	S_1	0.16-0.71	0.44	3.05	2.12	9.52×10^{-4}	16.50-41.22	30.40	1.35	Moderately polluted	19.02	Significant enrichment
	S_2	0.18-0.55	0.31	2.15	1.49	6.71×10^{-4}	22.50-55.20	40.10	1.47	Moderately polluted	25.08	Moderately severe enrichment
	S_3	0.40-1.44	0.91	6.31	4.37	1.97×10^{-3}	13.70-39.80	25.50	1.28	Moderately polluted	15.95	Significant enrichment
	S_4	0.21-1.85	0.83	5.75	3.99	1.80×10^{-3}	17.20-36.70	28.70	1.33	Moderately polluted	17.95	Significant enrichment
	S_5	0.16-0.66	0.41	2.84	1.97	8.87×10^{-3}	23.50-46.40	35.40	1.42	Moderately polluted	22.14	Moderately severe enrichment
N_2	S_1	0.08-0.43	0.24	0.29	0.20	9.15×10^{-5}	27.96-61.11	40.10	1.47	Moderately polluted	25.08	Moderately severe enrichment
	S_2	0.16-0.62	0.39	0.48	0.33	1.49×10^{-4}	22.50-50.20	38.30	1.45	Moderately polluted	23.96	Moderately severe enrichment
	S_3	0.21-0.84	0.52	0.64	0.44	1.98×10^{-3}	25.30-46.60	34.10	1.40	Moderately polluted	21.33	Moderately severe enrichment
	S_4	0.15-0.77	0.51	0.62	0.43	1.95×10^{-4}	17.20-42.70	30.40	1.35	Moderately polluted	19.02	Significant enrichment
	S_5	0.23-0.91	0.61	0.75	0.52	2.33×10^{-4}	22.30-46.80	34.80	1.41	Moderately polluted	21.77	Moderately severe enrichment
N_3	S_1	0.32-0.87	0.66	0.12	0.54	2.41×10^{-4}	22.20-51.23	35.10	1.41	Moderately polluted	21.96	Moderately severe enrichment
	S_2	0.15-0.82	0.53	0.10	0.43	1.94×10^{-3}	15.40-36.00	27.20	1.30	Moderately polluted	17.02	Significant enrichment
	S_3	0.15-0.89	0.49	0.09	0.40	1.79×10^{-4}	20.60-41.90	31.90	1.37	Moderately polluted	19.96	Significant enrichment
	S_4	0.43-1.40	0.88	0.17	0.71	3.22×10^{-4}	19.00-42.30	29.10	1.33	Moderately polluted	18.20	Significant enrichment
	S_5	0.43-1.12	0.71	0.13	0.58	2.60×10^{-4}	30.00-48.40	38.70	1.46	Moderately polluted	24.20	Moderately severe enrichment
N_4	S_1	0.31-0.75	0.51	0.26	0.18	7.96×10^{-5}	15.79-31.88	24.10	1.25	Moderately polluted	15.08	Significant enrichment
	S_2	0.21-0.62	0.48	0.24	0.17	7.49×10^{-5}	22.80-42.80	32.10	1.37	Moderately polluted	20.08	Moderately severe enrichment
	S_3	0.21-0.82	0.40	0.20	0.14	6.24×10^{-5}	27.80-49.80	37.80	1.45	Moderately polluted	23.65	Moderately severe enrichment
	S_4	0. 45-1.76	0.93	0.47	0.32	1.45×10^{-4}	23.30-38.00	30.50	1.35	Moderately polluted	19.08	Significant enrichment
	S_5	0. 32-1.32	0.81	0.41	0.28	1.26×10^{-4}	26.00-42.00	34.10	1.40	Moderately polluted	21.33	Moderately severe enrichment
N_5	S_1	0.06-0.70	0.30	0.15	0.10	4.68×10^{-5}	23.40-42.09	33.40	1.39	Moderately polluted	20.89	Moderately severe enrichment
	S_2	0.12-1.34	0.46	0.23	0.16	7.18×10^{-5}	18.60-41.40	37.30	1.44	Moderately polluted	23.33	Moderately severe enrichment
	S_3	0.16-0.67	0.39	0.20	0.14	6.08×10^{-5}	27.80-48.10	37.30	1.44	Moderately polluted	23.33	Moderately severe enrichment
	S_4	0.22-0.87	0.47	0.24	0.16	7.33×10^{-5}	22.30-37.20	39.40	1.47	Moderately polluted	24.65	Moderately severe enrichment
	S_5	0.41-1.20	0.69	0.36	0.24	1.08×10^{-5}	26.80-44.10	43.10	1.50	Moderately polluted	26.96	Moderately severe enrichment

(mean 0.49 mg/kg) [0.40mg/kg]. But the hierarchy has been changed for the corresponding soil samples. For soil samples collected from tomato field the hierarchy is: S_5 (mean 38.70 mg/kg) > S_1 (mean 35.10 mg/kg) > S_3 (mean 31.90 mg/kg) > S_4 (mean 29.10 mg/kg) > S_2 (mean 27.20 mg/kg); whereas the hierarchy for radish field is as follows: S_3 (mean 37.80 mg/kg) > S_5 (mean 34.10 mg/kg) > S_4 (mean 30.50 mg/kg) > S_2 (mean 32.10 mg/kg) > S_1 (mean 24.10 mg/kg).

Again, Arum leaves and respective soil exhibit the same concentration hierarchy for the five selected Upazila [Table 3]. The obtained hierarchy of Arum leaves [soil] is as follows: S_5 (mean 0.69 mg/kg) [43.10 mg/kg] > S_4 (mean 0.47 mg/kg) [39.40 mg/kg] > S_2 (mean 0.46 mg/kg) [37.30 mg/kg] > S_3 (mean 0.39 mg/kg) [37.30 mg/kg] > S_1 (mean 0.30 mg/kg) [33.40 mg/kg].

Both forms (organic and inorganic) of As is carcinogenic even in small concentration. Chronic exposure causes black foot disease (the most common and dangerous disease in Bangladesh), cancer, and tumor in the excretory system, membrane, kidney, lungs, etc. (Heikens, 2006; Chakraborti et al., 2010).

The concentration of arsenic in polished rice *(Oryza sativa)* varies and drastically exceeded the permissible range (0.20 mg/kg) for the selected five affected areas (FAO/WHO 2021). But, others selected agronomic crop samples do not exceed the permissible limit (1.00 mg/kg) prescribed by WHO (Das et al., 2004). On the other hand, the entire respective soil sample surpassed the permissible limit (10 mg/kg) approved by WHO (FAO/WHO 2021). Based on these outcomes [Table 3], this carcinogen (i.e., As) contaminates the mentioned polished rice grains. The maximum level of arsenic found in polished rice samples from the S_3 sampling station of Joypurhat district results from the excessive use of fertilizers, pesticides, and irrigation through arsenic-contaminated water. Nasrabadi et al. (2013) determined significantly higher As concentration downstream than upstream of Haraz River resulting from municipal/agricultural sewage, untreated discharged from the nearest coal mine, hot spring spas, etc., and safe for dermal exposure/limited ingestion of foodstuffs grown here. Besides, higher IAs concentration was obtained in a recent study that exposed severe risk for abiotic plus biotic factors of soil and consequent health risk by the food chain through biomagnifications (Khatun et al., 2021). Duxbury et al. (2003) reported IAs concentrations of 183 and 117 μg/kg in rice grain (with 14% water content) for the boro and aman seasons, respectively. However, a similar result has been reported by Das et al. (2004) in which they reported 0.136 mg/kg DW IAs availability in rice grain, which is far lower than our present findings. They also stated 0.09 –3.99 mg/kg DW and 0.07 –1.36 mg/kg DW of IAs for arum leaves and potatoes, respectively. Again, 15.68 mg/kg DW (mean) As concentration in the soil sample was also reported by Das et al. (2004), which was approximately half of our present report. This increasing IAs concentration is most alarming, possibly due to the extensive withdrawal of groundwater and irrigation by contaminated water for an extended period.

Inorganic arsenic concentrations in fish, water, and sediments samples

As seen in Table 4, the highest IAs concentration is obtained for Lata fish (range, 0.04-0.51 mg/kg) among the selected fish species; which is maximum for the R_5 location (mean, 0.34 mg/kg) and minimum for R_3 sites (mean, 0.20 mg/kg). The 2nd highest [mean highest and lowest concentration] IAs contamination (range, 0.01-0.56 mg/kg) [R_1, 0.41 mg/kg and R_3, 0.05mg/kg] is found in koi fish. Again, the 3rd peak [mean highest and lowest concentration] concentration level of IAs (range, 0.07-0.64 mg/kg) [0.40 mg/kg from R_4 and 0.14 mg/kg from R_3 sites] is found for Tangra fish wherein the lowest [2nd lowest] IAs concentration is noted for Spotted snakehead fish [Walking catfish] (range, 0.04-0.42 mg/kg) [0.04-0.75 mg/kg]. Table 4 clearly signifies that none of the fish samples exceeded the permissible recommendation by WHO (1.00 mg/kg of DW) (Das et al., 2004).

In a previous study in Bangladesh, the highest IAs concentration was obtained in Lata fish (same as the present study) followed by other selected fish species (Das et al., 2004).

Table 4. Concentrations of IAs and their associated Health Risk Assessment in fish samples collected from five selected locations

Sample Name	Selected river (Location)	Fish samples: Range of IAs content (mg/kg)	Fish samples: Mean of IAs content (mg/kg)	Health hazard assessment: ADI (µg/kg-BW/day)	Health hazard assessment: HQ	Health hazard assessment: CR
Walking catfish = N_7	Chiri (R_1)	0.04-0.21	0.11	0.12	0.09	3.65×10^{-5}
	Tulshi Ganga (R_2)	0.25-0.75	0.52	0.55	0.38	1.71×10^{-4}
	Haraboti (R_3)	0.03-0.06	0.04	0.04	0.03	1.37×10^{-5}
	Tulshiganga (R_4)	0.14-0.47	0.29	0.30	0.21	9.44×10^{-5}
	Choto Jamuna (R_5)	0.04-0.31	0.13	0.13	0.09	4.11×10^{-5}
Spotted snakehead fish = N_8	Chiri (R_1)	0.15-0.42	0.30	0.31	0.22	9.76×10^{-5}
	Tulshi Ganga (R_2)	0.08-0.25	0.15	0.16	0.11	4.88×10^{-5}
	Haraboti (R_3)	0.04-0.18	0.10	0.10	0.07	3.25×10^{-5}
	Tulshi Ganga (R_4)	0.09-0.40	0.21	0.22	0.15	6.90×10^{-5}
	Choto Jamuna (R_5)	0.14-0.29	0.21	0.22	0.15	6.83×10^{-5}
Lata fish= N_9	Chiri (R_1)	0.14-0.41	0.23	0.24	0.17	7.55×10^{-5}
	Tulshi Ganga (R_2)	0.18-0.47	0.32	0.33	0.23	1.03×10^{-4}
	Haraboti (R_3)	0.07-0.41	0.20	0.21	0.14	6.51×10^{-5}
	Tulshi Ganga (R_4)	0.08-0.31	0.21	0.22	0.15	6.83×10^{-5}
	Choto Jamuna (R_5)	0.04-0.51	0.34	0.35	0.25	1.11×10^{-4}
Koi fish = N_{10}	Chiri (R_1)	0.14-0.56	0.41	0.43	0.30	1.34×10^{-4}
	Tulshi Ganga (R_2)	0.14-0.28	0.21	0.22	0.15	6.77×10^{-5}
	Haraboti (R_3)	0.01-0.09	0.05	0.05	0.04	1.63×10^{-5}
	Tulshi Ganga (R_4)	0.18-0.40	0.30	0.31	0.22	9.70×10^{-5}
	Choto Jamuna (R_5)	0.19-0.34	0.25	0.26	0.18	8.20×10^{-5}
Tangra fish = N_{11}	Chiri (R_1)	0.09-0.31	0.21	0.22	0.15	6.96×10^{-5}
	Tulshi Ganga (R_2)	0.12-0.31	0.23	0.24	0.17	7.48×10^{-5}
	Haraboti (R_3)	0.07-0.28	0.14	0.15	0.10	4.56×10^{-5}
	Tulshi Ganga (R_4)	0.24-0.64	0.40	0.42	0.29	1.30×10^{-4}
	Choto Jamuna (R_5)	0.09-0.25	0.18	0.19	0.13	5.86×10^{-5}

Table 5. Concentration of IAs and their exposure consequence in surface water, sediments of the selected river

	Sample	Chiri (R_1)	Tulshi Ganga (R_2)	Haraboti (R_3)	Tulshi Ganga (R_4)	Choto Jamuna (R_5)
Health hazard Assessment	Surface water (Mean ± SD, mg/l)	0.21±0.02	0.43±0.01	0.10±0.02	0.72±0.24	0.52±0.10
	ADI (µg/kg-BW/day)	10.50	21.50	5.00	36.00	26.00
	HQ	35.00	71.67	16.67	120.00	86.67
	CR	15.75	32.25	7.50	54.00	39.00
Soil contamination level Assessment	Sediment, N_{13} (Mean ± SD mg/kg)	0.40±0.00	0.41±0.03	0.13±0.02	0.53±0.12	0.45±0.05
	I_{geo} range	-0.53	-0.52	-1.02	-0.41	-0.48
	I_{geo} based sediment quality	Unpolluted	Unpolluted	Unpolluted	Unpolluted	Unpolluted
	EF range	0.25	0.26	0.08	0.33	0.28
	Level of metal (IAs) enrichment	Low enrichment	Low enrichment	Low enrichment	Low enrichment	Low enrichment

From the Table 5, the ranking of mean IAs concentration for water [sediment] may be expressed as: R_4 (0.72 mg/l) [0.53mg/kg] > R_5 (0.52 mg/l) [0.45 mg/kg] > R_2 (0.43 mg/l) [0.41 mg/kg] > R_1 (0.21 mg/l) [0.40 mg/kg] > R_3 (0.10 mg/l) [0.13 mg/kg]. The obtained results indicate that all the water samples collected from five selected rivers exceed the tolerable limit (0.05 mg/l) recommended by the different international organizations (FAO/WHO) (Heikens, 2006; EPA 2001). In the present study, the highest IAs concentration is obtained for water collected from the R_4 location, 14 and 72 times greater than the Bangladeshi and WHO's standards of 0.05 and 0.01 mg/l, respectively. These reported values may be considered extremely high when compared to the maximum concentration of arsenic in the Kampong Cham watershed of Cambodia of 0.00237mg/l (Phan et al., 2010) or in Lake Awassa and Koka of Ethiopia of 0.003 mg/l (Dsikowitzky et al., 2013). However, our present results are well supported by some previous reports (Ali and Tarafdar, 2003; Das et al., 2004; FAO/WHO 2021; Nasrabadi and Bidabadi, 2013; Nasrabadi et al., 2015) in which they claimed the same level of IAs concentration in different parts of the world. Nasrabadi and Bidabadi (2013) found a significantly high amount of As (67-420μg/L) in drinking water collected from thirteen rural wells in Kurdistan, Iran, and reported surrounding volcanic activities as a major cause of As pollution. However, the sediment and water hierarchies remain the same for all the selected rivers. It indicates a close correlation for IAs contamination between sediment and water caused by the neighboring biotic plus abiotic environment. Thus, the increasing IAs concentration in water, besides destroying the aquatic environment, also devastates the whole food cycle (IRAC 1993; Hossain et al., 2021). Such a higher concentration of IAs in the soil of the affected areas can be attributed to the use of fertilizers, pesticides, irrigation of crops by underground water, deposition of sediment, diagenesis process, etc. (Schoof et al., 1999).

Heath hazard assessment for polished rice sample

The Accepted Daily Intake (ADI), Hazard Quotient (HQ), and Carcinogenic risk (CR) of trace IAs metals from different agronomic crops ingestion by a mature adult are presented in Table 3. The ADIs of IAs are assessed based on the mean IAs content of each food with the particular consumption followed by the typical consumer's body weight (Ahmed et al., 2016). Table 3 represents the ADI of selected Agronomic crops collected from the five different Upazilla of Joypurhat district. Highest ADI (6.31 μg/kg-BW/day) is obtained in polished rice samples obtained from the S_3 location. The ADI [range: 2.15- 6.31 μg/kg-BW/day] for polished rice samples significantly surpassed the WHO's recommended value (2.10 μg/kg-BW/day) (Ahmed et al., 2016), wherein the highest ADI value is approximately three (3) times higher than that of WHO's prescribed limit.

Same as ADI's values, HQ's values (range: 1.49 - 4.37) for polished rice samples also surpassed the safe limit, i.e., larger than 1. The peak HQ's value (4.37) is obtained for polished rice collected from the S_3 location, followed by the polished rice sample collected from the S_4 area (3.99). The lowest HQ's value (1.49) is found for the polished rice collected from the S_2 site, followed by the polished rice samples collected from the S_5 site (1.97). Table 3 presents the calculated carcinogenic risk (CR) values for the polished rice sample that lies within E-07 to E-04 for S_1 (9.52×10^{-4}) and S_2 location (6.71×10^{-4}) but CR > E-04 for S_3 (1.97×10^{-3}), S_4 (1.80×10^{-3}) and S_5 (8.87×10^{-3}) location. However, HQ values were noted above 1.0 for all polished rice samples, demonstrating that trace IAs levels in polished rice samples collected from the Joypurhat district were above the acceptable range for non-carcinogenic human health alarms.

Heath hazard assessment for different vegetable, fish, and water samples

Other ADI values for selected vegetable samples lie within the permissible limit [Table 3]. For potato samples, maximum [minimum] ADI of 0.75 μg/kg-BW/day [0.29 μg/kg-BW/day] value is obtained for S_5 [S_1] location followed by S_3 [S_2] location (0.64 μg/kg-BW/day) [0.48 μg/kg-

BW/day] [Table 3]. For rest three selected vegetables [i.e. potato (range: 0.09 - 0.17 μg/kg-BW/day), radish (range: 0.20 - 0.47 μg/kg-BW/day) and arum leaves (range: 0.15 - 0.36 μg/kg-BW/day)] also lies within the WHO's advised value (2.10 μg/kg-BW/day).

From the Table 3, HQ values for all studied vegetables always lie below 1 as follows: Tomato (range: 0.40 - 0.71) > Potato (range: 0.20 - 0.52) > Radish (range: 0.14 - 0.32) > Arum leaves (range: 0.10 - 0.24). Table 3 shows the computed CR values for potato sample which also lies within E-07 to E-04 for every locations [$S_1 = 9.15 \times 10^{-5}$, $S_2 = 1.49 \times 10^{-4}$, $S_4 = 1.95 \times 10^{-4}$, and $S_5 = 2.33 \times 10^{-4}$] except for S_3 location [CR = 1.98×10^{-3} > E-04].

From Table – 4, along with the selected fish species, the highest ADIs are found in Lata fish; the maximum for R_5 location (0.35 μg/kg-BW/day) and minimum for R_3 sites (0.21 μg/kg-BW/day). The 2nd and 3rd peak ADI's are found for koi fish (range, 0.05 – 0.43 μg/kg-BW/day) and Tangra fish (range, 0.15- 0.42 μg/kg-BW/day), respectively. Whereas, the lowest [2nd lowest] ADI's value is noted for Spotted snakehead fish [Walking catfish] (range, R_3= 0.10 – R_1= 0.31 μg/kg-BW/day) [range, R_3= 0.04 – R_2 = 0.55 μg/kg-BW/day]. The descending order for HQ [CR] values (Table- 4) for all the selected fish species may be exhibited as follows: Lata fish (range, R_4 = 0. 15 – R_3 = 0.25) [range, R_4= 6.83×10^{-5}– R_3 = 6.51×10^{-5}] > koi fish (range, R_3= 0.04 – R_1 = 0.30) [range, R_3= 1.63×10^{-5}– R_1 = 1.34×10^{-4}] > Tangra fish (range, R_3 = 0.10 – R_4 = 0.29) [range, R_3 = 4.56×10^{-5} – R_4 = 1.30×10^{-4}] > Walking catfish (range, R_3 = 0.03 – R_2 = 0.38) [range, R_3 = 1.37×10^{-5}– R_2 = 1.71×10^{-4}] > Spotted snakehead fish (range, R_3 = 0.07 – R_1= 0.22) [range, R_3 = 3.25×10^{-5}– R_1 = 9.76×10^{-5}].

From Table- 5; the ascending order of ADI's {HQ}[TR] values for surface water for studied locations may be represented as follows: R_3 (5 μg/kg-BW/day) {16.65} [7.5] < R_1 (10.5 μg/kg-BW/day) {35} [15.75] < R_2 (21.5 μg/kg-BW/day) {71.67} [32.25] < R_5 (26 μg/kg-BW/day) {86.67} [39] < R_4 (36 μg/kg-BW/day) {120} [54].

However, ADI's values for all selected vegetables and fish remain within WHO's commendation (2.1 μg/kg-BW/day). It is a good sign for the regular consumer regarding these vegetables and fish ingestion. HQ values for all vegetables and fish samples are far lower than 1, indicating negligible hazards for habitual consumers. But CR values remain within the safety limit for all selected vegetables except for the potato collected from the S_3 location (Table 3). So, Except for potatoes from this specific region, others are secure for consumption. Again, health risk assessment for surface water indicates some severe consequences. Although river water is not directly drunk by habitual people, urban people sometimes use it for household chores, irrigating, drinking their household animals, etc. Thus, a severe risk may arise as the above activities easily accommodate carcinogenic arsenic into the food chain. And later easily disrupt our food chain through bio-accumulation and then bio-magnification.

Soil contamination level Assessment

The assessed I_{geo} and EF values are shown in Table 3 and Table 5 for every sampling station. The hierarchy for I_{geo} [EF] for soil samples collected from corresponding rice plow land of different sampling station may be represented as follows (From Table 3): S_2 (1.47) [25.08] > S_5 (1.42) [22.14] > S_1 (1.35) [19.02] > S_4 (1.33) [17.95] > S_3 (1.28) [15.95].

The order of I_{geo} [EF] in case of potatoes assembled from selected sites is as follows (Table 3): S_4 (1.35) [19.02] < S_3 (1.40) [21.33] < S_5 (1.41) [21.77] < S_2 (1.45) [23.96] < S_1 (1.47) [25.08]. The hierarchy for I_{geo} [EF] against soil samples collected from tomato field is: S_5 (1.46) [24.20] > S_1 (1.41) [21.96] > S_3 (1.37) [19.96] > S_4 (1.33) [18.20] > S_2 (1.30) [17.02]; whereas the hierarchy for radish field is S_3 (1.45) [23.65] > S_5 (1.40) [21.33] > S_2 (1.37) [20.08] > S_4 (1.35) [19.08] > S_1 (1.25) [15.08]. Again, Arum leaves respective soil exhibit the hierarchy for I_{geo} [EF] is as follows [Table 3]: S_5 (1.50) [26.96] > S_4 (1.47) [24.65] > S_2 (1.44) [23.33] > S_3 (1.44) [23.33] > S_1 (1.39) [20.89]. But, Lowest I_{geo} [EF] values are obtained for river sediment (Table 5); which may be represented as follows: R_3 (-1.02) [0.08] < R_1 (-0.53) [0.25] < R_2 (-0.52) [0.26] < R_5 (-0.48) [0.28] < R_4 (-0.41) [0.33].

As seen in Table 3, the I_{geo} range confirmed around 100% of the agronomic fields are moderately polluted by various anthropogenic activities. Additionally, EF values confirmed that moderately severe enrichment occurred in approximately 65% agronomic field, and significant enrichment occurred in the rest 35% agronomic field. Again, I_{geo}-based analysis ensures sediment quality in which no anthropogenic pollution occurs in the sediment sample; for some cases, too minute natural contamination occurs. In addition, calculations of EF values also confirmed low to moderate enrichment of As. The difference in contagion level between agronomic field and river sediment may be due to the easy accumulation of IAs, which is facilitated by heavy irrigation using IAs-contaminated water (FAO/WHO 2021).

CONCLUSIONS

This study analyzed the IAs content in major foodstuffs, soil, sediment, and water collected from the Joypurhat district. The present study confirms that the IAs contents in all the foodstuffs lie within the FAO/WHO's recommended value except the polished rice sample. On the other hand, significant amounts of IAs are found in the soil, sediments, and water samples of the entire Joypurhat district. Health risk consideration ensures that non-carcinogenic risks for polished rice consumption and other foodstuffs are secured for ingestion. Additionally, untreated river waters are at a vulnerable state for severe cancer and non –carcinogenic health risk. Selected sampling stations are exclusively polluted by the enrichment of significant to moderately severe IAs caused by different anthropogenic actions. This type of situation arises from the unconscious and excessive withdrawal of contaminated groundwater for an extended period without considering the substitution of uncontaminated irrigating water sources like dug wells, rainwater, deep tube wells, etc. This terrible mistake leads to horrible groundwater contamination. Consequently, it may be proven an awful threat and a drastic source of destruction for surrounding inhabitants. Proper monitoring, implementation of appropriate law, sufficient human funds, modern water treatment plants, awareness, and knowledge regarding IAs contamination might be a pre-solution of upcoming health risks in Joypurhat district as well as entire Bangladesh.

ACKNOWLEDGMENTS

The authors sincerely thank to the department of Glass and Ceramic Engineering (GCE), Bangladesh University of Engineering and Technology (BUET), Dhaka, Bangladesh, for providing outstanding laboratory facilities.

GRANT SUPPORT DETAILS

The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interest regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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