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Accumulation of arsenic and other metals in soil and human consumable foods of Meherpur district, southwestern Bangladesh, and associated health risk assessment

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Abstract

Groundwater and soil contaminated with arsenic (As) are significant environmental health concerns worldwide and have become a serious health issue for millions of Bangladeshis. This contaminated groundwater is commonly used for Boro rice production during the dry season in Bangladesh, leading to long-term As deposition in soils. The consequences of long-term irrigation with As-contaminated groundwater for bioaccumulation in food crops and hence dietary exposure to As and other metals are a threat to the resident. Highly arsenic (292 μg L^{-1}) contaminated groundwater used to irrigate in the study area increases the As content (mean: 24.5 mg kg^{-1}) in irrigated soil than non-irrigated soil (mean: 8.43 mg kg^{-1}). The high enrichment factor (mean: 16.8) and geo-accumulation index (mean: 2.33) revealed that the soil of that particular area is strongly contaminated by As. In paddy, roots showed the maximum As concentration (mean: 139.8 mg kg^{-1}) followed by straws (mean: 5.25 mg kg^{-1}) and grains (mean: 1.23 mg kg^{-1}) where 85–95% As exist in the roots of paddy, and only 0.8–1% As translocate to the harvesting part of the rice in the study area. In food samples, two rice grains (R-1, R-3) and a drumstick (FS-5) exceed the Indian standard of As concentration (As: 1.1 mg kg^{-1}) in food. According to World Health Organization, the metals concentrations in all food samples exceeded the permissible limit except for Cu and Pb in red amaranth; Cr, Cd, and Pb in guava; and Cu and Cd in drumsticks. Higher values of bioaccumulation factor (BAF: 2.94) and the net translocation coefficient (NTC: 6.17) indicate As-accumulation in food from adjacent contaminated soil. Daily metal intake from food consumption suggests that the heavy metal(loid)s like As, Ni, and Pb represent high concentrations and exceed the daily ingestion limits (As: 3 μg kg^{-1} day^{-1}, Ni: 1.3 μg kg^{-1} day^{-1}, and Pb: 3.57 μg kg^{-1} day^{-1}) developed by the Food and Agriculture Organization and European Food Safety Authority. Carcinogenic ($>10^{-6}$) and non-carcinogenic (HQ > 1) health risks of As, Pb, Cd, and Cu suggest that the community was at potential health risk according to the United States Environmental Protection Agency.

Keywords Arsenic, Soils and crops, Bioaccumulation index, Hazard quotient, Carcinogenic risk, Human exposure

Introduction

Arsenic (As), a group-1 carcinogen is considered as one of the most significant global environmental toxicants which naturally occurs in rocks and soils [57, 78]. Although it is frequently present as a component of minerals, it is incredibly hard to find as a pure element. The primary As-bearing minerals are realgar (As_4S_4),

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arsenopyrite (FeAsS), niccolite (NiAs), and orpiment (As_2S_3), but it can also be found bonded to iron, manganese (oxy-)hydroxides, and sulfides in sedimentary rocks and soil [72, 74]. The levels of arsenic in foods generally reflect normal accumulation from the environment [80]. The problem of As contamination in agricultural soils occurs frequently and is likely to get worse in the near future. High concentrations of As in groundwater and surface soil have been reported in many countries of the world including Bangladesh, India, China, and Vietnam [42, 77]. There have been reports of high As concentrations in groundwater up to 2621 g L^{-1} [19, 20] and paddy soils with a range between 0.1 and 950 mg kg^{-1} [40, 86]. Domestic sewage, industrial wastewater, and irrigation utilizing As-enriched groundwater may be the main sources of As contribution to the soil-crops environment that influences food production all over the world [42]. High levels of As in soil pore water and crops have been associated with irrigation using As-enriched groundwater and the subsequent reductive dissolution of iron (Fe) (oxy)hydroxide minerals [108]. It has been observed that irrigation groundwater contains As, which accumulates in soil and not only affects agricultural growth but also puts food safety at risk across the food chain [85]. The increasing quantity of As in groundwater may endanger more than 200 million people worldwide, primarily in Asia [63, 84]. In South and Southeast Asia, especially in Bangladesh, India, Cambodia, Vietnam, and Taiwan, a major pathway of human exposure to As is via drinking water and intake from rice [40, 91]. Therefore, As contamination in drinking water and food is considered as one of the most serious issues associated with public health in Bangladesh [68]. Currently, in Bangladesh, 62 districts out of 64 have been affected by As and face a serious threat to public health, and 97% of Bangladesh's rural population relies on tube wells as their primary source of drinking water, and the country's entire supply of these wells is contaminated with As [1]. Together, 85 million people are at risk, and several thousand of them have shown symptoms of arsenicosis. Paddy rice, the main staple food for the inhabitants, is also irrigated with that As-contaminated groundwater, which causes a gradual increase of As in the soil. Therefore, few studies have reported that the As concentration in Bangladesh's groundwater, soil, and plants is higher than the permissible limit and that 85–95% of the total As in rice and a vegetable was inorganic [29].

Bangladesh is mostly situated in the Bengal Basin, formed by the three mighty Ganga–Brahmaputra–Meghna (GBM) river systems. The Ganga–Meghna–Brahmaputra (GMB) river basin is at risk due to the presence of high concentrations of arsenic in groundwater [26]. This sedimentary basin was developed by the deposition of significant amounts of sediments containing As, primarily from the Himalayas, during the Pleistocene and Holocene periods. In the fan deposit areas and the Holocene alluvium, arsenic is eroded from these sediments and entered groundwater aquifers. Although it is unclear how exactly As leaches into groundwater, most researchers agree that the As contamination source is geological, but there is still controversy over identifying the source. The source of As in the groundwater of Bangladesh remains obscure [17]. To date, three significant theories have reported the mechanisms of As contamination in underground water in the GMB basin: oxidation of As-bearing pyrite, reductive dissolution of FeOOH and MnOOH, and competitive exchange of phosphate anions in the aquifer [1, 2, 50, 90, 111]. The second mechanism has been more acceptable in the last couple of years [73].

Massive extraction of groundwater contaminated with As for irrigation has led to high concentrations of As in top soils [52]. Arsenic contamination of the soil raises the amount of As that accumulates in crops [58]. According to European Community (EC), the maximum permissible limit of arsenic for agricultural soils is 20 mg kg^{-1} and a high concentration of arsenic is toxic for plants, gradually reducing crop yields [23, 35, 66]. Natural As concentrations in soils are approximately 5 mg kg^{-1} globally but can be elevated depending on the nature of the soil and anthropogenic loading of As from different chemicals and pesticides [48]. Contaminated soil is the primary pathway to exposure to poisonous metals in the human body. Human dietary products like crops, vegetables, and fruits grow in contaminated soils capable of translocating metals from soil to crops, resulting in a probable human health hazard.

Geological and geochemical studies revealed that the release of As in various parts of the earth is caused by both anthropogenic activities and natural rock or mineral weathering processes [10]. The situation is exacerbated by the fact that As, once present in groundwater and soil, can be bioaccumulated by terrestrial and aquatic biota, which are either consumed by humans or absorbed into cattle feed [51]. The uptake of As in plants from irrigated water and soils is large enough to cause potential health hazards due to extensive contamination by As. Chronic ingestion of heavy metals can lead to a deleterious health hazard for a human being, even in a trace amount [11]. Arsenic is phytotoxic in high quantities and is not necessary for plant growth. Typical As concentrations are 1–1.7 mg kg^{-1} for plant shoots growing in unpolluted soils [34]. Soil–plant exposure to As can occur in humans through As transfer [25]. Some common vegetables have regulatory limits on their As content, such as Poland's standard for fresh vegetables is 0.2 mg kg^{-1}, China's standard for rice, beans, and vegetables is 0.5 mg kg^{-1} [59] and

Japan's is 1.0 mg kg^{-1} for spinach, tomatoes, and cucumbers. The European Commission and the US Food and Drug Administration proposed a maximum As-acceptable limit of 0.1 mg kg^{-1} for rice [36]. Aside from drinking arsenic-contaminated water, residents are also exposed to arsenic through dietary intakes of foods and vegetables, particularly rice cultivated on tainted fields in the region [33, 81]. Arsenic accumulated in rice from contaminated groundwater and soils is still considered the major dietary contributor to As intake for people [79].

Arsenic exposure has many adverse human health effects. Nearly all human body systems are impacted by exposure to arsenic, but the cardiovascular, neurological, hepatic, renal, hematological, endocrine, respiratory, and reproductive systems are the most severely affected by arsenic [65, 68, 84]. In addition, exposure to As during pregnancy increases the risk of infant death, spontaneous abortion, stillbirth, preterm birth, low birth weight, delayed child growth, a compromised immune system, a lower IQ, neurotoxicity, and neurodevelopmental impairment [102]. Chronic poisoning symptoms include skin diseases such as keratosis and possibly amputation due to gangrene. In Bangladesh, one of the most typical effects of long-term exposure to arsenic is skin lesions, i.e., arsenicosis, or hardening of the skin with lots of black and white spots [43]. High concentrations of Pb and Cd in food cause nerve, skeletal, circulatory, enzymatic, endocrine, and immunological system damage, hypertension, as well as pulmonary and renal impairment in the human body [45].

Most of the published paper is on As concentration in groundwater, soil, and crops, but there is little information on the bioaccumulation factor and net translocation coefficient of As from soil to crops and the associated carcinogenic health risk in the study area. Although some studies have suggested arsenic toxicity in rice cultivation, there have been very few investigations on its transfer from soil to rice and vegetables. Arsenic accumulation in food crops, notably rice and vegetables irrigated with As-contaminated groundwater, has been extensively observed in recent years [75, 76]. Little work has been conducted to investigate the translocation mechanism of As from root to grain in that As hotspot area where the As-affected people are found more. Moreover, only some authors have published a paper on As in soil and paddy. Still, no one has published an article on As and other metals in consumable foods and their impact on human health in this arsenic hotspot zone. To assess the health and environmental concerns, it is essential to investigate the transfer of arsenic from soil to edible parts of rice and vegetables in the study area. The current research work aimed to (1) assess the contamination level of As and other metals in soils and crops in the study area, (2) identify the possible sources and correlation among the metals with multivariate approaches, (3) measure the transfer of As from soils to the edible parts of various vegetables and rice grains, and (4) assess the human health risk associated with As-contaminated rice and vegetable consumptions.

Materials and methods

Site description

This research area is located in the Gangni Sub-District of Meherpur, Bangladesh, and is known as Bholadanga. This location is 15 km distance from Meherpur city and is situated on the boundary of the Meherpur-Kushtia-Chuadanga district. Agricultural activities are the primary source of livelihood for local inhabitants. It is located amid north and southwestern Bangladesh between the graphical coordinates of 23°51′00″ to 23°51′15”north latitudes and 88°51′35″ to 88°51′50″ east longitudes (Fig. 1). It is bordered on the north by Doulatpur Upazila, on the east by Mirpur Upazila, on the south by Alomdanga Upazila, and the west by Meherpur Sadar Upazila. The Mathabhanga is a transboundary river between India and Bangladesh that begins on the right bank of the Padma near Munshiganj, Kushtia District. Like the rest of Bangladesh, the study area has a tropical Indian Monsoon climate with a tropical wet and dry, or savanna, climate. The district's yearly temperature is 28.33 ℃ (maximum 39 ℃ in April and minimum 10.5 ℃ in January), and it is 0.59% higher than Bangladesh's averages. The study area is situated at an elevation of 23 m (76 feet) above sea level, whereas the topographic gradient runs north to south [101]. The majority of groundwater flows in the same direction as the topographic slope.

Sampling

Groundwater sample was collected from a nearby shallow tube well used to irrigate the agricultural land and village area. Before sampling, an electric pump was run for about 2–3 min to obtain fresh groundwater. A previously cleaned 500-mL plastic bottle was used to collect groundwater and then acidified with concentrated HNO_3 (1 mL). Soil samples ($n = 8$) and human consumable food samples ($n = 9$), along with paddy, fruits, and vegetables, were collected randomly from the nearby agricultural land and village area. Soil samples at 0–15 cm depth were collected (250 g each for different analyses) from adjacent fields of food samples. To create a homogeneous composite sample, triplicate soil samples were collected at random from each site and stored in a polythene bag. Four rice plants (RP) and five vegetables and fruits (FS) with edible parts were collected randomly from each field. All the food samples were kept in clean air-protected polyethylene bags with an icebox for carrying to the

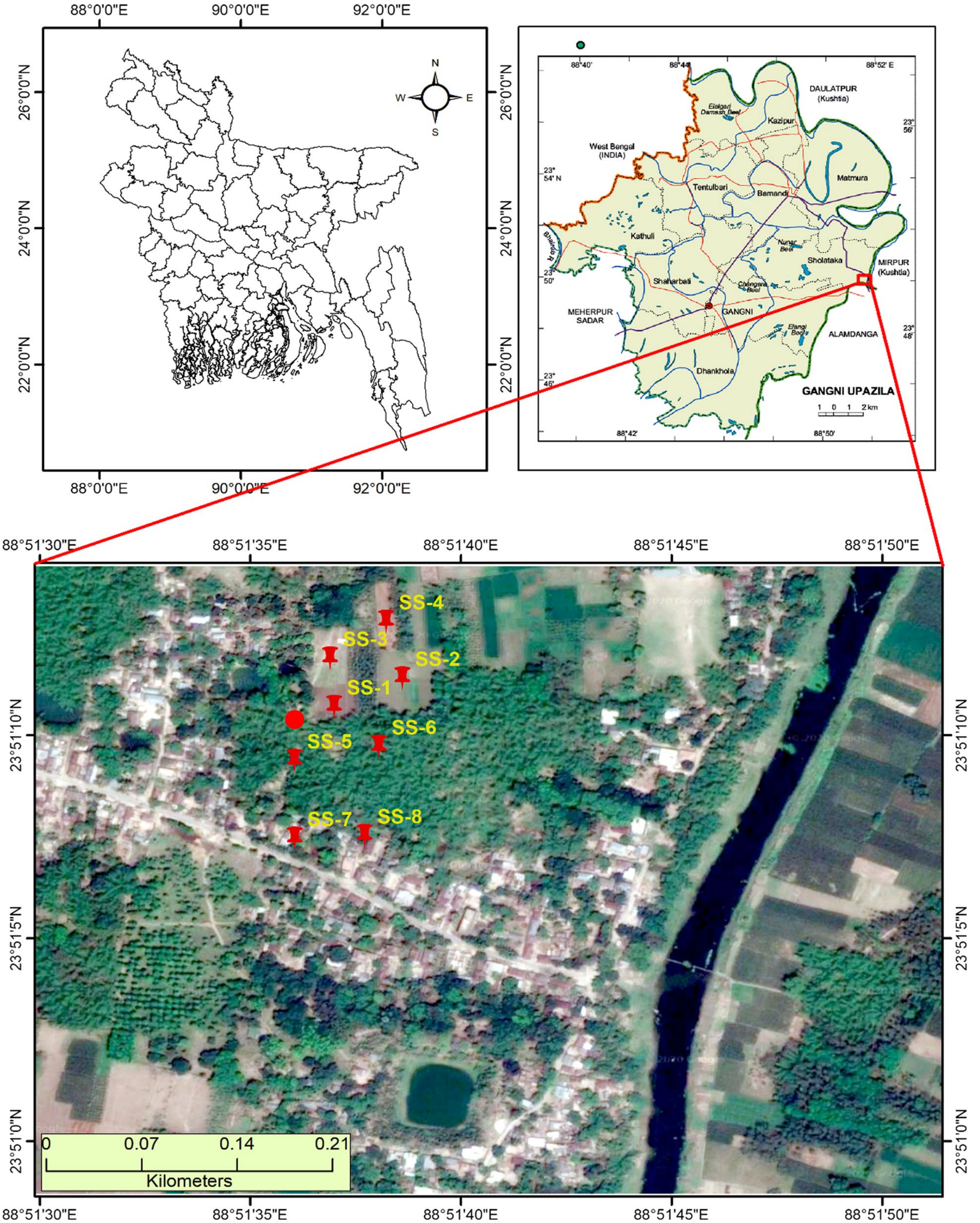


Fig. 1 Location map of the study area with soil sampling points at Meherpur, southwestern Bangladesh (red circle: shallow tube well contaminated with arsenic)

laboratory and stored at 4 ℃ until analysis. At the time of sampling, biodata (age, body weight, and sex) and food consumption rates and behaviors of the local people were collected to obtain individual As exposure assessments. A detail of the collected food samples is shown in Table 1.

Sample preparation

After collecting, the soil samples in the laboratory were dried up in the air, ground and passed through a 2-mm sieve, and finally stored in plastic pots. All soil samples were then oven-dried at 105 ℃, and the collected triplicate samples were carefully mixed entirely to get a composite sample. Then, prepared soil samples were kept in polyethylene bags for analysis. To extract the total metal concentration, about 10 g of dry soil sample was weighed into a 100-mL glass beaker. Then, 20 mL of concentrated HNO_3 was added to the samples, and the suspension was heated for approximately 2 h at 120 ℃ in a hot plate. Then, 5–10 mL of concentrated $HClO_4$ was added and again digested at 100 ℃ until it was a clear solution. The solution was then filtered through a Whatman™ 41 (150 mm) filter paper in the calibrated volumetric flask (100 mL) through multiple washing of the glass beaker and finally, it was stored in the plastic bottle for chemical analyses.

All food samples were rinsed with distilled water and washed with 0.05 M HCl solution to remove outside metals adhered in the free dust and soil particles on the root, straw, and grain (paddy). All crops, vegetable, and fruit samples were then dried up in the oven for 48 h at 110 ℃ to remove moisture. The food samples were then crushed to make powder. For metal content determination, about 10 g of dried powder samples were weighed in a 100 mL beaker and gradually heated up to 500 ℃ in a muffle furnace for ashing. After complete ashing, the samples were digested with 10 mL HNO_3 and 5 mL $HClO_4$ at around 100 ℃ in a hot plate and then filtered after cooling in a 50-mL volumetric flask and stored at 4 ℃ for analysis.

Sample analysis

Metal concentrations in soil and food samples were analyzed at the Institute of National Analytical Research and Service (INARS), Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka, Bangladesh, which is an ISO/IEC 17025:2017 accredited laboratory. The concentrations of As, Cu, Zn, Fe, and Mn in the soil and food samples were measured using an atomic absorption spectrometer (models: Spectra AA220 and AA240FS, Varian, Australia) equipped with electro-thermal temperature controller and flames, whereas Pb, Cd, Co, Ni, and Cr were determined using a Zeeman atomic absorption spectrometer (model: GTA 120-AA240Z, Varian, Australia) equipped with a graphite furnace. The certified reference materials (CRM) of metals of 1000 ± 4 mg L^{-1} stock solution (Fluka Analytical, Sigma-Aldrich, Germany) were used to construct the calibration curves, and the samples were measured against the prepared curves. All samples were measured in duplicate and the mean concentrations with a relative standard deviation (RSD) of less than 5% were considered for reporting.

Analytical quality control

Throughout the study period, from sample collection to laboratory analysis, the quality of the analysis was carefully monitored. For sample preparation, analytical-grade HNO_3 and $HClO_4$ from Merck Germany, and deionized water (electrical conductivity < 0.2 μS cm^{-1}, resistance 18.2 MΩ cm at 25 ℃) were used. All glassware used for the analysis, such as the pipette and volumetric flask, were calibrated and cleaned prior to use by soaking in 10% (v/v) HNO_3 overnight and then rinsing with deionized water. For sample weighing, a digital electrical balance (GR-200, A&D Company Limited, Tokyo, Japan) was used. The atomic absorption spectrometer was calibrated using the certified reference material (CRM) of the elements. The analytical accuracy and precision were ensured through the replicate measurement of CRM and the samples where the RSD was less than 5%. The measurement of sample blank and quality control standards

Table 1 List of collected food samples from Meherpur district of southwestern Bangladesh with descriptive identification

Vernacular name	English name	Scientific name	Family	Soil sample code	Food sample code	Number of samples
Dhan	Paddy	*Oryza sativa*	Graminae	SS (1–4)	RP (1–4)	4
Peyara	Guava	*Psidium guajava*	Myrtaceae	SS-5	FS-1	2
Lal shak	Red amaranth	*Amaranthus gangeticus*	Amaranthaceae	SS-7	FS-2	2
Dalim	Pomegranate	*Punica granatum*	Punicaceae	SS-6	FS-3	2
Kochu	Arum	*Colocasia esculenta*	Araceae	SS-8	FS-4	2
Sajina	Drumstick tree	*Moringa oleifera*	Moringaceae	SS-5	FS-5	2

were performed sequentially. The spike recovery in the analysis of metals was in the range of 96–104%. The detection limit for the analyzed metals were 0.2, 67.7, 3.1, 2.1, 2.2, 6.2, 7.5, 0.4, 0.3, and 1.2 µg/L for As, Fe, Mn, Co, Ni, Cu, Zn, Cr, Cd, Pb, respectively. Further details about quality assurance and quality control are described in our previous works [3, 28, 87].

Data analysis

Statistical analysis and software used

Analytical data were statistically analyzed with IBM SPSS Statistics software (Version 20, IBM Corporation, USA). To identify the source(s) of metals, multivariate statistical analyses such as principal component analysis (PCA), hierarchical cluster analysis (HCA), and correlation analysis were used. The results of HCA were expressed with a dendrogram. For spatial distribution, mapping, and other analytical calculations, Arc-GIS (version: 10.3.1), Adobe Illustrator (version: 2017), and Microsoft Excel 2013 were used usually.

Assessment of soil contamination

The enrichment factor (EF) and geo-accumulation (Igeo) index are the most often used methods for determining soil contamination degree. Enrichment factor and Igeo were used to assess the status of elemental contamination in the studied soil.

Enrichment factor (EF)

The most often used approach for estimating the impact of anthropogenic input on soil contamination is an enrichment factor (EF) for metal contamination levels above uncontaminated background levels [9, 103]. This study has potentially explained the metal associations and enrichment pattern throughout the period [14]. According to Taylor [93], the enrichment factor for each specific metal in the sample is typically calculated using the following relationship:

$$\mathrm{EF} = \frac{\left(\frac{\mathrm{Metal}}{\mathrm{Fe}}\right) sample}{\left(\frac{\mathrm{Metal}}{\mathrm{Fe}}\right) Background}.$$

Iron (Fe) was considered as the reference metal for geochemical normalization. The degree of soil contamination using enrichment factors can be classified into seven classes such as 0–1 (no enrichment), 1–3 (small enrichment), 3–5 (medium enrichment), 5–10 (medium to acute enrichment), 10–25 (acute enrichment), 25–50 (very acute enrichment), and greater than 50 (extremely acute enrichment).

Geo-accumulation index (I_{geo})

Muller [55] introduced the equation to represent the geo-accumulation index (Igeo), which is commonly used to measure additional metal concentrations in soil:

$$\mathrm{Igeo} = \frac{\mathrm{Log}_2\,(C_n)}{1.5\,(B_n)},$$

where Cn refers to the metal concentration from analysis and Bn is the geochemical background value of that metal. Muller [56] classified the geo-accumulation index as follows: 0 (uncontaminated), 0–1 (uncontaminated to moderately contaminated), 1–2 (moderately contaminated), 2–3 (moderately to strongly contaminated), 3–4 (strongly contaminated), 4–5 (strongly to extremely contaminated), and ≥ 5 (extremely contaminated). For the calculation of EF and Igeo by Krauskopf and Bird [41], the crustal assemblage was used as the background values.

Degree of metal contamination in foods

Bioaccumulation factor (BAF) of metals

Metals can enter the human body through three routes: natural or anthropogenic metal loads on soil, soil to food crops, and food crops to humans. The BAF is the potential ability of a plant species to transfer or accumulate a specific metal from adherent soil to edible sections of plants and can be described as the concentration ratio of metal in the plant material to that in the relevant soil [53, 78, 103]:

$$\mathrm{BAF} = \frac{\mathrm{C}_{\mathrm{Plant}}}{\mathrm{C}_{\mathrm{soil}}},$$

where Cplant and Csoil represent metal concentrations in green plants and soil, respectively, on a dry weight basis. BAF can control the metal's exposure to humans via the food chain [15].

Net translocation coefficient (NTC)

Previously, the term "translocation coefficient" (TC) was used to describe a plant's ability to absorb and translocate elements from water and soil via their root to the consumable part [49]. It was estimated on a dry weight basis for each soil–plant sample using the equations listed below [92, 104]:

$$\mathrm{TC\,root\,/\,soil} = \mathrm{Croot\,/\,Csoil},$$

$$\mathrm{TC\,straw\,/\,root} = \mathrm{Cstraw\,/\,Croot},$$

$$\mathrm{TC\,grain\,/\,straw} = \mathrm{Cgrain\,/\,Cstraw},$$

where Csoil, Croot, Cstraw, and Cgrain are the total concentrations of metal in soil, root, straw, and grain, respectively.

Daily intake of metals (DIM)

The daily ingestion of metals (DIM) typically depends on the amount of a particular metal in the food sample and the ingestion rate of that respective food. The DIM for adults was calculated by the following relationship [54]:

$$\mathrm{DIM} = \frac{\mathrm{C_{Metal \times W_{Food}}}}{\mathrm{B_w}},$$

where Cmetal is the metal concentration in food samples, Cwood is the average ingestion rate per day of the respective food, and Bw is the average body weight. The average daily ingestion of rice (grain), vegetables, and fruit for adults was considered to be 372 g, 274 g [112], and 200 g per person per day, respectively, while the average body weight for adults in the study was considered as 65 kg.

Health risk assessment

Non-carcinogenic risk

Non-carcinogenic risk assessments are often used to determine the possible health consequences of pollutants using the hazard quotient (HQ) [94]. The HQ values for each heavy metal can thus be measured through food consumption by nearby residents, and calculations were done using the conventional assumptions for an integrated USEPA risk study, as shown below [89, 96, 97]:

$$\mathrm{HQ} = \frac{\mathrm{EF} \times \mathrm{ED} \times \mathrm{FIR} \times \mathrm{MC}}{\mathrm{RfD} \times \mathrm{BW} \times \mathrm{AT}} \times 10^{-3},$$

where EF is the exposure frequency (365 d year^{-1}); ED is the exposure duration refers to life expectancy (72 years for Bangladesh); FIR is the food ingestion rate (g d^{-1}). The daily ingestion of rice, vegetables, and fruits is considered to be 372, 274, and 200 g, respectively [112]. MC is the metal concentration of every individual sample (mg kg^{-1},RfD oral reference dose (mg kg^{-1} d^{-1}). RfD value is 0.003, 1.5, 0.001, 0.004, 0.02, 0.04, and 0.3 for As, Cr, Cd, Pb, Ni, Cu, and Zn, respectively [99, 100]. BW is the average body weight (adult) in the study area. AT refers to the average lifetime for non-carcinogenic health hazards which is EF*ED = 365*72 = 26,280 days. HQ value of less than one suggests that daily exposure to a given metal through food consumption is unlikely to induce non-carcinogenic health impacts, whereas an HQ value larger than one indicates that chronic health concerns may develop in the exposed population [6].

Carcinogenic risk

Carcinogenic risk (CR) refers to an individual's lifetime risk of developing cancer as a result of exposure to a potential carcinogen. Cancer risk from As, Pb, and Cd exposure over a lifetime was calculated using the cancer slope factor (CSF) given by the USEPA [97]. A carcinogenic hazard value greater than 10^{-6} indicates that the people in the respective area were at high carcinogenic risk according to USEPA [98]:

$$\mathrm{CR} = \frac{\mathrm{EF} \times \mathrm{ED} \times \mathrm{FIR} \times \mathrm{MC} \times 10^{-}3}{\mathrm{BW} \times \mathrm{AT}} \times \mathrm{CSF}$$

or,

$$\mathrm{CR} = \frac{\mathrm{ADD} \times \mathrm{CSF}}{1000}.$$

CSF is the carcinogenic slope factor, which measures the upper-bound possibility of a single person developing cancer due to lifetime exposure to a specific level of probable carcinogen through an ingestion route. The CSF value of As, Pb, and Cd is 1.5, 0.0085, and 15 mg kg^{-1} day^{-1}, respectively [98].

Results and discussion

Contamination assessment of heavy metals in soil

The analytical results of metal(loid)s concentration in soil samples of the research area are shown in Table 2. The range of arsenic (As) concentration in irrigated land is 16.6–40.2 mg kg^{-1} (mean ± SD: 24.5 ± 10.9) and in non-irrigated/partially irrigated land is 4.54 to 10.3 mg kg^{-1} (mean ± SD: 8.43 ± 2.64). Except for all other metals, the concentration of As (mean ± SD: 16.5 ± 11.3) was found higher value than the average concentration of As (4 to 8 mg kg^{-1}) in soils of Bangladesh [4, 107]. The rest of the metal concentration shows average crustal values, which are naturally present as an earth material. Heavy metal(loid)s contamination of agricultural soil is one of the most serious obstacles to sustainable development, particularly in developing nations like Bangladesh. The degree of soil contamination in the study area is represented in Table 3. The enrichment factor (EF) is a widely used statistic for measuring how much the amount of an element in a sampling medium has risen due to human activity relative to normal natural abundance. Results of the enrichment factor (EF) of Co, Cd, and Cu show no enrichment ($\mathrm{EF} < 1$). Fe, Mn, Zn, and Pb show slight enrichment ($1 \leq \mathrm{EF} < 3$), and the results of arsenic show acute enrichment ($10 \leq \mathrm{EF} < 25$). EF value ranges from 0.05 to 1.5, revealing a natural process, i.e., the metal supply is entirely from the crustal materials whereas, values greater than 1.5 indicate metals are mostly from the anthropogenic origin [110]. Significant enrichment of arsenic suggests that the soils were contaminated, primarily due to anthropogenic activity like pumping groundwater from the arsenic-bearing aquifer for irrigation purposes. The metal enrichment factor of less than 1.5 suggested that these metals did not significantly contaminate the soils in the studied area and that their small

Table 2 Metal concentration (mg kg^{-1}) in soil samples of Meherpur district, southwestern Bangladesh

Sample ID	Description	As	Fe	Mn	Co	Ni	Cu	Zn	Cr	Cd	Pb
SS-1	Irrigated soil	17.5	26255.4	577.3	11.4	28.4	23.9	73.0	19.0	0.12	10.0
SS-2		23.8	29137.8	656.0	12.2	30.3	26.6	78.0	25.4	0.16	11.0
SS-3		40.2	38356.4	825.2	17.9	46.8	42.9	87.2	35.2	0.13	16.3
SS-4		16.6	19114.3	411.6	8.23	20.3	16.5	54.5	13.0	0.06	9.01
Mean		24.5	28216.0	617.5	12.4	31.5	27.5	73.9	23.2	0.12	11.6
SD		10.9	7965.80	171.9	4.03	11.1	11.1	13.8	9.50	0.04	3.25
SS-5	Non-irrigated soil	4.54	22304.0	415.2	8.78	22.3	22.3	118.3	13.2	0.11	10.9
SS-6		9.10	23315.0	510.4	11.0	27.5	22.9	82.4	18.1	0.08	9.29
SS-7		9.77	25042.2	494.4	12.0	28.6	24.3	66.2	18.6	0.07	9.57
SS-8		10.3	23119.4	515.7	10.6	26.2	26.4	74.0	17.4	0.06	10.0
Mean		8.43	23445.2	483.9	10.6	26.2	24.0	85.2	16.8	0.08	9.94
SD		2.64	1151.20	46.70	1.35	2.75	1.82	23.0	2.47	0.02	0.70
Descriptive statistics											
Mean	Both irrigated and non-irrigated soil	16.5	25,830.7	550.7	11.5	28.8	25.7	79.2	20.0	0.10	10.8
Maximum		40.2	38,356.4	825.2	17.9	46.8	42.9	118.3	35.2	0.16	16.3
Minimum		4.54	19,114.3	411.6	8.23	20.3	16.2	54.5	13.0	0.06	9.01
Range		35.7	19,242.1	413.6	9.64	26.5	26.4	63.8	22.2	0.10	7.29
SD		11.3	5853.6	136.7	2.94	8.01	7.62	18.7	7.27	0.04	2.35

Table 3 Results of enrichment factor (EF) and geo-accumulation index (*Igeo*) of soil samples of Meherpur district, southwestern Bangladesh

Sample ID	As		Fe		Mn		Co		Cu		Zn		Cd		Pb	
	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}
SS-1	**18.5**	**2.70**	1	−1.51	1.16	−1.30	0.87	−1.72	0.83	−1.79	1.99	−0.52	1.14	−1.32	1.47	−0.96
SS-2	**22.7**	**3.14**	1	−1.36	1.18	−1.12	0.83	−1.62	0.83	−1.64	1.91	−0.43	1.37	−0.91	1.45	−0.82
SS-3	**29.1**	**3.90**	1	−0.97	1.13	−0.79	0.93	−1.07	1.02	−0.94	1.62	−0.27	0.85	−1.21	1.63	−0.26
SS-4	**24.1**	**2.62**	1	−1.97	1.14	−1.79	0.86	−2.19	0.78	−2.33	2.04	−0.95	0.78	−2.32	1.81	−1.11
SS-5	**5.70**	**0.75**	1	−1.75	0.97	−1.78	0.79	−2.09	0.91	−1.89	3.79	0.17	1.23	−1.44	1.88	−0.84
SS-6	**10.8**	**1.75**	1	−1.68	1.15	−1.48	0.95	−1.76	0.89	−1.59	2.52	−0.35	0.86	−1.91	1.53	−1.07
SS-7	**10.8**	**1.85**	1	−1.58	1.03	−1.53	0.96	−1.64	0.88	−1.76	1.88	−0.67	0.70	−2.10	1.46	−1.03
SS-8	**12.4**	**1.93**	1	−1.70	1.17	−1.47	0.91	−1.83	1.04	−1.65	2.29	−0.50	0.65	−2.32	1.67	−0.96
Mean	**16.8**	**2.33**	1	−1.57	1.12	−1.41	0.89	−1.74	0.90	−1.70	2.26	−0.44	0.95	−1.69	1.61	−0.88

Bold figures indicate significant values

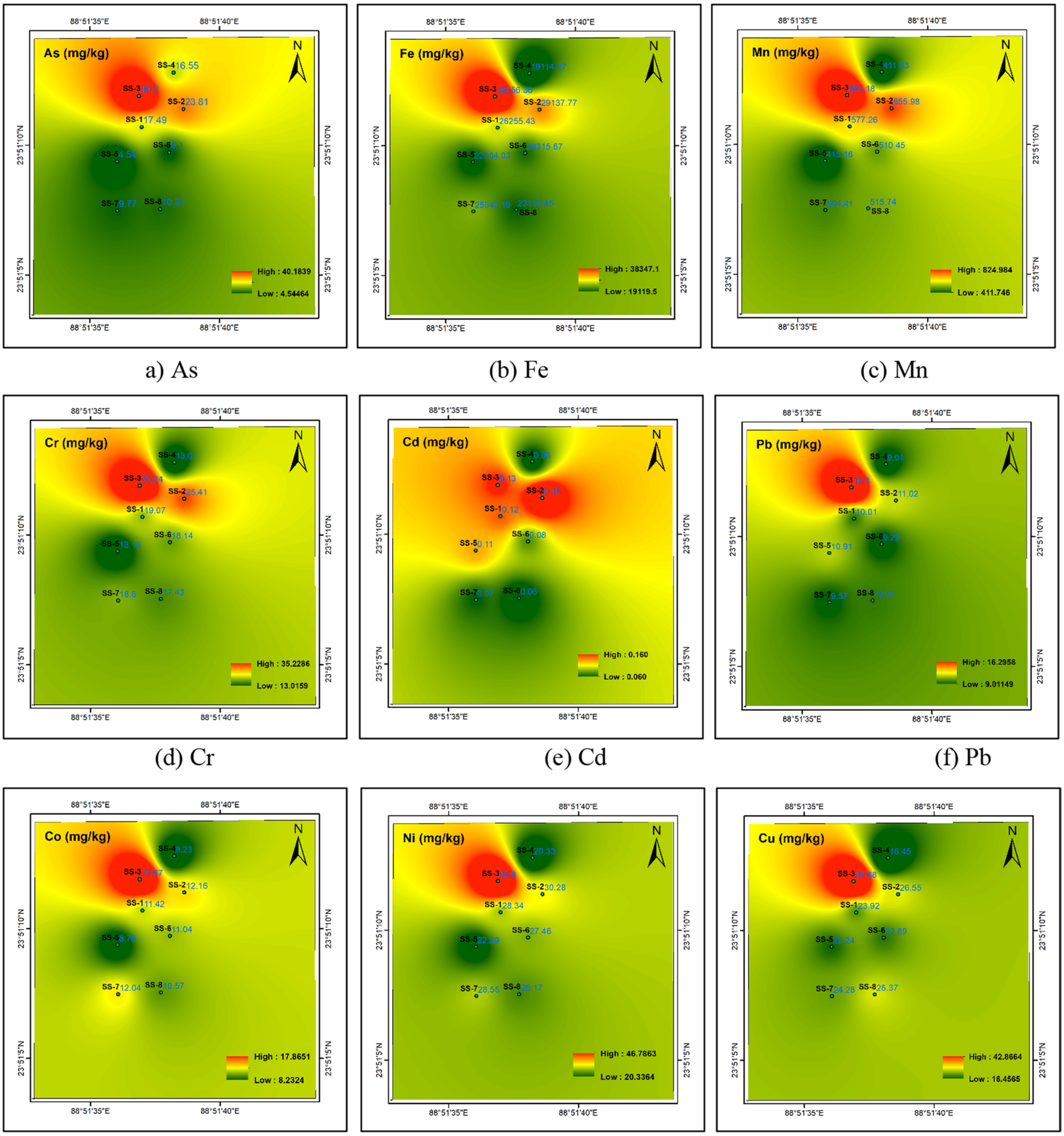
accumulation is entirely natural in origin. By comparing the current concentration to the background level, the geo-accumulation index (Igeo) was utilized to assess the level of heavy metal and metalloid components in the sediment. The results of the geo-accumulation index (Igeo) for Fe, Mn, Co, Cu, Zn, Cd, and Pb show particularly uncontaminated (Igeo ≤ 0), and As show moderately to strongly contaminated ($2 < \text{Igeo} < 3$), which occurs due to use of high As-contaminated groundwater. The As load on soil in the study area is a result of anthropogenic activities such as the use of wood preservatives in electric poles, the overuse of As-based fertilizers and pesticides in agriculture, and irrigation with As-contaminated groundwater [5, 12, 37, 67].

Spatial distribution maps of trace metals were developed to identify the spatial signature of trace metals in soil. For all trace metals, concentrations are higher in the northern portion among the sampling points SS-1, SS-2, SS-3, and SS-4. All of the samples above were collected from irrigated land where staple food rice was grown all year. Groundwater contains trace metals that accumulate on the soil over time. Trace metals concentration is lower in the southern portion of the study area among the sampling points SS-5, SS-6, SS-7, and

SS-8, primarily in the residential area where local people used to cultivate seasonal fruits and vegetables that are partially irrigated, explaining why metals concentration is lower in the southern portion than the northern portion (Fig. 2).

Contamination assessment of heavy metals in food

The analytical results of the metal concentration of collected food samples are presented in Table 4. A comparison of those metal accumulations with food standards established by WHO [24] found that only rice grain (R-1 to R-4) arsenic concentration is considered instead of rice



a) As (b) Fe (c) Mn

(d) Cr (e) Cd (f) Pb

(g) Co (h) Ni (i) Cu

Fig. 2 Spatial distribution of heavy metals in soil sample in the study area

Table 4 Heavy metals concentration (mg kg^{-1}) in food (rice grain and vegetables) samples of Meherpur district, southwestern Bangladesh

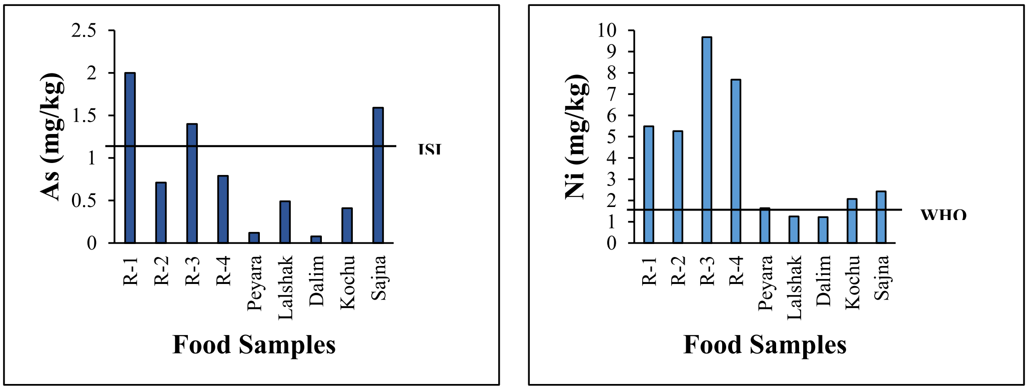
Sample ID	As	Fe	Mn	Co	Ni	Cu	Zn	Cr	Cd	Pb
R-1	2.00	636.0	88.4	0.23	0.96	2.89	11.5	1.30	0.06	0.61
R-2	0.71	101.5	98.6	< 0.2	1.83	1.67	21.8	1.80	0.13	1.43
R-3	1.40	108.9	133	< 0.2	0.62	2.15	11.8	0.79	0.06	0.53
R-4	0.79	238.6	133.7	0.03	0.81	2.42	14.7	1.00	0.07	0.58
FS-1	0.12	188.5	1.00	0.18	1.64	10.8	12.8	0.89	0.06	0.75
FS-2	0.49	775.2	0.95	0.29	1.25	5.68	12.6	1.21	0.20	0.92
FS-3	0.08	49.60	0.28	< 0.2	1.22	12.8	29.6	1.64	0.24	1.80
FS-4	0.41	575.4	1.67	0.2	2.07	18.3	16.9	1.91	0.11	1.15
FS-5	1.59	139.1	0.55	< 0.2	2.43	6.90	21.3	2.00	0.14	1.55
Average	0.84	312.5	50.9	0.19	1.43	7.07	17.0	1.39	0.12	1.04
Maximum	2.00	775.2	133.7	0.29	2.43	18.3	29.6	1.91	0.24	1.55
Minimum	0.08	49.60	0.28	< 0.2	0.62	1.67	11.5	0.79	0.06	0.53
Range	1.92	725.7	133.5	0.27	1.81	16.6	18.1	1.12	0.18	1.02
SD	0.68	272.5	61.0	0.10	0.61	5.78	6.12	0.46	0.07	0.47

plants. Two rice grains (R-1, R-3) and Sajna (drumstick) exceed the Indian standard (As: 1.1 mg kg^{-1}) [7] limit for As in human consumable foods (Fig. 3a). For a nickel, all food samples exceed the Indian standard (Ni-1.5 mg kg^{-1}) value except Lalshak (red amaranth) and Dalim (pomegranate) (Fig. 3b). Except for Lalshak (red amaranth) and Sajna (drumstick), all the samples exceed the WHO limit (10 mg kg^{-1}) for Cu in food (Fig. 3c) [105]. All the samples exceed the WHO standard (1 mg kg^{-1}) for Cr and Pb except Peyara (Guava) (Fig. 3d and f). For all paddy (grain) samples (R-1to R-4) and Dalim (pomegranate), Cd concentration exceeds the WHO limit (0.2 mg kg^{-1}) for food samples (Fig. 3e). Comparison of As concentration in soil, rice, and vegetable/fruits with relevant literature data is presented in Table 5 which is the most essential to understand the status of regional arsenic contamination level.

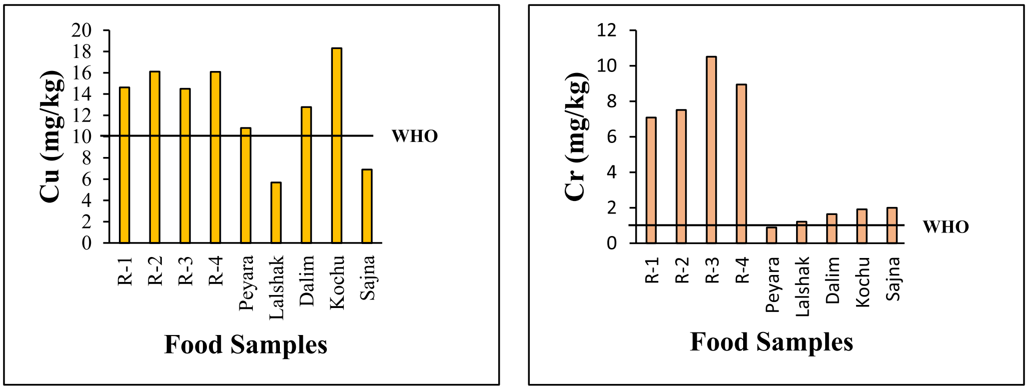
Multivariate statistical analysis

Multivariate statistical approaches, such as principal components analysis (PCA), hierarchical cluster analysis (HCA), and correlation analysis are excellent tools for identifying and categorizing the many causes of soil metal pollution [87]. These analyses are commonly used to anticipate heavy metal variability and its governing factors, as well as to emphasize the impact of human activities on soil enriched with heavy metal [44]. Tables 6 and 7 show the findings of the PCA of metal contents in soil and food (paddy) samples, respectively. In this study, two principal components were identified from the analytical data of soil samples, which narrated the cumulative variance of 68.166 and 90.532%, respectively (Fig. 4a and b). In food samples (paddy), two principal components were also extracted, which explain the cumulative variance of 68.071 and 97.063%, respectively (Fig. 5a and b). Two principal components (PCA-1 and PCA-2) were noted for soil samples and food samples individually. In PCA-1 of soil samples, As, Co, Ni, Cu, Fe, Mn, Cr, and Pb showed positive loading, whereas only Zn showed a negative value. The positive loadings of trace metals on soil suggest that human activity is to blame for the accumulation in the study area, which is most likely derived from irrigation water. Factor loadings larger than 0.71 are generally regarded as outstanding, while those less than 0.32 are regarded as extremely poor [22].

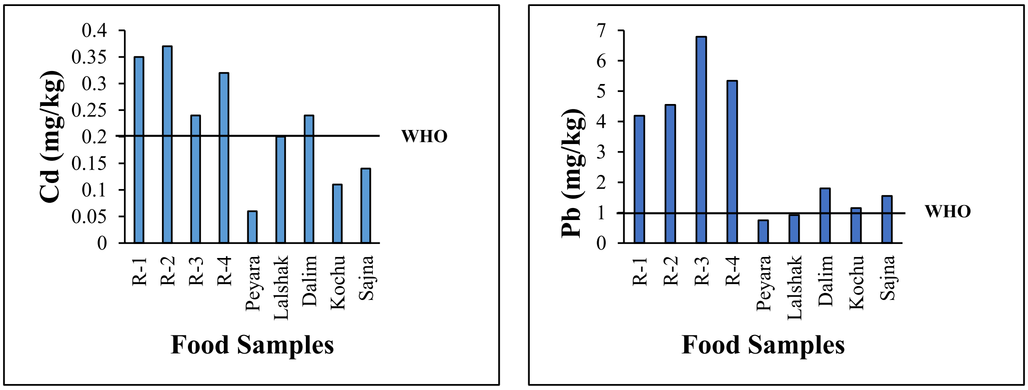
In PCA-1, an eigenvalue greater than 2 with a total variance of 68.166% and significant loadings of As (0.940), Fe (0.956), and Mn (0.974) revealed that the source of those trace elements in the soil sample is the same [64]. In PCA-2, Zn, Cd, and Mg showed positive loading, which also suggests anthropogenic loading on soil due to the use of different types of fertilizer [39]. The highest values of Zn (0.963) and Mg (0.897) in PCA-2 introduce phosphate and magnesium sulfate fertilizers used for agricultural practice in that area. Organic and phosphate fertilizers contain higher amounts of Pb and Cd, respectively [46]. Pb and Cd showed almost similar loading in both PCA-1 and PCA-2, which mainly came from both geogenic and anthropogenic origins. Total factor accumulation values greater than 0.75, 0.75–0.50, and 0.50–0.30 were graded as high, medium, and low, respectively [3]. In PCA-1 of food samples, Co, Ni, Fe, Mn, Cr, Pb, and Mg showed positive loading, which is almost similar to the PCA-1 of soil samples, while As and Mn are dominant in PCA-2. In both factors of PCA-1 and PCA-2, As



(a) As (b) Ni



(c) Cu (d) Cr



(e) Cd (f) Pb

Fig. 3 Diagram showing the graphical presentation of metal concentrations: (**a**) As, (**b**) Ni, (**c**) Cu, (**d**) Cr, (**e**) Cd, and (**f**) Pb for all the food samples collected from the study area. The solid line revealed WHO and ISI standard for metals in food

(PCA-1: 0.566; PCA-2: 0.817) and Mn (PCA-1: 0.586; PCA-2: 0.758) showed close positive loading, which suggests both natural and anthropogenic origins. The similar loadings of metals in soil and food samples in PCA-1 indicate homogeneity of origin. From PCA analysis, metal contents in entire soil and food samples are both geogenic and anthropogenic in origin because of the wide range of analytical testing of fertilizer products such as phosphate, micronutrient fertilizers, liming materials, insecticides, and pesticides that contain an elevated level of As, Ni, Cd, and Pb compared to other fertilizer types. The distribution of the same sorts of metals in soil samples and paddy samples was not analogous, which might be the result of distinct

Table 5 Comparison of arsenic concentration in soil, rice, and vegetable/fruits with relevant literature data

Sample area	As in groundwater (µg/L)	As in soil (mg/kg)	As in rice (mg/kg)	As in vegetable/fruits (mg/kg)	References
Meherpur	292	16.47	1.23	0.54	This study
Chandpur	210	21.35	0.41	1.88	Rokonuzzaman et al. [78]
Jessore	240	–	–	0.0004	Alam et al. [5]
Bogra	–	–	0.26	0.25	Islam et al. [31]
Faridpur	198	29.60	0.41	–	Norton et al. [61]
Noakhali	328	3.30	–	0.11	Rahman et al. [69]
India	131	52.60	0.62	–	Upadhyay et al. [95]
China	–	10.37	0.10	0.013	Jiang et al. [32]
Vietnam	480	–	–	0.87	Ngoc et al. [60]
Taiwan	–	184.6	0.20	–	[47]
Southwestern Bangladesh	87.3	13.0	0.23	–	Rahman et al. [70]

Table 6 Varimax rotated principal component analysis and communalities of the metals of the soil samples

Parameters	PCA-1	PCA-2	Communalities
As	**0.940**	−0.042	0.885
Co	**0.964**	0.135	0.947
Ni	**0.967**	0.175	0.965
Cu	**0.915**	0.295	0.924
Zn	−0.087	**0.963**	0.934
Fe	**0.956**	0.277	0.991
Mn	**0.974**	0.138	0.968
Cr	**0.978**	0.165	0.984
Cd	0.517	**0.545**	0.564
Pb	**0.853**	0.413	0.898
Mg	0.303	**0.897**	0.897
Eigenvalue	8.142	1.816	
Total variance %	68.166	22.366	
Cumulative % of variance	68.166	90.532	

Bold figures indicate significant values

Table 7 Varimax rotated principal component analysis and communalities of the metals of the food (paddy) samples

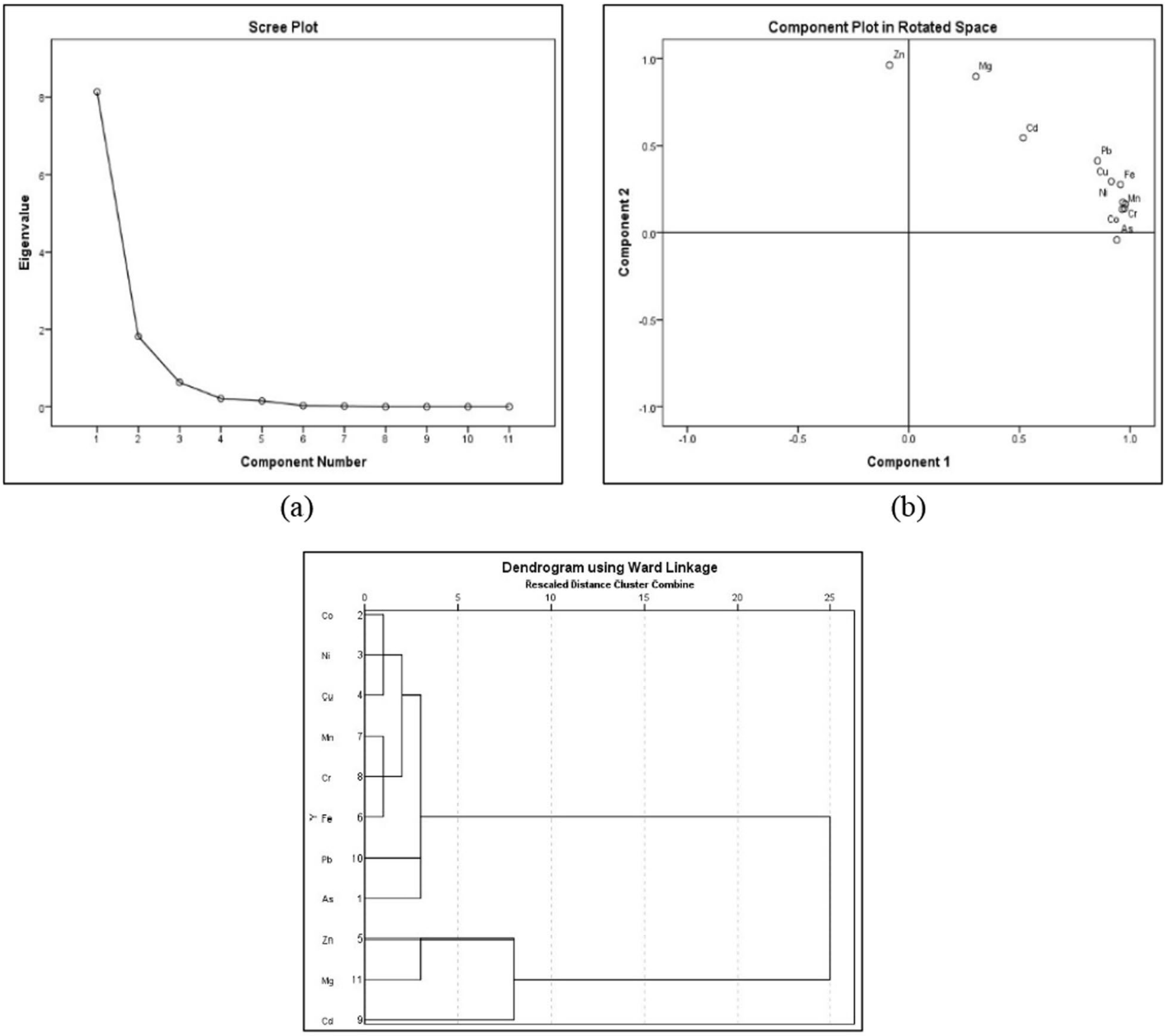
Parameters	PCA-1	PCA-2	Communalities
As	**0.566**	**0.817**	0.988
Co	**0.979**	0.185	0.993
Ni	**0.899**	0.432	0.995
Cu	0.028	**−0.984**	0.900
Zn	−0.964	**−0.257**	0.995
Fe	**0.871**	0.465	0.975
Mn	**0.586**	**0.758**	0.918
Cr	**0.952**	0.307	0.989
Cd	−0.810	**−0.581**	0.993
Pb	**0.934**	0.334	0.984
Mg	**0.966**	0.037	0.934
Eigenvalue	9.204	1.473	
Total variance %	68.071	28.992	
Cumulative % of variance	68.071	97.063	

Bold figures indicate significant values

emission and accumulation characteristics of the considered metals by plants from the source to the environment [15].

Moreover, hierarchical cluster analysis (HCA) was executed with Ward's methods to distribute the metals into different groups, representing the result with a dendrogram for soil samples (Fig. 4c) and food samples (Fig. 5c). For both soil and food samples, two clusters were identified with the Phenon line set to a rescaled distance of about 10 to show statistical similarity. In soil samples, Cluster-1 and Cluster-2 included Co, Ni, Cu, Fe, Mn, As, Pb, and Cr, and Zn, Mg, and Cd, which showed positive loadings on PCA-1 and PCA-2, respectively. It also exhibits the similarity between PCA and HCA. In food (paddy) samples, Cluster-1 showed positive loading of Fe, Ni, Cr, Pb, Co, Mg, As, and Mn, whereas Zn, Cd, and Cu are the dominant metals in Cluster-2. Only Cu and Mg showed dissimilarity between PCA and CA analyses in the paddy sample, which may happen due to the accumulation and resistance to the accumulation of metals in plant cells from different sources [15].

A mutual correlation among the studied metal concentrations in the soil and food samples was identified to determine their interrelationships, as presented in Table 8. In the case of soil samples, As showed strong positive correlations with Co ($r=0.831$), Ni ($r=0.854$), Cu ($r=0.792$), Fe ($r=0.871$), Mn ($r=0.905$), Cr ($r=0.903$), and Pb ($r=0.829$) and negative correlations with Zn ($r=-0.135$). Fe showed a strong correlation with Mn ($r=0.973$), Cr ($r=0.981$), and Pb



(a) (b)

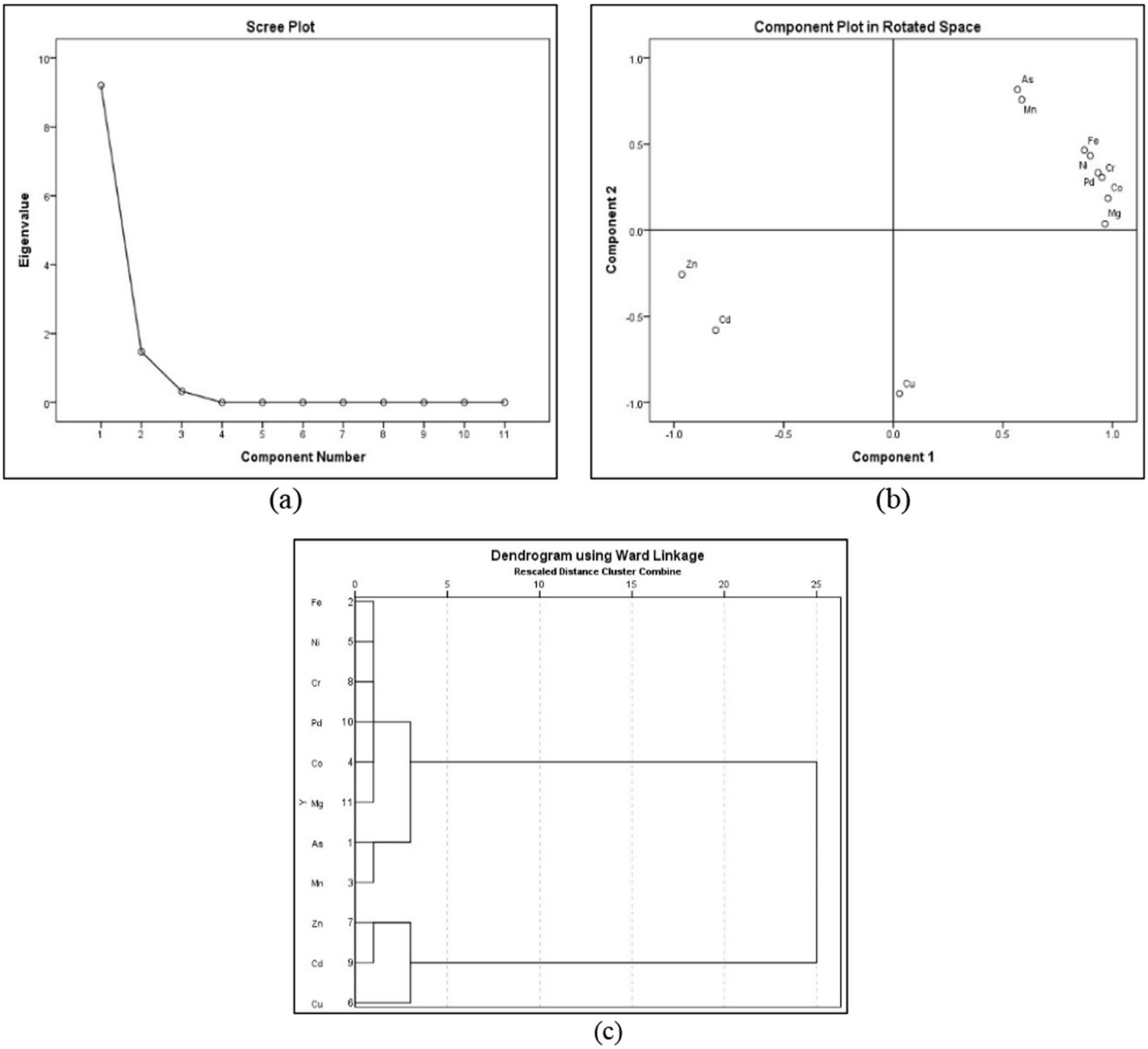
(c)

Fig. 4 PCA analysis with component in (**a**) scree plot, (**b**) rotated space, and (**c**) cluster analysis of metals in soil samples

(*r*=0.913). Homogeneous behavior among the metal groups revealed a similar source in origin [9]. In food samples, As showed a strong positive correlation with Co (*r*=0.942), Ni (*r*=0.972), Fe (*r*=0.978), Mn (*r*=0.983), Zn (*r*=0.796), Cr (*r*=0.963), and Pb (*r*=0.960) and an insignificant correlation with Cu (*r*=0.460). Fe showed a strong correlation with Mn (*r*=0.963), Cr (*r*=0.969), and Pb (*r*=0.970), which indicate similarity in soil and food samples as a common source.

Accumulation of arsenic in soils and plants

Agricultural activities frequently result in heavy metal deposition in soils, and this includes non-point sources of contaminated inputs such as fertilizers, insecticides, sewage sludge, organic manures, and composts [88]. Groundwater contaminated with As used for drinking purposes and the agricultural water supply is considered the major ingestion route of As to the human body and plants. Several hypotheses have been made to understand the origin (primary) and mobilization (secondary) causes of elevated arsenic in groundwater. Arsenic in the Bengal Delta, especially in GMB (Ganges–Brahmaputra–Meghna) flood plains, came from the Himalayas and the highlands in the neighboring areas. Sulfides washed out and accumulated in these GMB floodplain areas from the Himalayan arsenic-bearing pyrite. Contaminants



(a) (b) (c)

Fig. 5 PCA analysis with component in (**a**) scree plot, (**b**) rotated space, and (**c**) cluster analysis of metals in food (rice) samples

primarily affect the Holocene aquifer at depths ranging from 20 to 120 m, which cover southwest and south-central Bangladesh [71]. Arsenic can be found in Holocene sediments, which eventually mixed with groundwater as a result of microbial activity and the reductive dissolution of iron oxyhydroxides (FeOOH). A very low concentration of As in plants may have beneficial impacts on plant species [27]. Although the jury is still out on whether As is a natural element of some plants, it is widely accepted that As is not needed for plants [37]. The flooded method of irrigation is used in this region. As a result, a considerable amount of arsenic is available to both form adjacent soil and water to the rice plant. High concentration of As was found in root area, due to the synergistic effect of As from contaminated soil and groundwater [13]. The overall As content in water, soil, paddy roots, straw, and grain is presented in Table 9. A proposed model of arsenic circulation in an agronomical system modified from Sandberg and Allen [82], is shown in Fig. 6.

Metals transfer from soils to crops

The results of the bioaccumulation factor of metals showed that the BAF values for rice plants are higher than those for other vegetable samples shown in Table 10. For rice plants (RP), As, Cd, and Mn showed greater concentrations; Zn, Cu, Fe, Cr, and Pb showed moderate

Table 8 Pearson's correlation matrix among heavy metals concentration in soil and food samples

Parameters	As	Co	Ni	Cu	Zn	Fe	Mn	Cr	Cd	Pb
Soil samples										
As	1									
Co	**0.831**	1								
Ni	**0.854**	**0.995**	1							
Cu	**0.792**	**0.954**	**0.969**	1						
Zn	−0.135	0.055	0.106	0.246	1					
Fe	**0.871**	**0.969**	**0.977**	**0.954**	0.179	1				
Mn	**0.905**	**0.946**	**0.953**	**0.916**	0.045	**0.973**	1			
Cr	**0.903**	**0.967**	**0.971**	**0.932**	0.053	**0.981**	**0.987**	1		
Cd	0.564	0.479	0.500	0.477	0.406	0.662	0.658	0.617	1	
Pb	**0.829**	**0.861**	**0.901**	**0.940**	0.365	**0.913**	**0.848**	**0.871**	0.551	1
Food samples										
As	1									
Co	**0.942**	1								
Ni	**0.972**	**0.980**	1							
Cu	0.460	0.471	0.484	1						
Zn	**0.796**	**0.718**	**0.763**	0.565	1					
Fe	**0.978**	**0.989**	**0.990**	0.463	**0.743**	1				
Mn	**0.983**	**0.939**	**0.963**	0.511	**0.870**	**0.963**	1			
Cr	**0.963**	**0.963**	**0.979**	0.538	**0.865**	**0.969**	**0.984**	1		
Cd	0.637	0.588	0.586	0.380	**0.914**	0.598	**0.735**	**0.729**	1	
Pb	**0.960**	**0.963**	**0.977**	0.508	**0.857**	**0.970**	**0.969**	**0.992**	**0.717**	1

Bold figures indicate significant correlation (r > 0.7)

Table 9 Overall scenario of arsenic (mg kg^{-1}) content in irrigation inlets, soil, and paddy roots, straw, and grains in the study area

Arsenic in groundwater (contamination source)	Arsenic in soil		Arsenic in paddy			
	Sample ID	As	Sample ID	As		
				Root	Straw	Grain
Groundwater contains arsenic 292 μg L^{-1}	SS-1	17.5	RP-1	135.4	8.84	2.0
	SS-2	23.8	RP-2	74.2	3.30	0.71
	SS-3	40.2	RP-3	204.8	5.47	1.40
	SS-4	16.6	RP-4	144.8	3.37	0.79
Mean		24.5		139.8	5.25	1.23

concentrations; and Co and Ni showed low concentrations, which are moderately similar to their concentration in soil samples. The higher values of As for rice plants due to both soil As accumulated in previous years and As freshly introduced with irrigation water influence As uptake during rice growth [18]. Cadmium is the most accumulated metal and showed the highest BAF values among all the metals analyzed [83]. If the Cd enters the plant vessel, it interludes with the enzymes and takes the place of Zn, which results in an easy transfer of this metal from soil to the consumable part of the plant species compared to Zn [53]. Cd showed high accumulation in all the samples except Peyara without any visual effects. Cadmium transfer is highly susceptible due to its significant toxicity. Although soil samples were found to be low to moderately contaminated with Cd, with higher BAF values for rice plants and vegetable samples. For vegetable samples, Cu and Zn showed higher concentrations than all other metals. Comparatively low to moderate BAF values for Cr, Co, Fe, and Ni are probably due to their low mobility and phytoavailability, expressing their strong sorption in soils, which make lowers the free ion concentrations in the soil solution and makes them less available for plant accumulation [83]. The overall results

Table 10 Bioaccumulation factors (BAF) of metals from adjacent soil to collected food crop samples

Soil sample ID	Crop sample ID	As	Fe	Mn	Co	Ni	Cu	Zn	Cr	Cd	Pb
SS-1	RP-1	**8.36**	0.29	1.55	0.11	0.19	0.61	0.68	0.37	**2.92**	0.42
SS-2	RP-2	**3.28**	0.22	1.00	0.14	0.17	0.60	0.61	0.30	**2.94**	0.41
SS-3	RP-3	**5.27**	0.42	1.34	0.18	0.21	0.34	0.45	0.30	**1.85**	0.42
SS-4	RP-4	**9.00**	0.65	2.35	0.32	0.38	0.98	0.81	0.69	**5.33**	0.60
SS-5	FS-1	0.03	0.008	0.002	0.02	0.07	0.49	0.11	0.07	**0.50**	0.07
	FS-5	0.35	0.006	0.001	NA	0.11	0.31	0.18	0.15	**1.27**	0.14
SS-6	FS-3	0.09	0.002	0.0005	NA	0.04	0.56	0.36	0.09	**3.00**	0.19
SS-7	FS-2	0.05	0.03	0.002	0.03	0.04	0.23	0.19	0.07	**2.86**	0.01
SS-8	FS-4	0.04	0.02	0.003	0.02	0.08	0.70	0.23	0.11	**1.83**	0.11
Mean		**2.94**	0.18	0.69	0.12	0.14	0.54	0.40	0.24	**2.50**	0.26
SD		3.74	0.23	0.89	0.11	0.11	0.23	0.25	0.20	**1.38**	0.20

Bold figures indicate significant values

NA not applicable

strongly expressed that plant samples, especially rice plants, are mainly contaminated with As, Cd, and Mn, and other vegetable samples are also contaminated with cadmium, which showed a higher BAF value for all the samples except Peyara (guava).

The results of the net translocation coefficient (NTC) of arsenic for rice plants are almost similar to the transfer factor of arsenic in rice plants shown in Table 11. NTC soil/root has been thought to signify a plant's ability to accumulate a specific element. The translocation coefficient between soil and root (soil–root) showed a higher value than root–straw and straw–grain. On this basis, the NTC soil/root in the current study's data might be interpreted as indicating that rice has a greater capacity for As uptake. Such a high value (mean: 6.17) of TC root/soil for rice plants indicates arsenic sinks on soil and that soil arsenic can accumulate through the plant root to the harvesting part in the study area. The overall analytical and statistical results strongly indicate that food samples from Bholadanga village in Meherpur district, southwestern Bangladesh, are badly contaminated with As.

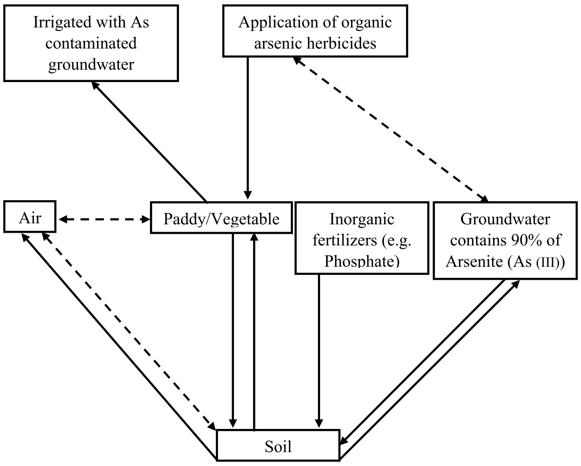


Fig. 6 A proposed model for the arsenic cycle in an agronomical ecosystem in the study area. Source: modified from [82]

Health risks of local people

In the research area, humans were exposed to As toxicity through consumable food and drinking water. The schematic diagram of As exposure to the human body is shown in Fig. 7. The daily intake of metals (Table 12) for rice and vegetables shows terrible value. Highly toxic metal(loid)s like As, Ni, and Pb represent the maximum value. They all exceed the daily ingestion limits of As (3 μg/kg bw/day) according to the benchmark dosage for a 0.5% increased incidence of lung cancer (BMDL0.5) of 3.0 μg/kg bw/day, which varied from 2 to 7 μg/kg bw/day [106], Ni (1.3 μg/kg/day), and Pb (3.57 μg/kg/day) for adults recommended by FAO/WHO [24] and EFSA [21]. The US Environmental Protection Agency (USEPA) has used HQ as another index for computing human dietary risk [30]. The risk for any non-carcinogenic health hazard through intake of arsenic-contaminated drinking

Table 11 Net translocation coefficient of As in rice plants (soil to grain) in studied area

Samples ID	Net translocation coefficient		
	TC $_{\text{root/soil}}$	TC $_{\text{straw/root}}$	TC $_{\text{grain/straw}}$
RP-1	7.74	0.07	0.23
RP-2	3.12	0.04	0.22
RP-3	5.10	0.03	0.26
RP-4	8.73	0.02	0.23
Mean	6.17	0.04	0.24
SD	2.55	0.022	0.02

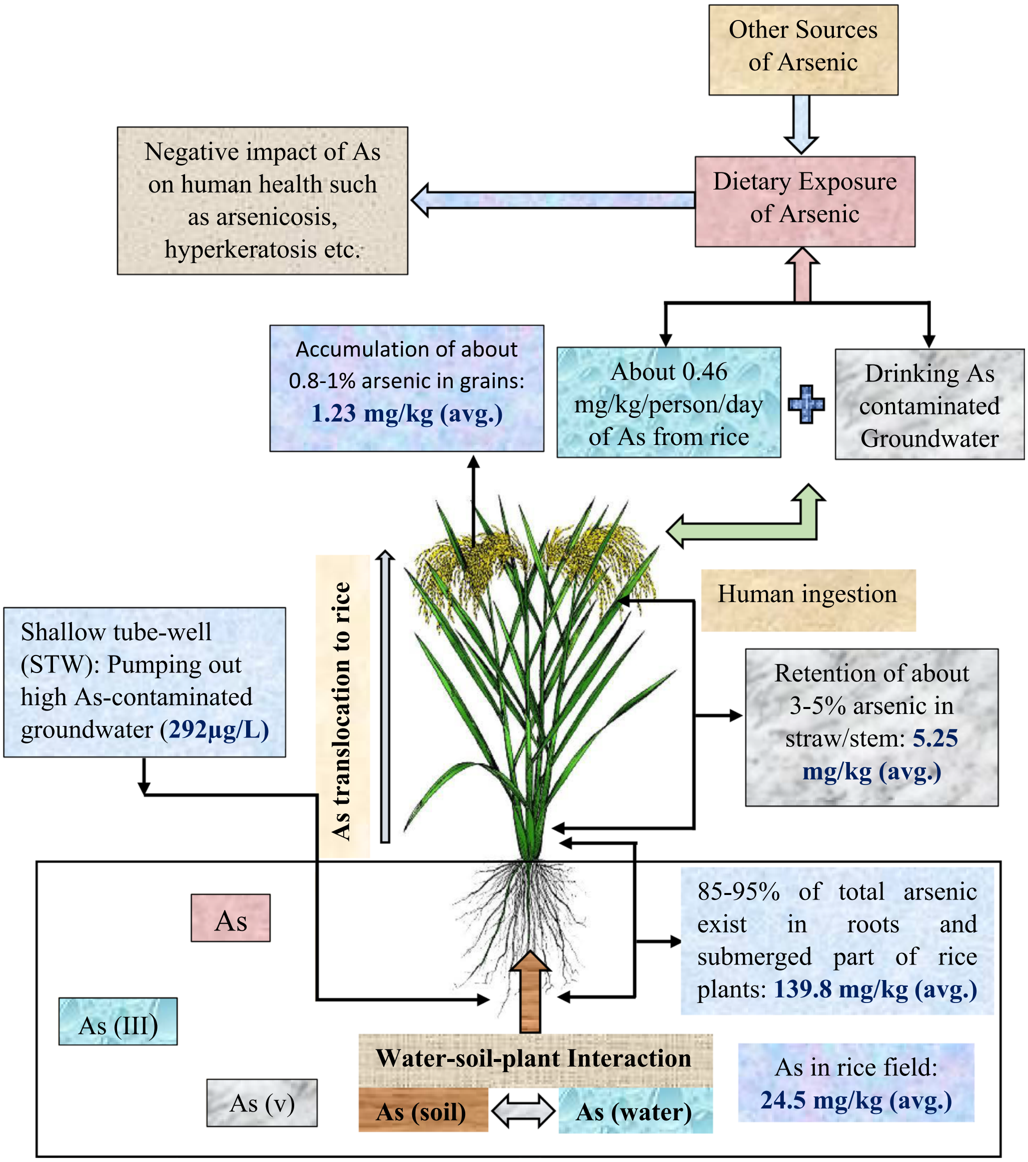


Fig. 7 Schematic diagram of human exposure to arsenic from direct consumption in the study area

water and foodstuffs is denoted when the hazard quotient (HQ) value is greater than 1 [8, 16, 33, 109]. The results of the HQ (Table 13) for contaminated metals in foodstuff revealed that the values of As in rice (RP-1 to RP-4) and drumstick (FS-5), Pb in rice (RP-2), pomegranate (FS-3), arum (FS-4), drumstick (FS-5), and Cu in arum (FS-4) ingestion rates were higher than 1 (HQ > 1; bold typed face), and it was noted that the local people in the research area seem to be exposed to the significantly non-carcinogenic health risk of As and Pb. Carcinogenic risk (CR) is used to calculate the likelihood of an individual developing cancer as a result of consuming contaminated food. The CR was calculated for As, Pb, and Cd according to the equation developed by USEPA [98] presented in Table 14. The carcinogenic risk value is greater than 10^{-6}, which suggests that the people in the vicinity are at high carcinogenic risk [62]. The carcinogenic risk from food consumption of the studied population is highly unacceptable and values were much beyond the safety standard value of 1×10^{-6} to 1×10^{-4} [100]. The results of CR for food indicate that for food samples, the values of As, Pb, and Cd are greater than 10^{-6} and the mean CR values for As, Pb, and Cd are 6.53×10^{-3}, 4.04×10^{-5}, and 7.72×10^{-3}, respectively, which suggest that the people of the study area are at high carcinogenic risk according to USEPA-2005. Priority must be given to focusing

Table 12 Daily intake of metals (µg day^{-1}) by food samples of Meherpur district, southwestern Bangladesh

Samples ID	As	Fe	Mn	Co	Ni	Cu	Zn	Cr	Cd	Pb
RP-1	**11.5**	3639.8	505.7	1.32	**5.50**	16.5	65.6	7.44	0.34	**3.49**
RP-2	**4.06**	580.7	564.3	NA	**10.5**	9.56	124.7	10.3	0.74	**8.18**
RP-3	**8.01**	623.0	761.1	NA	**3.55**	12.3	67.5	4.52	0.34	**3.03**
RP-4	**4.52**	1365.8	765.4	0.17	**4.64**	13.9	83.8	5.72	0.40	**3.32**
FS-1	0.37	579.8	3.08	0.55	**5.05**	33.3	39.4	2.74	0.18	**2.31**
FS-2	2.07	3267.9	4.00	1.22	**5.27**	23.9	53.2	5.10	0.84	**3.88**
FS-3	0.24	152.5	0.86	NA	**3.75**	39.3	91.0	5.05	0.74	**5.54**
FS-4	1.73	2425.4	7.04	0.84	**8.73**	77.2	71.2	8.05	0.46	**6.53**
FS-5	**6.70**	586.2	2.32	NA	**10.2**	29.1	89.6	8.43	0.59	**6.53**
Mean	4.40	1469	290.4	0.80	6.40	28.3	76.2	6.40	0.50	4.80
SD	3.78	1307.8	350.1	0.48	2.71	20.9	24.8	2.35	0.22	2.00

Bold figures indicate significant values

NA not applicable

Table 13 Hazard quotients (HQ) for individual metals caused by the consumption of foods

Sample ID	As	Cr	Cd	Pb	Ni	Cu	Zn
RP-1	**3.815**	0.005	0.343	0.873	0.275	0.413	0.219
RP-2	**1.354**	0.007	0.744	**2.046**	0.524	0.239	0.415
RP-3	**2.671**	0.003	0.343	0.758	0.177	0.308	0.225
RP-4	**1.507**	0.004	0.401	0.830	0.232	0.346	0.279
FS-1	0.123	0.002	0.185	0.577	0.252	0.832	0.131
FS-2	0.689	0.003	0.843	0.970	0.263	0.599	0.177
FS-3	0.081	0.003	0.738	**1.385**	0.188	0.982	0.303
FS-4	0.576	0.005	0.464	**1.212**	0.436	**1.930**	0.237
FS-5	**2.234**	0.006	0.590	**1.633**	0.512	0.727	0.299
Mean	1.450	0.004	0.517	1.143	0.318	0.708	0.254
SD	1.260	0.002	0.223	0.473	0.136	0.524	0.083

Bold figures indicate significant values

Table 14 Carcinogenic risk of As, Pb, and Cd due to direct consumption of foods

Sample ID	Carcinogenic risk		
	As	Pb	Cd
RP-1	1.72×10^{-2}	2.97×10^{-5}	5.10×10^{-3}
RP-2	6.09×10^{-3}	6.95×10^{-5}	1.11×10^{-2}
RP-3	1.20×10^{-2}	2.58×10^{-5}	5.10×10^{-3}
RP-4	6.78×10^{-3}	2.82×10^{-5}	6.00×10^{-3}
FS-1	5.55×10^{-4}	1.96×10^{-5}	2.70×10^{-3}
FS-2	3.11×10^{-3}	3.30×10^{-5}	1.26×10^{-2}
FS-3	3.60×10^{-4}	4.71×10^{-5}	1.11×10^{-2}
FS-4	2.60×10^{-3}	5.55×10^{-5}	6.90×10^{-3}
FS-5	1.01×10^{-2}	5.55×10^{-5}	8.85×10^{-3}
Mean	6.53×10^{-3}	4.04×10^{-5}	7.72×10^{-3}
SD	5.34×10^{-3}	1.60×10^{-5}	3.17×10^{-3}

on the carcinogenic risks to the populations residing in the study area, as they are at high risk of developing skin, bladder, and lung cancers as a result of arsenic exposure pathways [38]. Aside from drinking water, the potential health risk of arsenic from rice was a substantial threat in arsenic-affected areas [81].

Conclusions

Analytical results from this research indicate that the availability of As in irrigation water led to significant enrichment of As in the irrigated soil, mainly on the upper surface (0–10 cm), and through translocation processes or bioavailability of As gets into the human consumable food chain. This study also suggests gradual As poisoning in irrigated soil has increased. Local people in the study area are possibly affected by high As contamination through drinking water and ingestion of food for a couple of years and increased probability of health risk. High soil As is found in irrigated land of the northern portion whereas the southern portion is less contaminated. Strong correlations between dissolved Fe, As, and Mn in soil suggest that high elemental affinity is present between Fe, Mn, and As and from which As can strongly accumulate over the soil. According to principal component and cluster analysis, metal concentrations in soil are natural (As, Co, Ni, Cu, Fe, Mn) and anthropogenic (Zn, Cd, and Mg). Soil contamination indices like enrichment factor and geo-accumulation index indicate As is the dominant metalloid that contaminates soil in the study area. Among the food samples, paddy contains the highest As concentration compared to other vegetables or fruit samples. A higher value of BAF (Bioaccumulation Factor) and NTC (Net translocation coefficient) indicates plants absorb metals from adjacent soil. The toxicity of metals in humans is determined by their daily ingestion rate. Daily intake of metals shows harmful metal(loid)s like As, Ni, Cr, and Pb are the highest accumulated elements in the human body through the consumable food in the affected areas. Arsenic and Pb show the maximum potential hazard quotient from the food sample. The mean values of As and Pb for HQ are greater than 1 which indicates local people are at non-carcinogenic health risk from the intake of contaminated food. Individual carcinogenic health risks from As, Pb, and Cd indicate that the people in the Bholadanga village were at high carcinogenic risk and which exceed the cancer risk value (10^{-6}) recommended by the United States Environmental Protection Agency (USEPA). However, it is suggested to install tube wells at a higher depth (> 150 m) to get As-free groundwater, and also surface water could be used for irrigation purposes, which is free from As.

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Author contributions
MSR: methodology, formal analysis, investigation, writing—original draft preparation. AHMSR: conceptualization, methodology, investigation, data curation, writing—reviewing and editing, supervision. MABS: methodology, formal analysis, investigation, data curation, validation, writing—reviewing and editing, resources, supervision. MAA: resources, writing—reviewing and editing. MH: writing—reviewing and editing. All authors read and approved the final manuscript.

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