Heliyon 7 (2021) e06309

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

Appraisal of probabilistic human health risks of heavy metals in vegetables from industrial, non-industrial and arsenic contaminated areas of Bangladesh

Md. Morshedul Haque [a,*], Nahin Mostofa Niloy [a], Md Akhte Khirul [b], Md. Ferdous Alam [c,d], Shafi M. Tareq [a,*]

[a] *Hydrobiogeochemistry and Pollution Control Laboratory, Department of Environmental Sciences, Jahangirnagar University, Dhaka, 1342, Bangladesh*
[b] *Training Institute for Chemical Industries (TICI), Bangladesh Chemical Industries Corporation (BCIC), Narsingdi, 1611, Bangladesh*
[c] *Graduate School of Symbiotic System Science and Technology, Fukushima University, 1 Kanayagawa, Fukushima, 960-1296, Japan*
[d] *Institute of Nuclear Science and Technology, Atomic Energy Research Establishment, Dhaka, 1349, Bangladesh*

A R T I C L E I N F O

Keywords:
Heavy metals
Common vegetables
Estimated daily intake (EDI)
Human health risks
Probabilistic health risk

A B S T R A C T

Monitoring of heavy metal content in commonly consumed vegetables is of high priority for food safety, and public health risk assessment. Vegetables were collected from industrial, non-industrial, arsenic contaminated region and one of popular vegetable markets of Bangladesh for analyzing heavy metals (As, Cd, Pb, Cu and Zn) using Atomic Absorption Spectroscopy (AAS) with standard digestion procedure. Results showed significant variations of heavy metal content among vegetables and most of cases the metals (except Cu and some of Zn) revealed several times higher concentrations than that of maximum permissible level (MPL) values, which indicated the vegetables were contaminated through either natural or anthropogenic activities. The dietary intake of metals are responsible for association of health risk that evaluated by target hazard quotient (THQ), hazard index (HI), and target carcinogenic risk (TR) calculations. Estimated daily intake (EDI) for all metals were below the maximum tolerable daily intake (MTDI) values of all vegetables. The THQs for single metals were less than 1 (except As and Pb for few vegetables), indicating the inhabitant would not possess health hazard for single metal through vegetables consumption. However, the total target hazard quotient (TTHQ) of all metals were >1 (except Cu and Zn for industrial and non-industrial sites), suggesting potential health risk. HI values were found more than 1 (36.24 for industrial site, 16.74 for non-industrial site, and 15.03 for local market) representing the selected vegetables intake might be affected quality of food safety of densely populated Bangladesh. The probabilistic risk, individual, and total cancer risk (TR) for As and Pb were exceeded the threshold level (10^{-4}) and safe limit (10^{-6}), respectively, indicating peoples who have been consuming these vegetables long time, they might be exposed by lifetime cancer risk. Sensitivity analysis revealed that the metal concentration has high influence on carcinogenic risks.

1. Introduction

Food safety is a prime concern for public health because it is one of the main sources of nutrition for humans. Food consumption might be the key pathway for the exposure of human beings to pollutants including toxic elements. More than 90% of exposure to public health occurred by ingestion compared to other routes of exposure like dermal and inhalation [1]. Different types of vegetables play a significant role in human nourishment and fresh vegetables may make up an enormous extent of a healthy human diet as vital sources of nutrients, minerals, and fiber. The dietary estimation of vegetables utilized in human diets is considerably reliant on their environmental source and systems of farming, as well as local particularity in applied farming expertise. Vegetables are key for pedaling many chemical elements, especially trace elements, in natural environments due to their distinctive part in primary production. Unorganized and rapidly growing urban developments in developing countries have polluted the environment. Agricultural products especially vegetables might be up taken these pollutants from soil and might have a high concentration of heavy metals which are unsafe for public health [2, 3, 4, 5]. There have several influencing factors that might be affected the

* Corresponding authors.
E-mail addresses: morshedulhaq@gmail.com (Md.M. Haque), smtareq@yahoo.com (S.M. Tareq).

https://doi.org/10.1016/j.heliyon.2021.e06309
Received 18 July 2020; Received in revised form 27 November 2020; Accepted 15 February 2021

concentration of heavy metals in vegetables which included not only environmental pollution but also climate, nature of the plant growing soil, and plant maturity during the time of harvesting and biotransformation factors [3].

Vegetables can be exposed to heavy metals due to either natural or anthropogenic activities. Naturally occurring metals comes from crusted materials, gases, and particulate matter from volcanos and continental dust [6], where environmental induced metal concentration is relatively low compare to anthropocentric. The most important and common sources of metals in the vegetables are from anthropogenic actions like long-term use of the copious amount of pesticide and fertilizer, linear, point, and surface emission of metal [7] from industrial activities. Bangladesh is a rapidly growing developing country where fast industrialization and unorganized urbanization are a common phenomenon. The vegetables might be accumulated high concentrations of metals cultivated in fields that are located near the source of heavy metals pollution like industrial areas [8]. The prolonged intake of unsafe amounts of heavy metals through foodstuff promotes accumulation in different parts of the human body i.e., brain, kidney, liver, etc [9]. Due to its toxic, persistence and non-biodegrade nature exhibit carcinogenic, teratogenic, and mutagenic effects on human health. It was recommended that biotoxic effects due to heavy metals rely on the oxidation state and concentration, mode of deposition, and sort of sources [6, 10].

This study covers four types of vegetable samples which were collected from the industrial, non-industrial site, Arsenic contaminated sites, and one of the wholesale vegetable markets. Municipal or Industrial wastewater for use of irrigation purposes is a common practice in the major cities of some continent [11] like Asia, Latin America, and Africa. Wastewater irrigation is regular a scenario in Bangladesh that convey significant levels of toxic heavy metal that create an unsafe consumption of agricultural soil [12]. In Bangladesh, agriculture contributing 19.6 percent to the national GDP [13], which is one of the dominating sectors in the country's economy. In this country, some of 90 kinds of vegetables are being grown and the main meal of people's would contain boiled rice with vegetables [14]. So there has a great chance of people to expose to heavy metals from vegetables. To the best of the author's knowledge, this is the first work that considered three vegetable farming and one trading site together, conversely, there have been a limited number of systematic studies on heavy metals in vegetables of Bangladesh from different individual areas [3, 8, 14, 15, 16, 17, 18].

The accumulation of metals from the earth's crust to the edible portions of the vegetables is one of the major pathways to detrimental health effects. Therefore, investigation of the heavy metal concentrations (e.g., As, Pb and Cd) of locally cultivated common vegetable species are subjects of great importance. The key focus of this study was to inspect the levels of heavy metal of common vegetables that reach consumers as a function of different cultivation sites, e.g., industrial, non-industrial, and arsenic contaminated areas. Moreover, the objective of the study was to evaluate the probabilistic human health risks of heavy metals from commonly consumed vegetables of Bangladesh using Monte Carlo simulation methods. Outcomes of present work could provide necessary suggestions for the farmer on how to use optimize the number of agrochemicals (e.g., fertilization, herbicides, and pesticides), and choose the right location of agricultural lands for cultivation.

2. Materials and methods

2.1. Study area

This study was carried out in industrial, non-industrial, Arsenic contaminated sites and a local wholesale vegetable market in Dhaka and Faridpur region. The vegetables were collected randomly from study regions based on their availability. The industrial site was nearby Dhaka Export Processing Zone (DEPZ) which is situated under Dhamsana Union of Savar Upazila (Figure 1a). This zone is an enormous industrial area encompassed a huge number of foreign and local industries. The major industries (textiles, leather, printing, and dyeing, fertilizer, pharmaceutical, etc.) that are produced and discharged their effluents mostly without any prior treatment. The contaminated water disperses through an open drain system, later on, the water is used for irrigational purposes in the adjacent areas [8].

The non-industrial site was around Sutipara Union, Dhamrai Upazila (Figure 1b), which is located about 50 km northwest of Dhaka City. The main profession of the study area's people is agriculture where main crops are seasonal vegetables e.g., brinjal, carrot, amaranth, cabbage, etc. The agriculture of this area fully depends on rainwater and groundwater as well. We cannot found any industry around 10km from our sampling site during the sampling.

Arsenic is a well-established human carcinogen and that enter the body through ingestion of water and food [19, 20]. This study collected vegetables from the two most arsenic prone region in Bangladesh that are Faridpur Upazila (Figure 1c) [21] and Singair Upazila, Manikganj (Figure 1d) [22] for investigation of carcinogenic risk. Both sampling areas consist of a significant amount of arsenic in groundwater and soil. The vegetables were collected directly from an agricultural field where groundwater is the main source of irrigation.

For the investigation of basket vegetables, this study collected samples from a local wholesale vegetable markets named Savar Kacha Bazar under Savar Municipality, Savar (Figure 1e). This is one of the biggest vegetable market in the Savar Upazilla where mass people are directly or indirectly dependent on the market. Since it is a densely populated and industrial area, the maximum number of industrial workers meet their daily vegetable needs from this market unconsciously, whether it is harmful or beneficial to their health. So there is a great chance to be affected by heavy metals if daily consumed vegetables contain higher concentrations and/or concentrations above the minimum permissible limit (FAO/WHO/DoE).

2.2. Vegetable sampling

This study collected 10 vegetable species [*Solanum melongena* (Brinjal), *Spinacia oleracea* (Spinach), *Solanum tuberosum* (Potato), *Solanum lycopersicum* (Tomato), *Amaranthus paniculantus* (Red Amarnath), *Amaranthus viridis* (Green Amarnath), *Lagenaria siceraria* (Bottle Gourd), *Vigna sesquipedalis* (Yardlong bean), *Cucurbita maxima* (Pumpkin), *Daucas carota* (Carrote)] from each site for analysis of heavy metals. The sample selection and periodization were done based on the food habit of the people of study areas. All samples were collected from direct field and one of the local markets then carried with zipper polyethylene bag to the laboratory. After collection, samples were cautiously rinsed with double distilled water, cut into small pieces then put in an oven at 70–80 °C until a constant weight was obtained [23]. The dried vegetables were crushed and pulverized with standard procedure and stored in a freezer until analysis was performed.

2.3. Metal analysis

Analytical-grade chemicals were used for the analysis of samples where desired solution preparation was performed by Mili-Q water. In a brief, accurately weighted (∼1 g) of ground samples were digested in a mixer of 15 mL concentrated nitric acid: sulphuric acid: perchloric acid in a 5:1:1 ratio at 80–85 °C until a clear solution was obtained [24]. Then, the digested solutions were filtered by Whatman 42 paper after chilled at room temperature and diluted to 50 ml with Mili-Q water, and blank samples were also prepared in the same way. An Atomic Absorption Spectroscopy (AAS, model: Shimadzu AA-6800, Japan) was used for the determinations of heavy metal concentrations. All the experiments were performed in the Institute of Nuclear Science and Technology (INST), Atomic energy research establishment (AERE) at Savar, Dhaka. The concentrations of selected heavy metals (As, Cd, Pb, Cu, and Zn) in the vegetable samples were measured by AAS (AA-6800, Shimadzu Corporation, Japan) under ideal analytical conditions. The detection limits of

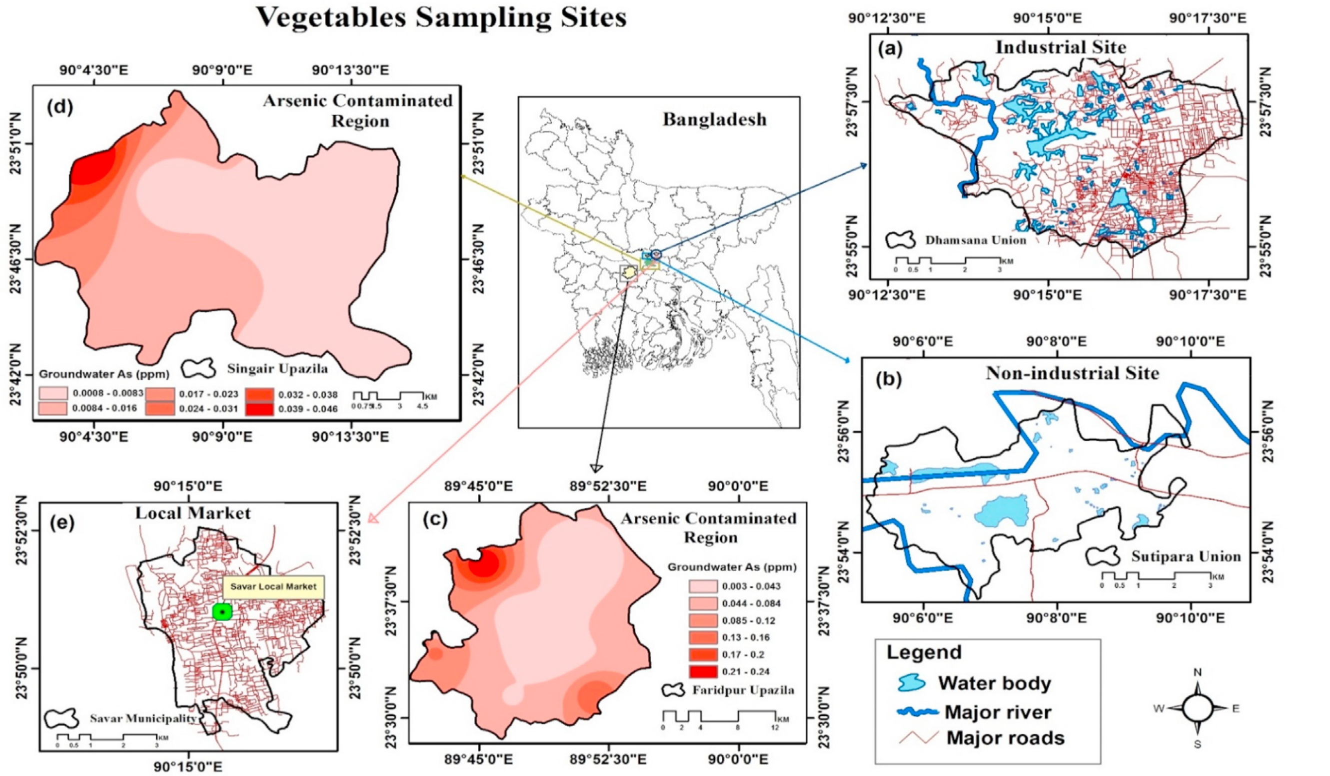

Figure 1. Geographical maps of sampling regions for (a) industrial site, (b) non-industrial site, arsenic contaminated region (c) Faridpur and (d) Singair Upazila, and (e) local market; the spatial distribution of As concentrations in groundwater was showed for two As contaminated sites and the data retrieved from previous study [38].

AAS were 25, 25, 50, 25, and 25 μg L^{-1} for As, Cd, Pb, Cu, and Zn, respectively. The AAS grade standard solutions and reagents were used for analysis which was soaring purity (99.99%). To prepare the working standards a 1000 mg L^{-1} standard stock solutions for each metal were used throughout the analysis. Standard reference materials (NBS-SRM 1573) was used for evaluation of accuracy and precision of the method and the heavy metal analysis results were found to be ±2% deviation of certified value. All the statistical analyses were performed by Microsoft Excel (Version 2013) and ArcGIS (Version 10.3) used for mapping.

2.4. Estimated daily intake (EDI)

The EDI of selected heavy metals was assessed by using an average metal concentration in samples, daily vegetable consumption rate, as well as a bodyweight of an individual. The calculation performed by following Eq. (1) [25].

$$EDI = \frac{FIR \times C}{BW} \tag{1}$$

In the above Eq. (1) the FIR indicates the food ingestion rate of a person (g $person^{-1}$ day^{-1}), C is the heavy metal concentrations in the vegetables (mg kg^{-1}) and BW indicates the bodyweight of adult populations. In this study, we considered the daily vegetable consumption rate of an adult person to be 130g for Bangladeshi people [14] and the average BW of an adult person was set to 60 kg [5, 26].

2.5. Health risk assessment

2.5.1. Non-carcinogenic risks

The non-cancer based health hazard for the people who consume metal-contaminated vegetables was evaluated by the Target Hazard Quotient (THQ) [27]. The THQ is the fraction of a single metal exposure level over a definite period (e.g., sub-chronic) to a reference dose (RfD) for that metal resultant from a comparable exposure period [5]. To evaluate the entire possible non-cancer-related effects from multiple heavy metals, the Hazard Index (HI) has been articulated depend on the health risk assessment of chemical mixtures of USEPA guidelines [27]. The THQ association with total target hazard quotient (TTHQ) and HI were evaluated by following Eqs. (2), (3), and (4), respectively [28].

$$THQ = \frac{Efr \times ED \times FIR \times C}{RfD \times BW \times AT} \times 10^{-3} \tag{2}$$

$$TTHQ = THQ\ metal\ 1 + THQ\ metal\ 2 + \ldots + THQ\ metal\ n. \tag{3}$$

$$HI = \Sigma \mathrm{TTHQ} = \mathrm{TTHQ}\ 1 + \mathrm{TTHQ}\ 2 + \ldots + \mathrm{TTHQ}\ \mathbf{n}. \tag{4}$$

In the above equations, *Efr* is the exposure frequency where this study considered 365 days per year; *ED* is the exposure duration, 70 years according's to USEPA [29]; *FIR* indicates the food ingestion rate of a person (g $person^{-1}$ day^{-1}); *C* is the heavy metal concentration in vegetables (mg kg^{-1}), *RfD* is the oral reference dose (mg kg^{-1} day^{-1}) which is 0.0003, 0.003, 0.0035, 0.04 and 0.3 mg kg^{-1} day^{-1} for As, Cd, Pb, Cu, and Zn, respectively [30]; *AT* is the averaging time for non-carcinogens (365 days $year^{-1}$ × number of exposure years, 70 years) and BW is similar to Eq. (1). If the THQ is less than 1 that indicates the comparative absence of health risks associated with the consumption of heavy metal contaminated vegetables. Conversely, if *THQ* is greater than or equal to 1 that indicates a significant human health risk [31].

2.5.2. Target carcinogenic risk (TR)

For cancer-causing agents, risks were assessed as the gradual possibility of a person developing lifetime cancer, because of exposure to that latent cancer-causing agent [27]. The target carcinogenic risk (TR) can be calculated as Eq. (5).

$$TR = \frac{Efr \times ED \times FIR \times C \times Csfo}{BW \times AT} \times 10^{-3} \tag{5}$$

In the above equation, *Efr, ED, FIR, C, BW,* and *AT* values and implication is similar to Eq. (2), and *Csfo* indicates the oral carcinogenic slope factor which was 1.5 and 8.5 $\times$ 10^{-3} $(mg^{-1}\ kg^{-1}\ day^{-1})^{-1}$ for As and Pb, respectively [30].

2.5.2.1. Monte Carlo Simulation. The probabilistic carcinogenic risk estimation was performed by using a Monte Carlo Simulation for carcinogenic metals exposure via vegetable ingestion. It is one of the recognized methods used to determine the variabilities and uncertainties of risk-based assessment [32, 33]. For the simulation, this study considered input variable [metals concentration (As and Pb), EF, ED, FIR, and BW from Eqn 5] were modeled as specific probability distribution function, average time (AT), and cancer slope factor (*Csfo*) were modeled as a point estimate. Each simulation was carried by 10,000 random trails of every input variable for ensuring the trustworthiness of the outcomes. In this study, the mean, median, 5th, and 95th percentiles of the cancer risks for As and Pb were extracted from the *TR* probability distribution. Besides, a sensitivity analysis was used to determine the values of the input variables that can affect risk approximation in a given set of assumptions [34]. Finally, the probability of risk and sensitivity analysis were performed by Crystal Ball software version 11.1.2.4 created by Oracle Co.

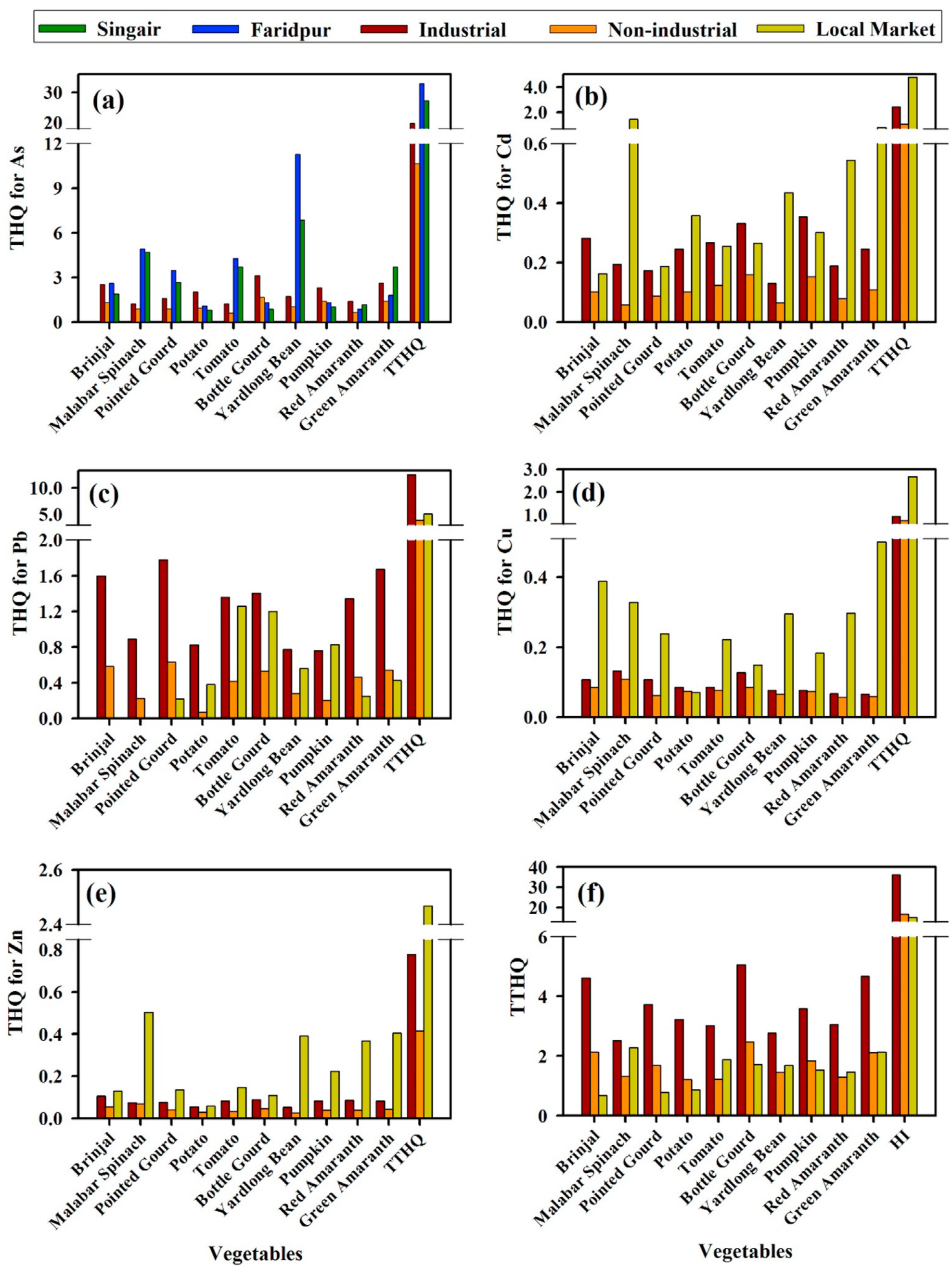

Figure 2. Target hazard quotient (THQ) and total target hazard quotient (TTHQ) of (a) As, (b) Cd, (c) Pb, (d) Cu, and (e) Zn; and (f) total metal THQ and hazard Index (HI) due to consumption of vegetables.

3. Results and discussion

3.1. Heavy metal concentration in vegetables

The analyzed heavy metal (As, Pb, Cd, Cu, and Zn) concentrations of different vegetables are listed in Table 1. The metals concentration in vegetables showed significant variability among the species and within the species. The variability of heavy metal concentrations of vegetables depends upon the climatic conditions, growth rate and period, and absorption and accumulation capacity of vegetables [35, 36], and metal concentration of the soil of cultivated land as well as irrigation water.

Arsenic (As) exposure is of significant concern in the present era because of its deleterious human health effects including dermatological, hematologic disorders (leukopenia, anemia, and eosinophilia), neurologic and neurobehavioral disorders, carcinoma, and so on [4]. The average concentration of As was 0.27 ± 0.09 mg kg^{-1} (range: 0.15–0.55 mg kg^{-1}) for an industrial site, whereas the lowest and highest value was observed at 0.17 mg kg^{-1} (Tomato) and 0.43 mg kg^{-1} (Bottle Gourd), respectively. The average As concentration of non-industrial site vegetables were 0.15 ± 0.05 mg kg^{-1} (range: 0.06–0.24 mg kg^{-1}) whereas the lowest and highest concentration was 0.08 mg kg^{-1} (Tomato) and 0.23 mg kg^{-1} (Bottle Gourd), respectively. As concentrations in market vegetables were below the detection limit (BDL) for all the samples. The arsenic-contaminated areas showed a wide variation of As content, the average concentration of As for the Faridpur region was 0.46 ± 0.43 mg kg^{-1} (range: 0.05–1.68 mg kg^{-1}) and Singair was 0.38 ± 0.28 mg kg^{-1} (range: 0.06–1.01 mg kg^{-1}). The lowest As values were 0.12 mg kg^{-1} (Red Amaranth) and 0.11 mg kg^{-1} (Potato) for Faridpur and Singair region, respectively, and the highest values were 1.56 mg kg^{-1} and 0.95 mg kg^{-1} showed in Yardlong Bean for both arsenic contaminated areas. The As concentration of individual vegetables in every sampling site was exceeded the maximum permissible level (MPL) except tomato in the non-industrial site. Average As content in vegetables were 2.7, 1.5, 4.6, and 3.8 times higher than the recommended level [28] for industrial, non-industrial, Faridpur, and Singair, respectively. The As content of this study was a significant level higher than any other previous investigation in Bangladesh [3, 14, 35, 37], especially arsenic-contaminated areas where the highest concentration was found 1.56 mg kg^{-1} in Faridpur's Yardlong Bean samples. Previous study on the Arsenic prone region reported that, the average As concentration of 14 different vegetables was found 0.28 mg kg^{-1} (range: 0.25–0.38 mg kg^{-1}) [38] which was significantly lower than present studied Arsenic prone regions. The elevated level of As might come from the use of As contaminated water (ground or surface) for irrigation [39, 40, 41], As content soil [42], or application of pesticides and fertilizers [43].

Cadmium (Cd) is one of the endocrine-disruption chemical (EDC) which may respond to the development of skeletal damage (osteoporosis), severe kidney damage, chronic renal failure, and also cancer (breast and prostate cancer) [44]. The average concentration of Cd was 0.33 ± 0.10 mg kg^{-1} (range: 0.17–0.53 mg kg^{-1}) for industrial site whereas the lowest and highest value was 0.18 mg kg^{-1} (Yardlong Bean) and 0.49 mg kg^{-1} (Pumpkin), respectively. All the vegetable samples in the industrial area exceeded the MPL (0.05 mg kg^{-1}) which was set by FAO/WHO [28]. The non-industrial site vegetable samples showed a lower range of Cd concentration compares to the other two study sites. In this site, the average Cd value was 0.14 ± 0.05 mg kg^{-1} (range: 0.07–0.25 mg kg^{-1}) and the lowest and highest value was 0.08 mg kg^{-1} (Malabar Spinach) and 0.22 mg kg^{-1} (Bottle Gourd), respectively. All of the vegetable's Cd levels were surpassed the MPL (0.05 mg kg^{-1}). The lowest and highest Cd level of local market vegetable sample was 0.22 mg kg^{-1} (Brinjal) and 2.01 mg kg^{-1} (Malabar Spinach), respectively. The average Cd value was 0.66 ± 0.54 mg kg^{-1} (range: 0.10–2.13 mg kg^{-1}) where all the sample crosses the MPL. The average Cd concentration for industrial site, non-industrial and local market vegetables were 6.6, 2.8, and 13.2 fold greater than the MPL [28], respectively. Islam et al. [16] reported that the Cd concentration of vegetables growing in different locations of Dhaka city was ranged from 0.03 to 0.32 mg kg^{-1}, which was significantly lower than this study (industrial and local market) average Cd concentration except for non-industrial site. On the other hand, the Cd content in Samta village vegetables was ranged from 0.01 (Papaya) to 0.22 mg kg^{-1} (Ghotkol) [14] that was significantly comparable with non-industrial site vegetables of the present study. The reported value of Alam et al. [14] for Cd was a significant level lower than the present study industrial site and local market vegetables. Additionally, the Cd value of industrial areas vegetables at Dhaka was ranged from 1.03 (Cabbage) to 4.65 mg kg^{-1} (Lady's Finger) [8] and that was 6–8.8 fold lower than this study industrial site. Furthermore, the Cd of a local vegetable market of Rajshahi city was ranged from 0.15 mg kg^{-1} (Pointed Gourd) to 1.75 mg kg^{-1} (Red Amaranth) [35] and that was some of the lower than the present study local market vegetables.

Lead (Pb) toxicity is an especially insidious danger with the capability of causing several organ damages in the human body including the endocrine system, kidneys, hematopoietic system, central nervous system, and reproductive system [45]. The Pb concentrations were observed from 0.81 mg kg^{-1} to 3.93 mg kg^{-1} with an average of 2.00 ± 0.64 mg kg^{-1} for industrial site vegetables whereas the lowest and highest value was 1.23 mg kg^{-1} (Pumpkin) and 2.87 mg kg^{-1} (Pointed Gourd), respectively. The Pb concentration of all vegetables at the industrial site was some fold higher than the MPL (0.10 mg kg^{-1}) and the average concentration was 20 fold higher than MPL [28]. The non-industrial site samples showed quite lower Pb concentration than the industrial and local market samples. The range of Pb concentrations of non-industrial site vegetables was 0.07–1.36 mg kg^{-1} with an average value of 0.64 ± 0.30 mg kg^{-1}, whereas the lowest and highest concentration was 0.11 mg kg^{-1} (Potato) and 1.02 mg kg^{-1} (Pointed Gourd), respectively. Pb concentrations of vegetable samples in the non-industrial area exceeded the MPL (0.10 mg kg^{-1}) and the average value was 6.4 fold higher than MPL (0.10 mg kg^{-1}). The lowest and highest concentrations of Pb in the local market samples were 0.35 mg kg^{-1} (Pointed Gourd) and 2.03 mg kg^{-1} (Tomato), respectively. The average Pb concentration of market vegetables was observed 1.03 ± 0.66 mg kg^{-1} and the range was BDL–2.20 mg kg^{-1} and also the value was 10.3 folds higher than the MPL (0.10 mg kg^{-1}). This study observed that the Pb content in vegetables of different sites has several folds higher than that of other comprehensive investigations in a different region of Bangladesh [16]. The local market vegetables Pb concentration (BDL–2.20 mg kg^{-1}) has several times lower than those reported in central market vegetables in Rajshahi City, Bangladesh (1.38–10.43 mg kg^{-1}) [35]. The range of Pb content in the industrial site of this study was also 3 times lower than that of previously studied industrial areas (2.28–11.84 mg kg^{-1}) [8]. Another studies showed that the mean Pb content was 0.02 mg kg^{-1} (range: 0.01–0.06 mg kg^{-1}) in seven different vegetables from entire Bangladesh [3], 0.5 mg kg^{-1} (range: 0.2–1.2 mg kg^{-1}) in twelve different vegetables from Patuakhali District [5] and 0.49 mg kg^{-1} (range: 0.02–1.40 mg kg^{-1}) in vegetables grown in Bogra district [37].

Copper (Cu) is a necessary nutrient that is vital for several physiological and biochemical functions. Insufficient Cu can result disrupt of metalloenzymes incorporation including in carbohydrate metabolism, hemoglobin formation, and cross-linking of collagen, hair keratin and, elastin [46]. However, a surplus amount of Cu has been related to cellular and tissue damage (Wilson disease) with a variety of deleterious effects and human diseases [4]. The results of this study revealed that the Cu concentrations of the industrial site ranged from 0.47 to 3.49 mg kg^{-1} with an average value was 1.72 ± 0.44 mg kg^{-1}. The lowest and highest Cu content was 1.22 mg kg^{-1} (Green Amaranth) and 2.43 mg kg^{-1} (Malabar Spinach), respectively. The average Cu concentration of the non-industrial site was 1.35 ± 0.28 mg kg^{-1} and the obtained data ranged from 0.67 to 3.05 mg kg^{-1}. For this site, the lowest and highest Cu content was found 1.05 mg kg^{-1} (Red Amaranth) and 2.01 mg kg^{-1} (Malabar Spinach), respectively. The average Cu concentration of the local market vegetables was 4.93 ± 2.28 mg kg^{-1} with a range of 1.3–9.48 mg kg^{-1}. The lowest and highest Cu values for this market

Table 1. Concentrations of heavy metals (mg kg^{-1} fresh weight) in commonly consumed vegetables collected from industrial sites, non-industrial sites, arsenic contaminated sites and a local market in Bangladesh.

Vegetables	As					Cd			Pb			Cu			Zn		
	Industrial	Non-Industrial	Local Market	Arsenic Contaminated Areas		Industrial	Non-Industrial	Local Market	Industrial	Non-Industrial	Local Market	Industrial	Non-Industrial	Local Market	Industrial	Non-Industrial	Local Market
				Faridpur	Singair												
Brinjal	0.35 ± 0.07	0.18 ± 0.04	BDL	0.36 ± 0.01	0.26 ± 0.07	0.39 ± 0.09	0.14 ± 0.02	0.22 ± 0.02	2.58 ± 0.86	0.94 ± 0.23	BDL	1.98 ± 0.47	1.57 ± 0.76	7.16 ± 0.04	14.7 ± 3.14	7.45 ± 2.04	17.86 ± 0.92
Malabar Spinach	0.17 ± 0.05	0.12 ± 0.08	BDL	0.68 ± 0.08	0.65 ± 0.07	0.27 ± 0.03	0.08 ± 0.01	2.01 ± 0.18	1.44 ± 0.43	0.36 ± 0.08	BDL	2.43 ± 1.06	2.01 ± 1.04	6.05 ± 0.68	10.3 ± 2.19	9.45 ± 1.12	69.75 ± 0.91
Pointed Gourd	0.22 ± 0.08	0.12 ± 0.07	BDL	0.48 ± 0.09	0.37 ± 0.06	0.24 ± 0.06	0.12 ± 0.03	0.26 ± 0.07	2.87 ± 1.06	1.02 ± 0.34	0.35 ± 0.08	1.99 ± 0.84	1.12 ± 0.83	4.41 ± 0.19	10.4 ± 2.75	5.55 ± 0.47	18.73 ± 1.80
Potato	0.28 ± 0.09	0.13 ± 0.05	BDL	0.15 ± 0.06	0.11 ± 0.05	0.34 ± 0.08	0.14 ± 0.02	0.50 ± 0.20	1.33 ± 0.37	0.11 ± 0.04	0.62 ± 0.17	1.56 ± 0.46	1.36 ± 0.49	1.30 ± 0.00	7.52 ± 0.85	3.89 ± 0.31	7.99 ± 1.67
Tomato	0.17 ± 0.02	0.08 ± 0.02	BDL	0.59 ± 0.05	0.51 ± 0.29	0.37 ± 0.09	0.17 ± 0.04	0.35 ± 0.23	2.19 ± 1.12	0.67 ± 0.16	2.03 ± 0.15	1.56 ± 0.76	1.43 ± 0.71	4.10 ± 0.25	11.2 ± 1.42	4.52 ± 1.53	20.38 ± 1.41
Bottle Gourd	0.43 ± 0.12	0.23 ± 0.01	BDL	0.18 ± 0.04	0.12 ± 0.05	0.46 ± 0.07	0.22 ± 0.03	0.37 ± 0.08	2.27 ± 1.21	0.85 ± 0.31	1.94 ± 0.37	2.37 ± 1.42	1.57 ± 0.28	2.75 ± 0.16	12.2 ± 2.73	6.54 ± 2.76	15.04 ± 1.77
Yardlong Bean	0.24 ± 0.04	0.14 ± 0.03	BDL	1.56 ± 0.12	0.95 ± 0.06	0.18 ± 0.01	0.09 ± 0.01	0.60 ± 0.04	1.25 ± 0.53	0.45 ± 0.08	0.91 ± 0.11	1.42 ± 0.82	1.21 ± 0.43	5.45 ± 0.26	7.28 ± 0.47	3.34 ± 0.98	54.01 ± 14.30
Pumpkin	0.32 ± 0.10	0.19 ± 0.02	BDL	0.18 ± 0.04	0.14 ± 0.03	0.49 ± 0.04	0.21 ± 0.05	0.42 ± 0.15	1.23 ± 0.42	0.33 ± 0.07	1.34 ± 0.37	1.42 ± 0.47	1.35 ± 0.79	3.38 ± 0.19	11.2 ± 2.49	5.22 ± 2.54	30.98 ± 8.84
Red Amaranth	0.19 ± 0.02	0.09 ± 0.01	BDL	0.12 ± 0.07	0.16 ± 0.03	0.26 ± 0.03	0.11 ± 0.00	0.75 ± 0.20	2.17 ± 0.76	0.75 ± 0.11	0.41 ± 0.09	1.25 ± 0.85	1.05 ± 0.38	5.49 ± 0.33	11.76 ± 1.12	5.44 ± 2.62	50.99 ± 16.46
Green Amaranth	0.36 ± 0.06	0.19 ± 0.03	BDL	0.25 ± 0.06	0.51 ± 0.06	0.34 ± 0.09	0.15 ± 0.02	1.11 ± 0.12	2.70 ± 1.18	0.87 ± 0.28	0.69 ± 0.10	1.22 ± 0.75	1.08 ± 0.37	9.24 ± 0.34	11.2 ± 1.75	5.87 ± 1.89	58.06 ± 12.66
Minimum	**0.17**	**0.08**	**-**	**0.12**	**0.11**	**0.18**	**0.08**	**0.22**	**1.23**	**0.11**	**0.35**	**1.22**	**1.05**	**1.3**	**7.28**	**3.34**	**7.99**
Maximum	**0.43**	**0.23**	**-**	**1.56**	**0.95**	**0.49**	**0.22**	**2.01**	**2.87**	**1.02**	**2.03**	**2.43**	**2.01**	**9.24**	**14.70**	**9.45**	**69.75**
Range	**0.15–0.55**	**0.06–0.24**	**-**	**0.05–1.68**	**0.06–1.01**	**0.17–0.53**	**0.07–0.25**	**0.10–2.13**	**0.81–3.93**	**0.07–1.36**	**BDL - 2.20**	**0.47–3.49**	**0.67–3.05**	**1.3–9.48**	**6.81–17.84**	**2.36–10.57**	**6.81–70.66**
Mean	**0.27 ± 0.09**	**0.15 ± 0.05**	**-**	**0.46 ± 0.43**	**0.38 ± 0.28**	**0.33 ± 0.10**	**0.14 ± 0.05**	**0.66 ± 0.54**	**2.00 ± 0.64**	**0.64 ± 0.30**	**1.03 ± 0.66**	**1.72 ± 0.44**	**1.35 ± 0.28**	**4.93 ± 2.28**	**10.78 ± 2.16**	**5.73 ± 1.78**	**34.18 ± 21.55**
FAO/WHO [28]	**0.10**					**0.05**			**0.10**			**40**			**20***		

Data are presented as mean ± SD (n = 3) and BDL = below detection limit.

* FAO/WHO [48]

vegetables was 1.3 mg kg^{-1} (Potato) and 9.24 mg kg^{-1} (Green Amaranth), respectively. In general, the Cu contents of analyzed three site vegetables were lower than MPL value (40 mg kg^{-1}) [28]. The Cu concentration has been described in the ranged of 8.30–34.3 mg kg^{-1} in an industrial area of Dhaka [47] and 0.3–32 mg kg^{-1} in the Paira Riversides vegetable of the Patuakhali [5]. Shaheen et al. [3] reported that the Cu concentration in vegetables that of Bangladesh representative samples were showed 5.93 mg kg^{-1} (range: 2.25–9.72 mg kg^{-1}). Another study reported that the mean Cu content was 1.7 mg kg^{-1} with a range of 0.09–3.72 mg kg^{-1} in thirteen different vegetables from Bogra [37], and 21 mg kg^{-1} (range: 2.1–86 mg kg^{-1}) in vegetables grown in Noakhali district [17].

Zinc (Zn) is also an essential nutrient like Cu and these two metal's functions and effects are quite similar [4]. The concentrations of Zn was ranged from 6.81 to 17.84 mg kg^{-1} with a mean of 10.78 $\pm$ 2.16 mg kg^{-1} for the industrial site whereas the lowest and highest value was 7.28 mg kg^{-1} (Yardlong Bean) and 14.7 mg kg^{-1} (Brinjal), respectively. The average Zn concentration of this site was 1.8 times lower than the MPL value (20 mg kg^{-1}) set by FAO/WHO [48]. The average Zn concentration of the non-industrial site vegetables was 5.73 $\pm$ 1.78 mg kg^{-1} (range: 2.36–10.57 mg kg^{-1}). The lowest and highest value was 3.34 mg kg^{-1} (Yardlong Bean) and 9.45 mg kg^{-1} (Malabar Spinach), respectively. The non-industrial site average Zn concentration was 3.5 folds lower than the MPL [48]. The local market vegetable sample showed a significantly higher range of Zn concentration compares to the other two study sites. The average Zn concentrations in the market vegetables were 34.18 $\pm$ 21.55 mg kg^{-1} (range: 6.81–70.66 mg kg^{-1}), where the mean value was 1.7 folds higher than the MPL value [48]. The lowest and highest Zn concentration was observed 7.99 mg kg^{-1} (Potato) and 69.75 mg kg^{-1} (Malabar spinach), respectively. A comprehensive study on vegetables of Bangladesh reported that the Zn concentration was ranged from 0.07 to 4.75 mg kg^{-1} in market vegetables [3], 16.30–119 mg kg^{-1} in industrial areas of Dhaka [47], and 19.54–42.06 mg kg^{-1} in the DEPZ area, Dhaka [8]. Rahman et al. [17] reported that the average Zn value among the leafy edible vegetables was 59.6 mg kg^{-1} (range: 21.4–182.9 mg kg^{-1}) and non-leafy edible vegetables were 44.3 mg kg^{-1} (range: 17.2–122.3 mg kg^{-1}) which was a significantly higher concentration than this study.

3.2. Daily intake of metals

The health hazard or risk of a group of populations depends on the route and degree of exposure. So it is essential to assess the degree of exposure by identifying the pathways of pollutants to target populations. In general, ingestion, inhalation, and dermal contact are the main routes of metal exposure to humans. Among those routes, food chain or ingestion is the most considerable pathway. The present study considered the ingestion pathway for As, Cd, Pb, Cu, and Zn which is presumed to be vegetable consumption. The EDI values of selected metals were calculated according to the mean concentration of metals in vegetables and the particular consumption rate of a person. The EDI and the maximum tolerable daily intake (MTDI) [49] values of investigated metals are shown in Table 2.

For the industrial sites, total EDI for As, Cd, Pb, Cu, and Zn were 5.92 $\times 10^{-03}$, 7.24 $\times 10^{-03}$, 4.34 $\times 10^{-02}$, 3.73 $\times 10^{-02}$, and 2.33 $\times 10^{-01}$ mg day^{-1}, respectively. The mean EDI values of the industrial site showed the following increased order: As < Cd < Cu < Pb < Zn. On the other hand, the EDI for As contaminated two sites (Faridpur and Singair) showed a lower value than MTDI for each vegetable as well as the total value of every site. For the non-industrial site, total EDI for As, Cd, Pb, Cu, and Zn were 3.19 $\times 10^{-03}$, 3.10 $\times 10^{-03}$, 1.38 $\times 10^{-02}$, 2.98 $\times 10^{-02}$, and 1.24 $\times 10^{-01}$, respectively. The mean EDI values of the non-industrial site showed the following increased order: Cd < As < Pb < Cu < Zn. The local market samples total EDI for Cd, Pb, Cu, and Zn were 1.43 $\times 10^{-02}$, 1.79 $\times 10^{-02}$, 1.07 $\times 10^{-01}$, and 7.41 $\times 10^{-01}$, respectively. The mean EDI showed the following increased order: Cd < Pb < Cu < Zn for the local market. The EDI of all the metals for the three

Table 2. Estimated daily intake (EDI) of heavy metals from commonly consumed vegetable samples and maximum tolerable daily intake (MTDI) for the Bangladeshi population.

Vegetables	As				Cd			Pb			Cu			Zn		
	Industrial	Non-Industrial	Faridpur	Singair	Industrial	Non-Industrial	Local Market	Industrial	Non-Industrial	Local Market	Industrial	Non-Industrial	Local Market	Industrial	Non-Industrial	Local Market
Brinjal	7.58×10^{-04}	3.90×10^{-04}	7.80×10^{-04}	5.63×10^{-04}	8.45×10^{-04}	3.03×10^{-04}	4.86×10^{-04}	5.59×10^{-03}	2.04×10^{-03}	-	4.29×10^{-03}	3.40×10^{-03}	1.55×10^{-02}	3.19×10^{-02}	1.61×10^{-02}	3.87×10^{-02}
Malabar Spinach	3.68×10^{-04}	2.60×10^{-04}	1.47×10^{-03}	1.41×10^{-03}	5.85×10^{-04}	1.73×10^{-04}	4.36×10^{-04}	3.12×10^{-03}	7.80×10^{-04}	-	5.27×10^{-03}	4.36×10^{-03}	1.31×10^{-02}	2.23×10^{-02}	2.05×10^{-02}	1.51×10^{-01}
Pointed Gourd	4.77×10^{-04}	2.60×10^{-04}	1.04×10^{-03}	8.02×10^{-04}	5.20×10^{-04}	2.60×10^{-04}	5.61×10^{-04}	6.22×10^{-03}	2.21×10^{-03}	7.63×10^{-04}	4.31×10^{-03}	2.43×10^{-03}	9.56×10^{-03}	2.25×10^{-02}	1.20×10^{-02}	4.06×10^{-02}
Potato	6.07×10^{-04}	2.82×10^{-04}	3.25×10^{-04}	2.38×10^{-04}	7.37×10^{-04}	3.03×10^{-04}	1.07×10^{-03}	2.88×10^{-03}	2.38×10^{-04}	1.33×10^{-03}	3.38×10^{-03}	2.95×10^{-03}	2.83×10^{-03}	1.63×10^{-02}	8.43×10^{-03}	1.73×10^{-02}
Tomato	3.68×10^{-04}	1.73×10^{-04}	1.28×10^{-03}	1.11×10^{-03}	8.02×10^{-04}	3.68×10^{-04}	7.65×10^{-04}	4.75×10^{-03}	1.45×10^{-03}	4.40×10^{-03}	3.38×10^{-03}	3.10×10^{-03}	8.89×10^{-03}	2.43×10^{-02}	9.79×10^{-03}	4.42×10^{-02}
Bottle Gourd	9.32×10^{-04}	4.98×10^{-04}	3.90×10^{-04}	2.60×10^{-04}	9.97×10^{-04}	4.77×10^{-04}	7.96×10^{-04}	4.92×10^{-03}	1.84×10^{-03}	4.19×10^{-03}	5.14×10^{-03}	3.40×10^{-03}	5.95×10^{-03}	2.64×10^{-02}	1.42×10^{-02}	3.26×10^{-02}
Yardlong Bean	5.20×10^{-04}	3.03×10^{-04}	3.38×10^{-03}	2.06×10^{-03}	3.90×10^{-04}	1.95×10^{-04}	1.30×10^{-03}	2.71×10^{-03}	9.75×10^{-04}	1.96×10^{-03}	3.08×10^{-03}	2.62×10^{-03}	1.18×10^{-02}	1.58×10^{-02}	7.24×10^{-03}	1.17×10^{-01}
Pumpkin	6.93×10^{-04}	4.12×10^{-04}	3.90×10^{-04}	3.03×10^{-04}	1.06×10^{-03}	4.55×10^{-04}	9.04×10^{-04}	2.67×10^{-03}	7.15×10^{-04}	2.89×10^{-03}	3.08×10^{-03}	2.93×10^{-03}	7.32×10^{-03}	2.43×10^{-02}	1.13×10^{-02}	6.71×10^{-02}
Red Amaranth	4.12×10^{-04}	1.95×10^{-04}	2.60×10^{-04}	3.47×10^{-04}	5.63×10^{-04}	2.38×10^{-04}	1.63×10^{-03}	4.70×10^{-03}	1.63×10^{-03}	8.81×10^{-04}	2.71×10^{-03}	2.28×10^{-03}	1.19×10^{-02}	2.55×10^{-02}	1.18×10^{-02}	1.10×10^{-01}
Green Amaranth	7.80×10^{-04}	4.12×10^{-04}	5.42×10^{-04}	1.11×10^{-03}	7.37×10^{-04}	3.25×10^{-04}	2.39×10^{-03}	5.85×10^{-03}	1.89×10^{-03}	1.49×10^{-03}	2.64×10^{-03}	2.34×10^{-03}	2.00×10^{-02}	2.43×10^{-02}	1.27×10^{-02}	1.21×10^{-01}
Total EDI	**5.92×10^{-03}**	**3.19×10^{-03}**	**9.86×10^{-03}**	**8.19×10^{-03}**	**7.24×10^{-03}**	**3.10×10^{-03}**	**1.43×10^{-02}**	**4.34×10^{-02}**	**1.38×10^{-02}**	**1.79×10^{-02}**	**3.73×10^{-02}**	**2.98×10^{-02}**	**1.07×10^{-01}**	**2.33×10^{-01}**	**1.24×10^{-01}**	**7.41×10^{-01}**
MTDI [49]	**0.13**	**0.021**	**0.21**	**30**	**60**											

Table 3. Human carcinogenic risk (TR) due to consumption of As and Pb through commonly consumed vegetables.

Vegetables	As				Pb		
	Industrial	Non-Industrial	Faridpur	Singair	Industrial	Non-Industrial	Local Market
Brinjal	1.14×10^{-03}	5.85×10^{-04}	$1.17 \times 10^{-0}3$	8.45×10^{-04}	4.75×10^{-05}	1.73×10^{-05}	-
Malabar Spinach	5.53×10^{-04}	3.90×10^{-04}	2.21×10^{-03}	2.11×10^{-03}	2.65×10^{-05}	6.63×10^{-06}	-
Pointed Gourd	7.15×10^{-04}	3.90×10^{-04}	1.56×10^{-03}	1.20×10^{-03}	5.29×10^{-05}	1.88×10^{-05}	6.48×10^{-06}
Potato	9.10×10^{-04}	4.23×10^{-04}	4.88×10^{-04}	3.58×10^{-04}	2.45×10^{-05}	2.03×10^{-06}	1.13×10^{-05}
Tomato	5.53×10^{-04}	2.60×10^{-04}	1.92×10^{-03}	1.66×10^{-03}	4.03×10^{-05}	1.23×10^{-05}	3.74×10^{-05}
Bottle Gourd	1.40×10^{-03}	7.48×10^{-04}	5.85×10^{-04}	3.90×10^{-04}	4.18×10^{-05}	1.57×10^{-05}	3.56×10^{-05}
Yardlong Bean	7.80×10^{-04}	4.55×10^{-04}	5.07×10^{-03}	3.09×10^{-03}	2.30×10^{-05}	8.29×10^{-06}	1.67×10^{-05}
Pumpkin	1.04×10^{-03}	6.18×10^{-04}	5.85×10^{-04}	4.55×10^{-04}	2.27×10^{-05}	6.08×10^{-06}	2.46×10^{-05}
Red Amaranth	6.18×10^{-04}	2.93×10^{-04}	3.90×10^{-04}	5.20×10^{-04}	4.00×10^{-05}	1.38×10^{-05}	7.49×10^{-06}
Green Amaranth	1.17×10^{-03}	6.18 × 10-04	8.13×10^{-04}	1.66×10^{-03}	4.97×10^{-05}	1.60×10^{-05}	1.27×10^{-05}
Total	$\mathbf{8.88 \times 10^{-03}}$	$\mathbf{4.78 \times 10^{-03}}$	$\mathbf{1.48 \times 10^{-02}}$	$\mathbf{1.23 \times 10^{-02}}$	$\mathbf{3.69 \times 10^{-04}}$	$\mathbf{1.17 \times 10^{-04}}$	$\mathbf{1.52 \times 10^{-04}}$

sampling sites was lower than the MTDI, according's to Shaheen et al. [3] and JECFA [49].

3.3. Health risk assessment

3.3.1. Non-carcinogenic health risk

Human health risks due to the consumption of metals polluted vegetables by adult inhabitants were the assessment based on THQ. The THQs of the five investigated metals were shown in Figure 2. The results showed that the THQ of some of the analyzed heavy metals was lower than 1 (except As, Cd, and Pb for some vegetables in all sampling sites), indicating that exposure of a single metal through vegetable ingestion did not pretense a significant health hazard [Figure 2(a-e)]. However, the THQ values of As was higher than 1 for all the vegetables in the industrial site whereas the non-industrial sites five vegetables [e.g., Brinjal (1.30), Bottle Gourd (1.66), Yardlong Bean (1.01), Pumpkin (1.37), and Green Amaranth (1.37)] demonstrated higher THQ values (>1), indicating human health might be posed significant non-cancer-related health risks due to As exposed vegetable consumption. The Arsenic contaminated sites THQ values showed significant variability among the vegetables where the THQ of As for the Faridpur region every sample was higher than 1 [except, Red Amaranth (0.87)] and also the Singair region every sample was higher than 1 [except, Potato (0.79) and Bottle Gourd (0.87)] (Figure 2a). The THQ values of Cd was lower than 1 for all the vegetables in the three sampling site except one [e.g., Malabar Spinach (1.45)] in the local market (Figure 2b). However, the THQ of Pb was higher than 1 for some vegetables in the industrial site [Brinjal (1.59), Pointed Gourd (1.78), Tomato (1.36), Bottle Gourd (1.41), Red Amaranth (1.34), and Green Amaranth (1.67)] and local market [e.g., Tomato (1.26) and Bottle Gourd (1.20)], indicating intake of Pb through vegetables possess potential non-cancer health risk (Figure 2c).

The total target hazard quotients (TTHQ) analysis shows the combined health risk for consumption of multiple heavy metals (Figure 2f). The TTHQ values of the determined metals (except Cu and Zn in the industrial and non-industrial sites) from entire analyzed vegetables were greater than 1, that indicating the people might be at potential non-carcinogenic risk if they consume all of those vegetables in their diet. Besides, the TTHQ of metals of all the analyzed vegetables were showed following descending order: Bottle Gourd > Brinjal > Green Amaranth > Malabar Spinach > Pointed Gourd > Potato > Pumpkin > Red Amaranth > Tomato > Yardlong Bean for industrial, non-industrial, and local market sampling sites [Figure 2(f)]. The TTHQ for Faridpur and Singair region was found greater than 1 which indicating As contaminated regions people might be possessed non-cancer risk due to consumption of local vegetables. The TTHQ for all vegetables (except Brinjal, Potato, and Pointed Gourd for the local market) was found higher than 1, indicating the possible significant health hazard of its consumption. However, TTHQs of vegetables were less or equal to 1, indicating no or slight possible risks in the intake of these vegetables.

The HI articulates the combined non-carcinogenic effects of multiple metals. In Figure 2f, HI values through particular vegetable consumption were 36.24, 16.74, and 15.03 for industrial, non-industrial, and local market sampling sites, respectively, where all the HI was greater than 1. The relative influences of As, Cd, Pb, Cu, and Zn to HI were 54.41, 6.66, 34.22, 2.57, and 2.15 %, respectively, for industrial sites and 63.43, 6.17, 23.48, 4.45, and 2.47%, respectively, for non-industrial sites. Conversely, the relative contribution of Cd, Pb, Cu, and Zn to HI for the local market was 31.66, 34.07, 17.78, and 16.42 %, respectively. The results showed

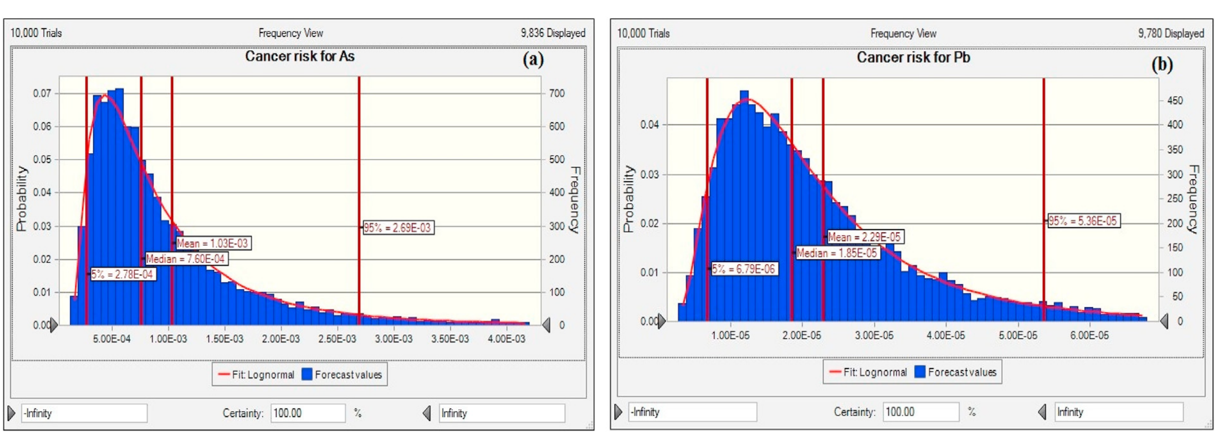

Figure 3. Predicted probability distribution results of the target carcinogenic risk (TR) for (a) As and (b) Pb.

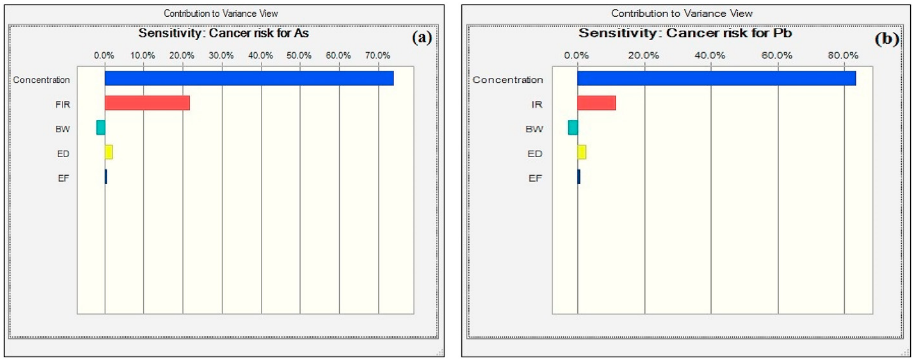

Figure 4. Sensitivity analysis on the target carcinogenic risks for (a) As and (b) Pb.

that, As and Pb were the main metals causative to the significant health hazard, with Cd being secondary and Cu and Zn being the least important metals.

3.3.2. Carcinogenic risks

As and Pb are well-organized carcinogenic agents categorized by International Agency for research on cancer (IARC) [50]. Many types of cancer could be resulted due to chronic exposure to carcinogens like As and Pb [9]. The calculated carcinogenic risks (TRs) of As and Pb through the investigated vegetables are presented in Table 3.

USEPA suggested that the safe limit for cancer risk is TR $< 10^{-6}$ (1 chance in 1,000,000 lifetime exposure) and threshold or unacceptable cancer risk limit is TR $> 10^{-4}$ (1 chance in 10,000 lifetime exposure) [30, 51]. The TR of As ranged from 5.53×10^{-04} to 1.40×10^{-03} for the industrial sites, 2.60×10^{-04} to 7.48×10^{-04} for the non-industrial sites, 3.90×10^{-04} to 5.07×10^{-03} for the Faridpur region, and 3.58×10^{-04} to 3.09×10^{-03} for the Singair region. Results showed that TR for As was higher than the threshold cancer risk limit ($>10^{-4}$) of all the studied vegetables in three sampling sites which indicating people consuming these vegetables are exposed to high lifetime cancer risk. Conversely, the TR of Pb ranged from 2.27×10^{-05} to 5.29×10^{-05} for the industrial sites, 2.03×10^{-06} to 1.88×10^{-05} for the non-industrial sites, and 6.48×10^{-06} to 3.74×10^{-05} for the local market. The TR of Pb for most of the studied vegetables in three sampling sites exceeded the safe limit ($>10^{-6}$) (except Malabar Spinach, Potato, Yardlong Bean and Pumpkin for the non-industrial sites and Pointed Gourd and Red Amaranth for the local market) but within the acceptable limit (10^{-6} to 10^{-4}) [5,52]. But the results revealed that people are at risk of cancer due to exposure to Pb through the consumption of Pb enriched vegetables in the long run [51]. The TR of studied vegetables were showed following order: Bottle Gourd > Brinjal > Green Amaranth > Malabar Spinach > Pointed Gourd > Potato > Pumpkin > Red Amaranth > Tomato > Yardlong Bean for all the sampling sites (Table 3). Moreover, the cumulative target cancer risk (total TR) of all studied vegetables for sampling sites were exceeded the threshold limit ($>10^{-4}$), indicating there have a significant cancer risk if people intake all of these vegetables.

3.3.2.1. Probabilistic health risk and sensitivity assessment. The cancer risks from ingestion of metals contaminated vegetables were evaluated using the TRs. The probability carcinogenic risks of As and Pb were studied using the Monte Carlo Simulation method (Figure 3a and b). Results showed that the mean probability of TR for As and Pb were 1.03×10^{-03} and 2.29×10^{-05}, respectively. Whereas the median values of TR for As and Pb were 7.60×10^{-04} and 1.85×10^{-05}, respectively with 100% certainty. The 5th and 95th percentile values were found 2.78×10^{-04} and 2.69×10^{-03} for As and 6.79×10^{-06} and 5.36×10^{-05} for Pb. According to the USEPA [51] guideline, the mean and 95th percentile values of As were exceeded the threshold value ($>10^{-4}$) which indicates that about 95% of people would experience high potential cancer risk from vegetable consumption. Additionally, the median and 5th percentile values of As were exceeded the safe limit ($<10^{-6}$). Conversely, the mean, median and 95th percentile values for Pb showed greater value than ($<10^{-6}$) which also suggesting 95% of people of the study area cross the safe limit boundary and they might have a chance of cancer risk in a lifetime for the consumption of Pb contaminated vegetables [30, 35] although the value was within in acceptable position (10^{-4} to 10^{-6}) [5]. Furthermore, only 5% population would not experience Pb-induced cancer risk for vegetable consumption. Moreover, As and Pb can be regarded as the priority heavy metals due to their carcinogenic risks.

The importance of the input variables involved in the TR calculation was assessed by sensitivity analysis [33, 34]. The results revealed that As and Pb concentration is the most important factors on the TR values for both heavy metals (Figure 4a and b). For As induced TR calculation, concentration, food ingestion rate (FIR), exposure duration (ED), and exposure frequency (EF) revealed the positive influences with the percentage of 73.8%, 21.7%, 2.0%, and 0.4%, respectively. While only body weight (BW) showed negative impacts with the percentage of -2.0% for carcinogenic risk (TR) calculation (Figure 4a). On the other hand, for the Pb induced TR calculation, concentration (83.2%), FIR (11.3%), ED (2.4%), and EF (0.4%) showed positive impacts, and only BW (-2.4%) revealed negative impact. However, this study indicates that metal concentration is significantly responsible for cancer risk estimation.

4. Conclusion

The concentrations of As, Cd, Pb, Cu, and Zn in the commonly consumed vegetables varied significantly as a function of plant species and growth locations. The findings of this study indicated that the metals concentration of the vegetables also remained higher than the respective MPL except for Cu and Zn. The EDI for every single metal of vegetables showed a relatively lower value compared with a tolerable limit. The non-carcinogenic health risk suggests that the THQ of individual metals like As, Cd, and Pb showed the potential human risk for some vegetables in all studied sites. Although most of the metals THQ <1 (including Cu and Zn) indicates the consumer would not pose a health risks. Whereas the TTHQ of all metals was >1 except Cu and Zn for the industrial and the non-industrial sites that indicate potential health risk for vegetable consumption. The As and Pb induced TR due to vegetable ingestion revealed both unacceptable (10^{-4}) and acceptable (10^{-4} to 10^{-6}) health risk, respectively. On the other hand, probabilistic health risk revealed 95% of people in the study area have a significant chance of cancer risk due to consumption of high As content vegetables. Overall, this study suggested that an interval monitoring is needed for the control and prevention of heavy metals contamination as well as ensuring food safety for Bangladeshi populations.

Declarations

Author contribution statement

Md. Morshedul Haque and Nahin Mostofa Niloy: Performed the experiments; analyzed and interpreted the data, wrote the paper

Md. Akhte Khirul and Md. Ferdous Alam: Contributed reagents, materials, analysis tools.

Shafi M. Tareq: Conceived and designed the experiments, wrote the paper.

Funding statement

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Data availability statement

Data included in article/supplementary material/referenced in article.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

Acknowledgements

The authors would like to acknowledge the Institute of Nuclear Science and Technology (INST), Atomic energy research establishment (AERE), Dhaka and Department of Environmental Sciences, Jahangirnagar University for providing logistic and technical support. The authors would also like to extend great gratitude to Farah Tasneem, Scientific Officer, INST, AERE, Dhaka for her kind support and suggestions.

References

[1] S.A. Mansour, M.H. Belal, A.A.K. Abou-Arab, M.F. Gad, Monitoring of pesticides and heavy metals in cucumber fruits produced from different farming systems, Chemosphere 75 (2009) 601–609.
[2] M.H.H. Ali, K.M. Al-Qahtani, Assessment of some heavy metals in vegetables, cereals and fruits in Saudi Arabian markets, Egypt, J. Aquat. Res. 38 (2012) 31–37.
[3] N. Shaheen, N.M. Irfan, I.N. Khan, S. Islam, M.S. Islam, M.K. Ahmed, Presence of heavy metals in fruits and vegetables: health risk implications in Bangladesh, Chemosphere 152 (2016) 431–438.
[4] P.B. Tchounwou, C.G. Yedjou, A.K. Patlolla, D.J. Sutton, Molecular, Clinical and Environmental Toxicology, Springer Basel, Basel, 2012.
[5] M.S. Islam, M.K. Ahmed, M. Habibullah-Al-Mamun, Determination of heavy metals in fish and vegetables in Bangladesh and health implications, Hum. Ecol. Risk Assess. 21 (2015) 986–1006.
[6] J. Manzoor, M. Sharma, K.A. Wani, Heavy metals in vegetables and their impact on the nutrient quality of vegetables: a review, J. Plant Nutr. 41 (2018) 1744–1763.
[7] A. Zwolak, M. Sarzyńska, E. Szpyrka, K. Stawarczyk, Sources of soil pollution by heavy metals and their accumulation in vegetables: a review, water, Air. Soil Pollut. 230 (2019).
[8] J.U. Ahmad, M.A. Goni, Heavy metal contamination in water, soil, and vegetables of the industrial areas in Dhaka, Bangladesh, Environ. Monit. Assess. 166 (2010) 347–357.
[9] A. Jan, M. Azam, K. Siddiqui, A. Ali, I. Choi, Q. Haq, Heavy metals and human health: mechanistic insight into toxicity and counter defense system of antioxidants, Int. J. Mol. Sci. 16 (2015) 29592–29630.
[10] J.N. Duruibe, M.O.C. Ogwuegbu, J.N. Egwurugwu, Heavy metal pollution and human biotoxic effects, Int. J. Phys. Sci. 2 (2007) 112–118.
[11] N. Gupta, D.K. Khan, S.C. Santra, An assessment of heavy metal contamination in vegetables grown in wastewater-irrigated areas of titagarh, West Bengal, India, Bull. Environ. Contam. Toxicol. 80 (2008) 115–118.
[12] M.K. Ahmed, S. Ahamed, S.M. Rahman, R.M. Haque, M.M. Islam, Heavy metals concentration in water , sediments and their bioaccumulations heavy metals concentration in water , sediments and their bioaccumulations in some freshwater fishes and mussel in dhaleshwari river , Bangladesh, Terr. Aquat. Environ. Toxicol. 3 (2009) 33–41.
[13] BBS(Bangladesh Bureau Of Statistics), Yearbook of Agricultural Statistics-2017, 2018. Dhaka.
[14] M.G.M. Alam, E.T. Snow, A. Tanaka, Arsenic and heavy metal contamination of vegetables grown in Samta village, Bangladesh, Sci. Total Environ. 308 (2003) 83–96.
[15] R. Parvin, A. Sultana, M.A. Zahid, Detection of heavy metals in vegetables cultivated in different locations in chittagong, Bangladesh, IOSR J. Environ. Sci. Toxicol. Food Technol. 8 (2014) 58–63.
[16] M.A. Islam, D. Romić, M.A. Akber, M. Romić, Trace metals accumulation in soil irrigated with polluted water and assessment of human health risk from vegetable consumption in Bangladesh, Environ. Geochem. Health 40 (2018) 59–85.
[17] M.M. Rahman, M. Asaduzzaman, R. Naidu, Consumption of arsenic and other elements from vegetables and drinking water from an arsenic-contaminated area of Bangladesh, J. Hazard Mater. 262 (2013) 1056–1063.
[18] M.S. Sultana, S. Rana, S. Yamazaki, T. Aono, S. Yoshida, Health risk assessment for carcinogenic and non-carcinogenic heavy metal exposures from vegetables and fruits of Bangladesh, Cogent Environ. Sci. 3 (2017) 1–17.
[19] H.K. Das, A.K. Mitra, P.K. Sengupta, A. Hossain, F. Islam, G.H. Rabbani, Arsenic concentrations in rice, vegetables, and fish in Bangladesh: a preliminary study, Environ. Int. 30 (2004) 383–387.
[20] S. Kapaj, H. Peterson, K. Liber, P. Bhattacharya, Human health effects from chronic arsenic poisoning–A review, J. Environ. Sci. Heal. Part A. 46 (2011) 677–679.
[21] S.M. Tareq, S. Safiullah, H.M. Anawar, M.M. Rahman, T. Ishizuka, Arsenic pollution in groundwater: a self-organizing complex geochemical process in the deltaic sedimentary environment, Bangladesh, Sci. Total Environ. 313 (2003) 213–226.
[22] M.M. Rahman, R. Sultana, M. Shammi, J. Bikash, T. Ahmed, M. Maruo, M. Kurasaki, M.K. Uddin, Assessment of the status of groundwater arsenic at singair upazila, Manikganj Bangladesh; Exploring the correlation with other metals and ions, Expo, Health 8 (2016) 217–225.
[23] G. Tiwari, S. Wang, J. Tang, S.L. Birla, Analysis of radio frequency (RF) power distribution in dry food materials, J. Food Eng. 104 (2011) 548–556.
[24] S.E. Allen, H.M. Grimshaw, A.P. Rowland, Chemical analysis, in: P.D. Moore, S.B. Chapman (Eds.), Methods Plant Ecol., Blackwell, Oxford, 1986, pp. 285–344.
[25] USEPA, Supplementary guidance for conducting health risk assessment of chemical mixtures, in: Risk Assess. Forum Tech. Panel [EPA/630/R-00/002], United States Environmental Protection Agency, Washington, DC, 2000.
[26] FAO, Arsenic contamination of irrigation water, soil and crops in Bangladesh: risk implications for sustainable agriculture and food safety in Asia, UN FAO Rep. 20 (2006) 1–46.
[27] USEPA, Risk assessment guidance for superfund, in: Human Health Evaluation Manual Part A, Interim Final I, 1989. EPA/540/1e89/002. Washington, DC, USA.
[28] FAO/WHO, joint FAO/WHO food standards programme Codex Committee on Contaminants in foods, in: Food CF/5 INF/1. Fifth Session. The Hague, The Netherlands, 2011. http://www.fao.org/input/download/report/758/REP11_CFe.pdf.
[29] USEPA, Technical Support Document for Water Quality-Based Toxics Control. EPA/505/2-90-001, 1991. Washington, DC, USA.
[30] USEPA, Risk Based Screening Table, Composite Table: Summary Tab 0615, 2015 http:/www2.epa.gov/risk/risk based screening table generic%0Atables.
[31] X. Wang, T. Sato, B. Xing, S. Tao, Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish, Sci. Total Environ. 350 (2005) 28–37.
[32] USEPA, Guiding Principles for Monte Carlo Analysis, 1997. Washington, DC, USA.
[33] C. Qu, K. Sun, S. Wang, L. Huang, J. Bi, Monte Carlo simulation-based health risk assessment of heavy metal soil pollution: a case study in the qixia mining area, China, Hum. Ecol. Risk Assess. 18 (2012) 733–750.
[34] M. Bodrud-Doza, S.M.D.U. Islam, M.T. Hasan, F. Alam, M.M. Haque, M.A. Rakib, M.A. Asad, M.A. Rahman, Groundwater pollution by trace metals and human health risk assessment in central west part of Bangladesh, Groundw. Sustain. Dev. 9 (2019) 100219.
[35] N. Saha, M.R. Zaman, Evaluation of possible health risks of heavy metals by consumption of foodstuffs available in the central market of Rajshahi City, Bangladesh, Environ. Monit. Assess. 185 (2013) 3867–3878.
[36] J. Pandey, U. Pandey, Accumulation of heavy metals in dietary vegetables and cultivated soil horizon in organic farming system in relation to atmospheric deposition in a seasonally dry tropical region of India, Environ. Monit. Assess. 148 (2009) 61–74.
[37] M.S. Islam, M.K. Ahmed, M. Habibullah-Al-Mamun, M. Raknuzzaman, The concentration, source and potential human health risk of heavy metals in the commonly consumed foods in Bangladesh, Ecotoxicol. Environ. Saf. 122 (2015) 462–469.
[38] S.M. Tareq, Arsenic and fluorescent humic substances in the ground water of Bangladesh: a public health risk, in: S.J. S.B.T.-H, A.T. Flora (Eds.), Handb. Arsen. Toxicol., Academic Press, Oxford, 2015, pp. 73–93.
[39] H.M. Anawar, J. Akai, K.M.G. Mostofa, S. Safiullah, S.M. Tareq, Arsenic poisoning in groundwater: health risk and geochemical sources in Bangladesh, Environ. Int. 27 (2002) 597–604.
[40] R.B. Neumann, A.P. St. Vincent, L.C. Roberts, A.B.M. Badruzzaman, M.A. Ali, C.F. Harvey, Rice field geochemistry and hydrology: an explanation for why groundwater irrigated fields in Bangladesh are net sinks of arsenic from groundwater, Environ. Sci. Technol. 45 (2011) 2072–2078.
[41] M.M. Haque, N.M. Niloy, O.K. Nayna, K.J. Fatema, S.B. Quraishi, J.-H. Park, K.-W. Kim, S.M. Tareq, Variability of water quality and metal pollution index in the Ganges River, Bangladesh, Environ. Sci. Pollut. Res. 27 (2020) 42582–42599.

[42] M.S. Sultana, Y.N. Jolly, S. Yeasmin, A. Islam, S. Satter, S.M. Tareq, Transfer of heavy metals and radionuclides from soil to vegetables and plants in Bangladesh, in: Soil Remediat. Plants, Elsevier, 2015, pp. 331–366.
[43] M.S. Islam, M.K. Ahmed, M. Habibullah-Al-Mamun, S. Masunaga, Trace metals in soil and vegetables and associated health risk assessment, Environ. Monit. Assess. 186 (2014) 8727–8739.
[44] L. Järup, Hazards of heavy metal contamination, Br. Med. Bull. 68 (2003) 167–182.
[45] H. Abadin, A. Ashizawa, Y.-W. Stevens, F. Llados, G. Diamond, G. Sage, M. Citra, A. Quinones, S.J. Bosch, S.G. Swarts, Toxicological Profile for Lead, Agency for Toxic Substances and Disease Registry (US)., Atlanta (GA), 2007.
[46] WHO/FAO/IAEA, Trace Elements in Human Nutrition and Health, World Health Organization, Geneva, 1996.
[47] M. Islam, M. Hoque, Concentrations of heavy metals in vegetables around the industrial area of Dhaka city, Bangladesh and health risk assessment, Int. Food Res. J. 21 (6) (2014) 212.
[48] FAO/WHO, Food Standards Program, Codex Alimentarius Commission (FAO/WHO), Joint FAO/WHO Food Standards Program 1st, XVII, Codex Alimentarius, 1984.
[49] JECFA, Summary and Conclusions of the 61st Meeting of the Joint FAO/WHO Expert Committee on Food Additives (JECFA), JECFA/61/SC, Rome, Italy., 2003.
[50] F.H. Tani, S. Barrington, Zinc and copper uptake by plants under two transpiration rates. Part I. Wheat (Triticum aestivum L.), Environ. Pollut. 138 (2005) 538–547.
[51] USEPA, Riskbased Concentration Table, 2010 www.epa.%0Agov/reg3hwmd/risk/human/index.htm.
[52] M. Fryer, C.D. Collins, H. Ferrier, R.N. Colvile, M.J. Nieuwenhuijsen, Human exposure modelling for chemical risk assessment: a review of current approaches and research and policy implications, Environ. Sci. Pol. 9 (2006) 261–274.