#### RESEARCH



# Elevated Uptake and Translocation Patterns of Heavy Metals in Different Food Plants Parts and Their Impacts on Human Health

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#### Abstract

Irrigation with contaminated wastewater is a common practice in cultivation of crops and vegetables in many developing countries due to the scarcity of available fresh water. The present study has investigated the transfer and mobilization trends of heavy metals in different crops and vegetables plants grown in contaminated soil and waterbody. The translocation patterns of metals from polluted sources into different organs of plants bodies such as roots and edible parts and associated health risks have been evaluated simultaneously. Total of 180 different environmental samples including food plants, agricultural soil, and irrigation water were collected and analyzed. Heavy metal concentrations (Fe, Ni, Mn, Pb, Cu, Cd, As) in water, soil, and different parts of crops and vegetable plants were compared with the permissible levels reported by FAO/WHO, EU, and USEPA. Different metals contents within the food plants were found to be in the order of Fe > Mn > Ni > Cu > Pb > Cd > As. Pollution load index (PLI) data indicate that soil is highly polluted with Cd as well as moderately contaminated by As and Cu. Bioconcentration factor (BCF) analysis showed excessive accumulation of some heavy metals in crops and vegetables. Target hazard quotient (THQ) and target carcinogenic risk (TCR) analysis data showed higher carcinogenic and non-carcinogenic risks for both adult and children from the consumption of metal-contaminated food items. The results of metal pollution index (MPI), estimated daily intake (EDI), and hazard index (HI) analyses demonstrated the patterns of metals pollution in different food plants.

Keywords Wastewater irrigation · Metal pollution · Crop and vegetables · Accumulation · Mobilization · Health risks

# Introduction

Heavy and toxic metals contamination in waterbodies, agricultural soil, and different agricultural products such as crops, vegetables, and fruits has become a major pollution concern nowadays in many developing countries. Crops and vegetables offer essential nutrients such as vitamins, proteins, carbohydrates, minerals, and fibers along with

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other essential micronutrients and trace metals to human diet [1]. Regular consumption of a variety of fruits, crops, and vegetables is highly recommended by "Guidelines for Americans 2015–2020" for entire lifespan, even in the time of pregnancy [2]. Standard qualities of crops and vegetables with essential nutrients originate from their plants characteristics and growth media. Plants can easily take up both essential and non-essential elements over a wide range of quantities from their growth mediums such as soil and water [3]. Any contamination in water and soil directly impacts plants growth, quality, and nutrient availability in different parts of plants bodies. Among different toxic chemical species found in modern agricultural system, the metallic contaminants have been appeared as serious and emerging pollutants in recent years [4–6]. Unplanned industrial establishments, release of toxic industrial effluents, and excessive use of various agrochemicals, e.g., insecticides, pesticides, and fertilizers, as well as natural disasters are the common sources of metal pollution in waterbodies, soil,

and plants grown in developing countries [4, 7-10]. Other anthropogenic activities responsible for metals contamination include mining and uncontrolled release of domestic wastewater [11-13]. Metals toxicities in agricultural soil largely depend on soil pH, soil structure, soil parent materials, and soil cation exchange capacity [4, 7, 14, 15]. Crops and vegetables plants can readily uptake different metallic species through their root systems, leading to the accumulation of toxic metals in plants tissues when they are being cultivated in contaminated growth environment [6]. Depending on the chemical forms and biochemical interactions, different plants tissues can retain chemical compounds during bioaccumulation processes [5].

Heavy metals are non-biodegradable and generally categorized as essential (copper, zinc, iron, manganese, selenium, and cobalt) and toxic (arsenic, cadmium, lead, mercury, nickel, etc.). They can exist in different terrestrial environmental components such as water, soil, and plants. Toxicity in the respective environmental sources is realized after a long period of accumulation [16-18]. It has been well known that surface waterbody and soil pollution occurs because of industrial wastes, soil structures, municipal sewages, agricultural runoff including fertilizers, pesticides, domestic wastes [4, 14, 15, 19, 20]. However, because of toxicity, abundance, persistence, and subsequent accumulation in aquatic habitats, significant research is ongoing focusing on heavy metal contamination in aquatic environment, as well as in different agricultural products which directly impact agroecosystem [21-23]. Different metals from these polluted waterbodies, soil, and sediments can be transported to human body through the food chains and thus severely affect in many physical, chemical, and biological functions such as lower the energy levels, damage DNA, and change the gene expression [20].

Bangladesh is a small country whose economy is largely dependent on the agricultural products. A wide variety of crops, vegetables, and fruits are grown in different areas within the country throughout the year. However, as a developing country, there have been rapid expansion of industrialization and urbanization activities occurred in recent years. These have caused the transformation of significant arable land areas into industrial zones in Bangladesh. Unfortunately, in most cases, these industrialization, urbanization, and other infrastructural developments underwent without any proper planning and guidelines. Most of the industrial and urbanization processes discharge their complex liquid wastes directly into the surrounding environment without any prior treatment or partial treatments and thus contaminate waterbodies, arable soil, and plants in the respective areas [10]. In addition, wastewater irrigation, frequent occurrence of floods and other natural disasters, and the excessive uses of different agrochemicals have caused the distribution and mobilization of toxic environmental pollutants from point

sources to the non-contaminated areas in the country [24]. Due to the scarcity of available agricultural lands, a large variety of crops and vegetables species are grown near different industrial areas in Bangladesh. This may result in the accumulation and deposition of various heavy and toxic metals in different food plants bodies grown in contaminated waterbodies and soil within the respective areas [4, 10]. Literature reports on metals contaminants in soil and food plants grown in different countries around the world show the serious concern among scientific communities about the impacts of metals pollution in agroecosystem [15, 25, 26]. Necessary data have been documented and reported in the literatures from various countries regarding the impacts of industrial and agrochemical pollution in food chain and their growth media [7, 25–28]. Bangladesh is an agricultural and riverine country. A good number of crops and vegetables such as rice, wheat, egg plants, green bean, lady's finger (okra), spinach, and bitter gourd are cultivated in the available arable lands near different industrial areas of Bangladesh. These crops and vegetables plants are at a serious risk of accumulating excessive metals in the different parts of their bodies from contaminated soil and waterbody. However, very limited information is available on the level, trend, and risk evaluation of bioavailable fraction and the transfer factor of heavy and toxic metals into crops and vegetables grown near industrial areas of Bangladesh [29]. Therefore, the present study aimed to investigate the accumulation and mobilization patterns of heavy metals from contaminated waterbody and arable soil into crops and vegetables grown around industrial areas of Bangladesh. This research also focused on translocation trends of toxic metals in different parts of crops and vegetables plants bodies as emerging contaminants in agroecosystem. In addition, this study concentrated on the assessment of various human health risks associated with the consumption of various metal-contaminated crops and vegetable species.

This study was carried around Dhaka Export Processing Zone (DEPZ) located at Savar Upazilla in Dhaka district of Bangladesh. DEPZ is the largest industrial area in Bangladesh, and it is located near (about 32 km away) the capital city Dhaka. Different crops and vegetables are grown randomly throughout the year in the available lands around DEPZ area where wastewater has been used in irrigation and cultivation of various food plants. All crops and vegetables samples were collected from the different plants' fields within the study area. Soil and irrigation water samples were collected simultaneously with the plants from the respective crops and vegetables fields and nearby waterbodies. After collection, food plants samples were washed, cleaned, and dried properly. Acid digestion method was used to process and digest all soil, crops, and vegetables samples with the application of appropriate mixture of Conc. HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, and HClO<sub>4</sub> solutions following the standard established procedures reported in literatures [10, 30]. The solutions obtained from the digestion of different soil and plant samples were analyzed for heavy and toxic metals by using atomic absorption spectrophotometric method (AAS). The degree of toxicity induced by heavy metals in soil and various food plants was assessed by the analyses of different pollution indices such as metal pollution index (MPI) and pollution load index (PLI) [31, 32]. Metal transfer from soil to plants and their mobilization in different plants parts were evaluated by the analysis of bioconcentration factor (BCF) [33]. Human health risk analyses were carried out by the determination of estimated daily intake (EDI), target hazard quotient (THQ), hazard index (HI), and total carcinogenic risk (TCR) associated with the consumption of different metal-contaminated crops and vegetables species [11, 12, 31].

# **Materials and Methods**

#### **Study Area**

The present study covers the area from 23.81650° N to 23.94880° N latitude and from 90.24390° E to 90.26870°E longitude of Savar Upazilla in the Dhaka district of Bangladesh (Fig. 1). Savar Upazilla is the fastest growing industrial area in Bangladesh which is situated near the capital city of Dhaka. Different environmental samples were collected from various agricultural fields located around the Dhaka export processing zone (DEPZ) in Savar Upazilla of Dhaka district (Fig. 1). DEPZ is the largest industrial area in Bangladesh, and it is situated about 32 km away from the capital city Dhaka. DEPZ accommodates about 400 industries including textile, dyeing, plastic, steel and metal processing, semiconductor materials, lather, tanning, paper, pharmaceuticals, fertilizer, and electroplating [34]. These industries continuously discharge their complex toxic effluents with a partial treatment or without any prior treatment into nearby waterbody within a lake which is known as Ashulia Lake. Farmers in this area cultivate a good number of crops and vegetables in the available lands around the Ashulia Lake which is located at close vicinity of these industrial establishments. These food plants are being grown up in contaminated soil and irrigated with toxic effluents mixed wastewater bodies from the lake (Fig. 1). These polluted crops and vegetables later transport to Dhaka city and millions of people in the capital city consume them. Various crops and vegetables such as rice, wheat, potato, brinjal, lady's finger, spinach, bean, bitter gourd, and cucumber are cultivated in the study area throughout the year following the monsoon season and winter season. Some food plants are grown in monsoon season and other are grown in the winter season. Most of the farmers in Bangladesh are not well educated. Generally, they are highly reluctant to follow

any specific standard protocols in their agricultural activities. Farmers dig soil in the available agricultural lands and make them suitable to grow various food plants. In most cases, they did not follow specific protocols to use fertilizer, pesticides, and other agrichemicals in cultivation. Farmers in the study area randomly grow different crops and vegetables in the available lands, and they extensively use fertilizers, pesticides, and other agrochemicals in growing crop and vegetables in the study area without following any specific protocols and thus contaminate arable soils and the respective food plants grown in there. The complex toxic effluents from various industries located in DEPZ within Savar Upazilla area are being released into an open drain which ends up mixing into waterbody of Ashulia Lake. Farmers use this mixed wastewater in irrigation and cultivation of various crops and vegetables species in the respective study area.

#### **Environmental Samples Collection**

Six different common crops and vegetables plants grown in industrially contaminated study area were selected for this research which were rice (Oryza sativa), brinjal or eggplant (Solanum melongena), lady's finger (Abelmoschus esculentus), water spinach (Ipomoea aquatica), red spinach (Amaranthus dubius), and green spinach (Spinacia oleracea). All crop and vegetable plants (rice, brinjal, lady's finger, water spinach, red spinach, green spinach), soil, and irrigation water were collected in August 2022 from the study area during the monsoon season, following the standard reported procedures [35, 36]. Random sampling procedures were used, and ten replicates were taken (at least 5 m away from each other) for each sample. Food plants and soil samples were collected from different agricultural plant fields located around Ashulia Lake in the study area (Fig. 1). During the environmental sample collection, the whole plants bodies of different crops and vegetables were collected from various plants fields. Soil samples were collected simultaneously with the food plants from the same sampling points within the respective agricultural fields where the plants were being taken. Water samples were collected from the Ashulia Lake water body which was used in irrigation (Fig. 1). All water samples were collected using the precleaned and presterilized HDPE bottles, homogenized, and carried out to the laboratory, filtered, acidified with  $HNO_3$  (pH < 2) and preserved into refrigerator (4 °C) for further analysis. Crops and vegetables samples were collected from the inner part of the agricultural land to eliminate the edge effect. The whole crops and vegetables plant bodies were collected by using plastic bags and the samples were taken to the laboratory, washed by tap water followed by distilled and deionized water. Different plants parts such as roots and edible parts were separated from the whole plants bodies, and they were dried in sunlight for 48 h and then dried in oven at 70–80



Fig. 1 Map of the study area

°C until a constant weight was achieved. The dried samples were ground by an electrical grinder (made of stainless steel) and passed through a 2 mm sieve, and finally, the powdered samples were saved in polythene zip bags and preserved in a refrigerator (4  $^{\circ}$ C) for metal analysis. Soil samples were collected simultaneously with the plants from the same sampling points with 15 cm depth from the surface in each case. After collection, the soil samples were carefully transported

to the laboratory for processing. They were initially dried in sunlight 24 h followed by oven drying at 105 °C for 6 h to remove all the moisture contents. The soil samples were ground by stainless steel made electrical grinder, sieved through a 2-mm nylon mesh to remove large debris, stones, and pebbles. The resulting soil samples were preserved in appropriate sealed containers for further analysis. Total of 180 different environmental samples including food plants, arable soil, and irrigation water were collected and analyzed during the present study. There were 60 samples of food plants (six different crop and vegetable plants: 10 samples of each type of food plants), 60 arable soil samples from six different crop and vegetable plant fields, and 60 samples of irrigation water.

## **Digestion of Crops, Vegetables, and Soil Samples**

Water samples were processed by conventional digestion method. 100 mL of water sample was transferred to a conical flask. 10 mL conc. HNO<sub>3</sub> and 0.5 mL H<sub>2</sub>SO<sub>4</sub> were also added into the flask, and the liquid mixture was heated until the volume reduced to about 5 mL. The resulting mixture was filtered using Whatman no. 41 filter paper, transferred to a HDPE bottle, and diluted to 100 mL with distilled and deionized water and preserved for metal analysis. Wet digestion method was used to process and digest all crops, vegetables, and soil samples [10, 30]. 0.5 g of each soil, crop, and vegetable sample was digested with 14 mL of acid mixture (conc. HNO<sub>3</sub>,  $H_2SO_4$ , and  $HClO_4$ ). The ratio of acids in the mixture was maintained as  $HNO_3:H_2SO_4:HClO_4 =$ 5:1:1 [10, 30]. Each environmental sample with acid mixture was heated on a hot plate until the volume of the mixture reduced to 5 mL. The resulting liquid mixture was cooled to room temperature, filtered to HDPE bottle, and diluted up to 100 mL with distilled and deionized water, which was then sealed and preserved for metal analysis. Blank digestion was also performed following the similar procedure using the same distilled and deionized water used for the dilution or original samples.

# **Quality Assurance and Quality Control**

All reagents and chemicals used in this study were in analytical grades. Glasswares were made up of high-quality quartz glass and steel apparatus were made of stainless steel. Analytical grades chemicals and reagents were purchased from Merck, Germany, and used without any further purifications. Pre-cleaned and pre-sterilized HDPE bottles were used to collect and preserve water and digested crops, vegetables, and soil samples. High-quality polythene bags were used for the transportation and primary preservation of the soil, crops, and vegetable samples. Standard stock solutions of known concentrations of different metals were purchased from Merck, Germany, and appropriate dilute solutions were prepared from the respective standard stock solutions by adding doubled-distilled and deionized water. These standard solutions of the metals with different concentrations were used to make standard calibration curves. Different factors (midpoint standard checks, blanks, calibration curve, and spiked samples) were inspected to ensure quality control, quality assurance, and calibration curve construction. All heavy metal concentrations were measured in milligrams per kilogram of dry weight for soil and plant samples and milligrams per liter for water samples. The standards reference material (SRM 1547) for soil and (CRM 042-050) for plants were used for precision and quality control of the analyses of all elements. Recovery of the metals were found within the range of 98.54 to 99.35%. Some environmental samples (plants and soil) were also analyzed by using the Inductively Coupled Plasma Mass Spectrophotometric (ICP-MS) method to cross-check the analyses of AAS and the corresponding data were well agreed with the results obtained from the AAS method.

# Heavy Metals and Arsenic Analyses in Crops, Vegetables, Soil, and Irrigation Water

Heavy metal and arsenic concentrations in soil, crops, vegetables, and water samples have been assessed by using an atomic absorption spectrophotometer (PerkinElmer Analyst 300, USA). Six different heavy metals (Cu, Fe, Pb, Cd, Ni, Mn) and one metalloid (As) have been investigated in all samples where the heavy metals were determined by using the flame AAS method, and for arsenic measurement, the hydrate generated AAS method was used. The detection limits of different heavy metals and arsenic were 0.006, 0.005, 0.04, 0.016, 0.05, 0.015, and 0.005  $\mu$ g kg<sup>-1</sup> for Cu, Mn, Pb, Cd, Fe, Ni, and As, respectively. The wavelengths used in analyses of heavy metals and arsenic were 324.80, 279.50, 217.00, 228.80, 248.30, 232.00, and 193.70 nm for Cu, Mn, Pb, Cd, Fe, Ni, and As, respectively.

# **Statistical Analysis**

Reliable statistical software was used to analyze the concentrations of heavy metals found in irrigation water, soil crops, and vegetable samples of the study area. Microsoft Excel 2013 was used to calculate the mean, standard deviation, and other health-related parameters. Pearson's correlation package SPSS (version 16.0) was used for source analysis of heavy metals found in different environmental samples.

#### **Bioconcentration Factor (BCF)**

Bioconcentration factor (BCF) sometimes known as transfer factor (TF) provides significant information on the quantities of heavy metals which are transferred from contaminated soil to vegetables/crops or contaminated waterbodies to vegetables/crops. It was first reported by Kloke *et al.* in 1984 [37]. BCF can be expressed as the ratio of metal concentrations in plant and the corresponding metal contents in agricultural soil when the metal transfer from soil to plant is considered. In case of metal mobilization from contaminated water to plant, it is the ratio of heavy metal concentration in plant and that observed in water [33, 38, 39]. The following equations were used to determine the degree of BCF in plants from corresponding contaminated soil and waterbodies:

$$BCF_{soil to plant} = \frac{C_{plant}}{C_{soil}}$$
$$BCF_{water to plant} = \frac{C_{plant}}{C_{water}}$$

#### **Metal Pollution Index (MPI)**

Metal pollution load index represents the total metal content in any specific environmental sample [40]. The concept of MPI was first reported by Usero *et al.* in 1997 [40]. It (MPI) explains the combined effect of pollution in any specific sample exerted by all metals. It can be calculated by using the following equation:

MPI = 
$$(C_1 + C_2 + C_3 + \dots C_n)^{1/n}$$

In the above equation,  $C_1$ ,  $C_2$ ,  $C_3$ , ...,  $C_n$  represent the concentrations of different metals in a specific environmental sample [32, 41]. The higher MPI values indicate the larger heavy metal contents in any specific food item which results in the higher human health risk due to the consumption of that contaminated food item.

#### **Pollution Load Index (PLI)**

Pollution load index (PLI) was defined and reported by Tomlinson *et al.* in 1980 [42]. It is a significant parameter which is used to demonstrate the extents of pollution or degree of contamination in soil system within a study area [42]. PLI can be calculated by using the following equation:

$$PLI = \frac{C_i}{C_i}$$

The terms  $C_i$  and  $C_r$  denote the concentrations of heavy metal in the respective soil sample and the reference concentrations of heavy metal in soil, respectively [12, 31, 42]. Heavy metal threshold concentrations used as reference standards in the present study to determine PLI were 40,000 mg/kg for Fe, 50 mg/kg for Ni, 1000 mg/kg for Mn, 35 mg/kg for Pb, 30 mg/kg for Cu, 0.35 mg/kg for Cd, and 6 mg/kg for As, respectively [42, 43].

#### **Health Risk Assessment Methods**

Health risk assessments provide an efficient way of determining the possible human health risks caused by the presence of excessive level of different heavy metals in specific food items [44]. In the present study, different health risk factors such as THQ, HI, and TCR were determined to investigate the health risks associated with the consumption of the different metal-contaminated vegetable and crop items.

#### **Estimated Daily Intake (EDI)**

The magnitudes of EDI explain the quantity of heavy metals taken up by a person daily through the consumption of metal-contaminated food items. It can be calculated using the following formula:

$$EDI = \frac{C \times FIR \times EF \times ED}{BW \times AT}$$

In the above equation, C represents metal concentration; EF is the exposure frequency (365 d  $y^{-1}$ ); ED represents the exposure duration (30 years for adults and 15 years for children) [45]; BW is the average body weight (adult = 60 kg and children = 16 kg) [30]; AT is average time (ED×EF); FIR is the food ingestion rate of the respective food items. A survey was conducted on 50 families to find the FIR values of different crop and vegetable samples [46, 47].

#### **Target Hazard Quotient (THQ)**

Target hazard quotient sometimes referred as hazard quotient (HQ) is the non-carcinogenic risk caused by any specific metal through the consumption of a particular food item. The following equation is used to determine the magnitudes of THQ for different metals in the crop and vegetable samples [48]<sup>2</sup>

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

The term RfD denotes the oral reference doses for metals, and the level of RfD for different metals was 0.7, 0.02, 0.14, 0.004, 0.04, 0.0005, and 0.0003 for Fe, Ni, Mn, Pb, Cu, Cd, and As, respectively [16, 39, 48–51]. Appearances of THQ value less than 1 (THQ<1) indicate the absence of non-carcinogenic risks for human health through the consumption of food items, THQ equal to 1 (THQ=1) suggests that it is at the boundary level, and THQ data are quite alarming when the values get higher than the 1 (THQ>1) which demonstrate that heavy metal levels in food items pose serious threat of non-carcinogenic risks to human health [16, 52].

#### Hazard Index (HI)

Hazard index (HI) explains the health risks posed by a mixture of metals. The magnitudes of HI less than 1 indicate the absence of non-carcinogenic risks through the consumption of different food items. However, the presence of HI values greater than 1 for metals in crops and vegetables is a matter of serious concern as there must be health hazard issues with the designated food items. The following equations can be used to determine HI [50, 51, 53]:

$$HI = \sum TTHQ$$

 $= TTHQ_1 + TTHQ_2 + TTHQ_3 + \dots + TTHQ_n$ 

Here, the prefixes indicate different food items. TTHQ in the above equation can be determined by using the following equations:

$$TTHQ = \sum THQ$$

 $= THQ_1 + THQ_2 + THQ_3 + \dots + THQ_n$ 

In these cases, the prefixes indicate different heavy metals. It is noted that TTHQ is the sum of THQ values for different metals in individual sample whereas the HI is the sum of TTHQ values for all studied environmental samples [50, 51, 53].

#### **Target Carcinogenic Risks (TCR)**

Target carcinogenic risk (TCR) sometimes referred as lifetime cancer risk is the cancer forming risk caused by individual metal throughout the whole life due to the consumption of different metal-contaminated food species. In the present study, the magnitudes of TCR were calculated based on the carcinogenic slope factor (CFSo) and the equation provided in the USEPA Region III Risk-Based Concentration Table [43]. The magnitudes of carcinogenic slope factor (CFSo) for Ni, Pb, Cd, and As were 1.7, 0.0085, 6.1, and 1.5, respectively [39, 51]. The higher degree of TCR (TCR >1×10<sup>-4</sup>) indicates the greater potential risks of causing cancer in both adult and children from the consumption of various metal-contaminated food items [49, 54, 55]. The equation used to determine the magnitudes of TCR is given below [49, 54, 55]:

$$TCR = \frac{C \times FIR \times EF \times ED \times CFSo}{BW \times AT} \times 10^{-3}$$

#### Results

#### **Assessment of Heavy Metals and Arsenic in Soil**

Total of seven elements were determined in soil of various crop and vegetable fields. The results of different metals analyses in agricultural soil from six different plants fields are presented in Table 1. Iron concentration in soil was observed to be the highest (54,447.04 mg/kg) whereas cadmium concentration was found minimum (6.60 mg/kg) among all the metals studied in the soil samples (Table 1). Soil from rice field accumulated the highest amount of iron which was 54,447.04 mg/kg, and the extent of nickel was 85.47 mg/kg. Maximum arsenic concentration was14.01 mg/kg observed in the green spinach field. The lowest concentration of iron was found to be 11,810.93 mg/kg in soil from the lady's finger field whereas the minimum concentration of arsenic (12.68 mg/kg) was measured in soil of red spinach field and the highest manganese content was also found in the soil of rice field which was 138.03 mg/kg (Table 1). Soil from red spinach field showed the lowest concentration of both manganese (54.33 mg/kg) and lead (8.62 mg/kg) whereas soil from water spinach field accumulated the highest amount of lead (35.23 mg/ kg), copper (89.56 mg/kg), and cadmium (9.69 mg/kg), respectively. The lowest quantities of copper (12.55 mg/ kg) and cadmium (6.60 mg/kg) were realized in soil of brinjal and green spinach fields, respectively (Table 1). Heavy metals concentrations determined in soils from various plant fields were varied significantly among different metals and agricultural fields. Metals pollution in arable soils and their variations in different agricultural plants fields might have occurred from various sources such as emission of untreated toxic effluents from industries, excessive uses of fertilizers, insecticides, pesticides as well as repeated uses of wastewater in irrigation [18]. Metals toxicities and their differences in agricultural soils and the corresponding uptake by plants also depend on soil pH, soil textures, types of soils, soil parent materials, characteristics of pollutants deposited in soil layers from different sources, and soil cation exchange capacity [15, 18].

## Heavy Metals and Arsenic Accumulation in Crops and Vegetables

Six different heavy metals such as Fe, Ni, Mn, Pb, Cu, Cd and one metalloid, As, were determined in different parts of crops and vegetables plants. The results of heavy metals and metalloid analyses in various food plants are presented in Table 2 and Fig. 2. Crops and vegetables plants showed significant variations to accumulate and

**Table 1** Heavy metals and arsenic concentrations (mg kg<sup>-1</sup>) in arable soil collected from different crops and vegetables fields in the study area (mean  $\pm$  SD and range)

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Sample source (plant field) (n)	Sample type	Fe	Ni	Mn	Pb	Cu	Cd	As
Rice (10)	Soil	$54,447.04\pm427.53$ (54,018.52-54,873.58)	85.47±6.07 (79.21–91.34)	$138.30\pm10.88$ (127.27–149.03)	$17.34\pm1.03$ (16.18–18.14)	25.66±1.39 (24.19–26.96)	7.40±0.68 (6.73–8.09)	12.86±1.09 (11.79–13.96)
Brinjal (10)	Soil	$12,252.32\pm167.30$ (12,069.18-12,397.13)	$31.48\pm2.06$ (29.59–33.67)	$123.48\pm11.04$ (112.61–134.68)	$17.05\pm1.32$ (15.63-18.24)	$\frac{12.55\pm1.05}{(11.37-13.38)}$	$6.94\pm0.74$ (6.16-7.63)	$12.84\pm0.92 \\ (11.92-13.75)$
Lady's finger (10)	Soil	$11,810.93\pm126.84$ (11,678.81-11,931.72)	36.41±2.11 (36.18−37.38)	$66.19\pm4.46$ ( $61.74-70.66$ )	27.23±2.33 (24.98–29.63)	$47.48\pm4.37$ (43.19–51.92)	$8.49\pm0.84$ (7.96–9.46)	$13.01 \pm 0.88$ (12.18-13.93)
Water spinach (10)	Soil	$37,288.57\pm403.57$ (36,884.98 $-37,692.12$ )	$61.26\pm3.47$ (57.79-64.73)	82.45±6.03 (76.28–88.32)	$35.23\pm2.30$ (33.23-37.74)	$89.56\pm6.95$ (82.79–96.68)	$9.69\pm0.86$ (8.98–10.64)	$15.00\pm0.79$ (14.38–15.89)
Red spinach (10)	Soil	$\begin{array}{c} 43,921.91 \pm 336.31 \\ (43,560.37 - 44,225.46) \end{array}$	$14.25\pm1.05$ (13.19–15.28)	54.33±3.46 (50.91–57.82)	$8.62\pm0.97$ (7.92-9.73)	$\begin{array}{c} 42.17 \pm 3.59 \\ (38.29 - 45.37) \end{array}$	$8.28\pm0.88$ (7.28-8.94)	$12.68\pm0.75$ (12.02–13.49)
Green spinach (10)	Soil	$29,613.35\pm159.33$ (29,429.66-29,714.22)	$34.54\pm2.49$ (31.99–36.96)	$60.54\pm4.58$ ( $60.02-61.18$ )	$9.06\pm0.89$ (8.19-9.96)	$19.58\pm1.06 (18.39-20.42)$	$6.60\pm0.89$ (5.62–7.35)	$14.01 \pm 0.91$ (13.14-14.95)
Safe limit <sup>a</sup>		40000	50	1000	35	30	0.35	6
Safe limit <sup>b</sup>		NR	60	NR	250	100	0.6	40
Safe limit <sup>c</sup>		NR	50	2000	100	100	3	NR
<sup>a</sup> [43] <sup>b</sup> [66]								

<sup>c</sup>[63] *NR* not reported in the respective references

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le 2 Heavy n	at trace (a)

Table 2 Heavy met	als and arsenic c	concentrations (mg kg <sup>-1</sup> )	in different parts of crc	ps and vegetables plan	ts grown around indust	rial area of Savar, D	haka, Bangladesh (mean <sub>-</sub>	- SD and range)
Plant types $(n)$	Plant organs	Fe	Ni	Mn	Pb	Cu	Cd	As
Rice (10)	Root	$2938.82\pm60.04$ (2893.38 $-3006.89$ )	$103.33\pm6.89$ (97.74–111.02)	126.51±8.65 (117.87–135.16)	$15.19\pm1.14$ (13.92–16.13)	65.35±2.90 (62.27–68.02)	3.66±0.36 (3.27–3.97)	$1.92\pm0.11$ (1.81-2.02)
	Edible part	$509.74\pm20.27$ (489.12-529.63)	$16.83\pm1.34$ (15.55–18.23)	$26.24\pm1.10$ (25.01–27.13)	5.89±0.81 (5.30- 6.82)	$19.73\pm1.08$ (18.93-20.96)	2.75±0.28 (2.43–2.97)	$0.86\pm0.13$ (0.74-0.99)
Brinjal (10)	Root	$1137.41\pm68.76$ (1083.45-1214.83)	$28.43\pm2.47$ (26.38–31.17)	$55.93\pm3.99$ (51.79-59.76)	5.28±0.83 (4.43– 6.09)	$27.07\pm1.08$ (26.21-28.28)	1.86±0.26 (1.57–2.06)	$0.68\pm0.06$ (0.62-0.74)
	Edible part	$150.38\pm 14.75$ (138.07–166.73)	22.71±1.56 (21.18–24.29)	$21.06\pm1.86$ (19.03-22.67)	8.74±0.71 (7.99– 9.39)	$16.51\pm0.85 \\ (15.83-17.46)$	3.05±0.27 (2.78–3.31)	$0.93\pm0.09$ (0.82-0.99)
Lady's finger (10)	Root	$1085.29\pm75.71$ (997.86-1129.22)	$21.84\pm1.75$ (19.86-23.17)	$50.09\pm3.91$ (46.03-53.82)	5.53±0.64 (4.89– 6.16)	21.86±1.28 (20.81−23.29)	3.42±0.38 (3.07–3.82)	$0.75\pm0.06$ (0.70-0.82)
	Edible part	$125.58\pm16.34$ (109.88-142.50)	$30.33\pm3.25$ (26.92-33.39)	$92.99\pm6.25$ (86.59–99.07)	5.83±0.93 (5.10− 6.88)	$14.54\pm0.94 \\ (13.47-15.23)$	2.58±0.40 (2.15−2.93)	$1.33\pm0.11$ (1.26-1.46)
Water spinach (10)	Root	$1072.10\pm42.92$ (1023.87–1106.08)	$36.27\pm2.86$ (33.24-38.91)	$35.92\pm2.22$ (33.85–38.27)	6.35±0.67 (5.74– 7.07)	24.81±1.12 (23.84−26.03)	2.39±0.32 (2.14−2.75)	$1.03\pm0.13$ (0.94-1.17)
	Edible part	432.91±17.19 (414.98–449.26)	$26.16\pm1.38$ (24.78–27.53)	$66.20\pm3.14$ ( $62.93-69.19$ )	8.75±1.00 (8.02– 9.89)	$18.25\pm0.98 \\ (17.14-19.02)$	1.36±0.33 (1.02–1.68)	$0.84\pm0.09$ (0.74-0.92)
Red spinach (10)	Root	$1459.71\pm 69.45$ (1382.83-1517.93)	$\begin{array}{c} 126.88 \pm 9.95 \\ (117.89 - 137.57) \end{array}$	$90.07\pm5.49$ (84.96-95.88)	$26.05\pm1.24$ (24.89–27.36)	23.57±1.33 (22.26–24.92)	1.91±0.23 (1.74–2.17)	$\frac{1.88\pm0.08}{(1.82-1.97)}$
	Edible part	246.23±19.17 (225.27–262.87)	$20.23\pm2.49$ (17.49–22.36)	$374.23\pm7.29$ (367.29–381.83)	7.28±0.96 (6.48– 8.35)	$15.43\pm1.06$ (14.25–16.31)	1.00±0.35 (0.68–1.37)	$1.45\pm0.16 (1.29-1.60)$
Green spinach (10)	Root	$1906.71 \pm 46.41$ (1867.63-1958.00)	$44.77\pm2.66$ (41.83-47.02)	$52.42\pm4.11$ (48.49–56.68)	$15.48\pm1.04$ (14.33-16.37)	$16.43\pm0.84$ (15.83-17.39)	2.59±0.34 (2.31–2.97)	$1.51{\pm}0.09$ (1.41-1.59)
	Edible part	$510.31\pm 25.49$ (486.66-537.30)	$17.66\pm1.55$ (16.12-19.22)	$40.39\pm1.95$ (38.62-42.48)	6.69±0.93 (5.83– 7.67)	$11.20\pm0.90 \\ (10.18-11.89)$	2.23±0.20 (2.02−2.41)	$0.95\pm0.14$ (0.81-1.09)
Safe limit <sup>abcd</sup>		425 <sup>a</sup>	$0.5^{\circ}$	$500^{\mathrm{b}}$	$0.2^{\rm c}$	$20^{d}$	$0.3^{a}$	0.1 <sup>c</sup>
Safe limit <sup>e</sup>		NR	10	500	0.1 - 0.3	10-40	0.05-0.2	0.1
Safe limit <sup>f</sup>		NR	1.5	NR	2.5	30	1.5	NR
<sup>a</sup> [58] <sup>b</sup> [60]								
رتاً: [68] و[70]: [68]								
°[28] fr <u>so</u> i								
<i>NR</i> not reported in the	he respective re	ferences						

translocate different heavy metals in various organs of their bodies such as roots and edibles parts. In most cases, roots of crops and vegetables plants contained higher concentrations of heavy metals and metalloid than their respective edible parts. However, in some cases, metals concentrations were found higher in the edible parts than those observed in roots (Table 2). The highest concentration of heavy metal was recorded for iron (510.31 mg/kg) in green spinach whereas the lowest metal content was observed for arsenic (0.68 mg/kg) in brinjal. Out of seven elements studied, the accumulation of Fe was found to be the highest in all crops and vegetables samples and the lowest metal uptake by food plants was realized with arsenic and the extents of different metals were found to be in the order of Fe>Mn>Ni>Cu>Pb>Cd>As. Among the edible parts of different food plants, the highest concentration of Ni (30.33 mg/kg), Mn (374.23 mg/kg), Pb (8.75 mg/kg), Cu (18.25 mg/kg), Cd (2.58 mg/kg), and As (1.45 mg/kg) was found in lady's finger, red spinach, water spinach, brinjal, and red spinach, respectively. The lowest concentration of Fe (125.58 mg/kg), Ni (16.83 mg/ kg), Pb (5.89 mg/kg), As (0.68 mg/kg), Mn (19.03 mg/ kg), Cu (11.20 mg/kg), and Cd (1.00 mg/kg) was found in lady's finger, rice, brinjal, green spinach, and red spinach, respectively (Table 2).

# Concentrations of Heavy Metals and Arsenic in Irrigation Water

Out of seven elements studied, Fe concentration in irrigation water was found to be much higher than all other metals. The highest concentration of metal in water was 2.9468 mg/L which was observed with Fe and the lowest metal content was recorded for As (0.0022 mg/L). The overall order of different heavy metal concentrations in irrigation water was Fe>Mn>Ni>Cu>Pb>Cd>As. Different metals and As contents determined in all water samples were found to be in range of Fe (0.8678–2.9468 mg/L), Mn (0.1761–1.0697 mg/L), Ni (0.0523–0.9651 mg/L), Cu (0.0757–0.8317mg/L), Pb (0.0640–0.1757 mg/L), Cd (0.0531–0.0908 mg/L), and As (0.023–0.042 mg/L). However, arsenic contents in all water samples were much lower than the concentrations of other metals found in the respective samples.

# Pollution Level and Mobilization of Heavy Metals in Crops and Vegetables

Mobilization of any metal into plants organs from their growth media such as water, soil can be determined by bioconcentration factor (BCF) analysis. The magnitudes of BCF determined for different metals in various food plants are

presented in Fig. 3. In case of mobilization of metals from soil to food plants, the highest BCF value was recorded as 3.207 for Mn in red spinach and the lowest degree of BCF was 0.05 for As in both rice and spinach. Pollution load index (PLI) for studied heavy metals in soil samples collected from different crop and vegetable fields is shown in Fig. 4. The degrees of pollution load index (PLI) of different metals in crops and vegetables plants were found to be varied from 27.69 to 0.054. The highest BCF value was found for Cd in water spinach and the lowest extent of BCF was found for Mn in red spinach. The total pollution effect exerted by all the studied heavy metals into different crops and vegetables species was demonstrated by metal pollution index (MPI) which is depicted in Fig. 5. All the studied crops and vegetables items showed considerable extents of metal pollution index (MPI) which were ranged from 2.17 (found in brinjal) to 2.49 (observed in green spinach).

#### **Human Health Risk Assessment**

Health risk analyses were carried out by determining and evaluating the carcinogenic risks and non-carcinogenic risks associated with the consumption of metal-contaminated crops and vegetables. Total target hazard quotient (TTHQ) demonstrates the total non-carcinogenic risks due to the intake of any polluted crop or vegetable.

# **Estimated Daily Intake of Heavy Metals**

Estimated daily intake (EDI) describes the quantities of heavy metals intake by anybody through the consumption of different food items. In the present study, EDI of metals in both adult and children were investigated, and the corresponding data are presented in Table 3. The results indicated that total estimated daily intake (TEDI) values were much higher than maximum tolerable daily intake of all the metals studied. The magnitudes of EDI for children were found to be higher than those observed with adult for all metals (Table 3). The highest TEDI was recorded for children which was 7460.84 for Fe whereas the minimum value was 15.31 found in adult for As.

#### **Non-Carcinogenic Risk**

Non-carcinogenic risk associated with consumption of different metal-contaminated food items was investigated by determining the target hazard quotient (THQ) for different elements. THQ and TTHQ are the commonly used characteristic parameters for describing non-carcinogenic risk associated with each metal from individual food item. Total target hazard quotient (TTHQ) is the total non-carcinogenic risk exerted by all studied metals due to the consumption of any specific food item. Hazard index (HI) is another significant parameter



Fig. 2 Different heavy metals and arsenic concentrations estimated in various crops and vegetables



Fig. 3 The magnitudes of BCF for different metals in rice, brinjal, lady's finger, water spinach, red spinach, and green spinach, respectively

which can be used to evaluate the health risks involved with the intake of metal-contaminated food species. The magnitudes of THQ, TTHQ, and HI determined for both adult and children for different metal contents in various crop and vegetables are presented in Table 4. A large range of THQ values for different metals were obtained, and the maximum THQ value was recorded for Fe (54.30) in children for rice whereas the minimum value was recorded for Mn (0.03) in adult for water spinach (Table 4). The degrees of TTHQ also showed a large

variation which were in the range of 3.49 to148.47 (Fig. 6). The lowest TTHQ value was obtained for adult in spinach whereas the maximum value was found for children in rice. Hazard index (HI) values were found considerably higher for children than for adult indicating greater risk to children from the intake of contaminated crop and vegetable species.

#### **Carcinogenic Risk**

Target carcinogenic risk also known as target cancer risk (TCR) is the most widely used factor for carcinogenic risk measurement in human health. The results of TCR analysis in both adult and children showed possible carcinogenic risk to both adult and children from the consumption of different crop and vegetable items which were more or less contaminated with heavy metals. The maximum TCR value was recorded with children for Ni in rice which was 0.14130 whereas the lowest value was found with adult for Pb in lady's finger (0.00001). Total target carcinogenic risk (TTCR) values of all studied metals indicate that children are at higher risk for cancer than adult due to the consumption of polluted crop and vegetable species. The extents of TTCR for adults were in the range of 0.00080 to 0.75184 whereas in the case of children TTCR values were varied from 0.00114 to 1.06742. Any magnitudes of TTCR greater than zero



Fig. 4 Pollution load index (PLI) for different metals in soils of various crops and vegetables plants fields





indicate the possible carcinogenic health risks to both adults and children.

# Discussion

Due to scarcity of enough fresh water in many developing countries like Bangladesh, small and progressive level farmers often use untreated wastewater or partially treated wastewater in irrigation of crops and vegetables [56, 57]. Different metal concentrations found in irrigation water were compared with the permissible levels set up by FAO [58], Indian [59], and USEPA [43]. Fe concentrations in all irrigation water samples which were much higher than other metal contents measured in the respective samples, and the results of metals analysis in water were comparable with the data reported previously from similar studies [60]. All concentrations of Mn and Pb in water were found to be higher than permissible limits of India and USEPA with few exceptions. As concentrations in all water samples were found below the FAO standard permissible levels [58], Cd contents in irrigation water were higher than FAO and Indian standard threshold metal values. Appearance of higher cadmium contents in irrigation water bodies might be due to the direct discharge of industry and municipal wastes [60, 61]. All metal concentrations measured in wastewater used in irrigation were varied significantly, and they were found to be in the order of Fe> Mn>Ni> Cu>Pb>Cd>As. All Cu concentrations determined in irrigation water were higher than Indian standard whereas they were observed to be lower than the permissible limits of USEPA and FAO [43, 58]. Excessive appearance of Cu in water might arise from the sewage and industrial wastes. The discharges of tanning wastes could bring As to irrigation waterbodies [61, 62]. As concentrations measured in all water samples were considerably lower in comparison to other metal contents found in the respective source, the results of metal analysis in irrigation waterbody showed a good agreement with the metal data reported previously in literatures from similar studies [54].

All soil samples collected from six different agricultural fields showed the extents of Fe which were highest among the other metal concentrations (Table 1). This might be due to soil structures and parent materials available in soils of the respective plant fields [18]. Appearance of elevated Fe concentration in agricultural soil might also be related to higher Fe concentration in wastewater used in irrigation of crops and vegetables grown in the respective study area. After Fe, the maximum heavy metal contents in soil were observed for Mn which were varied from 54.33 to 138.30 mg/kg in different plant fields and followed the order of rice> brinjal> water spinach>lady's finger>green spinach>red spinach field (Table 1). Elevated Mn content in soil might be correlated to higher abundance of Fe in the respective soil profile systems [18]. All Ni concentrations (34.54 to 85.47 mg/kg) in soil from six food plant fields were observed to be below the permissible value of 50 mg/kg with the exception in rice and water spinach plant fields where the average Ni content was 85.47 and 61.26 mg/kg, respectively (Table 1). The magnitudes of Pb in soil were varied from 8.62 mg/ kg (rice field) to 8.62 mg/kg (red spinach field), and all Pb concentrations measured in agricultural soil of different food plant fields were observed to be below the permissible limit of 35 mg/kg as reported in literature [43]. Pb contents in soil might arise from soil structures, soil parent materials, traffic emission, industrial, and natural activities [10, 15, 18, 25]. Average Cu concentrations in soil of six food plant fields were in the range of 25.66 to 89.56 mg/kg which were considerably higher than the metal threshold limit value of 30 mg/kg [43]. However, Cu contents in soil of brinjal field (12.55 mg/kg) and green spinach plant field (19.58 mg/kg) were observed to be below the USEPA standard permissible limit [43]. Arsenic concentrations in all soil samples collected from six different plant fields were found to be

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Sampre	EUI													
	Cu		Pb		Fe		ïŻ		Mn		Cd		$\mathbf{As}$	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children
Rice	114.70	162.89	31.80	45.16	2008.00	2851.66	83.12	118.04	152.52	216.60	9.14	12.99	4.98	7.07
Brinjal	7.28	7.68	3.85	4.06	66.27	69.93	10.01	10.56	9.28	9.79	1.35	1.42	0.41	0.43
Lady's finger	4.24	6.40	1.70	2.57	36.59	55.25	8.84	13.34	27.09	40.91	0.75	1.14	0.39	0.59
Water spinach	3.97	5.62	1.91	2.70	94.30	133.39	5.70	8.06	14.42	20.40	0.30	0.42	0.18	0.26
Red spinach	4.50	6.79	2.12	3.20	71.74	108.34	5.89	8.90	50.76	76.66	0.29	0.44	0.42	0.64
Green spinach	55.46	78.76	35.94	51.04	1775.78	2521.87	284.81	404.47	732.31	1039.99	16.06	22.81	6.88	9.78
TEDI	151.66	214.03	54.51	77.09	3290.07	4697.25	145.86	206.47	293.46	421.44	16.27	22.86	7.82	11.05
MTDI	4.50 <sup>a</sup>		0.21 <sup>b</sup>		15 <sup>b</sup>		0.90 <sup>a</sup>		5 b		2.00 <sup>a</sup>		$0.13^{\rm b}$	
<sup>a</sup> [62] <sup>b</sup> [28]														

higher than the standard threshold value of 6 mg/kg, but they were found to be lower than the permissible limit of China [63] (Table 1). Excessive appearances of Cu and As contents in arable soil might be due to the agricultural activities related to the extensive uses of fertilizers, animal manures, and pesticides as well as industrial activities [4, 10, 18, 64, 65]. Cd concentrations in all soil samples collected from various crop and vegetable fields were in the range of 12.68 to 15 mg/kg which were significantly higher than the corresponding permissible limit values obtained from three different sources (Table 1) [28, 43, 66]. Cd is considered as one of the most toxic and mobile elements in terrestrial environment. Higher Cd concentrations in arable soil might be attributed to soil structures, anthropogenic, and natural activities [10, 18, 25]. Among seven elements studied in soil of different food plant fields, the lowest metal concentration in soil was recorded for Cd and the trend of other five metal contents in soil followed the order of Mn>Ni> Cu >Pb>As. Differences in the extents of various heavy metals in agricultural soil might be attributed to the types and chemical forms of metallic species that exist in soil and soil properties and characteristics within the respective area [4, 14, 18, 25]. Metal pollution in soils of arable lands might arise from various anthropogenic sources such as unplanned industrialization, urbanization, and agricultural activities as well as natural sources including rock weathering, parent materials in soil, and mining activities [4, 10, 14, 18, 67]. Fe, Mn, Pb, and Ni concentrations in most of the soil samples collected from different crop and vegetable fields were found to be lower than the recommended permissible levels [43, 66]. There were no established permissible values or threshold values for heavy metals and metalloids in soil and plants in Bangladesh. Therefore, all heavy metals and arsenic concentrations determined in different soil, crops, and vegetable samples during the present study were compared with the appropriate permissible limit values reported by EU, WHO, China, and India [59, 63, 68, 69]. The findings of different metal analyses in soil from different crop and vegetable fields were compared with the metal data obtained from similar studies conducted previously in different countries around the world and the results from the present study showed very good agreement with soil heavy metal data reported in literatures (Table 5) [14, 18, 25, 26].

To determine the toxicity levels of metals in different food plants, all heavy metals and arsenic contents found in different crops and vegetables in the present study were compared with the corresponding permissible limit values reported by FAO/WHO, EU, India, and other sources [28, 58, 59, 68–70]. Fe concentrations in some plants samples were higher than the permissible limit of FAO/WHO [58, 69, 70] (Table 2). Out of seven elements studied, uptake and accumulation of Fe were highest in the edible parts of all crop and vegetable plants which well agreed with the data reported

Table 4 THQ, TTHQ, and HI of different heavy metals and arsenic for adults and children associated with contaminated crops and vegetables

Samples	Adult								Childre	en						
	Cu	Pb	Fe	Ni	Mn	Cd	As	TTHQ	Cu	Pb	Fe	Ni	Mn	Cd	As	TTHQ
Rice	38.23	7.95	2.87	4.16	1.09	18.29	16.59	89.18	54.30	11.29	4.07	5.90	1.55	25.97	23.56	126.65
Brinjal	2.43	0.96	0.09	0.50	0.07	2.69	1.37	8.11	2.56	1.02	0.10	0.53	0.07	2.84	1.44	8.56
Lady's finger	1.41	0.42	0.05	0.44	0.19	1.51	1.29	5.32	2.13	0.64	0.08	0.67	0.29	2.27	1.95	8.04
Water spinach	1.32	0.48	0.13	0.28	0.10	0.59	0.61	3.53	1.87	0.67	0.19	0.40	0.15	0.84	0.86	4.99
Red spinach	1.50	0.53	0.10	0.29	0.36	0.58	1.41	4.78	2.26	0.80	0.15	0.44	0.55	0.88	2.13	7.23
Green spinach	18.49	8.99	2.54	14.24	5.23	32.12	22.95	104.55	26.25	12.76	3.60	20.22	7.43	45.62	32.59	148.47
HI								215.47	HI							303.93

**Fig. 6** The magnitudes of TTHQ in adult and children from the consumption of different crops and vegetables



previously in literature from similar studies (Tables 2 and 5) [71]. Elevated uptake of Fe by food plants might occur due to the contamination in their respective growth media such as soil and irrigation water. The lowest accumulation of element was observed for As in the edible parts of all crops and vegetables plants which was supported by the results of similar studies reported previously in literatures (Tables 2 and 5) [16, 28, 72]. All crops and vegetables showed Ni concentrations which were higher than the permissible levels of India, China, and EU [59, 63, 68]. Ni contents observed in different crops and vegetables in the present study were considerably higher than the metal data reported in literature from similar studies conducted previously in different food plants grown in Barcelona, Spain [16]; Beijing, China [72]; and Lahore, Pakistan [73] (Table 5). Appearances of higher nickel contents in the edible parts of crops and vegetables plants might be attributed to the continuous discharging of effluents from battery industry, nickel-plated jewelry industry, machine parts industry, steel, wire, and electrical part manufacturing industries into surrounding waterbodies and arable lands. Mn contents in all crops and vegetables samples were found to be higher than WHO/FAO permissible limit values [58, 69, 70] (Table 2). Comparison of the results

of Mn analysis in crops and vegetables with the data cited in the literatures from previous studies showed that Mn concentrations in different food items from the present study were higher than those observed in similar plant species grown in Lahore, Pakistan [73], but they were lower than those measured in different plant species grown in Mojo, Ethiopia [28]. Manganese might be transported into different crops and vegetable plants from soil and irrigation waterbodies which were being affected by toxic effluents emitted from manganese alloy production industry, ceramics production industry, and glass manufacturing industry. Application of manganese-containing agrochemicals, aluminum can production, and welding activity might also have caused the excessive emission of Mn into nearby environment. All the crops and vegetables showed Pb concentrations which were higher than FAO/WHO [58, 69, 70] and Indian [59] threshold values, and the findings of Pb analysis in different food plants were highly comparable to the results of similar studies conducted previously by other research groups in different countries [28, 49]. House painting, plumbing with lead pipes, lead bullets, and lead storage batteries are frequently used nowadays which might cause the Pb pollution in irrigation waterbodies and arable lands from where crops

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Study area	Heavy metal c	concentrations (mg	g/kg)												References
(count y)	Fe		Ni		Mn		Pb		Cu		Cd		As		
	Crops and vegetables	Soil	Crops and vegetables	Soil	Crops and vegetables	Soil	Crops and vegetables	Soil	Crops and vegetables	Soil	Crops and vegetables	Soil	Crops and vegetables	Soil	
DEPZ area (Bangla- desh)	328.27 (125.58– 510.31)	31,555.69 (11,678.81– 54,873.58)	24.65 (14.30– 49.00)	43.90 (13.19– 91.34)	70.84 (19.61– 174.23)	87.55 (50.91– 149.03)	6.86 (5.47– 8.75)	19.09 (7.92– 37.74)	14.52 (9.54– 19.73)	39.50 (11.37– 96.68)	2.17 (1.00– 3.05)	7.90 (5.62– 10.64)	1.04 (0.78– 1.45)	13.40 (11.79– 15.89)	This study
Mikail Stream Basin (Turkey)	NR	NR	NR	57.00 (20.00– 137.00)	NR	NR	NR	16.00 (10.00– 27.00)	NR	41.00 (6.00– 90.00)	NR	0.26 (0.003- 1.09)	NR	5.98 (0.44– 18.43)	Aytop et al. (2023) [7]
Khyber Pakh- tunkhwa (Pakistan)	NR	NR	3.51	450.00	NR	NR	0.40	23.00	NR	NR	0.27	2.70	0.20	2.00	Nawab et al. (2019) [14]
Faisalabad, Punjab (Pakistan)	NR	NR	0.04	0.68	NR	NR	0.19	13.4	0.02	2.34	0.06	3.21	NR	NR	Ghani et al. (2022) [26]
Khyber Pakh- tunkhwa Province (Pakistan)	NR	NR	3.62	38.00	NR	NR	0.52	3.21	NR	NR	0.26	0.70	0.22	0.43	Nawab et al. (2019) [14]
Amik Plain (Turkey)	NR	28,446.00 (10,341.00– 46,246.00)	NR	273.00 (43.00– 884.00)	NR	607.00 (278.00– 1002.00)	NR	36.00 (1.00– 65.00)	NR	30.00 (12.00– 52.00)	NR	0.20 (0.01 - 0.84)	NR	2.60 (0.01– 22.70)	Aytop et al. (2023) [18]
Beijing (China)	NR	NR	$\begin{array}{c} 0.053 \\ (0.001 - \\ 1.689) \end{array}$	NR	NR	NR	$\begin{array}{c} 0.046 \\ (0.001 - \\ 0.655) \end{array}$	NR	0.51 (0.024– 8.25)	NR	$\begin{array}{c} 0.010 \\ (0.001 - 0.101) \end{array}$	NR	$\begin{array}{c} 0.013 \\ (0.001 - \\ 0.479) \end{array}$	NR	Bo et al. (2009) [ <mark>72</mark> ]
Tamale Municipal (Ghana)	3.71 (3.04– 4.47)	NR	NR	NR	0.17 (0.06– 0.95)	NR	BDL (BDL- 0.04)	NR	0.07 (0.04– 0.09)	NR	0.05 (0.01– 0.07)	NR	NR	NR	Ametepey et al. (2018) [27]
Mojo (Ethiopia)	287.78	43,918.33	2.99	33.04	164.72	1730.00	5.60	36.87	12.85	25.73	1.06	5.03	3.83	24.28	Gebeyehu et al. (2020) [28]
Barcelona (Spain)	NR	NR	0.06	NR	2.25	NR	0.08	NR	1.33	NR	0.01	NR	0.0003	NR	Margenat et al. (2019) [ <b>16</b> ]

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NR not reported in the respective references

and vegetable plants might uptake and distribute elevated quantities of Pb in their different body parts. Automobile emission might also be the possible source of Pb pollution as the study area was located in the vicinity of a highway. Cu contents found in all crops and vegetables plants were below the tolerable limits of EU [68] and India [59]. However, the findings of Cu analysis in the experimental crop and vegetable species were comparable with the Cu accumulation data reported previously in literatures from several similar studies conducted in different countries such as Ethiopia, Pakistan, China, and Ghana [26–28, 71, 72] (Table 5). With some exceptions, Cd concentrations in all crops and vegetables were found to be higher than the tolerable limits of FAO/ WHO [58, 69, 70] and India [59] (Table 2). The results of Cd analysis in crops and vegetable plants were highly comparable to the Cd data reported previously in literatures from other research groups who conducted their studies in similar crops and vegetable plants [27, 74] (Table 5). Copper and cadmium might have introduced into the food plants from their excessive presences in the respective polluted waterbodies and arable lands due to the continuous discharge of effluents from different copper pipe manufacturing industry, cables, wires, copper cookware, plating, rayon, pesticides, paints, pigment paints, pigment alloys, coatings, and batteries as well as plastic manufacturing industries in the respective areas.

In most cases, the uptake and accumulation of heavy metals and arsenic were much higher in roots in comparison to the edible parts of crops and vegetables plants bodies which was expected as plants roots have direct contact with water and soil. Any contamination in plant growth media such as water and soil could severely impact the nutrient contents in their roots and their subsequent translocation in other parts of plant bodies [10, 75, 76]. However, in some cases, the opposite trend was found where the edible parts showed higher deposition of metals than the respective roots of the plant bodies (Table 2). In the case of lady's finger plants, Mn, Pb, Ni, and As concentrations were considerably higher in the edible parts in comparison to those observed in the respective roots. Mn and Pb contents in the edible parts of water spinach plants were much higher than the respective metal level found in roots. Significantly higher amount of Mn was detected in the edible parts of red spinach plants which was three times more than those observed in the corresponding root system (Table 2). Similarly, brinjal plants showed higher accumulation of Pb and As in their edible parts in comparison to the respective element concentrations measured in the roots. Plants could differ in their abilities to uptake different metals or metalloids from their growth media and to translocate them in the different organs of their bodies [77-79]. Mobilization and translocation of Cd and As from the roots to edible parts were much higher than other heavy metals irrespective of the crops and vegetable plant species studied in the present investigation. The uptake, accumulation, and translocation patterns of different heavy metals and arsenic found in various parts of the crops and vegetable plants in the present study were highly comparable to the results of metal analyses in similar plants grown in other countries (Table 5) [77–80].

The magnitudes of BCF for different metals were highly varied among various crops and vegetables species which might have occurred due to the variations in metal concentrations in soil of the respective plant fields as well as the differences in the uptake and accumulation capacity of different crops and vegetables species [25, 26, 78, 81]. According to the guidelines of WHO/FAO [58, 69, 70], heavy metal contamination may occur if the BCF values exceed 0.2. For rice, the highest and lowest BCF values were recorded for Cu and Fe respectively which indicate that this food item has higher tendency of accumulating Cu in comparison to Fe from the similar sources and the BCF contents of other elements followed the order of Pb>Cd>Mn>Ni>As (Fig. 3). Accumulation and mobilization of Ni in green spinach were higher (1.25) which indicated the greater risk of Ni pollution in rice grown in the respective study area and the pattern of BCF of different elements was in the order of Ni>Mn> Pb> Cu> Cd> As>Fe. Similarly, the magnitudes of BCF for various heavy metals and As in brinjal followed the order of Cu>Ni> Pb> Cd> Mn>As>Fe whereas in the case of lady's finger the degree of BCF contents for different elements was in the order of Mn> Ni>Cu> Cd>Pb>As>Fe, and for the water spinach, the order was observed as Pb> Cu> Mn> Ni>Cd> As>Fe (Fig. 3). The uptake and translocation trends of different heavy metals and arsenic were highly differed among the types of elements as well as crops and vegetables plants studied which was revealed from the differences in the orders of BCF. These findings suggest that there were significant potential differences among the affinities of different crops and vegetables for the uptake and deposition of various heavy metals in their characteristic organs. Higher level of heavy metal uptake by different crops and vegetables might be related to the plant types, growth of plant species, their potential abilities to uptake and distribute nutrients from their growth media such as soil and water [33, 82, 83]. Metals uptake by plants from agricultural soil body greatly depends on soil properties, soil pH, soil cation exchange capacity, parent materials in soil [4, 10, 14, 18, 25, 26, 33, 67].

Pollution load index (PLI) is a significant parameter which demonstrates the level of soil pollution regarding the excessive presence of metals and metalloids. The degree of PLI suggests the sustainability for crops and vegetables production and its preferable magnitudes are considered as PLI<1 [83]. In the present study, the magnitudes of PLI for different metals were greater than the recommended values (Fig. 4) which apparently indicated that soil from various crop and vegetable fields were highly contaminated with Cd as well as moderately polluted with the excessive presence of As and Cu. PLI analysis data for other metals suggest that soil of arable lands in the respective area was relatively safe from Mn, Pb, Ni, and Fe pollution (Fig. 4). On the other hand, MPI analysis data revealed the combined effects of all seven elements (Fe, Ni, Mn, Pb, Cu, Cd, and As) for their uptake and accumulation in each of the crops and vegetable plants and MPI data were found to be significantly varied among different food plant species (Fig. 5). Total estimated daily intake (TEDI) analysis effectively determines the exposure level of heavy metals associated with the consumption of different metal-contaminated crop and vegetable items [71]. The results of EDI and TEDI analyses indicate that both adults and children are at higher risks due to the consumption of metal-contaminated food items (Table 4). The TEDI data associated with adults for different metals were observed as Cu (47.9), Pb (449.9), Fe (348.8), Ni (491.4), Mn (218.2), Cd (16.5), and As (117.8) which were 2.8% (Cu), 26.6% (Pb), 20.6% (Fe), 29.1% (Ni), 12.9% (Mn), 1.0% (Cd), and 7.0% (As) of total heavy metal intake from the consumption of contaminated crops and vegetables. Similarly, in the case of children, the magnitudes of TEDI for different metals were observed as Cu (67.8), Pb (638.2), Fe (497.4), Ni (697.7), Mn (311.7), Cd (23.3), and As (167.1) respectively which were 2.8% (Cu), 26.6% (Pb), 20.7% (Fe), 29.0% (Ni), 13.0% (Mn), 1.0% (Cd), and 7.0% (As) of total heavy metal intake from the intake of respective crops and vegetables. It is noticeable that the percentages of total heavy metal intake for different metals in crops and vegetables are almost identical for both children and adults. The findings of EDI and TEDI analyses for both adult and children for their intake of seven elements from different crops and vegetables were comparable with the results of similar studies conducted previously by other research groups [6, 13, 35]. The magnitudes of THQ for Cu and Pb were found higher than the acceptable value 1 for both adult and children in all the studied crops and vegetables. THQ data obtained from the present study were much higher than those reported from similar studies conducted previously by several research groups [16, 45, 48]. Fe and Ni contents in all crops and vegetables showed relatively lower non-carcinogenic health risks to both adult and children as revealed from THQ data which were found below one. Exception was observed with green spinach where the THQ value was higher than 1 suggesting the possible health risk to both adult and children. THQ values for Mn with both adult and children were shown below 1 for most of the crop and vegetable items with the exceptions in rice and green spinach. THQ contents of Cd and As showed non-carcinogenic risk to adult and children for almost all the food items whereas the results of THQ were comparable with the findings of similar studies reported previously in literatures [16, 84, 85]. It has been apparent that rice and green spinach exerted the highest non-carcinogenic risks for both adult and children than other vegetables as revealed from the magnitudes of THQ (Table 4). The results of overall TTHQ and HI analyses indicated higher non-carcinogenic risks to both adult and children due to consumption of the different contaminated crops and vegetables items (Table 4 and Fig. 6). TCR data of metals in crops and vegetables were found in the range of 0.00001 to 0.48417 whereas the magnitudes of total target carcinogenic risk were varied from 0.00114 to 1.06742 which demonstrated the possible target cancer risks to both adults and children. Most of the target cancer risk (TCR) values for different metals in the experimental food plants were found higher than the recommended permissible levels [30]. The results of TCR analysis showed that children are at higher carcinogenic risk than adult. The TCR data obtained from the present study were more or less similar to the findings of previous studies as reported in literatures [74, 86]. Assessment of total target carcinogenic risks (TTCR) suggested that As, Pb, Ni, and Cd in crops and vegetables showed higher carcinogenic risks to both adult and children. Previous studies on TTCR analyses of metals in food plants also provided similar information regarding carcinogenic risks to human health as observed in the present investigation [74, 87, 88].

These significant research findings will potentially benefit the Department of Environment of Bangladesh and other developing countries to formulate necessary environmental policies and guidelines for industries as well as agricultural activities which may eventually reduce metal pollution in food chain and thus can save the health of millions of people. Higher magnitudes of BCF, the corresponding TCR values and  $THQ \ge 1$  of metals found from the present study suggested that crops and vegetable plants should not be cultivated in the contaminated study areas. The interesting results of this study indicate that all industries in Bangladesh should be equipped with proper effluent treatment plants (ETP) and they should operate those ETP units during the entire period of the manufacturing processes. These research findings will create necessary awareness among people about consequences of metal pollution in food chain and related carcinogenic as well non-carcinogenic health risks associated with the consumption of different metal-contaminated crop and vegetable items. Continuous monitoring of industrial activities should be performed by the Department of Environment (DoE) to minimize the discharge of untreated industrial effluents into surrounding environment. The outcome of this study emphasizes that long-term monitoring and evaluation of metal pollution in crops, vegetables, and other food plants grown in different areas of the country are highly necessary. Further study on the detailed impacts of metal toxicities on human health from the consumption of metal-contaminated food items can be considered in the near future. The interesting results of the present investigation may also benefit the researchers and policymakers of many developing countries to understand and evaluate the sources, mobilization patterns of metal toxicities into food chain, and their ultimate consequences in the health of millions of people who are highly exposed to various contaminated food items.

# Correlation Analysis of Heavy Metals in Crops and Vegetables

Pearson correlation analysis was performed to visualize the inter-relationship among the heavy metals' contents found in soil as well as crops and vegetables studied in the present investigation. This correlation analysis provided significant information about the source metals in arable soil and the corresponding accumulation properties of heavy metals in different food plants [85]. A significant correlation between heavy metal concentrations suggests that they have similar or almost identical accumulation properties. It also provides information on common sources of the respective elements [89, 90]. In the present study, Fe contents in agricultural soil of six crop and vegetable fields were observed to be positively correlated with Ni, Mn, Pb, Cu, Cd, and As (r > 0.1; p < 0.01), indicating their common availability in soil which might be originated from similar sources such as soil properties, parent materials in soil profiles, agricultural and industrial activities [4, 10, 14, 18, 33, 67]. Ni concentrations in soil were positively correlated to Mn and Pb (r > 0.2; p < 0.05). Similarly, Mn contents in soil were observed to be correlated with As and Pb measured in the respective environmental samples (r > 0.1; p < 0.01). Pb contents determined in soil were positively correlated to Cu, Cd, and As (r > 0.5; p < 0.05)suggesting that they might have appeared in arable soil from similar sources and activities [4, 14, 18, 33]. Similarly, the extents of Cu in soil showed positive correlation with Cd and As (r > 0.4; p < 0.05). However, negative correlations were found among Ni, Cu, Cd, Cd, As as well as Mn, Cu, Cd in soil collected from six different crop and vegetable fields. The positive collections among the metal contents in agricultural soil apparently indicated that they might have originated from identical sources like industrial emission, agricultural activities, soil textures, soil parent materials available in the study area [4, 10, 14, 18, 33, 67]. In most cases, negative correlations were found among the different metal contents found in the crops and vegetable plants. However, a strong positive correlation was observed between the extents of Mn and As measured in all crops and vegetables (r > 0.7; p < 0.01), which might be due to their coexistence with similar chemical forms in the respective plant growth media. The uptake of metals by plants from different sources is greatly dependent on plants characteristics including plants types, plants growth patterns, availability of nutrients in their growth media [6, 10, 12, 16, 33]. Plants are found to be highly differed in their abilities to uptake and accumulate metals from their growth media as well as translocate them in different organs of their bodies [6, 10, 12, 16, 33].

# Conclusion

Herein, different heavy metals (Fe, Ni, Mn, Pb, Cu, Cd) and arsenic contamination profiles in various crops and vegetables plants grown in industrially polluted areas have been demonstrated with the evaluation of associated health risks to adult and children. Elevated uptake and mobilization patterns of various heavy metals in roots and edible parts of various crops and vegetables plants have been investigated. Heavy metal analyses were also carried out in plants growth media such as soil and irrigation water. Transfer and translocation patterns of these heavy metals from water and soil to different parts of crops and vegetables plants bodies were analyzed effectively. Soil of different crop and vegetable fields were found to be polluted with Cu, Cd, and As whereas the excessive presence of Mn and Cd was observed in irrigation water. Accumulation of metals was found to be considerably higher in roots than edible parts of the respective food plants bodies. However, the translocation and deposition of some metals were observed to be higher in the edible parts which were Ni in brinjal; Mn in lady's finger, water spinach, and red spinach; Pb in brinjal, lady's finger, and water spinach; Cd and As in brinjal respectively which revealed the differences in the abilities and choices of food plants to uptake and mobilize different metals in the various parts of their bodies. Ni, Pb, Cd, and As contents in the edible parts of all crops and vegetables plants were significantly higher than the corresponding permissible limit values. Excessive appearances of Ni, Pb, Cd, and As in the edible parts of different crops and vegetables are quite alarming regarding the food safety and health issues. Pollution characteristic analysis by PLI revealed the metal pollution in soil (PLI > 1) whereas the magnitudes of MPI greater than one apparently indicated the metal contamination in the crops and vegetables species. Human health risk analyses by EDI, THQ, HI, and TCR showed the possible non-carcinogenic as well as carcinogenic risks to both adult and children from the consumption of different metal-contaminated crops and vegetables grown in the study areas. TTCR of metals have been found in the range of  $1.14 \times 10^{-3}$  to 1.06742 for children and  $0.8 \times 10^{-3}$  to 0.75184 for adult indicating that children are at higher carcinogenic risk than adult due to metals contamination in crops and vegetables. EDI of all heavy metals and arsenic in both adult and children from the consumption of metal-contaminated food items were much higher than the corresponding recommended levels. TTHQ data of metals determined for adult (3.53-89.18) and children (4.99–148.47) as well as hazard index (HI) values of metals for adult (215.47) and children (303.93) revealed that children are at higher threat of non-carcinogenic risks than adults from the intake of polluted crops and vegetables species. The significant findings of this study reflect the qualities and heavy metals characteristics in crops and vegetables grown in the vicinities of industrials areas with possible health risks of millions of people in developing countries like Bangladesh. These interesting results may be highly useful for making different environmental policies to control heavy and toxic metals pollution in food plants with the management of industrial effluents as well as food safety in many developing countries. The present investigation mainly focused on the accumulation, mobilization, and translocation of heavy metals and arsenic in different parts of six crops and vegetables plants grown in contaminated soil and water as well as assessed the corresponding human health risks. Further study on soil types, soil textures, and soil profiles in the agricultural lands of the study area is under active consideration which will provide better understanding of metal pollution in soil system. Future study on seasonal variations of metals pollution in all crops and vegetables grown in the contaminated areas is also necessary to save the health of millions of people in the respective areas. Regular monitoring of heavy and toxic metals status in crops and vegetables grown around different industrial areas of Bangladesh is needed in an urgent basis. The Department of Environment should formulate appropriate policies and guidelines and implement them to minimize the metal pollution in human food chain.

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#### Declarations

Consent for Publication Not applicable.

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