Contamination of Irrigation Water, Soil, and Vegetables by Heavy Metals in a Multi-industry District of Bangladesh

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Abstract

Heavy metal (Cr, Cu, Zn, As, Cd, and Pb) concentrations in irrigation water, soil, and vegetables were investigated to assess the contamination levels of farmland adjacent to a multi-industry district in Bangladesh, and their seasonal differences were examined. In the district, many factories of textile, dyeing, ceramics, paints, and agrochemicals, etc. exist and they release wastewater into the nearby irrigation canals. As a result, the heavy metal concentrations in irrigation water and vegetables were found to exceed permissible levels. While those of the soil was mostly lower than the permissible level. The Banglabazar, Kashimpur, and Chandra areas are heavily polluted with wastewater. The large concentrations of Pb and As in Banglabazar are from battery and metallurgy factories located in the area that discharge wastewater containing a lot of Pb. The Kashimpur area is highly contaminated with Cr and Cd, which are used to add color and pigment to garments, plastics, and agrochemicals. The Chandra area receives wastewater from fabric printing, agrochemical, pharmaceutical, poultry feed, and fish feed factories, which creates high Zn and Cu concentrations in the area. According to the several representative indices of soil contamination, soil of the study areas was assessed to be between uncontaminated and moderately contaminated. Among vegetables, the concentration was the highest in root vegetables. The highest concentration occurred in Zn and the lowest in Cd for irrigation water, soil, and vegetables, respectively. Every concentration was lower in the wet season than in the dry season for irrigation water, soil and vegetables. The lower concentration in irrigation water in the wet season was caused by the dilution of the water for rainfalls in the season. The lower concentration of soil in the wet season was due to the low absorption of heavy metals from the diluted irrigation water. The lower concentration of vegetables in the wet season was due to the lower absorption of heavy metals from the lower concentrated irrigation water and/or soil. The hierarchical cluster analysis between vegetables and irrigation water, and between vegetables and soil in heavy metal concentrations revealed that heavy metals of vegetables were absorbed from irrigation water in wet season, while were absorbed from soil in the dry season. For the high heavy metal concentrations in vegetables, a high bio-concentration factor of vegetables (mostly >20 %) might be responsible. Despite that the soil’s concentrations were in permissible level, the high vegetable’s concentrations such as 27 times, 22 times, and 14 times higher than the standard were observed in the concentrations of Pb, Cr, and As, depending on the area. Furthermore, the health risk index was calculated to assess the potential health risk by consuming vegetables. The results showed that the vegetables grown in the study areas were assessed unsafe for human consumption due to high heavy metals concentrations.

CHAPTER 1

General Introduction

1.1 Background of the study and previous studies

In the developing countries, farmland heavy metal contamination is a severe environmental problem due to acute persistence and toxicity level in the ecosystem (Ağca and Özdel, 2014; Khan et al., 2013). In recent decades, heavy metals and metalloids in water, soil and vegetables have highly increased due to variety of anthropogenic activities like the expansion of industrialization and urbanization (Malan et al., 2015; Islam et al., 2017, Chen et al., 2010). More specifically, the urban areas are polluted from industrial discharges (textile, dye, metallurgical, chemical plant, power plant, coal combustion etc.), traffic emissions (vehicle exhaust, brick kilns, brake lining wear, tire wear etc.) and municipal wastes (Du et al., 2015). In agricultural soil, the heavy metals are mainly originated from smelting, mining, textile, dyeing, vehicle exhaust, as well as the application of fertilizers and pesticides (Jiang et al., 2017). Accumulation of heavy metals in soil often causes ecosystem malfunction and soil/water degradation, in addition, toxic metals enter food chains from contaminated water, soil, and air, and subsequently cause food contamination, which poses high health risk to human and animal (He et al., 2015). In the terrestrial environment, plants form one of the major sinks for both essential and toxic elements, contaminated agricultural soil with high heavy metal concentrations would thus lead to the increase in heavy metal uptake by the plants grown in such soils, and this in turn, pose potential hazards to human and animal health caused by ingestion of the plants (Casadovela et al., 2007; Kidd et al., 2007; Bose and Bhattacharyya, 2008; Odlare et al., 2008; Achiba et al., 2009; Delbari and Kulkarni, 2011).

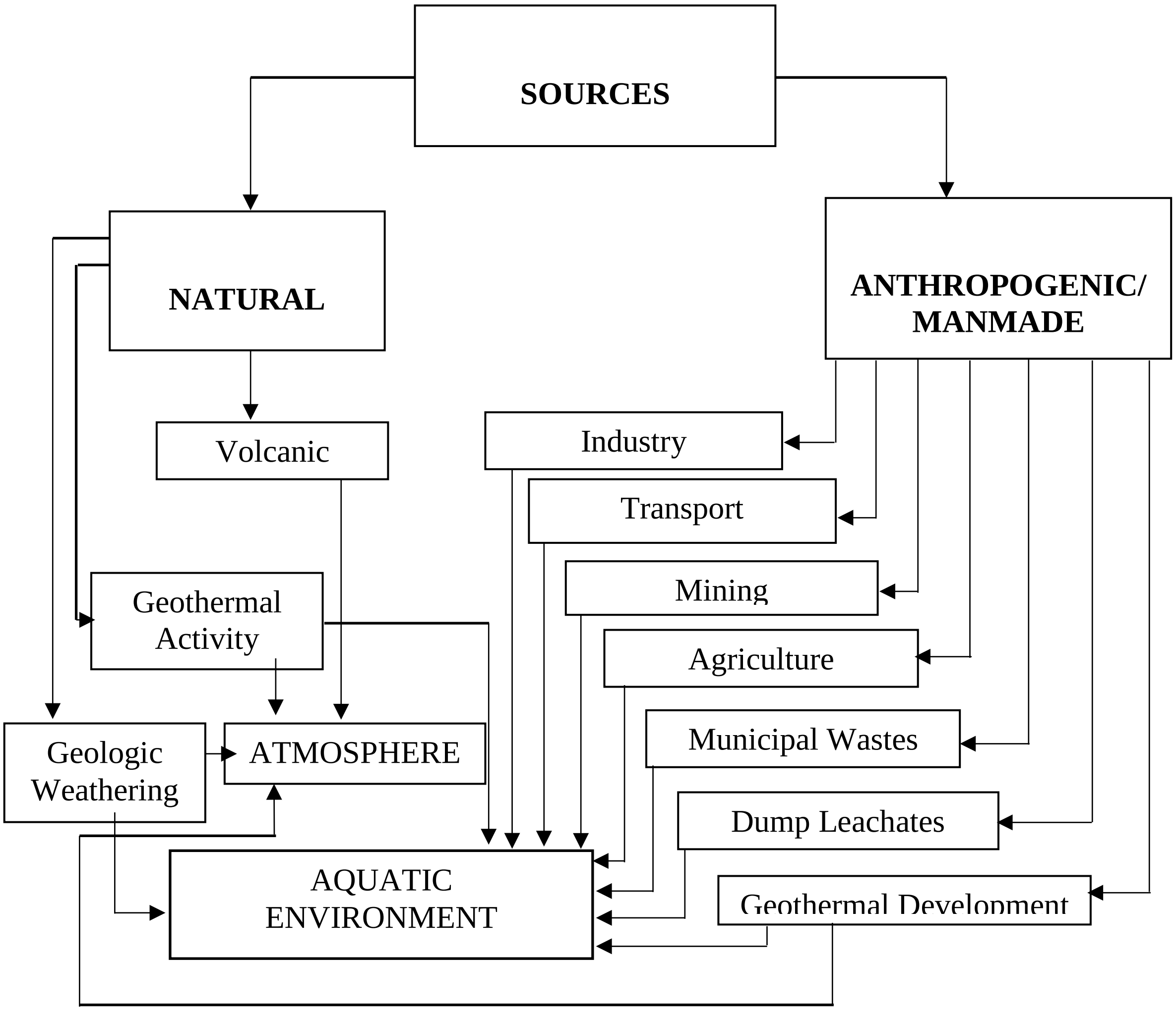

Fig. 1.1 Natural and anthropogenic sources of heavy metals (Foster and Charlesworth, 1996).

The environment may be contaminated with heavy metals as a result of natural activities, as well as from a variety of anthropogenic activities (Fig. 1.1). Weathering (physical, chemical, and biological) and transport processes can release elements from different rock.

The rates of elements released are controlled by numerous factors such as pH and temperature (Dill et al., 2001). The natural sources such as atmospheric deposition of airborne particles come from volcanic eruptions, smoke from forest fire, wind erosion, runoff, acid rain and oceanic spray. Heavy metals are also released from different anthropogenic or manmade activities, such as dumping of metallic wastes, mining activities, wastewater irrigation, sewage sludge, use of chemical fertilizers, manufacturing processes and fossil fuel combustion. The natural sources and sources from anthropogenic activities are shown in Fig. 1.1.

The scarcity of freshwater in the world directed towards the use of wastewater for irrigating the agricultural crops (Li et al., 2016). The irrigation water contaminated with industrial wastes causes significant heavy metal pollution in agricultural soil and crops, ultimately, increase human exposure to the pollutants. Faisal et al., (2014) analyzed the heavy metals (Fe, Mn, Zn, As, Cr, Pb, and Ni) of industrial effluents, river water, and groundwater in Savar industrial area of Bangladesh. The authors showed a multivariate analysis to identify different sources of heavy metals in water samples. Ahmed et al., (2012) reported that the surface water near Dhaka Export Processing Zone (DEPZ), Bangladesh was highly polluted by different heavy metals like Zn, Pb, Cu, Cr, and Ni which exceeded the permissible limit of drinking and surface water (world average). Numerous studies showed the soil and crops enrichment of heavy metals by industrial wastewater in the recent years. For example, Jiang et al., (2014) reported that heavy metal accumulation was potentially high in farmland caused by wastewater irrigation which was higher than fertilizer application and atmospheric deposition. Njoku et al., (2013) investigated soil pollution with zinc (Zn) in the industrial area of Lagos State, Nigeria. The soil was slightly polluted with Zn (84.04±0.02 µg/g) concentration which was higher than the concentration in the nonindustrial sites. They also reported that the concentration of Zn in soil was very much high in the dry season than in the wet season. In Europe, recently Nedelescu et al., (2017) showed the high Pb (566 mg/kg) and Zn (1143 mg/kg) concentrations in soil in an industrial area of Romania, where growing root vegetables (carrot) exceeded the maximum permissible limit for Pb and Cd. In another report, Hossen et al., (2017) collected 26 sediment samples from Tongi canal area, near Dhaka city of Bangladesh and showed a high degree of Zn, Pb, and Cu contamination. These contaminants were sourced from different industrial areas.

Arora et al., (2008) showed that if agricultural soils were irrigated with wastewater, the heavy metal concentrations will be significantly high in the edible parts of growing crops. They also mentioned that consumption of these wastewater-irrigated vegetables resulted in the accumulation of Mg, Zn, Cu, and Fe in the human body. Rahman et al., (2010) investigated the soil and rice plant contamination with heavy metals through industrial effluents in Dhaka, Bangladesh. The results revealed that rice plant was highly contaminated with Zn, Rb, Se, Sc followed by Cr and Cs. In addition, Real and Azam, (2017) collected rice, vegetables, and fish samples to measure Cr, Cd, Pb, As, Mg, Zn, and Ni concentrations in Bangladesh. The results showed that all heavy metals in food items, except Cd and Pb in vegetables, posed significant carcinogenic risks as compared to the EPA recommended level. Goni et al., (2014) revealed the heavy metal concentration and transfer factor were high in arable soil and crops, respectively in Dhaka Export Processing Zone (DEPZ) area of Bangladesh. The authors concluded that absorption of heavy metals by roots were significantly higher than the other plant parts.

Zhang et al., (2017), and Wenzel and Jockwer, (1999) reported that uptake of heavy metal by plants depends on the mobility and bioavailability of heavy metals which usually controlled by soil absorption and desorption features. Luo et al., (2011) showed that high concentrations of Cd (0.79 mg/kg) and Pb (0.38 mg/kg) were found in broccoli and lettuce, respectively, in an e-waste processing site of China due to heavy metal contamination in soil. Gupta et al., (2008) observed the exceeded level of Pb, Zn, Cd, Cr, and Ni concentrations in various vegetables collected from wastewater irrigated area in India. Ahmad and Goni, (2010) stated exceeded level of Zn, Pb, and Cu content in leafy vegetables in wastewater irrigated area in Bangladesh.

Some physiological changes were also observed in plants like the biomass and chlorophyll content of the plants progressively decreased with the increase of heavy metal content in agricultural soils, because of the destruction of marginal membranes of meristematic cells (Li et al., 2010). In another report, Ratul et al., (2018), evaluated that the potential health risk was observed due to the consumption of contaminated water irrigated vegetables in Narayangonj, Bangladesh. They concluded that Cr, Pb, and Ni concentration in vegetables exceeded the permissible limits, however, health risk index values for vegetables were < 1 indicated the absence of health risks associated with the consumption of contaminated vegetables.

1.2 Problem statement

In Bangladesh, food safety is a major health concern, where most of the foodstuffs are thought to be contaminated with carcinogenic (Pb, As, and Cd) and non-carcinogenic (Fe, Co, V, Cu, Zn, Cr, Mn, and Ni) heavy metals and not safe for ingestion (Sultana et al., 2017). Due to the unplanned industrialization and urbanization in the country, heavy metal contaminated wastewater is continuously released into water and soil, and as this result, crops grown there are polluted with heavy metals (Pb and Cd) which are consumed by many people through transportation and retailing of the crops (Ikeda et al., 2000). The consumption of contaminated products arising a serious health concern due to its potential harmful impacts (Sultana et al., 2014). When crops are cultivated in a contaminated soil, heavy metals can be easily accumulated in the edible parts of the crops that causes toxicity not only in crops but also in animals and human body through the food chain (Sharma et al., 2009). Once heavy metals are accumulated in different organs of human body, it is affected with assorted physiological disorders due to their non-biodegradable nature (Duruibe et al., 2007). Central nervous and blood systems, kidney, and bone in the human body could also be damaged by the accumulation of Cd and Pb (Liu et al., 2014). According to the World Bank report, in Bangladesh, about one million people are in a health hazardous situation due to heavy metal contamination especially by lead (The Daily Star, 2018). The report also showed that children are more vulnerable to neurological damage and IQ loss, further, the risk of miscarriage and stillbirth among pregnant women is still alarming.

The farmland of Gazipur District, Bangladesh was selected for the present study, where multiple industries, i.e. textile, dyeing, garment, pharmaceutical, ceramic, paint, packaging industries etc., dispose untreated wastewater into nearby waterbodies and which cause heavy metal contamination to the environment. Farmers use the contaminated irrigation water for planting crops (vegetables), which is the major cause of heavy metal contamination in vegetables in the district. Most of the vegetables are sold to the wholesale market in the capital Dhaka and are consumed by many people. The vegetables may be highly contaminated with heavy metals (Cr, Cu, Zn, Cd, As, Hg, Pd, Fe, and Ni) according to the previous studies (Ahmad and Goni, 2010; Lambert et al., 2000; Barakat, 2011; Sultana et al., 2011). However, the pollution of water, soil, and vegetables were not studied yet in both dry and wet season in a multi-industry area of Gazipur District, and the effect of water and soil contaminations on the vegetable contamination has not also studied yet. Therefore, it is reasonable to hypothesize that the level of heavy metals in irrigation water, soil and vegetables is a level of serious concern in identifying pollution status along with ecological risk and potential health risk to consumers.

1.3 Objective of the study

Therefore, this study intended to clarify the following points:

- ◊ How high are concentrations of heavy metals (Cr, Cu, Zn, As, Cd, and Pb) in irrigation water, soil, and vegetables in the wet and dry seasons in respective areas?

- ◊ How the contamination level of the respective elements is different between the dry and wet season, and among the areas (effect of multi-industry) in irrigation water, soil, and vegetables?

- ◊ How the irrigation water and/or soil affect vegetables in terms of heavy metal contamination in the two seasons, respectively?

- ◊ What are the effects of contaminated soil and vegetables on ecological risk and human health risk, respectively?

CHAPTER 2

Materials and Methods

2.1 Study area

Gazipur District (25°15′0″ N and 89°30′0″ E) is a suburban industrial area locating about 50 km away north from the capital Dhaka. The area is 1,806 km^2 with a population of 3,403,912 (BBS, 2011). The district situated on a Madhupur Tract agro-ecological region of Bangladesh with the dominant soils of deep to shallow red-brown terrace soils. Madhupur Tract has a terrace topography that not always flat but comprises of little curvy hills and ridges divided by a close array of shallow bides (valleys) (BBS, 2011). The soil conditions of this district vary widely and are often complex. The district has a flat lowland topography with an elevation of 4-24 m (Shapla et al., 2015). The soils are acid basin clays with nutrient-poor characteristics, i.e., deficient in organic matter, phosphate, nitrogen and lime (UNDP/FAO, 1988). Common landforms are uplands, dissected terraces associated with broad/ shallow, deep valleys and a dendritic drainage pattern is frequently observed in the district (Huq and Shoaib, 2013). In Gazipur District, the main food crops planted are paddy rice, jute, turmeric, mustard and vegetables (BBS, 2011). According to the Köppen-Geiger climate classification, Gazipur District belongs to tropical wet and dry/savannah climate zone with an annual average rainfall of 2,036 mm (Merkel, 2012). The wet season is from April-October and the dry season from November-March. The annual average air temperature is 25.8°C with the lowest in January (18.8°C) and the highest in August (28.8°C) on averages (Merkel, 2012).

The target areas for the present study in the district are Banglabazar, Kashimpur, and Chandra, where various small-scale factories are located. The main industries are different from area to area, i.e., in Banglabazar, textile, dye, battery, metallurgical and ceramics industries are located; in Kashimpur, plastic, garments and agrochemical industries, and in Chandra, pharmaceutical, agrochemical, fabric printing, poultry feed, and fish feed industries are located, respectively. In the respective areas, industrial wastewater is discharged from factories into nearby irrigation canals throughout the year. The selected industrial areas were compared with the non-industrial area (Bangladesh Agricultural Research Institute) of Gazipur Sadar Upazila from the previous study (Naser et al., 2018). The map of the study areas is shown in Fig. 2.1.

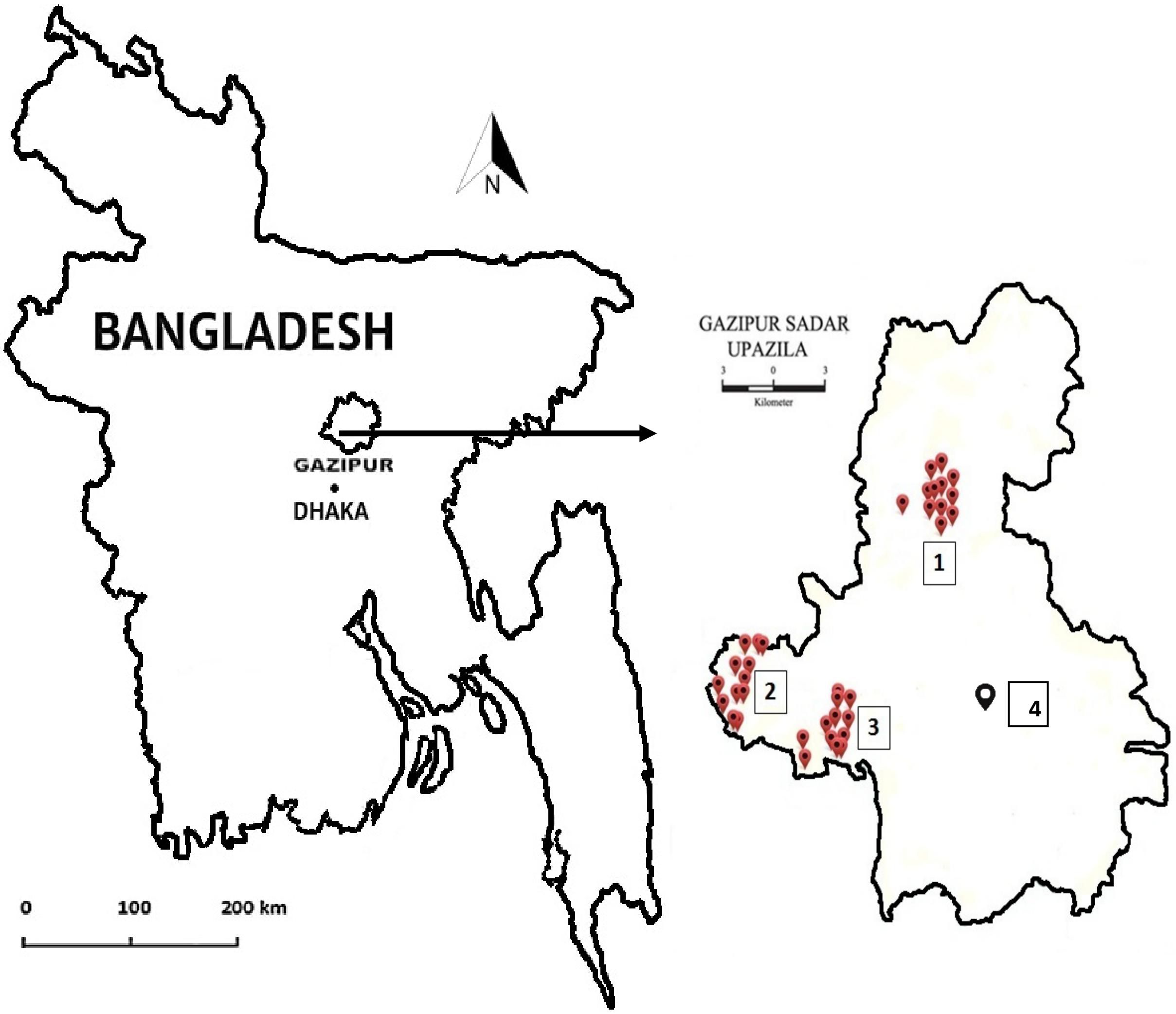

Fig. 2.1 Sampling locations of irrigation water, soil, and vegetables in Gazipur District, Bangladesh. 1. Banglabazar; 2. Chandra, 3. Kashimpur areas, and 4. Non-industrial area.

2.2 Sampling of irrigation water, soil and vegetables

Sampling of irrigation water, soil, and vegetables were carried out in two different times of wet and dry season. The first sampling was done in June-July, 2015 (wet season) and the second one was done in November-December, 2017 (dry season).

The irrigation water was sampled from irrigation canals at 12 points in each three targeted areas i.e. Banglabazar, Kashimpur, and Chandra as described in Fig. 2.1. Totally, the sampling was done at 36 points. In each area, sampling points were selected carefully to represent the situation of industrial location. At each sampling point, 500 ml of irrigation water was taken in an airtight sterilized plastic bottle and carried to the Bangabandhu Sheikh Mujibur Rahman Agricultural University (BSMRAU) laboratory. In BSMRAU laboratory, the water samples were carefully filtered through a paper filter (Whatman No. 42). After filtering, each water sample was stored in a 20 ml plastic bottle, where a drop of concentrated HNO_3 (65%) was added to make the pH <2 for preservation.

Similar sampling method to the irrigation water, soil was sampled from the three areas of Gazipur District. At each sampling point, five soil samples were taken from the surface soil. The sampling depth of soil was 0-15 cm that was decided based on the root depth and most active zone of maximum root concentration, and the most sensitive zone of erosion and atmospheric deposition (Malan et al., 2015; Neagoe et al., 2005). These five soil samples were subsequently added together and mixed thoroughly to make a composite soil sample for each sampling point. After that, the soil sample was packed into a Ziploc plastic bag and carried to the BSMRAU laboratory. In BSMRAU laboratory, the soil samples were broken into small parts to remove the debris like stones, plants etc. Then the soil samples were air dried for at least 7 days. After air drying, the soil samples were ground to fine powder with the help of a mortar and pestle and sieved through a 2 mm sieve. The soil samples were then stored in a Ziploc poly bags in a desiccator until analysis.

Different types of vegetables were selected from each of leaf, root and fruit vegetables for sampling, which are grown in the areas. These vegetables were consumed by the surrounding residents and supplied to the wholesale market of Dhaka capital as well. There are three agricultural seasons in Bangladesh, viz. Kharif 1 (March to July), Kharif 2 (July to October) and Rabi (October to March) (Diary Krishi, 2015). In the wet season (Kharif 1), five kinds of commonly grown summer vegetables were collected from nine vegetable beds in each area, and a total of 27 samples were collected. In the dry season (Rabi), nine commonly grown winter vegetables were collected from twelve vegetable beds, and a total of 72 samples were collected. Vegetables samples were taken from the places where the soil sampling was carried out. As similar to the soil samples mixing, five vegetable samples were mixed together to create a single composite sample for each vegetable species. After making composite samples, vegetables were packed in bags and brought to the BSMRAU laboratory. In BSMRAU laboratory, the vegetable samples were shaken gently by hand to remove soil and washed it with tap water followed by deionized water. The samples were then separated into root, stem, leaf and fruit parts. After that, the samples were air dried to remove the moisture and again packed in transparent Ziploc polyethylene bags for preservation in desiccator. In the analysis, the samples were dried again in a hot air oven at 60 °C for 48 hours until the constant weight was achieved. Finally, the dried samples were uniformly ground with a grinder and stored. The details of sampled vegetables are given in Table 2.1.

Table 2.1 Description of vegetables sampled from the study areas.

Season	**Sl. No.**	**Vegetables name**	**Scientific name**	**Vegetable type**
Wet season	1.	Taro	*Colocasia esculenta*	Leaf and root
	2.	Water spinach	*Ipomoea reptans*	
	3.	Helencha	*Enhydra fluctuans*	
	4.	Eggplant	*Solanum melongena*	Fruit
	5.	Sponge gourd	*Luffa acutangula*	
Dry season	1.	Radish	*Raphanus raphanistrum* subsp. *sativus*	Leaf
	2.	Amaranth	*Amaranthus lividus*	
	3.	Red amaranth	*Amaranthus gangeticus*	
	4.	Spinach	*Spinacia oleracea*	
	5.	Indian spinach	*Basella alba*	

	6.	Bottle gourd	*Lagenaria siceraria*	
	7.	Pumpkin	*Cucurbita moschata*	
	8.	Taro	*Colocasia esculenta*	Leaf and root
	9.	Yard long bean	*Vigna sesquipedalis*	Fruit

2.3 Sample analysis

For the pre-treatment/digestion of soil samples, the US EPA 3050B method (USEPA, 1996) was applied. The air-dried soil samples were mixed thoroughly to achieve the uniformity. From the mixed soil, 1 g (dry weight) was taken into a cleaned vessel for digestion, and 10 ml of 1:1 nitric acid (HNO_3) was added to it. The vessel was covered with a watch glass to reduce evaporation loss. The vessel was heated to 95 °C ± 5 °C on a hot plate and the mixture (soil and nitric acid) was refluxed for 10 to 15 minutes without boiling. The sample was allowed to cool, and 5 ml of concentrated nitric acid (65 % HNO_3) was added for refluxing the mixture again for 30 minutes. This reflux process was repeated many times until the reaction with HNO_3 was completed, which indicate no brown fumes appears from the mixture. The mixture solution was heated again at 95 °C ± 5 °C without boiling for two hours. After that, the solution was cooled, and 2 ml of double deionized water (Milli-Q Millipore 18.2 MΩ/cm resistivity) and 3 ml of 30 % hydrogen peroxide (H_2O_2) were added. The vessel was covered again with a watch glass, and returned it to the hot plate for peroxide reaction. The heating was continued until the effervescence subsides and then the vessel was cooled. After that, 1 ml of 30 % H_2O_2 was added, and the solution was heated again until the sample shows no further change. Finally, the mixture was continuously heated until the volume was reduced to approximately 5 ml. After cooling, the sample solution was quantified to 100 ml by adding double deionized water and then it was filtered with a 4μm paper filter (No. 5B; Advantec, Toyo Roshi Kaisha, Ltd., Tokyo, Japan).

For the pre-treatment/digestion of vegetable samples, the UWLAB method (UWLAB, 2005) was applied. Oven dried vegetable samples were mixed homogeneously, and 0.5 g of the sample was taken into 50 ml test tube. Five ml of concentrated nitric acid (65 % HNO_3) was added to the samples at room temperature for 2-3 hours. After the addition, the test tube was placed in the block heater and heated at 130 °C for 14-16 hours. During the heating, the tube was covered with plastic film to reduce water evaporation. After the heating, the tube was taken out from the block heater and film cover was properly removed. Then the sample was cooled for a few minutes. The mixture solution was heated again by adding 1 ml of 30 % hydrogen peroxide (H_2O_2) for 20-30 minutes and the process was continued two times until the mixture solution shows no further change. Finally, the mixture was cooled and quantified with double deionized water to 50 ml.

2.4 Instrumental analysis

Heavy metal concentrations (Cr, Cu, Zn, As, Cd, and Pb) in the irrigation water and the solutions from soil and vegetable digestion were analysed by inductively coupled plasma-mass spectrometry (ICP-MS; Agilent 7500 ce, Agilent Technologies, USA) at the Center of Advanced Instrumental Analysis, Kyushu University, Japan. The ICP-MS operating conditions were as follows: Nebulizer pump (0.15 rps), RF power (1550 W), RF Matching (1.64 V), Sampling depth (6.6 mm), Torch-H (-0.1 mm), Torch-V (-0.7 mm), Carrier gas (1.15 L/min). The detection limits of the target heavy metals were as follows: Cr, 0.02 µg/L; Cu, 0.03 µg/L; Zn, 0.06 µg/L; As, 0.05 µg/L; Cd, 0.04 µg/L; and Pb, 0.03 µg/L. The multielement standard W-X (Wako Pure Chemical Industries, Japan) for ICP-MS analysis was used to prepare calibration curves for the target elements. External standard solutions and extract solutions were diluted with 0.1 N HNO_3 to fit into the calibration range and reduce matrix interferences. Blank samples (0.1 N HNO_3) were run after every 6 determinations. A 1.0 µg/L multielement tuning solution (Agilent Technologies, USA) was used to cover a wide range of masses of trace elements. Every test was assessed using an internal quality approach and validated if the defined internal quality controls (IQCs) were satisfied.

2.5 Data analysis

Statistical analysis

The basic statistical parameters, and analysis of variance (ANOVA) on the heavy metal concentrations of irrigation water, soil and vegetables were calculated by Excel software (version 2016) and SPSS statistics software (version 21.0). Graphical presentation of data set was performed by Origin pro 8 software (Origin Lab, USA). The *t*-test and hierarchical cluster analysis (HCA) were implemented by SPSS statistics software (version 21.0).

Determination of bio-concentration factor (BCF)

Bioconcentration factor (BCF) is shown by the ratio of the trace metal concentration in plants to the concentration of the pollutant in surrounding environment (Ye et al., 2015). The BCF was calculated with the following equation.

$$\text{BCF}=\frac{\text{Heavy metal concentration in plant tissue (dry wt.)}}{\text{Heavy metal concentration in corresponding soil (dry wt.)}}$$

Following indexes related to the contamination are used.

Geo-accumulation index of soil

The contamination situation of soil can be assessed by geo-accumulation index (Müller, 1981; Ruiz, 2001). This index was calculated by the following equation.

$$I_{geo}=\log_2\frac{\text{CM (sample)}}{1.5 \text{ x CM (Background)}}$$

Where, $CM_{(Sample)}$ is the measured concentration of heavy metal in soil, $CM_{(Background)}$ is the background value (Kabata-Pendias, 2011) for same element and 1.5 is a multiplying factor.

The classification system of geo-accumulation index includes seven classes (Ruiz, 2001) $I_{geo} \leq 0$ uncontaminated, $0 < I_{geo} < 1$ uncontaminated to moderately contaminated, $1 < I_{geo} < 2$ moderately contaminated, $2 < I_{geo} < 3$ moderately to heavily contaminated, $3 < I_{geo} < 4$ heavily contaminated, $4 < I_{geo} < 5$ heavily to extremely contaminated, $5 \leq I_{geo}$ extremely contaminated.

Pollution load index of soil (PLI)

To assess the soil quality, an integrated approach of pollution load index of the heavy metals is calculated for detecting pollution which allows to compare the pollution levels between sites and between different times. The PLI is the multiplications of contamination factor of different heavy metals with the nth root (Islam et al., 2015).

$PLI = (CF_1 X CF_2 X CF_3 X……X CFn)^{1/n}$

Where, CF in contamination factor or single pollution index

Potential ecological risk index (RI)

The potential ecological risk index (RI) assesses the degree of heavy metal contamination in soil on the basis of toxicity of heavy metals and environmental response. The following equations are used to calculate the RI (Guo et al., 2010).

$C^i_f=C^i/C^i_n$

$E^i_r=T^i_rXC^i_f$

$RI=\sum_{i=1}^{n} E^i_r$

Where C^i_f is the contamination factor; C^i is the concentration of heavy metal in the soil; C^i_n is the reference value for the heavy metal (Kabata-Pendias, 2011); E^i_r is the monomial potential ecological risk factor; T^i_r is the heavy metal toxic response factor. The toxic response factors for Cr, Cu, Zn, As, Cd and Pb were 2, 5, 1, 10, 30, and 5, respectively (Guo et al., 2010; Islam et al., 2017). Indices and grades of potential ecological risk of heavy metals are given in Table 2.2.

Table 2.2 Indices and grades of potential ecological risk of trace metals pollution (Luo et al., 2007).

Potential ecological risk factor (E^i_r)	**Grade of ecological risk**	**Potential ecological risk index (RI)**	**Pollution degree**
$E^i_r < 40$	Low risk	$RI < 65$	Low risk
$40 \leq E^i_r < 80$	Moderate risk	$65 \leq RI < 130$	Moderate risk
$80 \leq E^i_r < 160$	Considerable risk	$130 \leq RI < 260$	Considerable risk
$160 \leq E^i_r < 320$	High risk	$RI \geq 260$	Very high risk
$E^i_r \geq 320$	Very high risk		

Daily Intake of heavy Metals (DIM)

The estimated daily intake of heavy metals through vegetables is calculated with the following equation.

$$\mathrm{DIM} = \frac{Cmetal\ X\ Cfactor\ X\ Dfood\ intake}{Baverage\ weight}$$

Where C_{metal}, C_{factor}, $D_{\mathrm{food\ intake}}$ and $B_{\mathrm{average\ weight}}$ represent the heavy metal concentrations in vegetables (mg/kg), conversion factor, daily intake of vegetables (Kg) and average body weight (Kg), respectively. According to the method of Rattan et al., (2005), the fresh to dry weight conversion factor of green vegetables is 0.085. The daily intake of vegetables for adult persons was considered as 0.1673 kg (BBS, 2016). The average body weight of adult person in Bangladesh was considered as 66.50 kg (Khadem and Islam, 2014).

Health Risk Index (HRI)

The health risk index is the quotient between daily intake of heavy metals (DIM) and their reference dose (R_fD), which is calculated as follows (Pierzynski et al., 2000).

$$\mathrm{HRI} = \frac{DIM}{RfD}$$

Where R_fD represents the oral reference doses, which are 0.003, 0.04, 0.3, 0.0003, 0.0005, and 0.004 mg/kg for Cr, Cu, Zn, As, Cd, and Pb, respectively (JECFA, 1993; USEPA, 2007). HRI> 1 means that the exposed population is in a potential health risk.

CHAPTER 3

Concentrations of Heavy Metals in Irrigation Water, Soil and Vegetables and their Seasonal Variations

3.1 Results of the concentrations

3.1.1 Irrigation water

The mean concentrations and range of heavy metals (mg/L) in the irrigation water of dry and wet seasons are shown in Table 3.1. The following characteristics were found in Table 3. In the dry season, out of six heavy metals, Zn had the highest concentration (17.49) in Chandra which ranged from 5.73 to 31.55 in the three industry areas. The lowest mean Zn concentration (12.81) was found in Banglabazar. The Cu showed the highest mean concentrations (6.85) in Chandra followed by Kashimpur and Banglabazar. In the study areas, the concentration of Cu ranged from 0.39 to 15.26. The Zn and Cu concentrations in the wet season showed the same trend with those of dry season i.e. the highest in Chandra and the lowest in Banglabazar, respectively.

The Cr concentration (mg/L) in the study areas ranged from 0.58 to 9.32 in the dry season. The maximum mean concentration (3.20) was observed in Kashimpur area whereas the minimum concentration (1.09) was in Banglabazar. In the dry season, Kashimpur area also revealed the highest mean concentration for Cd (0.023) whereas the lowest (0.012) was found in Banglabazar. The Cd concentration was found to range from 0.001 to 0.084 in all study sites. A similar trend with these Cr and Cd concentrations was found in the wet season i.e. the highest was observed in Kashimpur and the lowest was in Banglabazar, respectively.

Table 3.1 Mean heavy metal concentration of irrigation water (mg/L) and range of concentration (in parentheses) in dry and wet seasons in the three areas.

Season	**Area**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**
Dry	Bang.	1.09	1.81	12.81	0.63	0.012	2.04
		(0.58-1.99)	(0.39-4.36)	(5.73-19.34)	(0.30-1.57)	(0.001-0.041)	(0.69-4.43)
n = 12	Kashi.	3.20	5.15	15.34	0.50	0.023	0.58
in each area		(1.31-9.32)	(1.70-15.02)	(6.71-26.96)	(0.31-0.96)	(0.001-0.084)	(0.13-1.44)
	Chan.	2.13	6.85	17.49	0.44	0.022	0.83
		(0.93-7.88)	(1.39-15.26)	(6.88-31.55)	(0.09-0.75)	(0.001-0.056)	(0.31-2.61)
Wet	Bang.	0.45	1.43	8.6	0.62	0.007	1.75
		(0.17-0.87)	(0.58-2.18)	(7.23-10.4)	(0.25-1.58)	(0.002-0.015)	(0.58-4.14)
n = 12	Kashi.	0.92	4.33	9.86	0.20	0.009	0.35
in each area		(0.44-1.94)	(1.94-10.1)	(8.38-13.5)	(0.06-0.78)	(0.001-0.033)	(0.14-0.52)
	Chan.	0.85	6.04	13.7	0.16	0.009	0.36
		(0.23-1.41)	(1.24-13.30)	(7.51-22.9)	(0.03-0.61)	(0.002-0.031)	(0.25-0.50)
Permissible level [a]		0.1	0.2	2.0	0.1	0.01	5.0

Bang.: Banglabazar; Kashi.: Kashimpur; Chan.: Chandra; *n*: number of samples.

[a] The values show FAO standard (Ayers and Westcot, 1985)

The mean concentration (mg/L) of As (0.63) was the highest in Banglabazar area which ranged from 0.09 to 1.57 in all study areas in the dry season. The lowest mean of As concentration (0.44) was observed in Chandra. Banglabazar area also exhibited the highest Pb concentration in the dry season with the mean value of 2.04 and the lowest concentration (0.58) was found in Kashimpur. In all study areas, the Pb concentration ranged from 0.13 to 4.43. In the wet season, the trend was much more similar with that of dry season i.e. the highest Pb (1.75) and As (0.62) concentrations were found in Banglabazar.

The one-way ANOVA analysis was conducted to compare the mean differences of heavy metal concentrations in irrigation water between the dry and wet season, respectively. The results of one-way ANOVA for irrigation water are given in Table 3.2. The results showed the significant differences ($p < 0.05$) between heavy metal concentrations in the dry season i.e. null hypothesis rejected. The significant variations were also found in the wet season ($p < 0.05$).

Table 3.2 The one-way ANOVA results of heavy metal concentration in irrigation water in the dry and wet season.

Season	**Source of Variation**	**Sum of Squares**	**Degree of Freedom**	**Mean Square**	**F Value**	**Sig.**
Dry	Between Groups	5959.8	5	1191.9	97.6	<0.05
	Within Groups	2564.9	210	12.21		
Wet	Between Groups	3089.1	5	617.8	110.1	<0.05
	Within Groups	1178.2	210	5.6		

3.1.2 Soil

Heavy metal concentrations (mg/kg) in the farmland soil of the study areas in the two seasons are summarized in Table 3.3. A wide difference in soil heavy metal concentrations was observed in the soil in the dry season where the highest mean concentration was found for Zn (186.89) in Chandra followed by Banglabazar and Kashimpur. The concentration of Zn ranged from 82.34 to 290.85 in three industry areas. The lowest mean concentration of Zn (163.55) was found in Kashimpur. In the wet season, Zn concentration (150.7) was also high in Chandra.

The Cr concentration of soil ranged from 32.89 to 112.11 in the dry season. The highest mean concentration of Cr (70.20) was observed in Kashimpur area, whereas lowest mean concentration (65.64) was found in Banglabazar. The mean Cu (85.48) concentration was the highest in Kashimpur area with ranging from 26.80 to 128.28 in the three industrial areas. The lowest mean concentration of Cu (40.94) was found in Chandra area. The similar trend of the mean Cr and Cu concentrations were found in the wet season.

Table 3.3 Mean heavy metal concentration in soil (mg/kg) and the range of concentration (in parentheses) in dry and wet seasons in the three areas.

Season	**Area**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**
Dry	Bang.	65.64	57.10	169.24	16.83	0.338	36.17
		(32.89-86.23)	(36.25-100.33)	(92.72-290.85)	(8.56-19.94)	(0.026-0.582)	(20.87-53.22)
n = 12	Kashi.	70.20	85.48	163.55	11.89	0.278	32.79
in each area		(49.88-112.11)	(50.65-128.28)	(92.30-263.58)	(7.88-18.61)	(0.042-0.655)	(26.30-43.58)
	Chan.	66.78	40.94	186.89	10.53	0.261	34.24
		(49.89-87.78)	(26.80-118.77)	(82.34-261.89)	(5.72-15.05)	(0.098-0.502)	(27.14-45.14)
Wet	Bang.	48.98	28.93	146.5	9.75	0.188	30.79
		(39.51-62.75)	(17.22-38.22)	(85.9-262.8)	(5.01-18.11)	(0.042-1.014)	(25.02-41.68)
n = 12	Kashi.	56.69	40.37	145.2	8.39	0.165	26.32
in each area		(38.35-70.14)	(30.15-50.68)	(86.5-221.6)	(4.43-12.23)	(0.045-0.438)	(20.14-29.22)
	Chan.	51.12	34.50	150.7	6.48	0.185	29.68
		(43.07-56.67)	(21.67-43.96)	(87.6-252.7)	(4.56-9.21)	(0.034-0.356)	(24.49-41.02)
Permissible level [a]		100	100	200	5	1	60

Bang.: Banglabazar; Kashi.: Kashimpur; Chan.: Chandra; *n*: number of samples.

[a] The values show EU standard (Ministry of the Environment Finland, 2007)

The concentration of other heavy metals ranged from 20.87 to 53.22 for Pb, from 5.72 to 19.94 for As and from 0.026 to 0.655 for Cd in the study areas. Banglabazar area exhibited the highest Pb concentration (a mean of 36.17), whereas the lowest Pb concentration (a mean of 32.79) was observed in Kashimpur. The mean As (16.83) and Cd (0.338) concentration was also found high in Banglabazar area. The Chandra site exhibited the minimum mean concentration for As (10.53) and Cd (0.261). In the wet season, the mean highest concentrations of As (9.75), Cd (0.188), and Pb (30.79) were observed in Banglabazar area.

The one-way ANOVA was used to express the differences of the concentrations in different heavy metals of soil between the dry and wet season (Table 3.4). The results revealed that the soil heavy metal concentrations were significantly varied with each other ($p < 0.01$) in the dry season. In the wet season, soil heavy metal concentrations also showed significant variations with heavy metal species at 0.05 level.

Table 3.4 The one-way ANOVA results of the heavy metal concentrations of soil in the dry and wet season.

Season	**Source of Variation**	**Sum of Squares**	**Degree of Freedom**	**Mean Square**	**F Value**	**Sig.**
Dry	Between Groups	694159.8	5	138832	189.5	<0.05
	Within Groups	153819.8	210	732.5		
Wet	Between Groups	513878.9	5	102775.8	222.6	<0.05
	Within Groups	96955.9	210	461.7		

The soil heavy metal concentrations in the industrial areas were found to be high in comparison with those in non-industrial area located near to this study area of Gazipur Sadar Upazila. The latter concentrations were studied by Naser et al., (2018). In non-industrial area, the concentration of Pb ranged from 4.67 mg/kg to 5.07 mg/kg with a mean of 4.9 mg/kg, whereas the mean Cr concentration (12.03 mg/kg) ranged from 11.8 to 12.3 mg/kg in the same area.

In the dry season, the soil Pb concentration (mean value) was about 7.4 times (Banglabazar), 6.7 times (Kashimpur) and 7 times (Chandra) high in the industrial area compared to the non-industrial area. In the wet season, the mean Pb concentration was also high in the industrial area (Banglabazar, 6.3 times; Kashimpur, 5.4 times and Chandra, 6.1 times) than in the non-industrial area.

The Cr concentration in soil was also higher in industrial areas compared to non-industrial area, i.e. 5.5 times (Banglabazar), 5.8 times (Kashimpur) and 5.6 times (Chandra) higher in the dry season, whereas 4.1 times (Banglabazar), 4.7 times (Kashimpur) and 4.2 times (Chandra) higher in the wet season. The mean Cd (0.73 mg/kg) concentration ranged from 0.70 mg/kg to 0.78 mg/kg in the non-industrial area which showed a little higher (2.3 times in the dry season and 3.5 times high in the wet season) concentration compared to the industrial areas. The Cr, Cd and Pb concentrations in the non-industrial area were below the permissible limit of EU.

3.1.3 Vegetables

Heavy metal concentrations of the vegetables in the wet and dry seasons with their permissible limit were shown in Table 3.5. In the dry season, the heavy metal concentration in vegetables (mg/kg) ranged from 1.85 to 65.82 for Cr, from 6.93 to 35.47 for Cu, from 27.39 to 181.04 for Zn, from 0.14 to 8.69 for As, from 0.003 to 1.145 for Cd, and from 0.34 to 21.03 for Pb, whereas the concentrations and ranges were very low in the wet season for Cr (0.56 to 23.60), Cu (4.27 to 67.64), Zn (22.91 to 114.70), As (0.05 to 2.31), Cd (0.01 to 0.74) and Pb (0.78 to 18.04).

In the respective areas, the Chandra showed the highest mean concentration of Zn (66.84 mg/kg), Cd (0.392 mg/kg), and Pb (6.68 mg/kg) in the dry season, whereas only Zn (56.39 mg/kg), was the highest in the wet season in the same area. The mean highest Cr (22.60 mg/kg), Cu (15.66 mg/kg), and As (1.43) concentrations were observed in Banglabazar in the dry season vegetables, whereas the As (1.07 mg/kg), Cd (0.14 mg/kg), and Pb (8.10) concentrations were higher in the wet season. Kashimpur area exhibited the maximum concentration of Cr (8.04 mg/kg) and Cu (23.98 mg/kg) in the wet season only.

The one-way ANOVA was used to show the differences of concentrations in different heavy metals between the dry and wet season (Table 3.6). The results revealed that the difference in heavy metal concentrations was significant ($p < 0.01$) in the dry season. In the wet season, the difference in heavy metal concentrations also showed significant variations at 0.05 level.

Table 3.5 Mean and range (in parentheses) of heavy metal concentration (mg/kg) of vegetables in dry and wet seasons in the three areas.

Season	**Area**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**
Dry	Bang.	22.60	15.69	61.49	1.43	0.334	6.63
		(1.85-65.82)	(6.93-35.47)	(27.86-107.6)	(0.16-8.69)	(0.006-1.076)	(0.34-21.03)
n = 24	Kashi.	14.73	13.01	59.89	1.33	0.299	5.50
in each area		(2.76-46.81)	(7.06-20.68)	(27.39-97.61)	(0.14-5.76)	(0.003-1.053	(0.40-15.56)
	Chan.	20.71	15.66	66.84	1.01	0.392	6.68
		(3.60-62.71)	(7.59-27.58)	(30.53-181.0)	(0.14-2.26)	(0.003-1.145)	(0.36-14.89)
Wet	Bang.	4.34	12.91	54.13	1.07	0.143	8.10
		(0.56-14.07)	(8.31-20.3)	(24.46-114.7)	(0.17-2.31)	(0.025-0.739)	(1.52-18.04)
n = 9	Kashi.	8.04	23.98	44.82	0.63	0.097	5.03
in each area		(1.05-23.60)	(8.51-67.64)	(22.91-85.82)	(0.05-2.03)	(0.011-0.408)	(0.78-11.49)
	Chan.	4.39	12.85	56.39	0.69	0.147	4.98
		(0.58-12.95)	(4.27-18.92)	(35.66-108.4)	(0.07-1.91)	(0.014-0.374)	(2.38-11.01)
Permissible level [a]		1	30	20	0.1	0.2	0.3

Bang.: Banglabazar; Kashi.: Kashimpur; Chan.: Chandra; *n*: number of samples.

[a] The values show FAO/WHO standard (Islam and Hoque, 2014; Sultana et al., 2017; FAO/WHO, 2011)

Table 3.6 The one-way ANOVA results of heavy metal concentration in vegetables in the dry and wet season.

Season	**Source of Variation**	**Sum of Squares**	**Degree of Freedom**	**Mean Square**	**F Value**	**Sig.**
Dry	Between Groups	197401.6	5	39480.3	202.2	<0.05
	Within Groups	83174.9	426	195.2		
Wet	Between Groups	52207.9	5	10441.6	90.6	<0.05
	Within Groups	17980.2	156	115.3		

The cultivated vegetables in the studied industrial areas were found higher in heavy metal concentrations compared with the vegetables of non-industrial area of Gazipur Sadar Upazila (Naser et al., 2018). The observed Pb concentration (mean 0.92 mg/kg) ranged from 0.76 mg/kg to 1.08 mg/kg in non-industrial area, whereas the mean Cr concentration (2.4 mg/kg) ranged from 2.23 mg/kg to 2.52 mg/kg in the same area. In the dry season, the mean value of Pb concentration was about 7.2 times (Banglabazar), 6 times (Kashimpur) and 7.3 times (Chandra) higher in the industrial area than in the non-industrial area. The mean Pb concentration in the wet season was also found to be higher in the industrial area (Banglabazar, 8.8 times; Kashimpur, 5.5 times and Chandra, 5.4 times) followed by non-industrial area.

In the vegetables of industrial area, the Cr concentration exhibited higher values compared to non-industrial area, i.e. 9.4 times (Banglabazar), 6.1 times (Kashimpur) and 8.6 times (Chandra) higher in the dry season, whereas 1.8 times (Banglabazar), 3.4 times (Kashimpur) and 1.8 times (Chandra) higher in the wet season. The mean Cd (0.40 mg/kg) concentration ranged from 0.29 mg/kg to 0.48 mg/kg in the non-industrial area, which only showed higher concentration (1.3 times in the dry season and 4 times in the wet season) compared to the industrial areas. The Cr, Cd and Pb concentrations exceeded the permissible limit of FAO/WHO in the non-industrial area.

3.2 Assessment of heavy metals contamination of soil using representative indices

Heavy metals are natural constituents of soil which concentration varies with the parental materials (Barbieri, 2016). In the study areas, the heavy metal content in soil has been increased due to different anthropogenic activities like waste disposal. Due to these activities, soil health becomes hampared. The only determination of the heavy metals in the upper layer of the soil cannot provide overall indications about the state of soil contaminations, because it does not allow the distinction between natural background value and anthropogenic enrichment (Barbieri, 2016). For assessing soil contamination, some indices are used widely by many researchers (Islam et al., 2017; Islam et al., 2015; Aktaruzzaman et al., 2014). Here, the author described three indices of the geoaccumulation index (Igeo), pollution load index (PLI) and potential ecological risk index (RI) that are representative ones, by which soil contamination is classified into several degrees.

The calculated Igeo in dry and wet season for heavy metals of Banglabazar soil and their contamination intensity are illustrated in Fig. 3.1. In the dry season, the mean Igeo values for Cr (-0.48), Cu (-0.09), Cd (-1.11) and Pb (-0.25) exhibited zero class, indicating uncontaminated soil by these heavy metals. The mean Igeo value of Zn (0.61) exhibited positive values within the range of $0 < \text{Igeo} < 1$, indicating uncontaminated to moderately contaminated soil by Zn. Similarly, the positive Igeo value of As (0.69) representing uncontaminated to moderately contaminated soil by As.

In the wet season, Zn showed the positive mean Igeo value (0.38) within the range of 0 < Igeo < 1 (Igeo class 1) indicating uncontaminated to moderately contaminated soil by Zn. The mean Igeo values of other heavy metals like Cu, Cr, As, Cd and Pb were -0.88, -1.04, -0.15, -2.44 and -0.41, respectively, were within the range of Igeo ≤ 0 indicates uncontaminated soil by these heavy metals. For the differences between dry and wet season, the Igeo values in the wet season are significantly lower than the dry season for the respective heavy metals according to the paired t-test ($t = 2.8$–6.0; df = 11; $p < 0.05$).

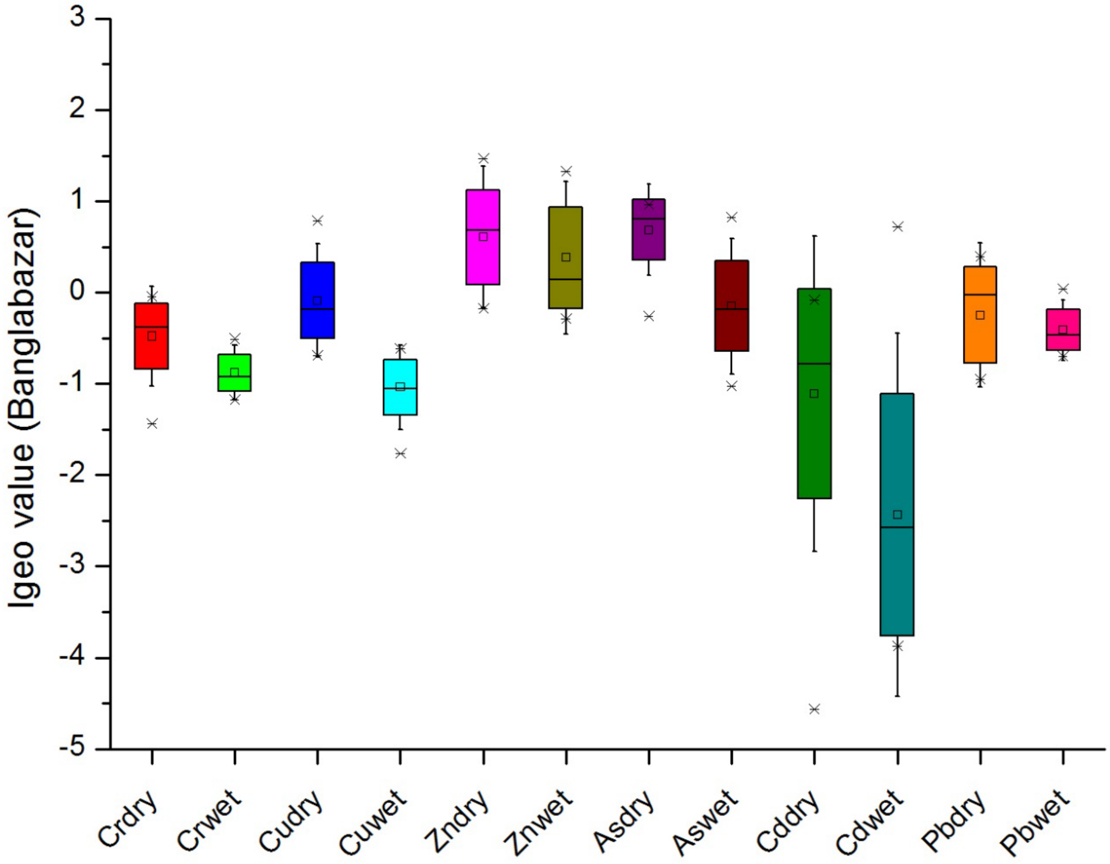

Fig. 3.1 Geoaccumulation index (Igeo) value of heavy metals in the dry and wet season soil of Banglabazar.

Fig. 3.2 presents the geoaccumulation index (Igeo) of the studied heavy metals in Kashimpur area in the dry and wet season. In the dry season, mean Igeo index demonstrated that Cr (-0.39), Cd (-1.52) and Pb (-0.32) have index value below zero which specifies uncontaminated soil by these heavy metals, whereas Zn (0.56), Cu (0.52) and As (0.16) has positive Igeo value (mean) between 0 and 1, and falls in class 1. This indicates that the area is in the level of uncontaminated to moderately contaminated by Zn, Cu and As in the dry season.

In the wet season, the soil of Kashimpur area showed the highest Igeo value for Zn (0.42) which is lower than dry season, and falls in class 1 ($0 < Igeo < 1$) indicating uncontaminated to moderately contaminated soil by Zn. On the other hand, the Igeo values for Cr, Cu, As, Cd and Pb were -0.68, -0.55, -0.35, -2.30 and -0.63, respectively, indicating uncontaminated pollution level. For the differences between dry and wet season on the respective heavy metals, the Igeo values in the wet season is significantly lower than in the dry season (according to the paired *t*-test, $t = 2.2–16.2$; df = 11; $p < 0.05$).

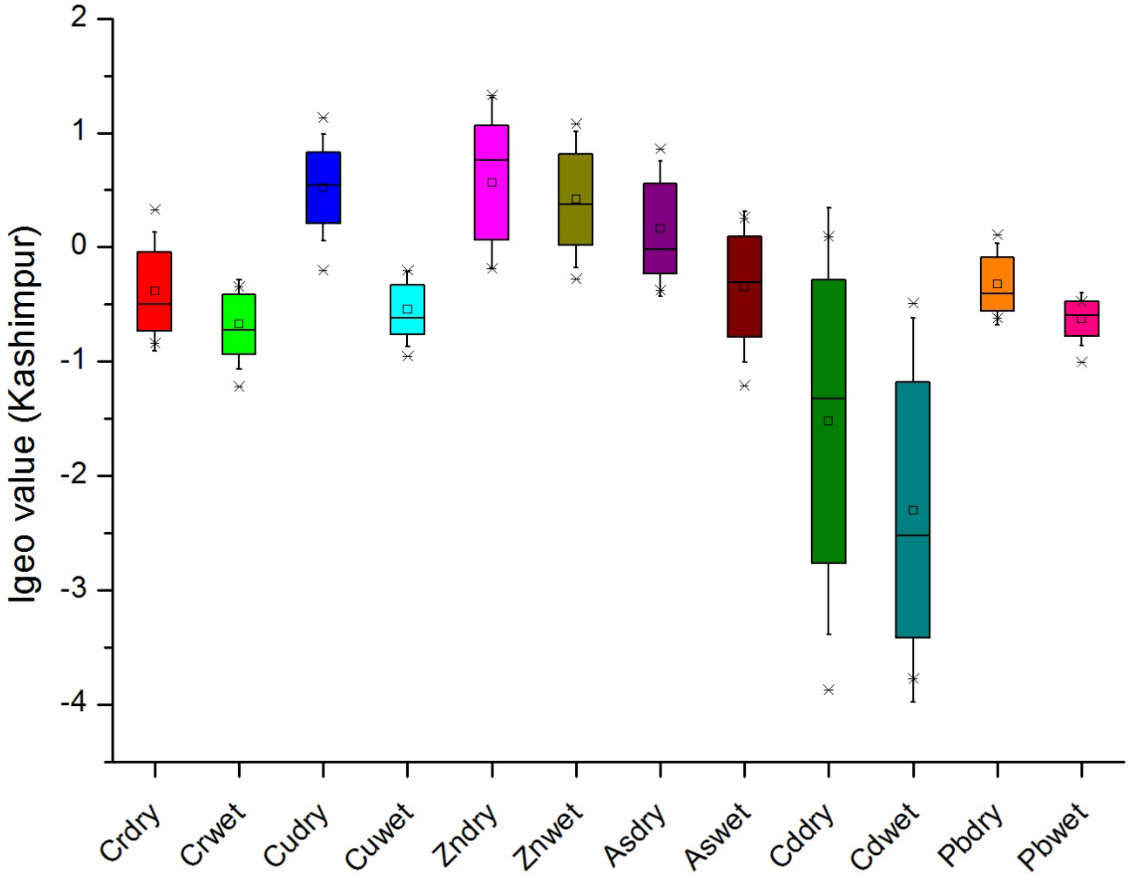

Fig. 3.2 Geoaccumulation index (Igeo) value of heavy metals in the dry and wet season soil of Kashimpur.

The geoaccumulation index of Chandra area in the dry and wet season are shown in Fig. 3.3. In the dry season, the highest value was observed for Zn (0.74) which included in Class 1 (0 < Igeo < 1), that soils are uncontaminated to moderately contaminated by Zn. However, the Igeo values of Cr (-0.45), Cu (-0.65), As (-0.01), Cd (-1.40) and Pb (-0.26) showed the values less than zero which indicates that soils are uncontaminated.

In wet season, the majority of the undertaken heavy metals like Cr (-0.81), Cu (-0.79), As (-0.69), Cd (-1.97) and Pb (-0.47) showed negative Igeo values indicating uncontaminated soil by these heavy metals. The Igeo value of Zn (0.43) demonstrating uncontaminated to moderately contaminated soil due to included in Class 1 (0 < Igeo < 1). Similar to Banglabazar and Kashimpur area, the Chandra area also exhibited lower Igeo values in the wet season than that in the dry season for the respective heavy metals, based on the paired *t*-test ($t = 3.8$–4.9; df = 11; $p < 0.05$).

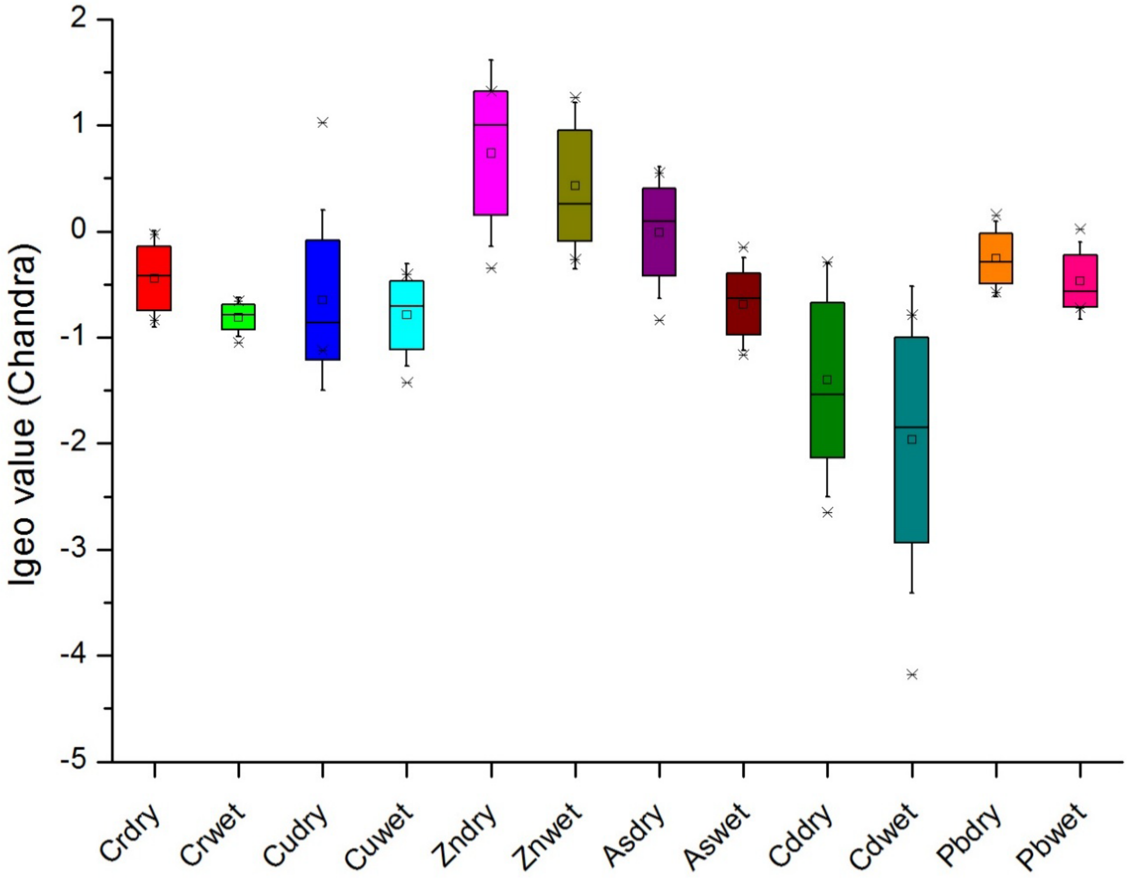

Fig. 3.3 Geoaccumulation index (Igeo) value of heavy metals in the dry and wet season soil of Chandra.

From the above, the Igeo values of heavy metals were found to be the highest for Zn followed by As among the other heavy metals in the three industrial areas, which might be due the higher concentration in soil especially for Zn and lower level in the background samples. On the other hand, the characteristics of lower Igeo values in the wet season showing the dilution effect of rainfall (in irrigation water) on the soil's low absorption of the heavy metals.

The overall toxicity and quality status of the soil samples were assessed by pollution load index (PLI). The calculated pollution load index (PLI) values of heavy metals in soils are concised in Fig. 3.4 which were ranged from 0.81 to 1.76 during dry and 0.65 to 1.66 during the wet season. However, the higher PLI (1.4 which is above 1) value in the dry season confirmed that the soils of the studied areas were slightly contaminated and/or polluted. In the wet season, PLI values are very close to baseline level i.e. (PLI = 1) indicated the entire areas are perfect. The PLI can afford some thoughtful understanding to the residents about the quality of the entire environment (Islam el al., 2015). Moreover, it correspondingly provides valuable evidence to the decision makers on the pollution or contamination status (Suresh et al., 2012). Higher PLI values were observed in the dry season which might be due to the effects of anthropogenic activities (especially industrial activities) with less rainfall effect by dilution of irrigation water on heavy metal absorption by soil.

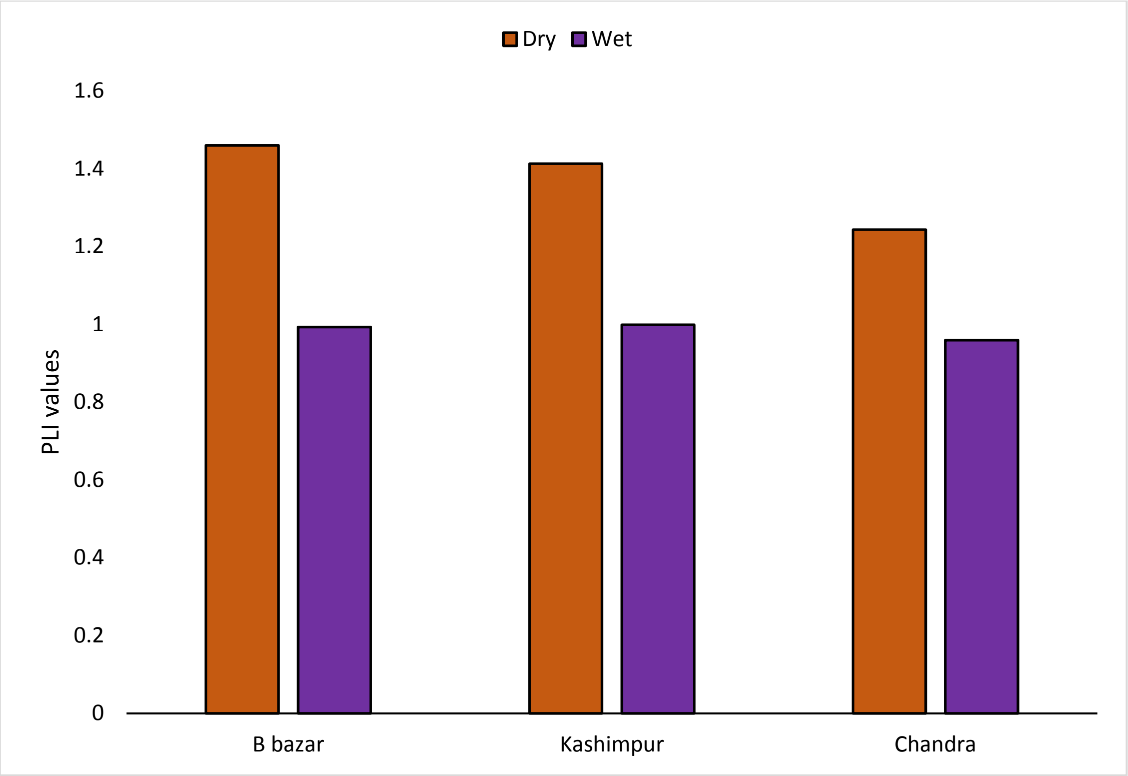

Fig. 3.4 Pollution load index (PLI) values of heavy metals in the dry and wet season soil of study areas.

The degree of heavy metal pollution based on their toxicity and environmental response was assessed to present the potential ecological risk index (RI). Table 3.7 showed the potential ecological risk factor (E^i_r), risk index (RI) and pollution degree of study areas in the dry and wet season. Considerable variations were observed for the potential ecological risk factor (E^i_r) of individual heavy metals which indicated ecological risk of heavy metals varied with the study areas. In the dry season, the E^i_r values of Cr, Cu, Zn, As, Cd and Pb were significantly lower than 40 (minimum grade of ecological risk) indicating that soils are low potential ecological risk in all study areas. But potential ecological risk index (RI) of Kashimpur and Chandra area was less than 65 indicates that low RI, whereas Banglabazar showed little higher RI value (68.05) indicating moderate risk. The mean value of E^i_r and RI indicates low potential ecological risk. Similar variations were observed in the wet season i.e. low potential ecological risk for the lower E^i_r values (< 40) and low RI for the lower RI values (< 65). According to the report of Islam et al. (2017), RI signifies the sensitivity of different biological communities to various toxic substances and shows potential ecological risk initiated by heavy metals. The highest E^i_r values were observed for Cd in the study areas which represent the significant contribution of Cd to the RI of the surrounding environment and may originate from manmade sources especially industrial activities (Luo et al. 2012). Overall, the range of RI is 36.87 to 68.05, indicating low to moderate ecological risk.

Table 3.7 Potential ecological risk factor, risk index and pollution degree of heavy metals in the dry and wet season soil of study areas.

Season	Area	Potential ecological risk factor (E^i_r)						Risk Index (RI)	Pollution degree
		Cr	Cu	Zn	As	Cd	Pb		
Dry	Bangla.	2.21	7.34	2.42	24.64	24.74	6.70	68.05	Moderate risk
	Kashi.	2.36	10.99	2.34	17.41	20.32	6.07	59.49	Low Risk
	Chan.	2.24	5.26	2.67	15.41	19.06	6.34	50.99	Low Risk
	Mean	2.27	7.86	2.47	19.16	21.38	6.37	59.51	Low Risk
Wet	Bangla.	1.65	3.72	2.09	14.27	13.82	5.70	41.25	Low Risk
	Kashi.	1.91	5.19	2.07	12.28	12.13	4.88	38.46	Low Risk
	Chan.	1.72	4.43	2.15	9.48	13.59	5.50	36.87	Low Risk
	Mean	1.76	4.45	2.11	12.01	13.18	5.36	38.86	Low Risk

Bang.: Banglabazar; Kashi.: Kashimpur; Chan.: Chandra.

3.3 Seasonal differences in the contamination of irrigation water, soil and vegetables

Box-and-whisker plot of heavy metal concentration of irrigation water in the wet and dry season was depicted in Fig. 3.5. From the result of Fig. 3.5, a clear difference was observed for heavy metal concentrations between the dry and wet season.

The mean concentrations of Zn (15.21 mg/L) and Cu (4.60 mg/L) were significantly higher followed by those of Cr (2.14 mg/L), Pb (1.15 mg/L) and As (0.52 mg/L) in the dry season. The lowest mean concentration was observed in Cd (0.02 mg/L). The magnitude of heavy metal concentration decreased in the order of Zn > Cu > Pb > Cr > As > Cd in the wet season, and Zn > Cu > Cr > Pb > As > Cd in the dry season, respectively. In the dry season, the mean Zn, Cu, Cr and As concentrations in the respective areas exceeded the permissible limit of FAO standard (Table 3.1). The Pb concentration was within the permissible limit of FAO, whereas Cd concentration was above the permissible limit of FAO (Ayers and Westcot, 1985) (Table 3.1).

In the wet season, the order of magnitude of heavy metal concentrations was much more similar to the dry season’s order, however, the concentrations itself were lower than in the dry season. The heavy metal concentrations of Cr, Cu, Zn, As, Cd, and Pb in irrigation water were about 3 times, 1.2 times, 1.4 times, 1.6 times, 2.3 times, and 1.4 times higher, respectively, in the dry season compared to the wet season.

Regarding the difference between dry and wet seasons, the concentrations in the dry season were observed to be significantly higher than in the wet season according to the paired *t*-test ($t = 2.4–5.0$, df = 35, $p < 0.05$). Even in the wet season, the concentration of each heavy metal exceeded the FAO standard except Pb and Cd. Therefore, the water is identified to be inappropriate to use for irrigation purposes in both seasons.

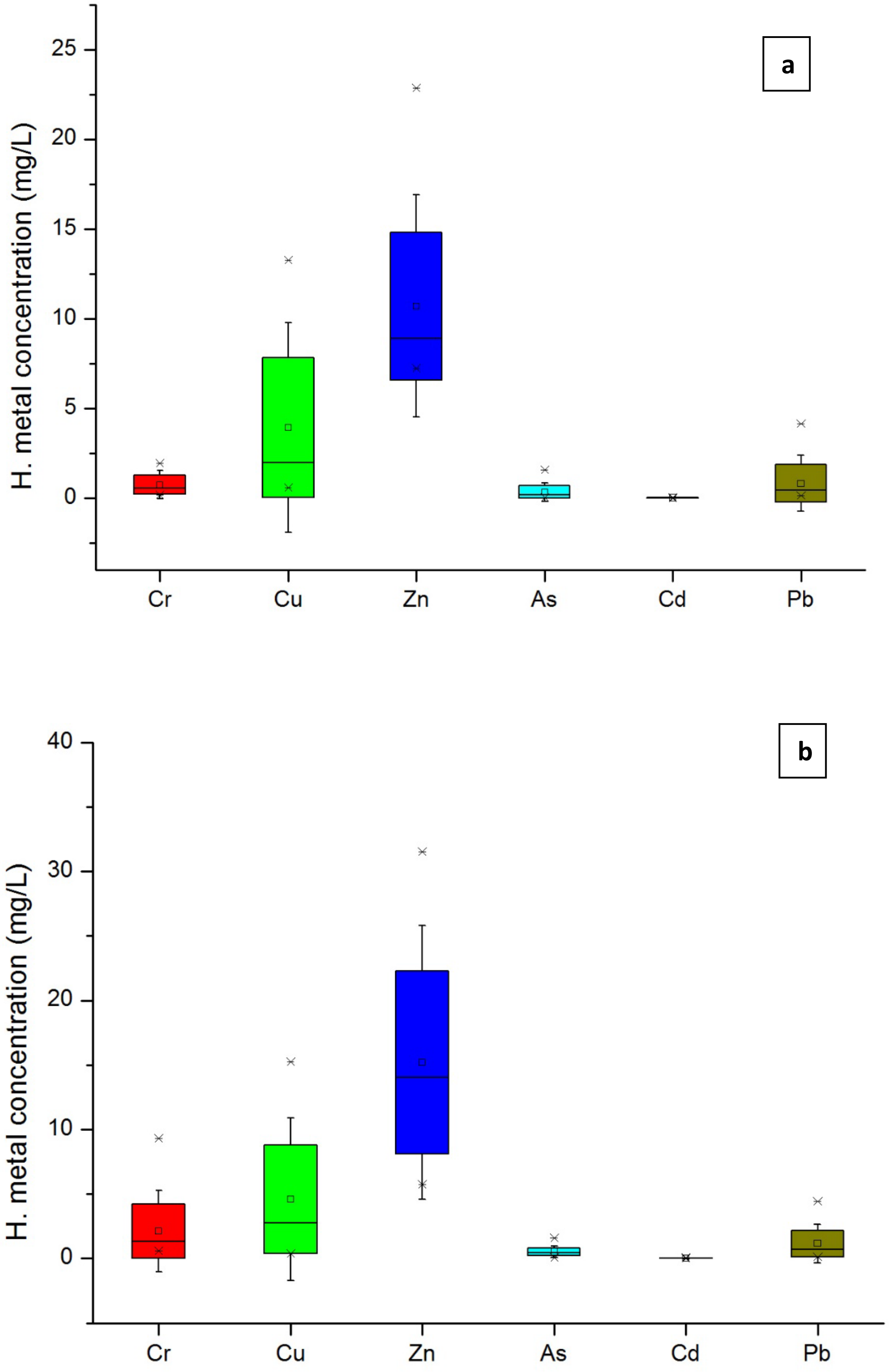

Fig. 3.5 Box-and-whisker plot of heavy metal concentration in irrigation water (a) wet season, (b) dry season.

The seasonal changes of the soil heavy metal concentrations were depicted in Fig. 3.6. A wide variation was observed in the dry and wet season's concentration of soil, where the highest was Zn (173.23 mg/kg in the dry season; 147.44 mg/kg in wet season) and the lowest was Cd concentration (0.29 mg/kg in the dry season; 0.18 mg/kg in the wet season). The second highest concentration was found for Cr followed by Cu, Pb, and As in both seasons. The order of magnitude of heavy metals in the dry season was Zn > Cr > Cu > Pb > As > Cd. In the wet season, the order of the magnitude of heavy metal concentrations was similar to that of the dry season, however, the concentrations were significantly lower than those of dry season. The heavy metal concentrations of Cr, Cu, Zn, As, Cd, and Pb were about 1.3 times, 1.8 times, 1.2 times, 1.6 times, 1.6 times, and 1.2 times higher, respectively, in the dry season compared to the wet season.

For the difference of the concentrations of soil between the two seasons, the concentrations in the wet season were found to be significantly lower than in the dry season in the respective areas according to the paired t-test ($t = 3.2–6.3$, df = 35, $p < 0.05$). In both seasons, all of the heavy metal concentrations of soil were below the permissible limit of EU and Indian standards (Ministry of the Environment Finland, 2007; Awashthi, 2000). Only As concentration in both seasons exceeded the permissible limit (Table 3.3). Therefore, the soil is considered to be appropriate for planting purposes.

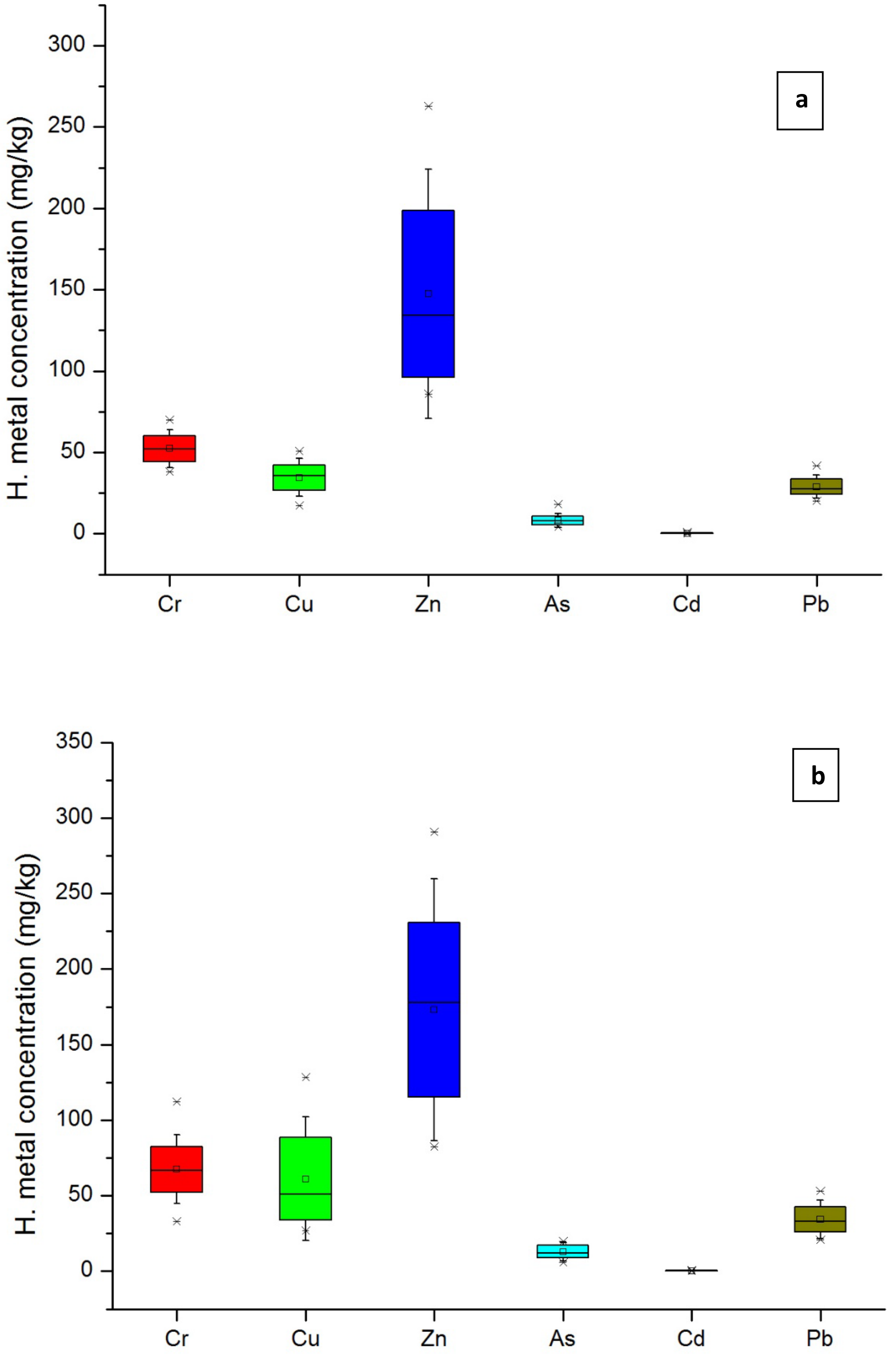

Fig. 3.6 Box-and-whisker plot of heavy metal concentration in soil (a) wet season, (b) dry season.

Seasonal differences (between dry and wet season) of heavy metal concentrations in vegetables are plotted through box-and-whisker plots in Fig. 3.7. The mean highest concentration of heavy metals was observed in Zn (62.74 mg/kg) in the vegetables of the dry season followed by Cr (19.35 mg/kg), Cu (14.79 mg/kg), and Pb (6.27 mg/kg). Consequently, the lowest concentration was observed in Cd (0.34 mg/kg) followed by As (1.26 mg/kg) in the same seasonal vegetables. In the wet season, some differences were observed, i.e. the maximum concentration of vegetables was found in Zn (51.78 mg/kg) followed by Cu (16.58 mg/kg), Pb (6.00 mg/kg), and Cr (5.59 mg/kg). In the dry season, the minimum concentration was found in Cd (0.13 mg/kg) followed by As (0.80 mg/kg).

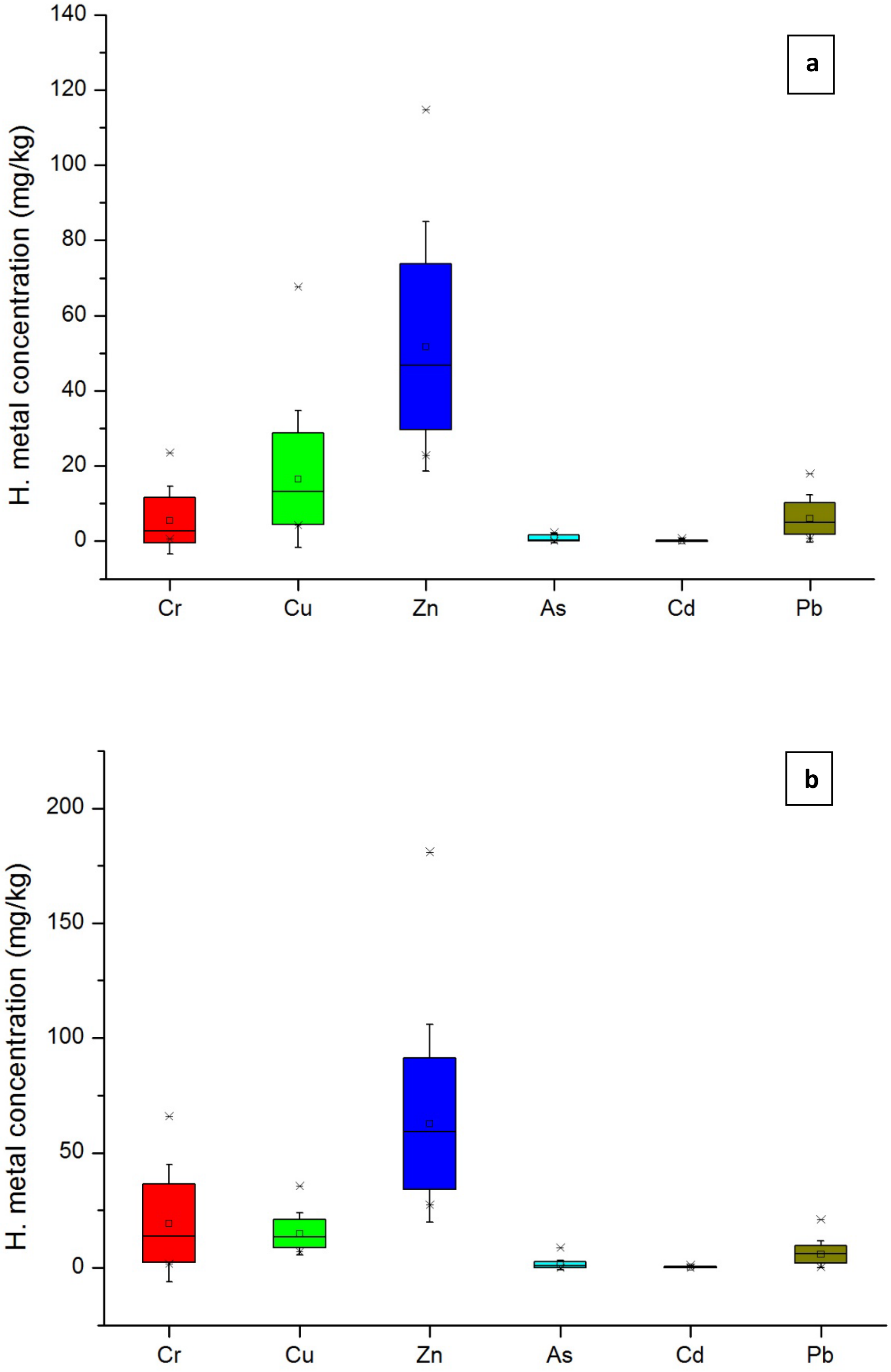

Fig. 3.7 Box-and-whisker plot of heavy metal concentration in vegetables (a) wet season, (b) dry season.

The order of the magnitude of heavy metals of vegetables in dry season was Zn > Cr > Cu > Pb > As > Cd. In the wet season, the order of the magnitude of heavy metal concentrations was Zn > Cu > Cb > Cr > As > Cd, and which were lower than those in the dry season. The comparison of the magnitude order of the concentrations of vegetables with those of soils and irrigation water revealed that vegetable's order in the dry season showed a very close match with the soil of dry season and the order of wet season's vegetables were closely related with the order of irrigation water in that season. The heavy metal concentrations of Cr, Cu, Zn, As, Cd and Pb of vegetables in the dry season were about 3.5 times, 0.9 times, 1.2 times, 1.6 times, 2.6 times, and 1.04 times, respectively higher than those in the wet season.

The heavy metal concentrations in vegetables were found to be significantly higher in the dry season than in the wet season (by paired t-test; $t = 0.18–5.15$, df = 26, $p < 0.05$). The concentration of heavy metals of vegetables exceeded the permissible limit of FAO in Cr, Zn, As, and Pb concentration in the wet season, and in Cr, Zn, As, Cd, and Pb concentrations in the dry season. The Cu concentration in both seasons satisfied this permissible limit, whereas Cd satisfied this limit only in the wet season for vegetables. Here, high concentrations such as 27 times, 22 times, and 14 times higher than the standard were observed in the concentrations of Pb, Cr, and As, depending on the area. Therefore, vegetables grown in these areas was identified as risky for human consumption.

3.4 Spatial distribution of the contamination of irrigation water, soil and vegetables

Industrialization is one of the factors contributing to heavy metal contamination in farmland. The three industrial study areas (Banglabazar, Kashimpur, and Chandra) were heavily contaminated by heavy metals with untreated wastewater discharged from various factories. When comparing the heavy metal concentrations between the areas, the extent of irrigation water contamination showed a significant variation with study areas according to the two-way ANOVA in both seasons (F = 4.06–8.74, df = 2, $p < 0.05$). Industrial situation is somewhat different from each other in each area (Banglabazar, Kashimpur, and Chandra). Effluent treatment plant (ETP) is supposed to be installed in each industrial unit for the proper treatment of the wastewater. However, only a few industries installed ETP and that is operated even occasionally. Most of the industries discharge industrial effluents directly into nearby canals and water bodies, and which are thought to be severely contaminated. The contaminated canal water is regularly used for irrigation purposes.

The results of Table 3.1 suggest that the irrigation water of Banglabazar area was highly contaminated with As and Pb. Similarly, the water of Kashimpur area was also contaminated highly by Cr and Cd, whereas the water of Chandra area showed highly contamination of Zn and Cu. In Kashimpur area, where Cr and Cd compounds are used to add color and pigments to garments and used in plastics and agrochemicals manufacturing, the water was highly contaminated with Cr and Cd. In Banglabazar area, paint, ceramics, textile, dye, metallurgical and battery factories are located and discharging As and Pb. In Chandra area, wastewater is discharged from fabric printing, pharmaceutical, agrochemical, poultry feed and fish feed factories, and irrigation water is to contain Zn and Cu.

The heavy metal concentrations of soil in Banglabazar, Kashimpur and Chandra areas showed insignificant variation, obtained from the result of two-way ANOVA. For all heavy metals, variations between mean concentrations were small, and those of minimum and maximum concentrations were as well. The relatively low heavy metal concentrations and lack of significant variations between them in soil may be ascribed that the heavy metal concentration of irrigation water was rather equalized between the areas in the process of heavy metal absorption from irrigation water to soil and from soil to vegetables. According to the result of Sharma et al., (2007), lack of significant variations in soil may be attributed to the plant uptake of heavy metals from soil during their growth and development. Similar variations were observed by Zakir et al., (2015) for Zn, Cr, Cu, and Pb concentrations in the soil.

For vegetables, the variations of the respective heavy metal concentrations between the study areas (Banglabazar, Kashimpur and Chandra) were found to be insignificant (according to the method of two-way ANOVA) indicating that the absorption ability of heavy metals by vegetables was not different with areas, which may be due to the insignificant variation of heavy metal concentration in the soil between these industrial areas.

3.5 Sources of heavy metals in irrigation water, soil, and vegetables

The vegetable fields of the study area received a significant amount of irrigation water from nearby canals through the narrow channel during the wet and dry seasons. These surface water are often used as the main source for irrigation water for cultivating vegetables. In the study area, unplanned and rapidly growing industrial factories discharged untreated wastewater into the canals from which the water is pumped up to irrigate croplands. Therefore, the surface water was severely polluted and serves as a reservoir to contain an elevated amount of toxic heavy metals. This is the reason for the high heavy metal concentration in the irrigation water at the sampling areas. Barakat, (2011) and Lambert et al., (2000) showed that industrial wastewater contained most common heavy metals such as Cr, Cu, Pb, Ni, Cd, Zn, and As with high toxicity and all of which posed environmental and health risk. According to the result of Ross, (1994), contamination of farmland soils with heavy metals through irrigation water is mainly due to be sourced from contaminated water from lakes, rivers, canals, channels or deep wells.

From the results of Fig. 3.5, the highest standard deviation of the heavy metal concentration was observed for Zn and Cu followed by Pb and Cr. Discharges of wastewater into canals were observed at many places affected by different factories, and the concentrations of heavy metal may vary depending on the places, i.e. type of wastewater, resulting in these tendencies. According to the result of Sharma et al., (2007), the manufacturing process of the product and the raw materials used in the industry are the sources of the heavy metals in wastewater. However, the mean concentration of each heavy metal in the wet season was lower than in the dry season, which may be due to the effect of rainfall in the wet season that diluted the heavy metal concentration of the irrigation water.

On the other hand, the order of the magnitude of heavy metal concentrations of the soil followed a similar order to that of the irrigation water, except for Cr and Cu in the dry season, and Cr, Cu, and Pb in the wet season (Fig. 3.5 and Fig. 3.6). Namely, the Zn concentration was the highest value, and the As and Cd concentrations were the lowest one. This similarity of the order in heavy metal concentrations between soil and irrigation water indicated that the soil contamination was mainly caused by the contamination of the irrigation water. In addition, the background value reported by Kashem and Singh, (1999) indicated that the increased heavy metals concentrations in the studied soil might be due to the frequent use of industrial wastewater-mixed water for irrigation. Similar results were observed by different authors in other industrial areas of Bangladesh (Sulatana et al., 2014; Naser et al., 2009; Ahmad and Goni, 2010). Lower concentrations of heavy metals for soil were found in the wet season than in the dry season indicated the effect of diluted irrigation water on the occurrence of lower concentration of heavy metals of soil in the wet season.

Regarding the order of the concentrations, Cr, Cu and Pb concentrations in the soil were different from those of irrigation water. Therefore, the involvement of some other factors (to differentiate the orders) are conceivable. For instance, adsorption and desorption with ion-exchange, precipitation, immobilization, mobilization, plant uptake etc. of heavy metals influence the distribution and retention characteristics of heavy metals in soil (Wuana and Okieimen, 2011; Levy et al., 1992). The chemical forms of heavy metals in soil might be affected by mobility, toxicity, and bioavailability of heavy metals (Shiowatana et al., 2001; Buekers, 2007). Dudal et al., (2005) stated that the mobility of heavy metals along with soluble organic matter might be affected by high-precipitation events. However, soil physical and chemical features influence the movement and bioavailability of trace metals, which depend not only on the trace metal concentration but also on environmental factors, adsorption-desorption characteristics of soil, precipitation reactions and the properties of heavy metals (Cuevas and Walter, 2004; Li et al., 2003). Those might differentiate the order of heavy metals between the soil and irrigation water.

Wastewater irrigation for growing crops is common practice in Bangladesh, especially near the industrial zones. Wastewater irrigation causes the excessive accumulation of heavy metals in farmland soils, which result not only in soil contamination but also in vegetables contamination and ultimately food quality and safety (Muchuweti et al., 2006; Arora et al., 2008). In the study areas, different types of industries disposed their wastewater to the adjacent canals, and the canal water is used by the local farmers for growing various vegetables seasonally. The use of contaminated irrigation water in the areas possibly increased the uptake and accumulation of heavy metals in the cultivated vegetables. The order of heavy metal concentrations in vegetables were as follows Zn > Cr > Cu > Pb > As > Cd in the dry season and Zn > Cu > Pb >Cr> As > Cd in the wet season.

In the dry season, the order of vegetables was very similar to the order of soils and dissimilar to the order of irrigation water (Figs. 3.5b, 3.6b and 3.7b). Similarly, in the wet season, the order of vegetables was close to the order of irrigation water (Figs. 3.5a, 3.6a and 3.7a). Some variations were observed among Cr, Cu and Pb concentrations in the order of irrigation water and vegetables in the dry season. The heavy metal concentrations in vegetables may be varied due to the differences in their morphology, kind of vegetables and physiology in the uptake, exclusion, accumulation, and retention of heavy metals (Singh et al., 2010). Moreover, lower concentrations of heavy metals were found in vegetables in the wet season than in the dry season, showing the effect of rainfall diluting and lowering the heavy metal concentration in irrigation water on the concentration of heavy metals in vegetables.

CHAPTER 4

Effects of Irrigation Water and Soil Contaminations on Vegetable Contamination

4.1 Relationship of heavy metals among irrigation water, soil, and vegetables

Hierarchical cluster analysis (HCA) was applied to clarify the effect of irrigation water and soil on vegetables in the heavy metal concentrations in the respective seasons, and the results (dendrograms) were shown in Fig. 4.1, Fig 4.2 and Fig. 4.3, respectively.

For the irrigation water in the dry season (Fig. 4.1a), As and Cd formed a cluster with the shortest distance (< 5) along with Pb, showing a close relationship between them. Cr and Cu linked with the cluster of As, Cd, and Pb with small distance (< 10), indicating their moderate relatedness. While in the wet season (Fig. 4.1b), As, Cd and Cr formed a cluster along with Pb and Cu, showing the similarity of clusters with those of the dry season. The Zn concentration had a weak relationship with the other heavy metals, because the concentration showed a large distance (25) different from the other heavy metal concentrations in both seasons.

For the soil, As and Cd formed a cluster with a shortest distance (< 5), showing a close relationship between both dry and wet seasons (Fig. 4.2). A cluster was formed between Cr and Cu with a small distance (< 5), and another cluster was formed among Pb, Cd and

As in the dry season (distance < 5) (Fig. 4.2a). In the wet season, Cu and Pb formed a cluster together with Cr (Fig. 4.2b). In both seasons, Zn, which has a long distance (25), was isolated from other heavy metals, showing a weak relationship of Zn with the other heavy metals.

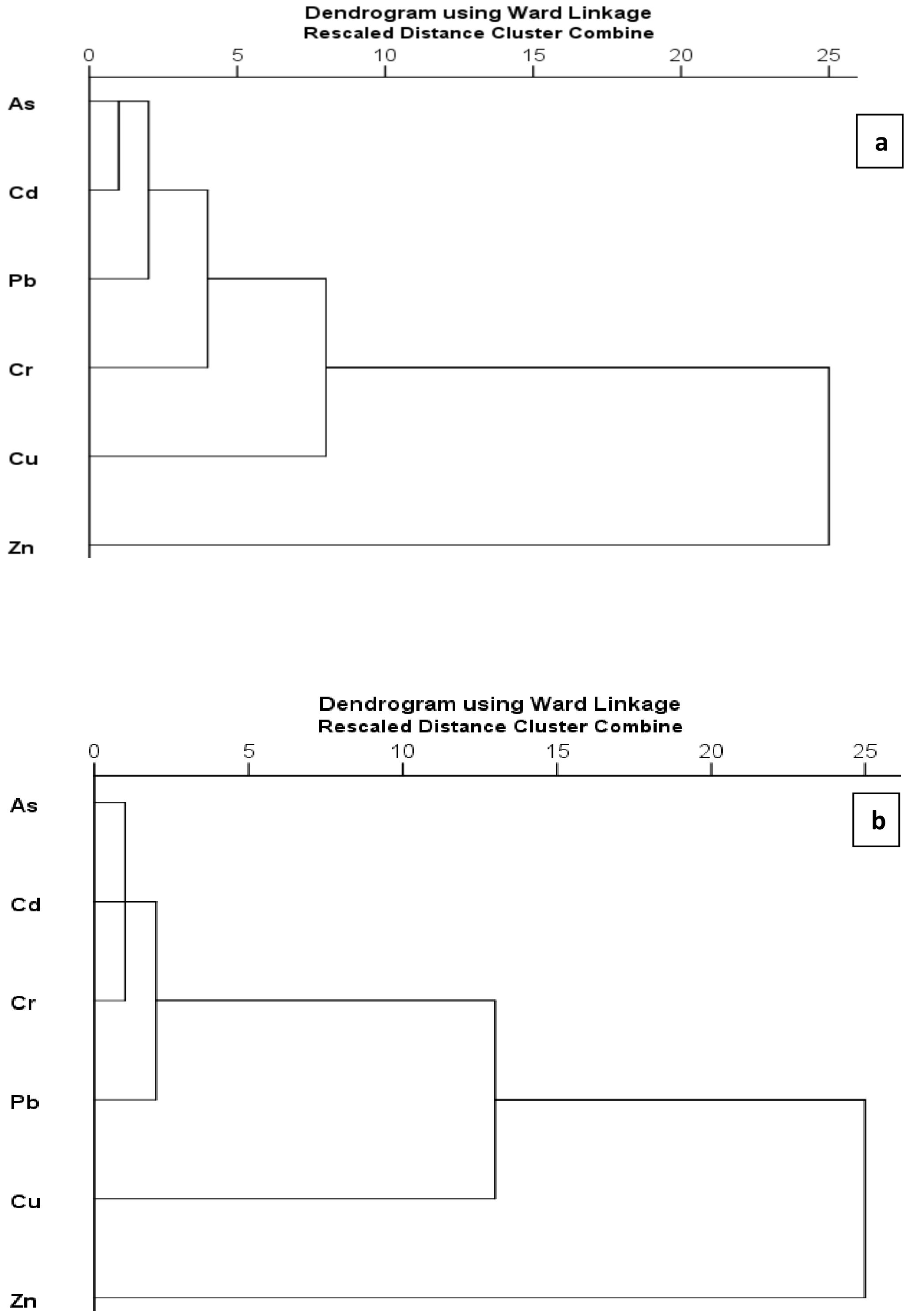

Fig. 4.1 Dendrogram of the Hierarchical Cluster Analysis for heavy metals in irrigation water (a) dry season, (b) wet season.

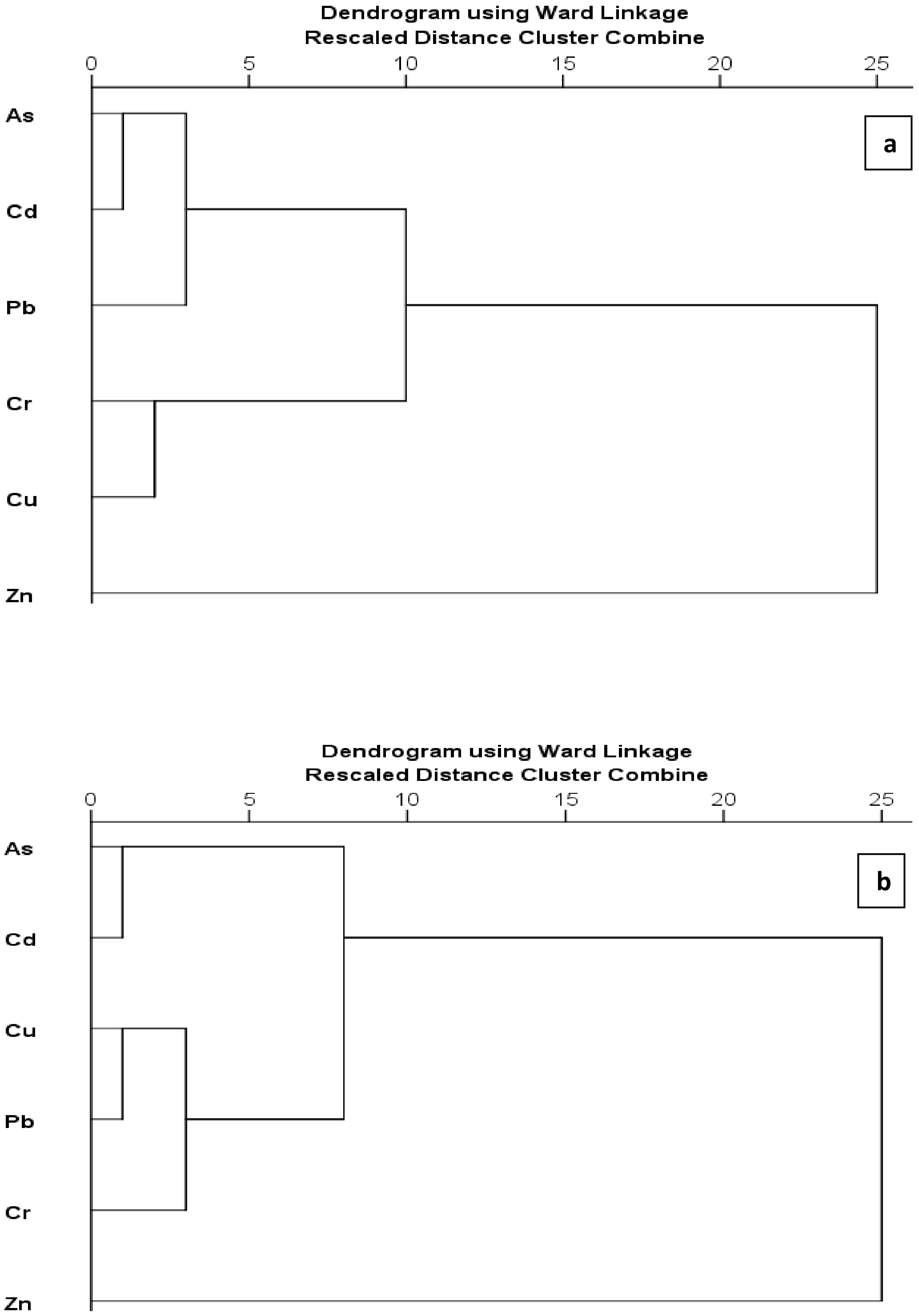

Fig. 4.2 Dendrogram of the Hierarchical Cluster Analysis for heavy metals in soil (a) dry season, (b) wet season.

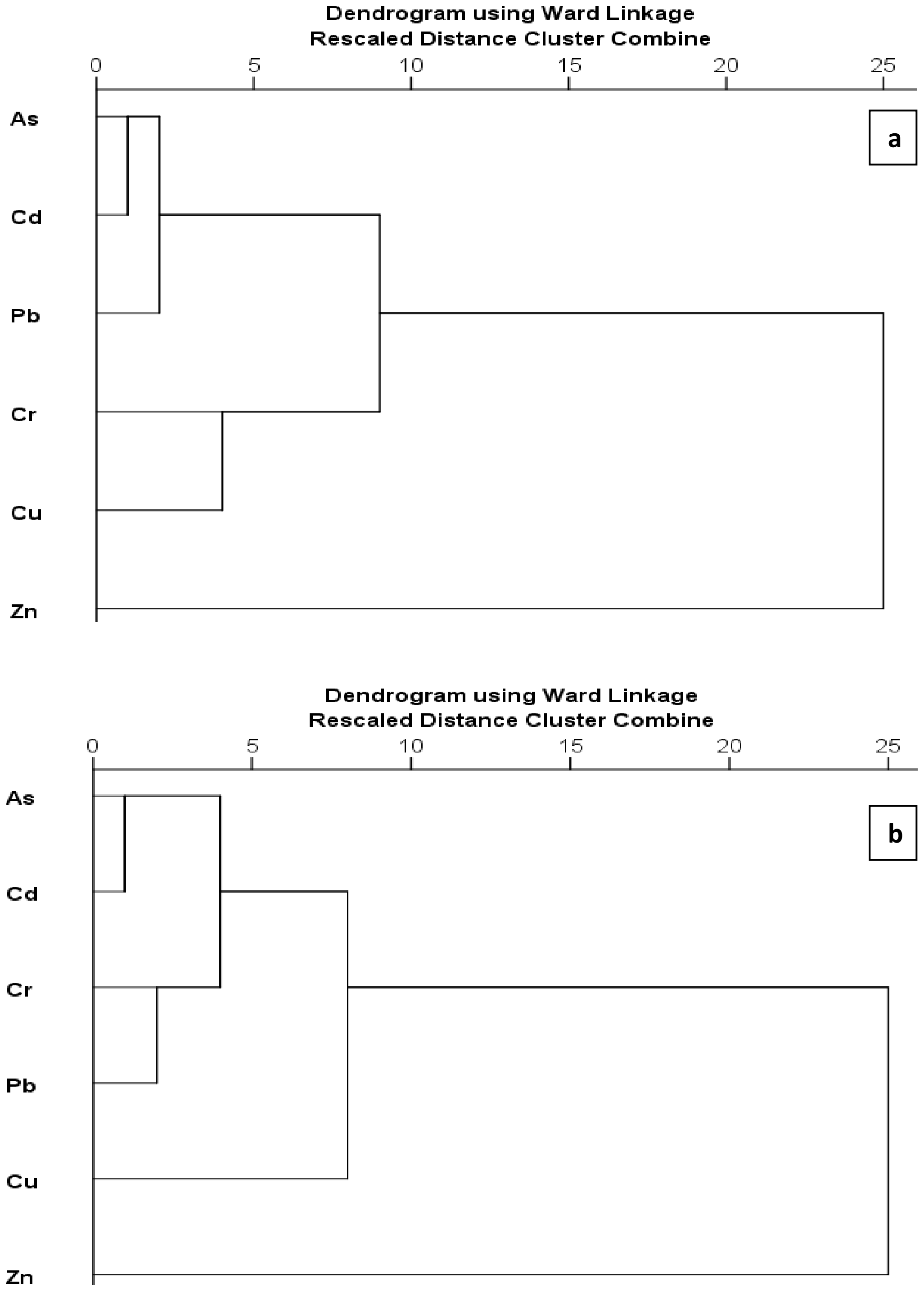

Fig. 4.3 Dendrogram of the Hierarchical Cluster Analysis for heavy metals in vegetables (a) dry season, (b) wet season.

For the vegetables, As and Cd showed a close relationship with each other and formed a cluster (distance < 5) in both dry and wet season (Fig. 4.3). Another cluster was formed between Cr and Cu in the dry season and between Cr and Pb in the wet season with a distance <5. In the dry season, the cluster between As and Cd was associated with Pb, forming another small cluster (distance < 5), and which formed a large cluster consisting of Cr-Cu (distance < 10) (Fig. 4.3a). In the wet season, the dendrogram is quite different from that of the dry season, i.e. the cluster of As-Cd was connected with another cluster of Cr-Pb and formed a large cluster in association with Cu (distance < 10) (Fig. 4.3b). The distance (25) of Zn from the cluster of other heavy metals in the dendrogram was long, indicating its weak relationship with the other heavy metals in both seasons.

To identify the effect of irrigation water or soil on the heavy metal absorption by vegetables, the dendrogram of vegetables was compared with those of irrigation water and soil in the respective seasons. In the dry season, the dendrogram of vegetables is significantly resembled with that of soil, which formed two different clusters (As-Cd-Pb and Cr-Cu) with shortest distances (< 5), whereas Zn showed longest distance (25) (Figs. 4.2a and 4.3a). This similarity of the dendrogram in the dry season suggested that heavy metals in vegetables were probably absorbed from the soil.

While in the wet season, the dendrogram of vegetables is significantly resembled with that of irrigation water, because the cluster of As-Cd in vegetables was similar with the cluster of As-Cd-Cr in irrigation water with the shortest distance (< 5) (Figs. 4.1b and 4.3b). At the later stage, As, Cd, Cr, and Pb in vegetables were linked together and showed similarity with irrigation water (distance < 5), whereas moderate and weak relatedness were found for Cu and Zn, respectively, in vegetables and irrigation water. Therefore, the heavy metals in vegetables in the wet season is thought to be absorbed from irrigation water.

In the wet season, the upper soil layers contribute sigificant soil moisture (more than 60 % of plant water requirements), whereas the contributed moisture is less than 50 % in the dry season (Sun et al., 2011). Therefore, the vegetables in the wet season are thought to absorb more moisture content than vegetables in the dry season, which indicates that the heavy metals in wet season vegetables accumulate mainly with the application of irrigation water. The lack of irrigation water in the dry season triggered the vegetables to uptake heavy metals from soil, which caused the water (and heavy metal) absorption from soil.

4.2 Heavy metals transfer from irrigation water to vegetables in the wet season

Heavy metal concentrations (mean values) for the irrigation water and vegetables in the wet season were depicted in Fig. 4.4 and Fig. 4.5 to show the transfer of heavy metals from irrigation water to vegetables. In irrigation water, the highest concentration was found for Zn followed by Cu concentration, whereas the lowest concentration was observed for Cd followed by As in the wet season. The vegetables heavy metal concentration also showed the highest concentration for Zn in the wet season which is similar to irrigation water, whereas Cu was the second highest. As like as irrigation water, the lowest concentration in vegetables was observed for Cd followed by As in this season.

In this season, the magnitude of heavy metal concentrations for irrigation water and vegetables decreased in the order of Zn > Cu > Pb > Cr > As > Cd, and Zn > Cu > Pb > Cr > As > Cd, respectively. To identify the heavy metal transfer in vegetables, the order of concentrations was compared and found similarity with the order of concentration of irrigation water in this season. This means that the heavy metals in vegetables were absorbed mainly from the irrigation water when contaminated irrigation water was applied to grow vegetables. Arora et al., (2008) mentioned that the application of contaminated wastewater for irrigation generally led to alter the physicochemical characteristics of soil and consequently trace metal uptake characteristics by the vegetables.

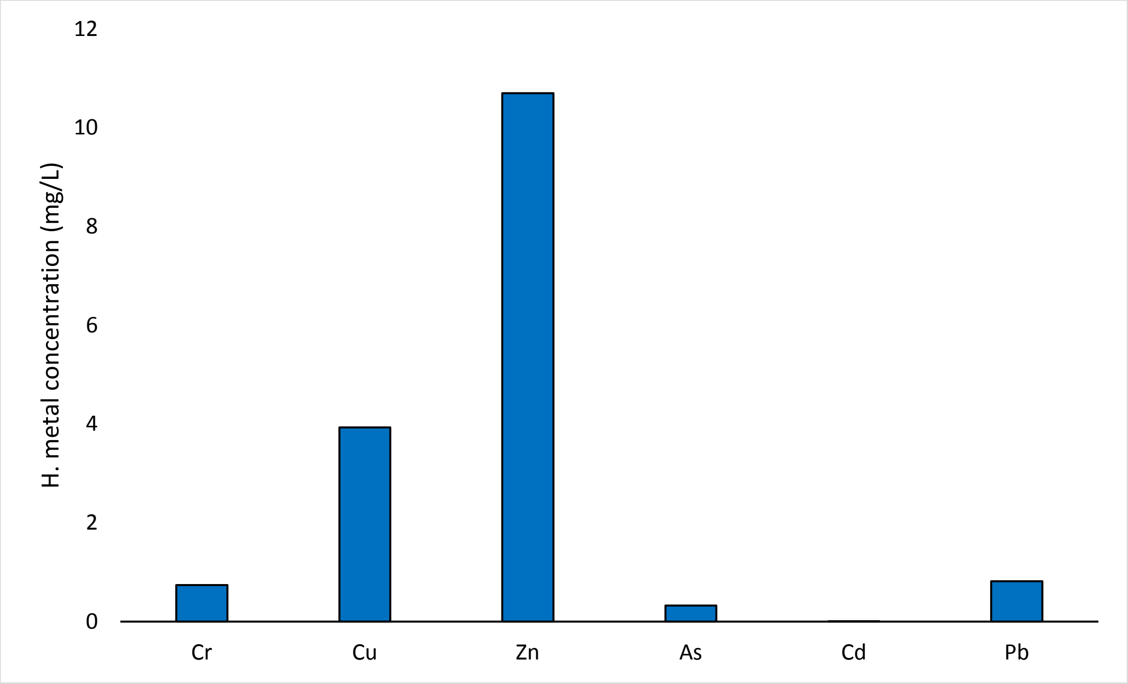

Fig. 4.4 Bar graph showing heavy metal concentration in irrigation water in the wet season.

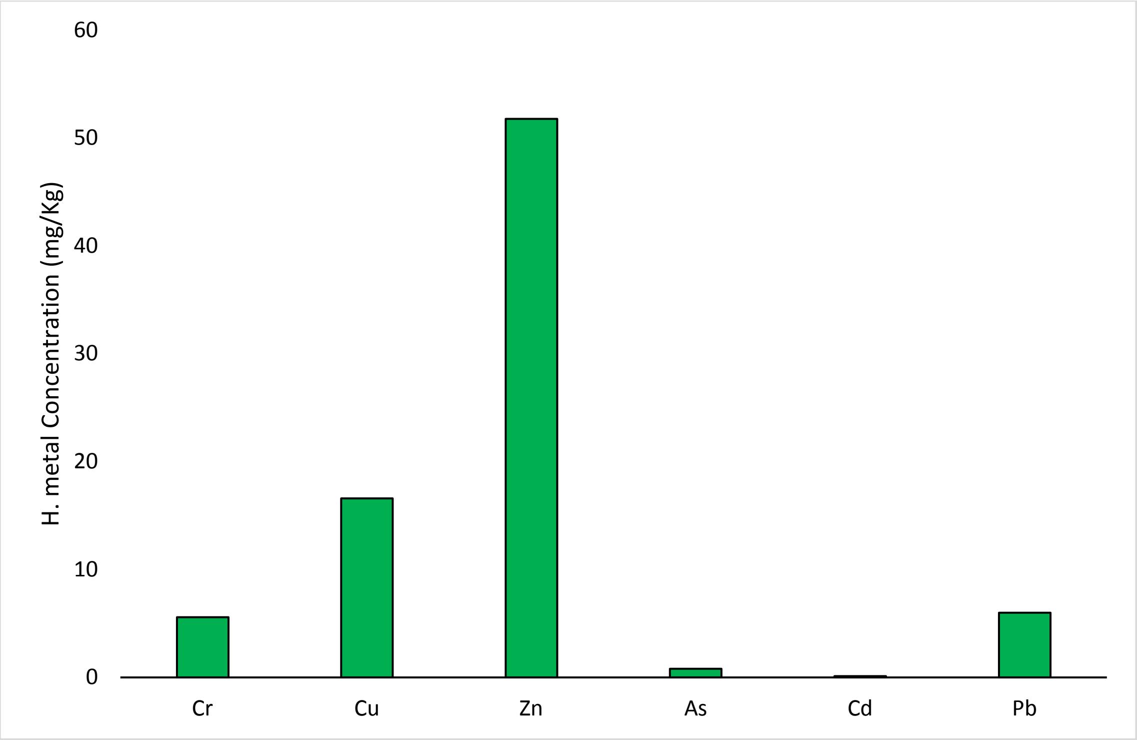

Fig. 4.5 Bar graph showing heavy metal concentration in vegetables in the wet season.

4.3 Heavy metals transfer from soil to vegetables in the dry season

The transfer of heavy metals from soil to vegetables is considered to be vital factor for human exposure via food consumption (Garg et al., 2014). Despite the soil heavy metal concentration was under permissible level, vegetable's heavy metal concentration exceeded its permissible level. For this reason, transfer ability of heavy metals from soil to vegetables might be large. Therefore, the bioconcentration factor (BCF) of heavy metals was examined in this section. BCF shows the transferability of heavy metals from soil to vegetables, and is calculated as the ratio of heavy metal concentration in vegetables to that in soil (based on dry weight) (Ye et al., 2015). BCF (shown in Table 4.1) is important in the dry season, because soil appears to be responsible for heavy metal accumulation in vegetables. The average of BCF values for heavy metal and vegetable are also shown in the Table 4.1.

BCF was the highest in Cd (1.18), followed by Zn (0.37) and then by Cr (0.27), while the lowest was As (0.09), according to the averages of BCF data. Excepting As and Pb, BCF was >0.20, showing high values that indicates a high heavy metal absorption from the soil. According to data of FAO/WHO (2011), the BCF of vegetables higher than 0.20 is thought to be contaminated highly by anthropogenic activities and have a high health risk. For the respective vegetables, radish and red amaranth (both 0.61) showed the highest value, followed by amaranth and spinach (both 0.47), according to their averages of BCF data. These vegetables (belonging to leafy vegetables) showed significant high absorption. Taro (0.44) (root and leaf vegetables) showed also a high absorption. These characteristics might contribute to the high heavy metal concentration that exceeded the permissible level.

Table 4.1 The mean bio-concentration factor (BCF) of heavy metals for the respective vegetables.

Division of vegetables	**Name**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**	**Ave.**
Leafy	Radish (*n* = 6)	0.66	0.53	0.41	0.11	1.77	0.15	0.61
	Amaranth (*n* = 12)	0.18	0.34	0.30	0.09	1.73	0.18	0.47
	Red amaranth (*n* = 6)	0.25	0.26	0.54	0.14	2.25	0.19	0.61
	Spinach (*n* = 6)	0.43	0.34	0.28	0.10	1.50	0.16	0.47
	Indian spinach (*n* = 6)	0.06	0.11	0.39	0.09	0.61	0.08	0.22
	Pumpkin (*n* = 6)	0.10	0.23	0.26	0.03	0.87	0.26	0.29
	Bottle gourd (*n* = 6)	0.22	0.25	0.28	0.05	1.06	0.24	0.35
Root and leafy	Taro (*n* = 18)	0.47	0.37	0.57	0.18	0.75	0.27	0.44
Fruiting	Yard long bean (*n* = 6)	0.05	0.17	0.28	0.01	0.11	0.01	0.11
	Ave.	0.27	0.29	0.37	0.09	1.18	0.17	

n, number of samples.

Therefore, every vegetable is identified to be contaminated by heavy metals with an alarming level. The variation of heavy metal concentrations in vegetables may be ascribed to those of soil in the dry season. The higher contaminations of heavy metals in the cultivated vegetables were due to plant uptake of the heavy metals from the soil.

4.4 Contamination level of heavy metals in each type of vegetables

Heavy metal concentrations in the vegetables varied widely with the type of vegetables and the areas, as shown in Fig. 4.6 and Fig. 4.7. According to the result of Fig. 4.6, the highest Zn concentration (161.36 mg/kg) was observed in taro root from Chandra in the dry season followed by Indian spinach (94.54 mg/kg) from the same area, and by radish leaf (103.35 mg/kg) from Banglabazar. The lowest Zn concentration was observed for pumpkin (28.79 mg/kg) in Kashimpur area. The highest Cr (63.55 mg/kg) and Cu (34.69 mg/kg) concentration was observed in radish leaf from Banglabazar; however, Banglabazar also showed the highest As (6.92 mg/kg) and Pb (17.23 mg/kg) in taro root. In Chandra area, red amaranth leaf showed the highest Cd (1.11 mg/kg) concentration in this dry season.

Regarding the total concentration of all heavy metals (dry season), taro root was the highest in the heavy metal concentrations in all three areas, followed by radish leaf, red amaranth leaf, taro stem and Indian spinach, whereas yard long bean and pumpkin were the lowest in the concentrations. The mean Zn concentration (all areas) was also high in taro root (113.89 mg/kg). Taro root also had an enormous amount of mean Cr (39.84 mg/kg), Cu (20.81 mg/kg), Pb (14.72 mg/kg), As (4.61 mg/kg) and Cd (0.31 mg/kg) concentrations, which exceeded the FAO permissible limit (Table 7). However, the highest mean Cr (48.98 mg/kg) and Cu (25.77 mg/kg) concentrations were found in radish leaf in all three areas, which are higher than the other leaf vegetables of red amaranth, amaranth, Indian spinach, and spinach. On the other hand, the lowest concentrations were observed in yard long bean and pumpkin for all heavy metals. The small standard error values of the vegetable concentration in the upper part of the bar graph (Fig. 4.6) indicate no significant differences between the areas.

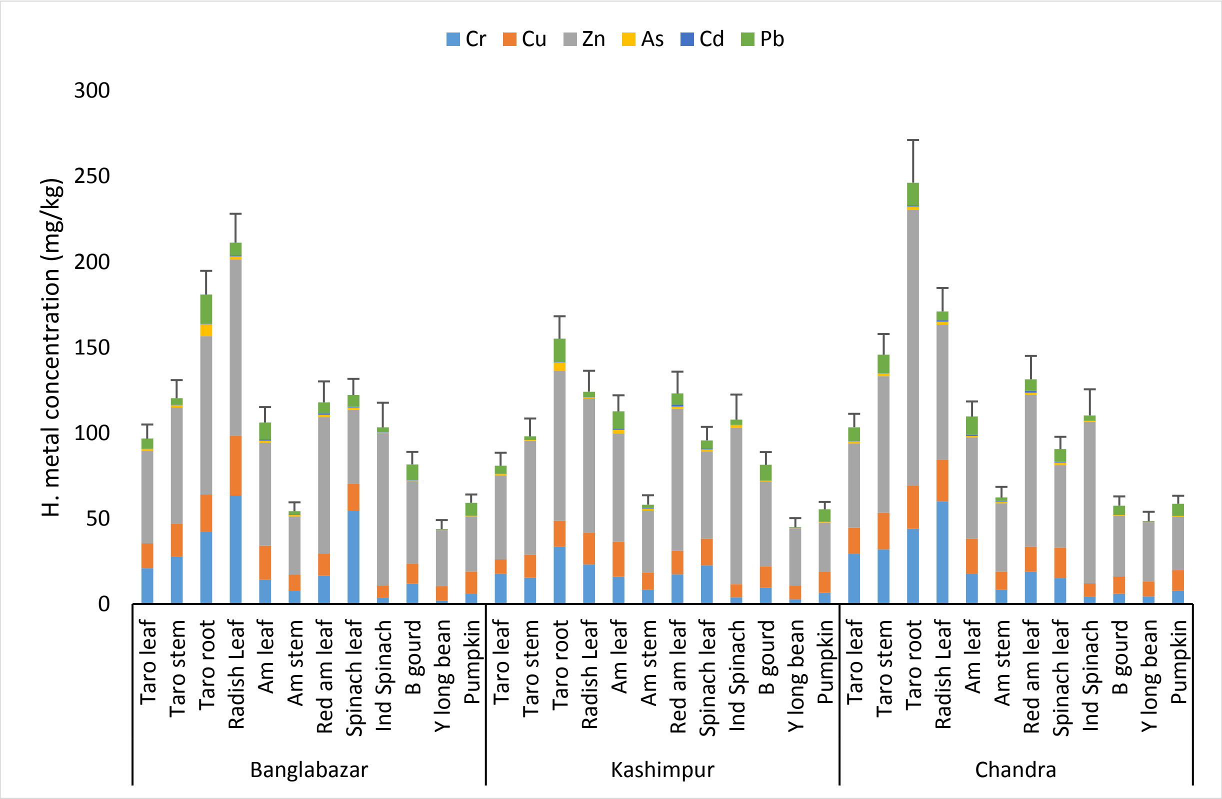

Fig. 4.6 Heavy metal concentrations in different vegetables of dry season in the respective areas.

In the wet season (Fig. 4.7), the highest Zn concentration (114.7 mg/kg) was observed in helencha from Banglabazar, followed by water spinach root (66.9 mg/kg) and water spinach shoot (61.1 mg/kg) from the same area, and taro root (66.7 mg/kg) from Chandra. Taro leaf contained the lowest Zn concentration (22.9 mg/kg) in the Kashimpur area.

The Cr and Cu concentrations in taro root and water spinach root were high in Kashimpur; however, the Pb and Cd concentrations were high in Banglabazar. In eggplant, the concentrations of As and Cd were high in Kashimpur but low in Banglabazar. On the other hand, the Cr and Cu concentrations were high in eggplant from Banglabazar. Sponge gourd contained high As and Cd concentrations in Chandra and low As and Cd concentrations in Kashimpur, whereas the Cu and Pb concentrations in sponge gourd were high in Banglabazar. Regarding the total concentration of all heavy metals, helencha contained the highest concentration in all three areas; the concentration in water spinach root was the second highest in all three areas; and the sponge gourd, eggplant and taro leaf concentrations were low in the three areas. The small standard error values of the vegetable concentrations in the upper part of the bar graph (Fig. 4.7) indicate no significant differences between the areas.

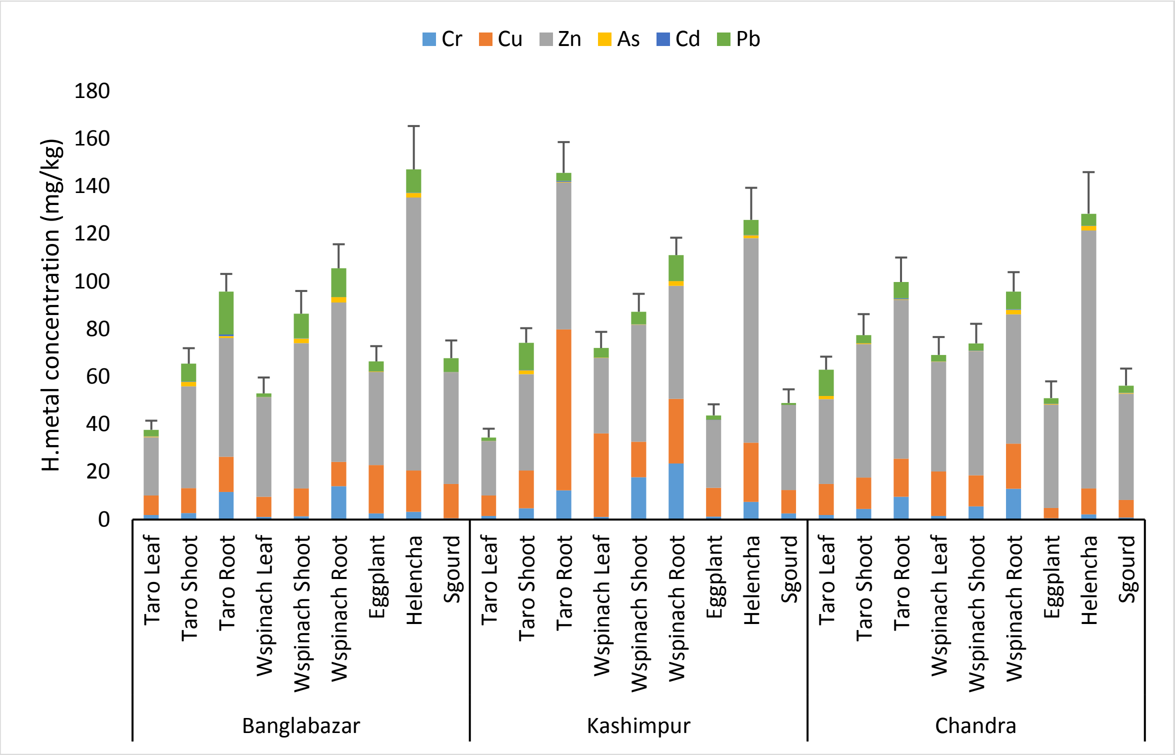

Fig. 4.7 Heavy metal concentrations in different vegetables of wet season in the respective areas.

In terms of the differences in root, leaf and fruit vegetables, the total heavy metal concentration was the highest in root vegetables, followed by leaf and then by fruit vegetables (Figs. 4.6 and 4.7). Root vegetables grown in the soil are thought to be more heavily contaminated than leaf and fruit vegetables that are grown in close contact with soil. Taro root has a short underground stem, while water spinach root is having roots developed from nodes. However, both plants grow in moist soils, making these vegetables easily contaminated with heavy metals, when the soil moisture is provided by irrigation water that contains high levels of heavy metals. In vegetables, heavy metal concentrations may be attributed due to the absorption and translocation of it within the plant body (Vousta et al., 1996). The absorption and translocation ability also depends on the kind of vegetables, differentiating the heavy metal concentrations among the vegetables (Islam and Hoque, 2014).

The root and leaf vegetables exceeded the FAO/WHO permissible limit for Cr, Zn, As and Pb concentrations in the wet season (Table 3.5). In the dry season, the root and leaf vegetables exceeded permissible limit for Cr, Zn, As, Cd and Pb concentrations. Therefore, root and leaf vegetables are found to be unsuitable for human consumption in both seasons.

4.5 Estimated daily intake of heavy metals and health risk assessment

The level of heavy metals toxicity to human being depends upon their daily consumption or intake (Singh et al., 2010). Long term exposure to heavy metals through the consumption of vegetables hamper biochemical process in humans (Ashwange et al., 2013). Thus, to assess the toxicity level of vegetables for human consumption in heavy metal concentrations, estimated daily intake of heavy metals (DIM) was calculated. The calculation method of estimated daily intake of heavy metals (DIM) was shown in chapter 2. DIM of heavy metals of vegetables grown in the dry and wet season is shown in Table 4.2 and 4.3, respectively.

In the vegetables of the dry season, the DIM values for Cr, Cu, Zn, As, Cd and Pb were ranged from 0.0007 to 0.0105, from 0.0016 to 0.0055, from 0.0066 to 0.0196, from 0.00004 to 0.00048, from 0.000001 to 0.000229, and from 0.00008 to 0.00189 mg/kg per day, respectively (Table 4.2). Among different vegetables in the dry season, radish showed the highest intake for Cr and Cu, whereas Indian spinach and red amaranth showed the highest intake for Zn and Cd, respectively. Taro indicates the high intake of As and Pb. The average DIM values of vegetables decreased in the order of Zn > Cr > Cu > Pb > As > Cd.

Table 4.2 DIM (mg/kg per day) in the respective vegetables of the dry season.

Division of vegetables	**Name**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**	**Total**
Leafy	Radish Leaf	0.0105	0.0055	0.0186	0.00030	0.000125	0.00111	0.036
	Amaranth	0.0026	0.0032	0.0104	0.00025	0.000111	0.00130	0.018
	Red Amaranth	0.0038	0.0030	0.0179	0.00029	0.000229	0.00139	0.027
	Spinach	0.0066	0.0035	0.0102	0.00029	0.000098	0.00136	0.022
	Indian spinach	0.0009	0.0016	0.0196	0.00020	0.000036	0.00062	0.023
	Pumpkin	0.0014	0.0027	0.0066	0.00009	0.000017	0.00155	0.012
	Bottle gourd	0.0020	0.0024	0.0095	0.00009	0.000024	0.00170	0.016
Root and leafy	Taro	0.0062	0.0037	0.0168	0.00048	0.000041	0.00189	0.029
Fruiting	Yard long bean	0.0007	0.0018	0.0072	0.00004	0.000001	0.00008	0.010
	Ave.	0.0038	0.0030	0.0130	0.0002	0.0001	0.0012	

Table 4.3 DIM (mg/kg per day) in the respective vegetables of the wet season.

Division of vegetables	**Name**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**	**Total**
Root and	Taro	0.0012	0.0040	0.0095	0.00016	0.000043	0.00156	0.016
Leafy	Water spinach	0.0019	0.0038	0.0107	0.00021	0.000019	0.00136	0.018
	Helencha	0.0009	0.0038	0.0220	0.00035	0.000029	0.00151	0.029
Fruiting	Eggplant	0.0003	0.0026	0.0079	0.00003	0.000016	0.00059	0.011
	Sponge gourd	0.0003	0.0023	0.0091	0.00003	0.000016	0.00069	0.012
	Ave.	0.0009	0.0033	0.0118	0.0002	0.00002	0.0011	

In the vegetables of the wet season (Table 4.3), the highest intakes of Cu, Cd and Pb were observed in taro. Helencha showed the highest intake for Zn and As, whereas water spinach exhibited the highest intake for Cr. The DIM values for vegetables in the wet season were lower than in the dry season, which may be due to the lower concentrations (heavy metals) in the wet season. The DIM values for Cr, Cu, Zn, As, Cd and Pb were ranged from 0.0003 to 0.0019, from 0.0023 to 0.004, from 0.0079 to 0.022, from 0.00003 to 0.00035, from 0.000016 to 0.000043, and from 0.00059 to 0.00156, respectively. The order of heavy metal intake for all vegetables are Zn > Cu > Pb > Cr > As > Cd.

The health risk index (HRI) calculated for the consumption of vegetables in the wet and dry season for the respective heavy metals, are presented in Table 4.4 and Table 4.5, respectively. The health risk index (HRI) is the ratio of daily intake of heavy metals (DIM) and reference dose (R_fD) (Pierzynski et al., 2000). In the dry season (Table 4.4), radish showed the highest HRI for Cr and Cu. Taro exhibited the highest HRI for As and Pb, whereas red amaranth and Indian spinach presented the highest HRI values for Cd and Zn. However, the highest HRI value (range) was observed for Cr (from 0.29 to 3.49) followed As (from 0.29 to 1.59), Cu (from 0.04 to 0.14) and Zn (from 0.02 to 0.07). The lowest mean HRI value was found for Cd which ranged from 0.002 to 0.459 followed by Pb (from 0.02 to 0.47). In total, leaf and root vegetables showed more than 1 HRI values. In the wet season (Table 4.5), taro indicated the highest HRI values for Cu, Cd and Pb. Helencha showed the highest HRI values for Zn and As, whereas water spinach showed the highest HRI value for Cr. HRI values for vegetables were lower in the wet season than in the dry season. The mean HRI values were found to be higher for As (0.53) followed by Cr (0.31), Cu (0.08) and Zn (0.04). The lowest HRI value was observed for Cd (0.05). In total, the leaf and root vegetables showed more than 1 HRI values.

Table 4.4 HRI for heavy metals in the respective vegetables in the dry season.

Division of vegetables	**Name**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**	**Total**
Leafy	Radish	3.49	0.14	0.06	1.00	0.250	0.28	5.22
	Amaranth	0.86	0.08	0.03	0.85	0.223	0.33	2.37
	Red Amaranth	1.26	0.07	0.06	0.98	0.459	0.35	3.18
	Spinach	2.20	0.09	0.03	0.95	0.197	0.34	3.81
	Indian spinach	0.29	0.04	0.07	0.67	0.072	0.15	1.29
	Pumpkin	0.48	0.07	0.02	0.29	0.034	0.39	1.28
	Bottle gourd	0.65	0.06	0.03	0.29	0.049	0.42	1.51
Root and leafy	Taro	2.08	0.09	0.06	1.59	0.082	0.47	4.38
Fruiting	Yard long bean	0.22	0.05	0.02	0.12	0.002	0.02	0.43
	Ave.	1.28	0.08	0.04	0.75	0.15	0.31	

Table 4.5 HRI for heavy metals in the respective vegetables in the wet season.

Division of vegetables	**Name**	**Cr**	**Cu**	**Zn**	**As**	**Cd**	**Pb**	**Total**
Root and	Taro	0.40	0.10	0.03	0.54	0.09	0.39	1.55
Leafy	Water spinach	0.63	0.09	0.04	0.69	0.04	0.34	1.83
	Helencha	0.30	0.09	0.07	1.17	0.06	0.38	2.08
Fruiting	Eggplant	0.11	0.07	0.03	0.12	0.03	0.15	0.50
	Sponge gourd	0.09	0.06	0.03	0.11	0.03	0.17	0.50
	Ave.	0.31	0.08	0.04	0.53	0.05	0.29	

To consider the health risk of any toxic heavy metals, it is necessary to estimate the exposure level of each heavy metal by quantifying its exposure sources to the target organisms (Khan et al., 2008). The food chain is one of the most important pathways among other pathways of pollutants to humans. If various heavy metal contaminated crops were consumed by people, the people face serious health risks. The vegetables produced in the study areas are supplied to local and wholesale markets of Gazipur district and Dhaka city, therefore, the calculated HRI for respective vegetables still deserves attention. The total HRI values for all vegetables were > 1 except for fruit vegetables (yard long bean, sponge gourd, and eggplant) in both seasons. Therefore, potential health risks for root and leaf vegetables in both seasons are a high concern. The heavy metals (Cr, Cu, Zn, As, Cd, and Pb) exposure through intake of these vegetables are considered to be a high health risk.

CHAPTER 5

General Conclusions

Following conclusions were derived from this study.

For irrigation water and vegetables, heavy metal concentrations exceeded their permissible levels, while in soil, nearly all heavy metals concentrations were below the permissible level. A significant effect of different factories that discharge different type of wastewater on the heavy metal concentrations was observed in irrigation water in each area, but not observed in soil and vegetables.

The order of heavy metal concentrations and hierarchical cluster analysis revealed that heavy metals in vegetables were sourced from the soil in the dry season, and from irrigation water in the wet season. In the wet season, heavy metal concentrations in irrigation water, soil, and vegetables were lower than in the dry season. For this reason, the followings were considered: rainfall occurs in the wet season, which dilutes the irrigation water, making the heavy metal concentrations in the irrigation water lower; the lower amount of heavy metals is adsorbed on soil particles due to the diluted irrigation water; and the diluted irrigation water contributes to the lower heavy metal concentrations in vegetables in the wet season. The heavy metal concentrations in vegetables exceeded the permissible levels, despite the heavy metal concentrations in the soil being under permissible levels, for which a high bio-concentration factor of vegetables was responsible.

The geoaccumulation index (Igeo) and pollution load index (PLI) values revealed that soil in the study areas were uncontaminated to moderately contaminated by the heavy metals in both seasons. From the ecological point of view, the study areas were in low risk to moderate risk according to the data of risk index (RI). The total health risk index (HRI) values for root and leaf vegetables were greater than 1 except for fruit vegetables in both seasons. So, the root and leaf vegetables are mostly expected to be high health risk. Vegetables in this area are shipped to the market and eaten by many people, however the vegetables were not suitable for food, and it was thought that some measures will be needed in the future.

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