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Heavy Metals in Commonly Consumed Root and Leafy Vegetables in Dhaka City, Bangladesh, and Assessment of Associated Public Health Risks

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Abstract

Background: Excess accumulation of heavy metals in frequently consumed food (e.g., vegetables, fruits) is a serious threat to human health. The concentration of heavy metals in four root vegetables and five leafy vegetables collected from the Kawran Bazar fresh vegetable market of Dhaka, Bangladesh, were analyzed. Average daily intake (ADI), hazard quotient (HQ), and hazard index (HI) were also estimated to assess the human health risks posed by the intake of heavy metals from the consumption of the studied vegetables. Apparent differences in the concentration of heavy metals present in different vegetables were observed.

Results: The presence and degree of contamination was assessed for chromium (Cr), cadmium (Cd), lead (Pb), nickel (Ni), copper (Cu), zinc (Zn), iron (Fe), and manganese (Mn). Concentrations of Cr, Cd, Ni, and Cu exceeded the maximum permissible limit (MPL) in some root vegetables. Cr, Cd, Pb, Ni, Cu, Zn, and Fe concentration were higher in most studied leafy vegetables. ADI was found to be lower than the maximum permitted tolerable daily intake in most cases except Cr (0.202 mg/person day) and Fe (19.681 mg/person day) for the consumption of turnip and mint, respectively. Hazard quotient of Cr for turnip (1.121) and Zn for spinach (1.104), as well as the hazard indices of turnip (1.541), mustard (1.663), spinach (2.113), coriander (1.925), and mint (2.834), exceeded unit value, signifying potential health hazard from the dietary intake of the studied vegetables.

Conclusions: Higher concentrations of heavy metals were found in leafy vegetables compared to the root vegetables. Hence, the consumption of leafy vegetables has higher potential health risks than root vegetables. This study suggests that regular monitoring of heavy metals in vegetables is essential to prevent health risks associated with consuming heavy metal contaminated vegetables.

Keywords: Heavy metals, Root vegetables, Leafy vegetables, Hazard quotient, Average daily intake, Hazard index

Background

Heavy metals have drawn much attention due to their ubiquity, toxicity at trace level, and persistence in the environment (Ali et al. 2019; Jia et al. 2018). The exposure of heavy metals in the environment is increasing with the expansion of urbanization and industrialization. Both geological sources and anthropogenic activities contribute to heavy metal contamination (Intawongse and

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Dean 2006; Wilson and Pyatt 2007). Significant sources of heavy metals can be from point sources (e.g., thermal power plants, leather, coal mines, goldmines, smelting, textiles, and electronics waste processing) and non-point sources (e.g., soil erosion, urban runoff, and open freight storage) (Rai et al. 2019). When soil or irrigation water becomes contaminated by heavy metals, it perturbs the environment as well as the ecosystem (Mahmud et al. 2017). Mainly, soils act as a sink for those heavy metals. Consequently, the heavy metals contaminated soil serves as the source of heavy metals and transports them to water, vegetables, animals, and humans (Mondol et al. 2019; Waheed et al. 2019). Having higher bioaccumulation tendencies, edible plants such as vegetables are more susceptible to heavy metal contamination through bio-uptake (Intawongse and Dean 2006). The consumption of heavy metals contaminated vegetables can cause several acute human health issues (Duruibe et al. 2007; Pan et al. 2016; Rehman et al. 2018; Singh et al. 2010).

The situation is more acute in underdeveloped and developing countries with almost no monitoring and enforcement of environmental regulations (Chowdhury et al. 2016). Despite its well-known health risks, farmers in many developing countries use municipal wastewater in irrigation (Mahmud et al. 2017; Raja et al. 2015). While resulting in higher yield when using municipal wastewater, it comes with inevitable dire consequences such as soil and vegetable contamination (Waheed et al. 2019) (Fig. 1). Additionally, easy and unregulated excess use of fertilizers and pesticides further contributes to heavy metal pollution (Rodríguez Martín et al. 2006; Wångstrand et al. 2007). Pesticides used in Bangladesh increased dramatically from 7350 metric tons in 1992 to 45,172 metric tons in 2010 (Akter et al. 2018). The lack of proper environmental monitoring and enforcement makes it easier to overlook the side effects.

In Bangladesh, vegetables are one of the most consumed staple foods. Therefore, if the vegetables are contaminated, they can put a vast population at risk. Many studies reported high heavy metals [e.g., chromium (Cr), cadmium (Cd), lead (Pb), nickel (Ni), copper (Cu), zinc (Zn), iron (Fe), and manganese (Mn)] content in different vegetables (Ali and Al-Qahtani 2012; Guerra et al. 2012; Huang et al. 2020; Sharma et al. 2007, 2009; Manzoor et al. 2018; Mishra et al. 2020; Paradelo et al. 2020). Mi et al. (2019) have observed 35 common Chinese cabbage genotypes and studied the accumulation of five different heavy metals (Cd, Cr, Hg, Pb, and As) (Mi et al. 2019). Their results indicated a higher accumulation of Cd in the cabbage than the others. Another study conducted in Iran also studied the concentration of Hg and Pb in ten different leafy vegetables and herbs and found a high

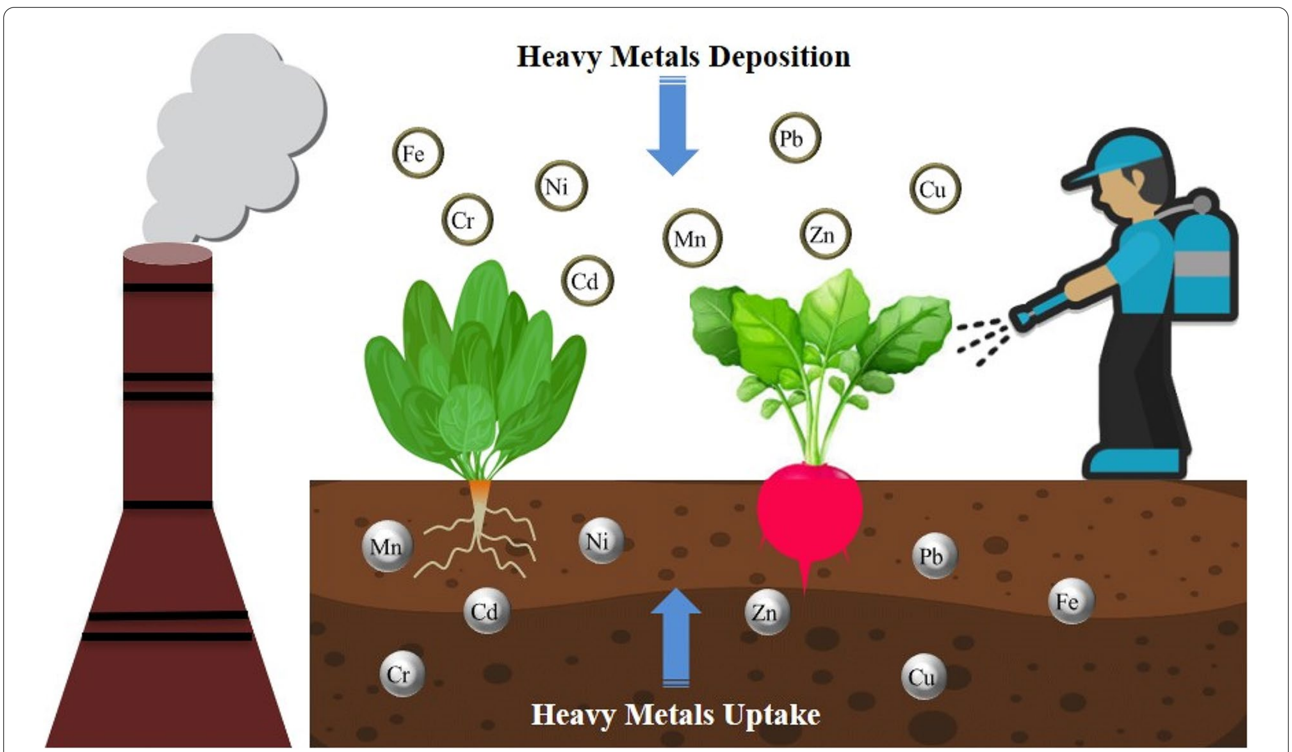

Fig. 1 Conceptual schematic of the uptake and deposition of heavy metals in root and leafy vegetables

level of contamination compared to WHO/FAO standards (Ghasemidehkordi et al. 2018). Sharma et al. (2007) also conducted a study to observe the heavy metals contamination due to wastewater irrigation in suburban areas of Varanasi, India (Sharma et al. 2007). According to their findings, the use of wastewater in irrigation has increased the contamination of Cd, Pb, and Ni in the edible portion of vegetables. The heavy metal uptake capacity of any specific vegetable depends on the climate, soil type, period of irrigation, atmospheric depositions, the concentration of heavy metals in soil, and the degree of maturity of the plants at the time of harvesting (Ghasemidehkordi et al. 2018; Voutsa et al. 1996). Therefore, studies conducted in other countries are not fully capable of representing the level of contamination in Bangladesh. Shaheen et al. (2016) studied ten commonly grown fruits and vegetables in Bangladesh (fruit, legume, root) for heavy metal contamination (Shaheen et al. 2016). The study showed that Cd in tomato and Pb in mango had exceeded the maximum allowable concentration set by WHO/FAO. This study primarily focused on fruit vegetables and analyzed very few leafy vegetables. It is evident from previous studies that leafy vegetables are most susceptible to heavy metal uptake than other types of vegetables (Huang et al. 2020). Besides, it also did not consider the effect of the harvesting period (early or late season harvesting) on the level of contamination. Since different vegetables have different mechanisms of accumulating heavy metals, other vegetable species may not present the same characteristics (Zhou et al. 2016). Therefore, the present research is designed to explore (i) the degree of heavy metals contamination in leafy and root vegetables, (ii) the effect of harvesting period (early, mid-time, or late season harvesting) on the degree of contamination, and (iii) public health risks associated with consumption of vegetables contaminated with heavy metals. Four species of root vegetables and five leafy vegetables commonly grown and consumed throughout Bangladesh were collected from the local market and analyzed to monitor heavy metal concentration. The concentration of chromium (Cr), cadmium (Cd), lead (Pb), nickel (Ni), copper (Cu), zinc (Zn), iron (Fe), and manganese (Mn) in those vegetables were determined. The hazard quotient (HQ) and the hazard indices (HI) associated with these heavy metals in different vegetables were also calculated to assess the public health risk.

Materials and methods

Study area and sample collection

The root and leafy vegetables were collected from the Kawran Bazar vegetable market, the biggest fresh produce market, which serves as the supply hub of vegetables for the entire Dhaka city. Therefore, the vegetables collected from this market represent the quality of the vegetables consumed by the residents of Dhaka city. Around 60 to 70 percent of the Dhaka city's demand of vegetables are meet up by vegetables grown in Manikganj, Tangail, Narsingdi, Munshiganj, Savar, Keraniganj and Dhamrai districts. Four commonly grown and popular native root and five leafy vegetable samples were collected in four phases with at least 14 days intervals for two months. Phasing out the sample collection over two months helps better understand the level of contamination at different harvesting periods (early, mid-time, and late harvesting). In each sampling phase, 3 replications for each sample of root and leafy vegetables were randomly purchased to represent a random resident's intake of heavy metals. To increase the reproducibility of the result a composite sample was prepared for the analysis by mixing the three replications. The root and leafy vegetables that are popularly consumed by the people of this country were selected for this study.

Sample preparation and preservation

The collected fresh vegetable samples were washed thoroughly with clean tap water and then washed three times using distilled water to wash out foreign particles and surface pollutants adhering to the surfaces. The cleaned samples were sliced into small pieces and air-dried in the open air for 2 h to eliminate excess moisture. The weight of each fresh vegetable sample was taken, then dried in an induction oven approximately at 65 °C for about 90 h and reweighed the dry weight. The moisture content was determined (Table 1). The dried vegetable samples were ground using pestle and mortar and sieved through a 0.20 mm screen to homogenize the samples. The

Table 1 Description of the vegetable samples collected from Kawran Bazar vegetable market, Dhaka, Bangladesh

Common name	Scientific name	Moisture content (%)
Root vegetables		
Beet	*Beta vulgaris*	88.0
Radish	*Raphanus sativus*	95.3
Carrot	*Daucus carota*	88.0
Turnip	*Brassica rapa*	91.0
Leafy vegetables		
Mustard	*Brassica campestris*	92.0
Cabbage	*Brassica oleracea*	93.0
Spinach	*Spinacia oleracea*	91.0
Coriander	*Coriandrum sativum*	93.4
Mint	*Mentha arvensis*	85.2

powdered samples were then preserved at room temperature until further analysis.

Digestion of the sample

A widely used acid-based digestion method was used for the plant sample digestion (Alain et al. 2021; Blum et al. 1996; Samuel and Babatunde 2021). Briefly, one gram of the oven-dried sample was weighed into a 100 mL beaker, followed by the addition of 10 mL of concentrated HNO_3 (analysis grade, Merck-Germany) and 2 mL concentrated $HClO_4$ (Merck-Germany). The digestion was carried out on a hot plate at 150–200 °C. Then, the digested samples were taken out and cooled down to room temperature. After cooling down, the samples were filtered and transferred into a volumetric flask filled with DI water up to 100 mL. This 100 mL sample stock solution was then stored in a plastic bottle for further analysis.

Analysis of the samples

The concentration of heavy metals such as chromium, cadmium, lead, nickel, copper, zinc, iron, and manganese in the extracts was analyzed using an Atomic Absorption Spectrophotometer (Varian AA 240). The measurements were made using a hollow cathode lamp of Cr, Cd, Pb, Ni, Cu, Zn, Fe, and Mn at wavelengths of 357.9, 228.8, 283.3, 232, 324.7, 213.9, 248.3, and 279.5 nm, having detection limits of 1.7963, 0.1986, 100, 3.5473, 20.0, 5.1260, 30.0, and 10.0 µg/L, respectively. The sensitivity was checked and maintained using the standard solutions intermittently.

Average daily intake (ADI)

Human health risks due to the transfer of contaminants to the human body are frequently assessed using many indices such as average daily intake (ADI), hazard quotient (HQ), and hazard index (HI) (Cui et al. 2004; Gall et al. 2015; Zhuang et al. 2009). The ADI was calculated using the equation below (Kacholi and Sahu 2018):

$$ADI = Av_{consumption} \times \%DW_{vegetable} \times C_{heavy\,metal} \quad (1)$$

where ADI is the average daily intake of heavy metal per person per day (mg/person day), $Av_{consumption}$ is the average daily consumption of vegetables per person per day (g/person day), % $DW_{vegetable}$ is the percentage of the dry weight of vegetables (%DW = [(100 − % moisture)/100]), and $C_{heavy\ metal}$ is the average heavy metal concentration of dry weight vegetable (mg/g). The average daily consumption of vegetables reported by the household income and expenditure survey of Bangladesh is 166.1 g per person as of 2011 (Ibrahim and Mollah 2011). The value 166.1 g/person day was therefore used in calculating the ADI values, and the average weight of a person was considered to be 60 kg (FAO/WHO 1993). If the ADI is above the maximum permissible daily intake value, it may cause various health hazards.

Hazard quotient (HQ)

Hazard quotient (HQ) is the ratio of exposure of a contaminant to the level at which there will be no expected health risk from the exposure of that contaminant. The HQ value of less than one is considered to pose no potential health risks. But when the HQ is higher than 1, it means potential health risks can be expected due to the exposure to any specific contaminant (Bermudez et al. 2011; Chary et al. 2008). The following equation has been used to calculate the hazard quotient proposed by (Granero and Domingo 2002):

$$HQ = ADI/R_f D \quad (2)$$

where ADI represents the average intake of vegetables per day (mg/kg day), and R_fD depicts the oral reference dose of the metal of interest (mg/kg/day). R_fD represents the tolerable daily exposure of any specific contaminant without any major risk of health effects throughout the lifetime of a person. The R_fD values for Pb (0.004), Zn (0.30), Cu (0.04), Cd (0.0005), Cr (0.0003), Ni (0.02), Fe (0.70), and Mn (0.14) in mg/kg day unit are set by the WHO/FAO (2013).

Hazard index (HI)

While hazard quotient (HQ) is used for assessing the health risks due to any single contaminant, hazard index (HI) is used to evaluate the potential health risks when a person is exposed to multiple contaminants at the same time. This is mainly because exposure to more than one contaminant has additive effects. Therefore, HI can be an effective tool to understand the potential health risks that are associated with human exposure to multiple contaminants simultaneously. Same as hazard quotient, an HI value of greater than 1 signifies potential health risks. The HI is the sum of the hazard quotients for each pollutant, as shown in the following equation (Kacholi and Sahu 2018):

$$HI = \sum_{i=1}^{n} HQ \quad (3)$$

Statistical analysis

The statistical analysis of the experimental data was performed using ANOVA (Analysis of Variance) and Duncan's Multiple Range Test (DMRT) in IBM SPSS (Version 20) as outlined by (Kwanchai A. Gomez 1984). The latter was used for testing the significance of differences

Table 2 Concentration of heavy metals in root vegetables

Heavy metals	Root vegetables	1st phase (mg/kg dw)	2nd phase (mg/kg dw)	3rd phase (mg/kg dw)	4th phase (mg/kg dw)	Range (mg/kg dw)	Mean (mg/kg dw)	Maximum permissible limit (MPL) (mg/kg dw)
Chromium (Cr)	Beet	0.60	0.90	1.40	1.90	0.60–0.90	1.20 a	2.30 (FAO/WHO 2011)
	Radish	2.00	1.20	0.80	2.10	0.80–2.10	1.53 a	
	Carrot	1.80	0.50	1.80	2.20	0.50–2.20	1.58 a	
	Turnip	**2.70**	0.80	0.90	**49.60**	0.80–49.6	**13.50 a**	
Cadmium (Cd)	Beet	0.10	0.20	**0.40**	0.10	0.10–0.40	**0.20 a**	0.20 (FAO/WHO 2001)
	Radish	**0.30**	0.10	**0.50**	0.00	0.00–0.50	**0.23 a**	
	Carrot	**0.50**	**0.40**	**0.30**	0.00	0.00–0.50	**0.30 a**	
	Turnip	0.10	0.20	0.00	0.00	0.00–0.20	0.08 a	
Lead (Pb)	Beet	0.00	0.00	0.00	0.00	0.00–0.00	0.00 a	0.30 (FAO/WHO 2011)
	Radish	0.00	0.00	0.00	0.00	0.00–0.00	0.00 a	
	Carrot	0.00	**3.00**	0.00	0.00	0.00–3.00	**0.75 a**	
	Turnip	0.00	**1.00**	0.00	0.00	0.00–1.00	0.25 a	
Nickel (Ni)	Beet	0.00	0.00	0.80	1.50	0.00–1.50	0.58 a	2.70 (FAO/WHO 2011)
	Radish	**3.30**	0.00	1.60	1.50	0.00–3.30	1.60 a	
	Carrot	0.00	0.00	0.60	0.90	0.00–0.90	0.38 a	
	Turnip	1.10	0.00	**4.50**	0.00	0.00–4.50	1.40 a	
Copper (Cu)	Beet	**11.40**	8.10	**19.30**	**16.80**	8.10–19.3	**13.90 b**	10 (FAO/WHO 2011)
	Radish	9.50	2.00	6.50	9.10	2.00–9.50	6.78 a	
	Carrot	2.80	3.60	4.10	4.20	2.80–4.20	3.68 a	
	Turnip	8.10	7.90	7.10	5.40	5.40–8.10	7.13 a	
Zinc (Zn)	Beet	**61.75**	35.99	**50.21**	**61.99**	35.99–61.99	**52.49 b**	50 (FAO/WHO 2011)
	Radish	**61.60**	11.77	48.65	**68.03**	11.77–68.03	47.51 b	
	Carrot	10.50	17.55	7.28	9.39	7.28–17.55	11.18 a	
	Turnip	**50.95**	24.09	42.76	45.37	24.09–50.95	40.79 b	
Iron (Fe)	Beet	138.10	103.00	133.70	446.40	103.00–446.40	205.30 a	450 (FAO/WHO 2007)
	Radish	292.30	37.40	82.30	248.20	37.40–292.30	165.05 a	
	Carrot	47.90	119.70	58.50	149.30	47.90–149.30	93.85 a	
	Turnip	96.50	150.50	41.80	85.30	41.80–150.50	93.53 a	
Manganese (Mn)	Beet	13.9	19.3	28.7	37.8	13.9–37.8	24.93 a	500 (FAO/WHO 2007)
	Radish	64.7	4.2	23.1	30.7	4.2–64.7	30.68 a	
	Carrot	10.3	11.4	13.5	21	10.3–21.0	14.05 a	
	Turnip	11.1	11.9	27.6	13	11.1–27.6	15.90 a	

*DMRT analysis has been performed separately for each heavy metal. Mean values with the same letter(s) in a column of an individual heavy metal state no significant difference from each other at a 5% level by DMRT. The values in bold exceeded the maximum permissible limit set by the FAO/WHO

between mean values. The 0.05 level of significance was chosen for statistical judgment.

Results and discussion

Heavy metal concentration in root vegetables

Concentrations of 8 different heavy metals (Cr, Cd, Pb, Ni, Cu, Zn, Fe, and Mn) in 4 frequently consumed root vegetable species are determined (Table 2). The root vegetables studied for heavy metal are beet, radish, carrot, and turnip. All heavy metal concentrations are expressed on a dry weight basis. Also, the effects of early (1st phase), mid-time (2nd and 3rd phases), and late (4th phase) harvesting on the metal concentrations are evaluated.

In the case of chromium (Cr), the highest concentration (49.60 mg/kg dw) was recorded in turnip that was harvested in the late-season (4th phase) among all four root vegetables. On the other hand, the lowest chromium concentration (0.50 mg/kg dw) was recorded in the carrot that was harvested in the mid-time season (2nd phase). The concentration of Cr was also found to be higher in turnip collected in the 1st and 4th phases. The overall concentration of Cr was found to be in turnip > carrot > radish > beet order. The very high Cr value

(49.60 mg/kg dw) made us concerned, and we did the analysis several times and found a similar value. This high value can be ascribed to the high use of preservatives at the end of the season. One study from Bangladesh also observed lower concentrations of heavy metals in carrots collected from 30 agro-ecological zones of Bangladesh (Shaheen et al. 2016). Among all four types of root vegetables, the only turnip exceeded the maximum permissible limit (MPL) over the season.

Average cadmium (Cd) concentration exceeded the MPL in all root vegetables except turnip. Our data also shows that beet, radish, and carrot harvested during the mid-time season (3rd phase) had higher heavy metal content than all other harvesting periods. On the contrary, late-harvested root vegetables resulted in having the lowest heavy metal concentrations. While the highest Cd concentrations were recorded in radish and carrot (0.50 mg/kg dw), turnip had the lowest concentration (0 mg/kg dw) overall. Some recent studies from Bangladesh also reported the Cd concentrations found to be 0.001–1.60 mg/kg in 12 vegetable species (Islam et al. 2015), 0.006–0.3 mg/kg in 16 vegetable species (Rahman et al. 2013), and 0.60 mg/kg in 5 vegetable species (Ahmad and Goni 2010).

Lead (Pb) contamination was relatively lower than the other heavy metals for the root vegetables. The lead was detected only in cases of carrot (3.0 mg/kg dw) and turnip (1.0 mg/kg dw) harvested during the 2nd phase and exceeded the MPL as well. Otherwise, there was no lead contamination. Pb concentration in five commonly grown vegetables (eggplant, chili, ladies' finger, tomato, and green cabbage) was reported by Ahmad and Goni (2010) to be 3.9 mg/kg (Ahmad and Goni 2010).

Nickel (Ni) contamination in the root vegetables was also within the maximum permissible limit except for radish (3.30 mg/kg dw) harvested in the early season and turnip (4.50 mg/kg dw) harvested in the mid-time (3rd phase) season. Surprisingly, all the root vegetables collected in 2nd phase did not contain any of those heavy metals. The highest Ni concentration was recorded in turnip (4.50 mg/kg dw) harvested in the mid-time (3rd phase) season. Recent literature reported Ni concentrations in vegetables from different regions of Bangladesh as 0.02–12 mg/kg in 12 vegetable species (Islam et al. 2015), 0.3–4.7 mg/kg in 16 vegetable species (Rahman et al. 2013), and 3.0 mg/kg in 5 vegetable species (Ahmad and Goni 2010).

Among all four root vegetables, the beet only exceeded the MPL of copper (Cu) set by the FAO/WHO. While the highest Cu concentration was recorded in beet (19.30 mg/kg dw) during the 3rd phase, radish had the lowest concentration (2.0 mg/kg dw) during the 2nd phase. A study from Bangladesh reported the Cu concentration in common vegetables (tomato, green chili, brinjal, bean, potato, onion, and carrot) in the range of 2.254–9.718 mg/kg (Shaheen et al. 2016).

The concentration of zinc (Zn) was also measured in all four species of root vegetables. During the early season harvesting, all except carrot exceeded the MPL. Maximum Zn concentration was found in radish (68.03 mg/kg dw) during the late harvesting season. The lowest concentration was found in carrots (7.28 mg/kg dw) during the 3rd phase of harvesting. The average Zn concentration only in the beet exceeded the MPL throughout the season. A previously mentioned study also found the lowest concentration of Zn in carrot 0.074–4.75 mg/kg (Shaheen et al. 2016). In another study, the median concentration of Zn in common vegetables (16 vegetable species) in Bangladesh was reported to be 50 mg/kg (Rahman et al. 2013).

Iron (Fe) concentration was significantly higher in all root vegetables concerning other heavy metals that are analyzed in this study. But under no circumstances the concentration of Fe exceeded the maximum permissible limit. The highest concentration of Fe was found in late-harvested beet (446.4 mg/kg dw), while the lowest was found in radish (37.4 mg/kg dw) harvested during the mid-time season (2nd phase). The overall concentration of Fe was found to be in beet > radish > carrot > turnip order. One study from Tanzania has analyzed four different vegetables (potato leaves, African spinach, ladies' finger, and brinjal) and observed a mean Fe concentration of 48.40–136.40 mg/kg (Kacholi and Sahu 2018).

Like Fe, manganese (Mn) also did not exceed the MPL in any of the root vegetables and was well below the MPL. Maximum Mn concentration was found in radish (64.7 mg/kg dw) during the early harvesting season. The overall concentration of Mn was found to be in radish > beet > turnip > carrot order. In a study, the median concentration of Mn in common vegetables in Bangladesh was reported to be 65 mg/kg (Rahman et al. 2013).

Heavy metal concentration in leafy vegetables

Five different leafy vegetables (mustard, cabbage, spinach, coriander, and mint) were studied for 8 different heavy metals (Cr, Cd, Pb, Ni, Cu, Zn, Fe, and Mn). Mean concentrations of the heavy metals in five studied leafy vegetables revealed significant variation ($p < 0.05$) for Cd, Ni, Zn, and Fe but revealed no variation for Cr, Pb, Cu, and Mn (Table 3). Table 3 summarizes the metal concentrations found in all leafy vegetable samples throughout the season.

Maximum Cr concentration was found in mint (16.7 mg/kg dw) harvested in the 3rd phase, while the lowest concentration was recorded in cabbage (0 mg/kg dw) harvested in the 2nd phase. The average

Table 3 Concentration of heavy metals in leafy vegetables

Heavy metals	Leafy vegetables	1st phase (mg/kg dw)	2nd phase (mg/kg dw)	3rd phase (mg/kg dw)	4th phase (mg/kg dw)	Range (mg/kg dw)	Mean (mg/kg dw)	Maximum permissible limit (MPL) (mg/kg dw)
Chromium (Cr)	Mustard	**7.70**	**2.70**	**5.50**	**2.90**	2.70–7.70	**4.70 a**	2.30 (FAO/WHO 2011)
	Cabbage	1.70	0.00	1.30	**2.40**	0.00–2.40	1.35 a	
	Spinach	1.40	**2.50**	1.70	**3.30**	1.40–3.30	2.23 a	
	Coriander	1.50	**2.50**	**5.00**	**6.50**	1.50–6.50	**3.88 a**	
	Mint	**2.60**	1.40	**16.70**	**4.00**	1.40–16.70	**6.18 a**	
Cadmium (Cd)	Mustard	**0.40**	**0.40**	0.00	0.00	0.00–0.40	0.20 a	0.20 (FAO/WHO 2001)
	Cabbage	0.00	**0.40**	0.00	0.00	0.00–0.40	0.10 a	
	Spinach	**0.90**	**0.60**	**0.50**	0.00	0.00–0.90	**0.50 a**	
	Coriander	**0.60**	**1.80**	**1.00**	**1.10**	0.60–1.80	**1.13 b**	
	Mint	**0.50**	**0.30**	0.00	0.00	0.00–0.50	0.20 a	
Lead (Pb)	Mustard	0.00	**5.00**	0.00	**1.80**	0.00–5.00	**1.70 a**	0.30 (FAO/WHO 2011)
	Cabbage	0.00	0.00	**3.00**	0.00	0.00–3.00	**0.75 a**	
	Spinach	0.00	**2.00**	**0.68**	0.00	0.002.00	**0.67 a**	
	Coriander	0.00	**3.00**	**5.00**	0.00	0.00–5.00	**2.00 a**	
	Mint	0.00	**2.00**	**6.00**	**1.00**	0.00–6.00	**2.25 a**	
Nickel (Ni)	Mustard	1.70	1.50	**6.50**	**3.20**	1.70–6.50	**3.23 b**	2.70 (FAO/WHO 2011)
	Cabbage	0.00	0.00	2.70	0.00	0.00–2.70	0.68 ab	
	Spinach	1.70	0.00	0.30	0.90	0.00–1.70	0.73 a	
	Coriander	1.70	2.50	**3.50**	**4.50**	1.70–4.50	**3.05 b**	
	Mint	0.50	0.00	**3.70**	1.60	0.00–3.70	1.45 ab	
Copper (Cu)	Mustard	5.70	5.30	**44.30**	**17.90**	5.30–44.3	**18.30 a**	10 (FAO/WHO 2011)
	Cabbage	1.50	3.10	8.40	4.30	1.50–8.40	4.30 a	
	Spinach	**11.40**	**14.40**	**10.10**	4.00	4.00–14.40	9.98 a	
	Coriander	17.40	8.70	**16.90**	**15.00**	8.70–17.40	**14.50 a**	
	Mint	**16.50**	**10.40**	**11.70**	**12.20**	10.4–16.5	**12.70 a**	
Zinc (Zn)	Mustard	43.57	49.27	**125.78**	**72.59**	43.57–125.78	**72.80 a**	50 (FAO/WHO 2011)
	Cabbage	21.06	19.63	**51.27**	12.25	12.25–51.27	26.05 a	
	Spinach	**233.82**	**125.38**	**184.99**	**197.80**	125.38–233.82	**185.50 b**	
	Coriander	32.57	**228.33**	**71.14**	**68.97**	32.57–228.33	**100.25 a**	
	Mint	35.56	46.00	**57.92**	**78.85**	35.56–78.85	**54.58 a**	
Iron (Fe)	Mustard	295.40	**916.70**	**1450.10**	**880.40**	295.40–1450.10	**885.65 b**	450 (FAO/WHO 2007)
	Cabbage	66.90	67.00	63.70	103.30	63.70–103.30	75.23 a	
	Spinach	177.10	**920.90**	**625.40**	**787.20**	177.10–920.90	**627.65 ab**	
	Coriander	**535.20**	**615.90**	**1588.70**	**2365.70**	535.20–2365.70	**1276.38 b**	
	Mint	**920.90**	**619.80**	**1198.10**	**463.60**	463.60–1198.10	**800.60 ab**	
Manganese (Mn)	Mustard	51	35.8	389.3	157.9	35.8–389.3	158.50 a	500 (FAO/WHO 2007)
	Cabbage	13.7	16.3	43.5	23.5	13.7–43.5	24.25 a	
	Spinach	177.9	33.4	130.4	194.9	33.4–194.9	134.15 a	
	Coriander	36.5	324.6	89.1	143.1	36.5–324.6	148.33 a	
	Mint	66.2	58.7	201.8	127.1	58.7–201.8	113.45 a	

*DMRT analysis has been performed separately for each heavy metal. Mean values with the same letter(s) in a column of an individual heavy metal state no significant difference from each other at a 5% level by DMRT. The values in bold exceeded the maximum permissible limit set by the FAO/WHO

concentration of Cr exceeded the MPL in mustard (4.70 mg/kg dw), coriander (3.88 mg/kg dw), and mint (6.18 mg/kg dw). Also, all the leafy vegetables harvested in the late season exceeded the MPL of Cr concentration. Besides, mustard has exceeded the MPL in all harvesting seasons.

Higher Cd contamination was observed during the first two phases of harvesting compared to the latter two. All the leafy vegetables harvested during the 2nd phase exceeded the MPL. The highest concentration of Cd was found in coriander (1.80 mg/kg dw) harvested in the 2nd phase. Also, the concentration of Cd in coriander exceeded the MPL in all harvesting seasons.

Though there was no Pb contamination found in early harvested leafy vegetables, the average concentrations of Pb exceeded the MPL in all leafy vegetables by the end of the season. Maximum Pb concentration was found in mint (6.0 mg/kg dw) harvested in the 3rd phase. Overall concentrations of Pb in the leafy vegetables are found to be in mint > coriander > mustard > cabbage > spinach order.

Contamination by Ni was relatively lower than the contamination by chromium, cadmium, and lead. During the first two phases, the Ni concentrations were below the MPL set by FAO/WHO. Considering all four phases, the average Ni concentration only in mustard (3.23 mg/kg dw) and coriander (3.05 mg/kg dw) exceeded the MPL. The highest concentration of Ni was recorded in coriander (4.50 mg/kg dw) harvested in the late season. Research from Sri Lanka also found that Ni, Cd, Cr, and Pb levels exceeded the MPL set by FAO/WHO (2001) in five different green leafy vegetables (Kananke et al. 2014).

Maximum Cu concentration was found in mustard (44.30 mg/kg dw) harvested in the mid-time season (3rd phase), while the lowest concentration was recorded in cabbage (1.50 mg/kg dw) harvested in the early season. Also, Cu contaminations were much higher in the second half of the harvesting season than in the first half. Considering all four phases, the average Cu concentration in mustard (18.3 mg/kg dw), coriander (14.5 mg/kg dw), and mint (12.7 mg/kg dw) exceeded the MPL. Besides, mint has exceeded the MPL in all harvesting seasons.

Zn has also demonstrated higher concentrations in the second half of the harvesting season than in the first half. The average Zn concentrations in all leafy vegetables except cabbage have exceeded the MPL. The highest concentration of Zn was found in spinach (233.82 mg/kg dw) harvested in the early season, while the lowest concentration was found in cabbage (12.25 mg/kg dw) harvested in the late season. The average concentration of Zn decreased in the order of spinach > coriander > mustard > mint > cabbage.

Fe concentration was significantly higher in all leafy vegetables concerning other heavy metals analyzed in this study. The highest concentration of Fe was found in late-harvested coriander (2365.7 mg/kg dw), while the lowest was found in cabbage (63.7 mg/kg dw) harvested during the mid-time season (3rd phase). Also, coriander and mint have exceeded the MPL in all harvesting seasons. One study reported that the average concentration of Fe, Pb, and Cd in the leafy vegetables collected from the Satkhira district of Bangladesh exceeded the permissible limit (Uddin and Dhar 2019). Overall concentrations of Fe have decreased in the order of coriander > mustard > mint > spinach > cabbage order.

Under no phases and conditions, Mn has exceeded the MPL in any leafy vegetables. Maximum Mn concentration was found in mustard (389.30 mg/kg dw) harvested in the mid-time season (3rd phase). The lowest concentration was found in cabbage (13.7 mg/kg dw) harvested in the early season.

The major sources of heavy metals in the studied root and leafy vegetables might be due to soil contamination. Besides, solid waste disposal, sludge applications, vehicular exhaust, and agrochemicals are the most prominent reasons behind soil contamination. Therefore, contamination of agricultural soils through these anthropogenic activities leads to excessive heavy metal uptake by vegetables, which in turn affects food quality and safety (Muchuweti et al. 2006). And the difference in heavy metals among different phases of sampling can be attributed to the land in which the particular vegetable had grown, probably having a different level of contamination, or at the end of the season when there is scarcity for a particular vegetable, the tendency to use preservative increases.

Assessment of public health risks

Average daily intake (ADI) of heavy metals

Heavy metals have a toxic impact, but the detrimental effect becomes apparent only when long-term consumption of contaminated vegetables occurs. Humans exposed to a chronic level of toxic heavy metals through the consumption of vegetables may have apparent impacts after years of exposure (Bahemuka and Mubofu 1999; Ikeda et al. 2000). Also, it has been reported that the consumption of food contaminated with heavy metals can significantly reduce essential body nutrients. This nutrient reduction can often result in growth retardation, immunological deficiency, disabilities, and a high prevalence of upper gastrointestinal cancer rates (Iyengar and Nair 2000; Türkdoğan et al. 2003).

The ADI of heavy metals was calculated according to the concentration of each metal in the vegetable samples. The mean ADI values and the Permitted Maximum Tolerable Daily Intake (PMTDI) of the studied metals via dietary intake of the studied root and leafy vegetables are represented in Table 4. The ADI values of the heavy metals from the consumption of studied root vegetables revealed no significant variation except for Cu. On the other hand, the ADI values of the heavy metals from the consumption of studied leafy vegetable samples varied significantly except for Pb. The ADI values of all the heavy metals were below the PMTDI except for Cr by consumption of turnip (0.202 mg/person day) and Fe by

Table 4 Average daily intake (ADI) of heavy metals (mg/person day)

Heavy metals	Average daily intake (mg/person day)							
	Cr	Cd	Pb	Ni	Cu	Zn	Fe	Mn
PMTDI	0.20 (National Research Council Subcommittee 1989)	0.046 (FAO/WHO 2003)	0.21 (FAO/WHO 2003)	0.30 (WHO 1996)	2.0 (FAO/WHO 2011)	20.0 (FAO/WHO 2011)	17.0 (FAO/WHO 2011)	5.0 (National Institute of Nutrition 2009)
Root vegetables								
Beet	0.024 a	0.004 a	0.000 a	0.011 a	0.277 b	7.692 a	4.092 a	0.497 a
Radish	0.012 a	0.002 a	0.000 a	0.012 a	0.053 a	2.627 a	1.288 a	0.239 a
Carrot	0.031 a	0.006 a	0.015 a	0.007 a	0.073 a	1.758 a	1.871 a	0.280 a
Turnip	**0.202 a**	0.001 a	0.004 a	0.021 a	0.107 a	4.384 a	1.398 a	0.238 a
Leafy vegetables								
Mustard	0.062 b	0.003 a	0.023 a	0.043 b	0.243 ab	8.371 ab	11.769 b	2.106 ab
Cabbage	0.016 a	0.001 a	0.009 a	0.008 a	0.050 a	2.478 a	0.875 a	0.282 a
Spinach	0.033 ab	0.007 ab	0.010 a	0.011 a	0.149 ab	19.865 b	9.383 ab	2.005 ab
Coriander	0.042 ab	0.012 b	0.022 a	0.033 ab	0.159 ab	10.187ab	13.992 b	1.626 ab
Mint	0.152 ab	0.005 ab	0.055 a	0.036 ab	0.312 b	11.451 ab	**19.681 b**	2.789 b

*DMRT analysis has been performed separately for root and leafy vegetables. Mean values with the same letter(s) in a column of root and leafy vegetables state no significant difference from each other at 5% level by DMRT within the root or leafy vegetables

consumption of mint (19.681 mg/person day). The highest contribution of different heavy metals into the ADI was reported through consumption of turnip (Cr), coriander (Cd and Pb), mustard (Ni), mint (Cu, Fe, and Mn), and spinach (Zn). It reveals that the transfer of heavy metals through leafy vegetables was more likely than the root vegetables. Also, coriander and mint are mostly contaminated by heavy metals than other leafy vegetables.

One study from Iran has reported the daily intake of heavy metals through the consumption of five different vegetables (leek, sweet basil, parsley, garden cress, and tarragon) (Maleki and Zarasvand 2008). Their results showed the daily dietary intake of Pb, Cu, Cr, and Cd through vegetable consumption was estimated at 2.96, 2.50, 1.72, and 0.07 mg/person day, respectively (Maleki and Zarasvand 2008). Another study estimated the daily intakes of Cd, Pb, and Cu for children were 0.311, 0.622, and 2.320 µg/ person day, whereas for adults were 0.182, 0.363, and 1.357 µg/person day, respectively (Naghipour et al. 2018).

Hazard quotient (HQ) and hazard index (HI)

Health risks associated with heavy metals intake through the consumption of food is often evaluated using hazard quotient (HQ) and hazard index (HI). The hazard quotient (HQ) has been recognized as a useful parameter for evaluation of risk associated with consumption of metal contaminated food crops (Zhuang et al. 2009), and hazard index (HI) is estimated as the sum of hazard quotients when more than one heavy metal is involved (Si et al. 2015). Hazard quotient (HQ) and hazard index (HI) value greater than 1 indicates a potential health risk to those consuming these vegetables.

From the results, the HQ value of all heavy metals in all root vegetables was below one except for Cr in Turnip (1.121) and Zn in spinach (1.104). It depicts that there are potential health risks due to Cr and Zn when consuming turnip and spinach, respectively. Figure 2 also shows the hazard indices associated with the consumption of different vegetables. Among the studied samples, turnip, mustard, spinach, coriander, and mint exceeded the safe hazard index value. Four out of the five leafy vegetables have HI values of more than 1. On the other hand, one out of 4 studied root vegetables were found to have an HI value higher than 1. The highest HI value was 1.54 for the consumption of turnip, and the lowest value was 0.362 for the consumption of radish among the root vegetables. Among the leafy vegetables, the highest HI value was 2.83, and the lowest value was 0.38 for the consumption of mint and cabbage, respectively. The hazard indices among all the vegetable samples studied decreased in the order of mint > spinach > coriander > mustard > turnip > beet > carrot > radish > cabbage.

The study showed that leafy vegetables pose more health risks than root vegetables (Fig. 2). The heavy metal concentrations in leafy vegetables, average daily intake, hazard quotient, and hazard index due to consumption of leafy vegetables were much higher than that of root vegetables. This finding complies with previous studies (Wagesho and Chandravanshi 2015; Zhou et al. 2016), stating that potential health risks associated with leafy vegetables are higher than the root vegetables. Higher

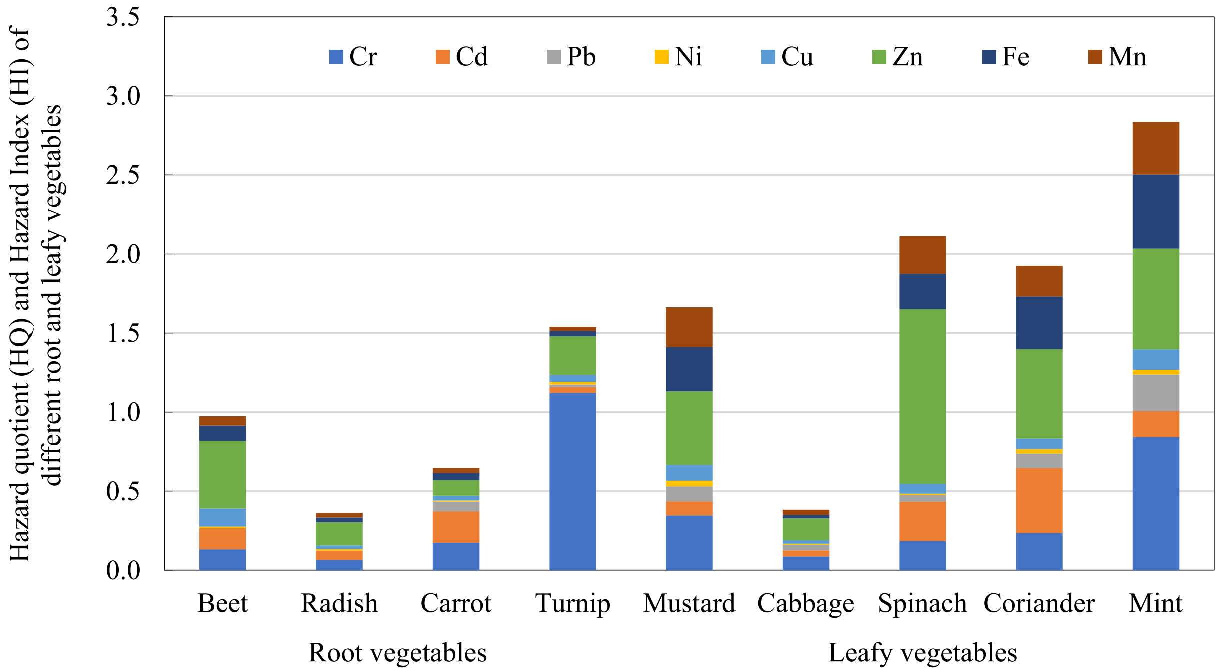

Fig. 2 Hazard quotient (HQ) and Hazard Index (HI) of different vegetable samples

accumulation of heavy metals in leafy vegetables may be due to leaves being the most active part of the vegetables performing photosynthesis (Marchiol et al. 2004; Perfus-Barbeoch et al. 2002; Zhou et al. 2013, 2016). Also, leafy vegetables are usually smaller in size and often closer to the ground than most other vegetables. That makes them more susceptible to heavy metals accumulation from the contaminated soils. Also, activities such as mining and smelting near the vegetation can facilitate heavy metals deposition on the vegetable leaves and stem as well (Zhou et al. 2016). Contamination through surface deposition may also happen during the transportation and sale of the vegetables.

Conclusions

The current investigation revealed that all the four root vegetables contain at least one of the six carcinogenic heavy metals (Cr, Cd, Pb, Ni, Cu, Zn) beyond the MPL. In contrast, the Fe and Mn level is below the MPL. In the case of leafy vegetables, only Mn was below the MPL in all five studied vegetables, whereas one or more of the other seven heavy metals (Cr, Cd, Pb, Ni, Cu, Zn, Fe) are exceeding the MPL. Surprisingly, though the root vegetables have the Fe content lower than the MPL, the leafy vegetables have a very high concentration. The ADI calculation showed the turnip has Cr, and mint has Fe values higher than the PMTDI. The HQ and HI study revealed that turnip among the root vegetables and mustard, spinach, coriander, and mint from the leafy vegetables are posing health risks to its consumers. The results revealed that the consumption of leafy vegetables has a higher potential health risk than root vegetables. Therefore, this study suggests the responsible authority to take proper steps in monitoring and identifying source of the heavy metals present in the commonly consumed root and leafy vegetables.

Abbreviations

AAS: Atomic Absorption Spectrophotometer; ADI: Average Daily Intake; ANOVA: Analysis of Variance; DMRT: Duncan's Multiple Range Test; FAO: Food and Agriculture Organization; HI: Hazard Index; HQ: Hazard Quotient; MPL: Maximum Permissible Limit; PMTDI: Permitted Maximum Tolerable Daily Intake; WHO: World Health Organization.

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Author contributions

RS: Investigation, data curation, visualization, writing-original draft preparation; RUT and KAH: Visualization, data curation, writing-original draft preparation, reviewing, and editing; ASC and MNM: Conceptualization, Methodology, project administration, writing-reviewing and editing. All authors read and approved the final manuscript.

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