



# Human Health Risk Assessment of Heavy Metals Through the Consumption of Common Foodstuffs Collected from Two Divisional Cities of Bangladesh

H. M. Zakir<sup>1</sup> · Q. F. Quadir<sup>1</sup> · M. Z. I. Mollah<sup>1</sup>

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## Abstract

This study was designed to evaluate heavy metal status in commonly consumed foodstuffs collected from two divisional cities, viz., Chottogram and Mymensingh of Bangladesh and to assess potential human health risks through the consumption of those foodstuffs. A total of 90 food samples were analyzed for Cu, Zn, Ni, Pb, Mn, and Fe contents by an atomic absorption spectrophotometer. The calculated per capita per day dietary intakes of Cu and Mn from rice and Mn's same value through spinach consumption in both cities were higher than the recommended daily allowances (RDA) for an adult human. Similarly, the computed daily metal intake (DMI) values of Pb for both males and females were also higher than the upper tolerable intake level (UTIL) through the consumption of rice collected from Mymensingh city. The average calculated hazard index (HI) values for males and females due to dietary intake of rice were 47.67 and 93.30 in Chottogram city and 97.98 and 192.04 in Mymensingh city, respectively. The present study identified rice as the most contaminated food item, followed by spinach, brinjal, and lentil. Still, dietary intakes of beef, broiler, and fish samples collected from both the cities were found as safe or less hazardous. Among the individual metals, Mn was the most dominant metal present in different foodstuffs collected from both the cities and contributed significant risk to the people. Finally, the study concluded that Bangladesh's food consumption patterns should be changed and advised to incorporate more proteins in regular diet charts by reducing carbohydrates.

**Keywords** Plant & animal origin foodstuffs · Metal contamination · Hazard index · RDA · PCA

## Introduction

Heavy metal contamination in foodstuff is a severe public health concern all over the world. In the meantime, both producers and consumers have recognized these metals' presence in foodstuffs as problematic for human consumption. The occurrence of these heavy metals in the field and farms, and then into the common foodstuffs of Bangladesh is associated with various types of anthropogenic activities like-rapid industrial growth, wastewater irrigation, sludge

application, use of metal-contaminated agrochemicals, toxic metal-rich fish and poultry feeds and inappropriate handling of food during storage and transport (Rana et al. 2014; Zakir et al. 2015, 2016, 2017; Hossain et al. 2017; Islam et al. 2020; Lamastra et al. 2018; Sarker et al. 2017). It has been reported that the concentrations of various heavy metals in different food items of our country are high, and the amount is sufficient to cause multiple health hazardous problems to the peoples (Aysha et al. 2017; Haque et al. 2018; Zakir et al. 2020a). Several diseases may develop due to intake of heavy metal-contaminated foodstuffs. Surplus Cu had been associated with liver damage, and Zn may produce adverse nutrient interactions with Cu (FDA 2001). On the other hand, some toxic metals like Pb and Cd are lethal for a human being even at the minute quantity (Llobet et al. 2003) and may induce urologic disorders (viz., renal tumors and kidney dysfunctions), softening of the bones, increase blood pressure and heart diseases risk (Ikem and Egienbor 2005).

Traditionally, cereals, vegetables, and fishes (both cultured and natural) are very common in Bangladeshi regular

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✉ H. M. Zakir  
zakirhm.ac.bau@gmail.com; zakirhm\_ac@bau.edu.bd

<sup>1</sup> Department of Agricultural Chemistry, Faculty of Agriculture, Bangladesh Agricultural University, Mymensingh-2202, Bangladesh

diet charts, but nowadays, cultured fishes and broiler captured most of the protein sources in our daily diet. Several studies (Aysha et al. 2017; Zakir et al. 2018; Akter et al. 2020) reported higher uptake or accumulation of different toxic metals in those foodstuffs. As a result, nowadays, heavy metals' accumulation in the human body from these common foodstuffs is a burning question. Thus, toxic metallic contaminated foodstuff is a crucial issue of food quality and safety assurance of Bangladesh. However, different international and national regulations on food quality have also lowered the acceptable limits of toxic metals in different foodstuffs (Radwan and Salama 2006), which will reduce the metallic contamination of the food chain through increased awareness of peoples.

In Bangladesh, evaluating the risks and benefits of consuming different foodstuffs is critical; being a key source of essential elements and nutrients. Thus, it is imperative to investigate the level of toxic elements in common foodstuffs to have insight into the safety of foodstuffs consumed by the country's people. Considering this fact, the present study deals with determining different heavy metals in commonly consumed foodstuffs and assessing their potential human health risk. The study was focused mainly on two divisional cities of Bangladesh, viz., Chottogram and Mymensingh. From a demographic point of view, Chottogram is a cosmopolite city where peoples of different regions living together. On the other hand, Mymensingh being another populated city where the demography is more or less homogenous. Both the cities are in close proximity to various chemical contamination, including their point sources of contamination.

## Materials and Methodology

### Selection of Foodstuff Samples

There were 2 (two) types of origin of foodstuff considered for collecting samples-(1) Plant origin such as rice (coarse and fine), lentil (*Lens culinaris*) (coarse and small), wheat flour, brinjal (*Solanum melongena*) and spinach (*Spinacia oleracea*), and (2) Animal origin such as beef, broiler (liver, muscle, and skin) and fish (abdomen and muscle). Furthermore, the present study selected two fish species, viz., tilapia (*Oreochromis niloticus*) and ruhi (*Labeo rohita*). Category in selecting foodstuffs was done to minimize variations among the peoples of different income groups. Fine rice and small lentil grains were collected as a choice of higher-income group peoples, while coarse grains are choice of lower-income group peoples, and both items are prevalent in Bangladeshi diet chart. Similarly, the tilapia fish is comparatively cheaper, which is chosen by the lower-income group peoples, while the ruhi fish is taken commonly by the

higher-income group peoples. On the other hand, the broiler is one of the most commonly consumed protein sources in our diet nowadays.

### Collection and Processing of Samples

A total of 45 (forty-five) samples of 15 food items, viz., rice (fine and coarse), lentil (small and coarse), wheat flour, spinach, brinjal, beef, broiler (liver, muscle, and skin), ruhi (muscle and abdomen) and tilapia (muscle and abdomen) were collected from each city. Three (03) samples for each foodstuff were collected from different vendors of each city to minimize heterogeneity. Thus, the present study handled a total of 90 (ninety) samples. Finally, the sample mass was reduced to about 500 g for further processing of this study. Samples were collected from Mymensingh city during November 2017 and from Chottogram city in January 2018. Collected samples were kept in individual polythene/zipper bag with specific marking and tagging. After collection, all samples were brought to the post-graduate laboratory of the Department of Agricultural Chemistry, Bangladesh Agricultural University, Mymensingh-2202, Bangladesh.

At the laboratory, plant origin samples were air-dried first and then oven-dried for 48 to 72 h at 50 °C temperature, and it was stopped when a constant weight was obtained for a particular sample. After proper drying, the samples were powdered using a grinding machine. In the case of fish, samples were cleaned first, and then it was separated into flesh (edible) and bone (non-edible). Fish flesh again separated into muscle and abdomen. Similarly, collected broiler samples were also separated into liver, muscle, and skin after cleaning. Then fish (muscle and abdomen), broiler (liver, muscle, and skin), and beef samples were dried in an electric oven (Model: ED 56, Binder, Germany) at 60–80 °C temperature, and it was stopped when a constant weight was obtained for each sample. After completing drying, all samples were ground/paste well separately using a mechanical grinder and stored in a plastic zip-lock bag with proper marking and labeling. Care was taken to avoid any contamination during the grinding process.

### Preparation of Extract for Chemical Analyses

Aqueous extracts for foodstuff samples were prepared following the wet oxidation method using a di-acid mixture ( $\text{HNO}_3:\text{HClO}_4 = 2:1$ ) as described by Singh et al. (1999), which is used for the determination of different heavy metals. For this purpose, exactly 1.00 g of powdered/fine paste sample was taken into a 250 mL conical flask and digested with 10 mL of the di-acid mixture at 180–200 °C temperature. Then, the content was cooled to about 25 °C temperature. After cooling, the flask was washed with double distilled water, and the content was collected into a 100 mL

volumetric flask as filtrate. Analytical reagent grade quality (Merck, Germany) acids were used to prepare the extracts.

### Determination of Heavy Metals Concentration in Samples

Six metals, viz., Cu, Zn, Pb, Ni, Mn, and Fe were selected for this study based on the previous published reports (Haque et al. 2018; Zakir et al. 2018, 2020a; Akter et al. 2020). The concentration of these metals in aqueous extracts of foodstuffs were measured by an atomic absorption spectrophotometer (AAS) (SHIMADZU, AA-7000, Japan), furnished with single element hollow-cathode lamp as a light source at the wavelength of 324.8, 213.9, 283.3, 232.0, 279.5 and 248.3 nm, respectively. For this purpose, the instrument was calibrated according to the manufacturer's recommendations. Maximum sensitivity with an air-acetylene flame was used to operate the instrument, and the concentration limit of detection for Cu and Zn was  $<0.01 \mu\text{g g}^{-1}$ , while for Pb, Ni, Mn, and Fe was  $0.01\text{--}0.09 \mu\text{g g}^{-1}$ . All glass apparatuses were soaked in 14%  $\text{HNO}_3$  for 24 h, and finally, they were washed well using distilled water to avoid contamination.

### Analytical Quality Control

In order to satisfy the defined internal quality controls (IQCs), each sample was made to run, including blank, a certified reference materials (CRM), and a known amount of particular metal is "spiked" into powdered coarse rice sample collected from Mymensingh city. The obtained results and calculated recovery percent are presented in Table 1. CRM collected from the National Metrology Institute of Japan (NMIJ) (Reference # 7502-a, white rice powder) were analyzed for the studied metals utilizing the same procedure as that used for different food samples to check the effectiveness of the digestion and analytical procedure, and the recoveries were within 87–110% of the certified values. On

the other hand, in the spiked sample, the recoveries were within 88.5–107.1% of the original sample values (Table 1).

### Measurement of the Quantity of Nutritional Important Metals

Among the metals studied, per capita per day intakes of Cu, Zn, Mn, and Fe from individual foodstuff were calculated using the following formula as most vitamin-mineral combinations contain small amounts of these metals and compared with their recommended daily allowance (RDA) as reported by Connie and Christine (2009).

$$\text{Metal intake (per capita per day)} = \frac{\text{FR} \times \text{C}}{1000}$$

where FR is the food intake rate ( $\text{mg person}^{-1} \text{day}^{-1}$ ), which is taken from the Household Income and Expenditure Survey of Bangladesh (HIES 2017), and C represents the concentration of metal in the foodstuff sample ( $\text{mg kg}^{-1}$ , fresh weight).

### Assessment of Human Health Risk

#### Estimation of Daily Metal Intakes (DMI)

The DMI ( $\text{mg kg-bw}^{-1} \text{day}^{-1}$ ) was calculated to assess metallic health risk due to the intake of common foodstuff samples using the following formula-

$$\text{DMI} = \frac{\text{FR} \times \text{C}}{\text{BW}}$$

where FR is the food intake rate ( $\text{mg person}^{-1} \text{day}^{-1}$ ), C represents the concentration of metal in foodstuff sample ( $\text{mg kg}^{-1}$ , fresh weight), and BW denotes the average body weight (70 and 50 kg for adult male and female, respectively) (BBS 2015).

**Table 1** Analytical results obtained for Certified Reference Material (CRM) and spiked samples along with per cent recovery

Heavy metal	CRM sample (Reference # 7502-a, white rice powder)			Spiked sample (powdered coarse rice grain of Mymensingh city)			
	Certified value ( $\mu\text{g g}^{-1}$ )	Observed value ( $\mu\text{g g}^{-1}$ )	Recovery (%)	Original amount ( $\mu\text{g g}^{-1}$ )	Added amount ( $\mu\text{g g}^{-1}$ )	Observed value ( $\mu\text{g g}^{-1}$ )	Recovery (%)
Cu	3.02	$2.81 \pm 0.060$	93	$5.90 \pm 0.39$	2.00	$8.06 \pm 0.32$	102.0
Zn	26	$22.62 \pm 0.041$	87	$20.26 \pm 3.58$	2.00	$21.52 \pm 2.54$	96.7
Ni	0.39	$0.414 \pm 0.008$	106	$0.30 \pm 0.52$	2.00	$2.13 \pm 0.26$	92.6
Fe	4.48	$4.93 \pm 0.071$	110	$48.32 \pm 15.2$	5.00	$57.11 \pm 5.55$	107.1
Mn	11.2	$10.08 \pm 0.085$	90	$10.22 \pm 3.37$	5.00	$15.97 \pm 2.74$	104.9
Pb	0.0043	bdl ( $<0.01$ )	–	$2.17 \pm 0.29$	2.00	$3.78 \pm 0.22$	88.5

bdl below detectable limit

### Calculation of Target Hazard Quotients (THQ)

The calculation of THQ was accomplished with the following formula as outlined by the US EPA (2010)

$$\text{THQ} = \frac{E_F \times F_D \times \text{DMI}}{R_{fD} \times \text{BW} \times T}$$

where  $E_F$  means exposure frequency;  $F_D$  stands for the exposure duration; DMI represents the daily metal intakes ( $\text{mg kg-bw}^{-1} \text{ day}^{-1}$ ); RfD signifies the oral reference dose ( $\text{mg kg}^{-1} \text{ day}^{-1}$ ); BW is the mean body weight (kg), and T indicates the mean exposure time for noncarcinogens ( $365 \text{ days year}^{-1} \times \text{number of exposure years}$ ).

### Calculation of Hazard Index (HI)

Different contaminants can cause similar adverse human health effects; hence, it is often appropriate to combine hazard quotients associated with different substances (Al-Jassir et al. 2005; Sharmin et al. 2020). Hazard index is used to estimate the potential human health risk when more than one heavy metal is consumed. HI was calculated as the sum of THQs (Ametepey et al. 2018; Zakir et al. 2020b).

$$\text{HI} = \text{THQ}_1 + \text{THQ}_2 + \text{THQ}_3 \dots \text{THQ}_n$$

### Principal Component Analysis (PCA)

Principal component analysis displays the correlation structure of a data matrix. It is designed to transform the original variables into a new one (axes), known as principal components (PC), lie along with maximum variance directions. PCs are linear combinations of the original variables (Sarbu and Pop 2005). In this study, the PCA calculation was performed using Minitab 17, a statistical and data analysis software package.

## Results and Discussion

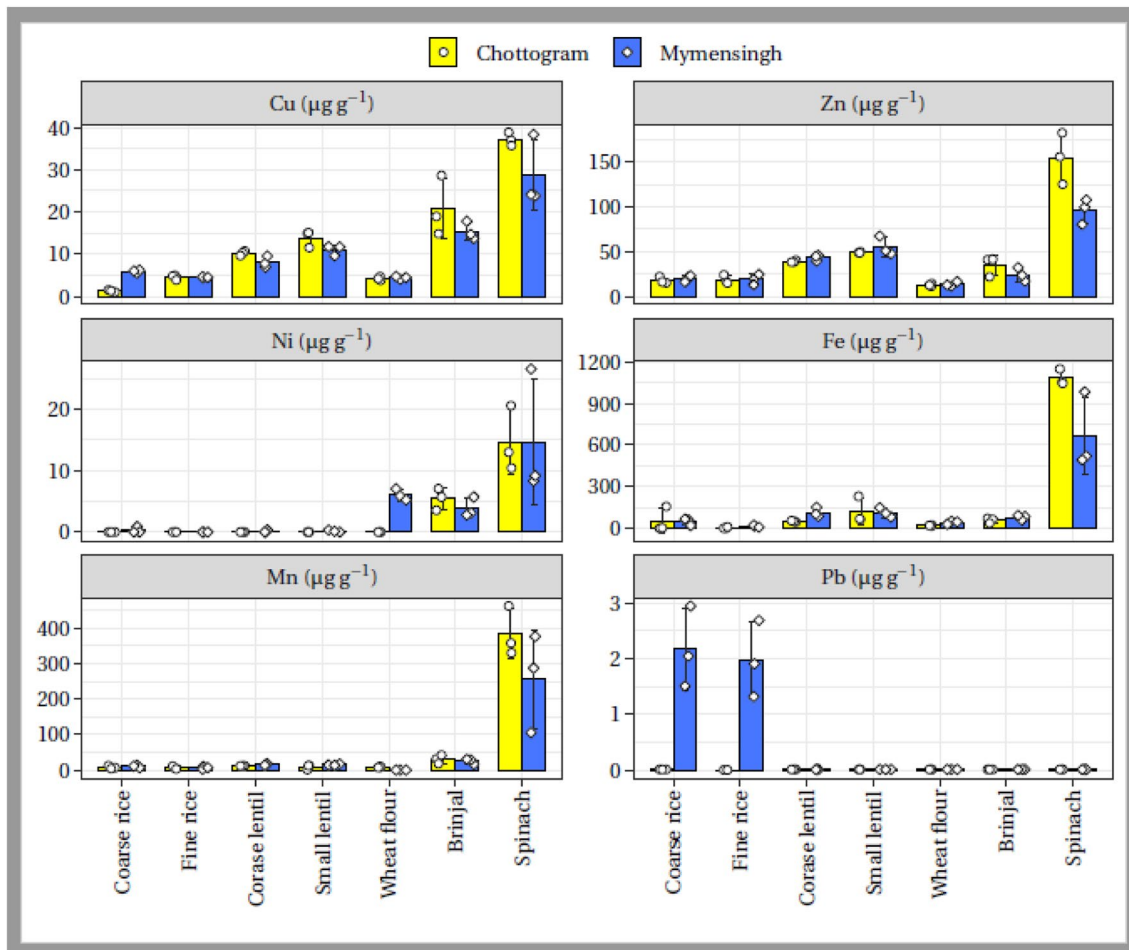
### Heavy Metal Status in Different Grains and Vegetables

In Bangladesh, anciently, rice has been cultivated all over the country, and it is the principal source of carbohydrate in our regular diet. Commonly consumed rice grains in Bangladesh can be classified as fine and coarse, and both types of grains were analyzed in this study. The content of Cu, Zn, Pb, Ni, Mn, and Fe in rice grain samples are presented in Fig. 1. It is apparent from Fig. 1 that rice grains

collected from Mymensingh city contained a comparatively higher amount of all studied metals. Notably, significantly higher Pb, Cu, and Ni quantities were obtained from coarse grain samples, usually consumed by the city's lower-income group peoples. However, Pb and Ni contents were  $< 0.01 \mu\text{g g}^{-1}$  in both types of rice grains collected from the Chottogram city. The amounts of Cu, Zn, and Pb in wheat flour samples collected from both the cities were almost equal (Fig. 1). Ni contents in wheat flour were  $6.02 \pm 0.15$  and  $< 0.01 \pm 0.00 \mu\text{g g}^{-1}$  in samples collected from the Mymensingh and Chottogram city, respectively, while Mn content was detected only in samples obtained from Chottogram city ( $8.31 \pm 2.43 \mu\text{g g}^{-1}$ ). Such variation in metal contents in the same food item between the cities might be due to the geographical pattern of cultivated soil, farming practices, amount and type of agrochemicals, water quality, handling, and processing of the crops.

Nevertheless, Pb contents were  $< 0.01 \mu\text{g g}^{-1}$  in wheat flour samples collected from both the cities. Iron and Mn contents in fine rice samples collected from both cities were almost the same. However, Fe content in coarse rice of Chottogram city was higher than that of Mymensingh city, and Mn content in the same rice was vice versa (Fig. 1). It can be seen from Table 2 that the average contents of all heavy metals in rice grain samples obtained in this study were lower than wheat flour. In contrast, Cu, Zn, Ni, and Fe contents in wheat flour samples were greater than the previously published results from other countries globally, including Bangladesh.

The recommended dietary allowances (RDA) for Zn and Mn are  $8.0$  and  $1.80 \text{ mg day}^{-1}$  for women,  $11.0$  and  $2.30 \text{ mg day}^{-1}$  for adult men, respectively (Connie and Christine 2009; FDA 2001). On the other hand, the RDAs for Cu and Fe are  $0.90$  and  $8.00 \text{ mg day}^{-1}$  for an average healthy adult human, respectively (Connie and Christine 2009). The calculated amounts (mg per capita per day) of nutritionally essential metals (viz., Fe, Zn, Cu, and Mn) in Bangladeshi diet through consumption of individual foodstuff of both cities along with the RDA are presented in Fig. 2. Considering the RDA values of Cu and Mn, it can be said that the consumption of rice collected from both the cities provides higher amounts of these metals per capita per day, which may be health hazardous. On the other hand, the consumption of coarse rice collected from both cities provides higher amounts of Fe than the RDA (Fig. 2), although the amount is very much lower than the upper tolerable intake levels (UTIL) ( $45.0 \text{ mg day}^{-1} \text{ person}^{-1}$ ) (FDA 2001), thus assumed to be safe. It can also be seen from Fig. 2 that considering RDA, per capita per day rice consumption rate in the Bangladeshi diet is sufficient to supply more than 50% of the daily requirement of Zn. However, the consumption of wheat flour collected from both cities provides a lower amount of those nutritionally essential metals than the RDA



**Fig. 1** The concentration of heavy metals in different grains (rice, lentil and wheat) and vegetable samples collected from Chottogram and Mymensingh cities of Bangladesh

(Fig. 2). However, their concentration (except Fe) in wheat flour was almost similar to rice, which is mainly due to per capita per day higher rice consumption rate (367.19 g for rice and 19.83 g for wheat) in the Bangladeshi diet (HIES 2017). The UTIL for an adult human being are set at 1.0 and  $0.24 \text{ mg day}^{-1} \text{ person}^{-1}$  for Ni and Pb, respectively (FDA 2001; Garcia-Rico et al. 2007). Considering these values, we can summarize that rice consumption collected from the Mymensingh city may be health hazardous in the context of Pb content but seems to be safe regarding Ni content. Similarly, wheat flour collected from both cities was also assumed to be harmless in the context of both Pb and Ni contents because the amounts of both metals were within the UTIL.

Lentil is another commonly consumed food in our diet, which is relatively cheaper than other protein sources, and due to this, it is sometimes called “poor man’s meat” in Bangladesh. Lentil samples collected from the Mymensingh city contained comparatively higher amounts of Zn, Mn, and Ni, while Cu content was higher in samples collected

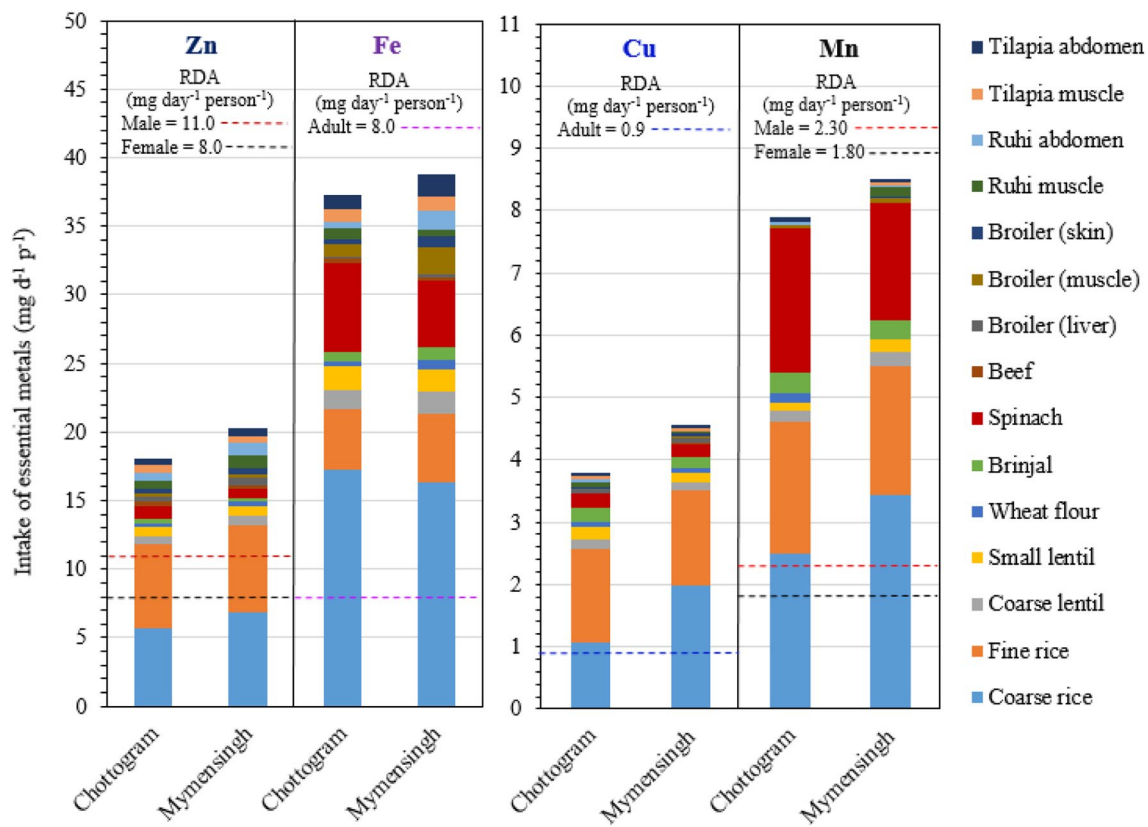
from the Chottogram city. However, Pb contents in lentils obtained from both cities and Ni content of the Chottogram city were  $<0.01 \pm 0.00 \mu\text{g g}^{-1}$  (Fig. 1). It is apparent from Fig. 1 that both types of lentils (coarse and small) contained a comparatively higher amount of Cu, Zn, and Mn than rice grains and wheat flours. In Bangladesh, per capita per day lentil consumption rate is 15.60 g (HIES 2017), which estimates that consumption of lentils collected from both the cities is not sufficient enough to supply these essential metals. However, they are a good source of these essential metals for human nutrition by considering RDA (Fig. 2).

Similarly, lentils collected from both cities assumed to be safe for dietary intake regarding Ni and Pb contents, considering the UTIL as reported by FDA (2001) and Garcia-Rico et al. (2007), respectively. Iron content in coarse lentil was higher in samples of Mymensingh city than that of Chottogram city. In contrast, in the case of small lentil, it was vice versa, and both types of lentil contained a comparatively higher amount of Fe than rice grains and wheat flour (Fig. 1). However, the calculated amount of Fe through

**Table 2** Levels of heavy metals in grains, vegetables, meat and fish samples in previously published results from different parts of the world including Bangladesh (data in parenthesis indicates this study average  $\pm$  SD)

Name of sample	Heavy metal ( $\mu\text{g g}^{-1}$ )							References
	Cu	Zn	Ni	Pb	Mn	Fe		
Rice	5.45 $\pm$ 1.11 (4.57 $\pm$ 0.34)	54.46 $\pm$ 21.27 (18.91 $\pm$ 4.48)	0.22 $\pm$ 0.04* (0.08 $\pm$ 0.13)	0.196 $\pm$ 0.16* (1.04 $\pm$ 0.16)	14.82 $\pm$ 6.33 (7.60 $\pm$ 3.83)	94.10 $\pm$ 15.1 (38.49 $\pm$ 14.1)	Zakir et al. (2020a), *Jafari et al. (2018)	
Lentil	2.53 $\pm$ 0.27 (10.78 $\pm$ 1.27)	91.03 $\pm$ 1.10 (46.61 $\pm$ 3.93)	1.91 $\pm$ 0.11 (0.07 $\pm$ 0.09)	0.52 $\pm$ 0.08 ( $<$ 0.01 $\pm$ 0.00)	72.47 $\pm$ 1.33 (12.51 $\pm$ 2.68)	107.37 $\pm$ 4.23 (105.97 $\pm$ 26.7)	Leshe and Tessema (2014)	
Wheat flour	1.47 $\pm$ 0.35 (4.32 $\pm$ 0.54)	6.34 $\pm$ 1.69 (12.95 $\pm$ 2.03)	0.198 $\pm$ 0.11* (3.01 $\pm$ 0.08)	0.107 $\pm$ 0.01 ( $<$ 0.01 $\pm$ 0.00)	11.30 $\pm$ 3.73 (4.15 $\pm$ 1.22)	9.39 $\pm$ 2.46 (30.82 $\pm$ 6.98)	Ludajic et al. (2015), *Bawiec et al. (2014)	
Brinjal	32.25 $\pm$ 0.96 (18.08 $\pm$ 4.61)	41.00 $\pm$ 1.15 (29.45 $\pm$ 9.40)	1.80 $\pm$ 0.08 (5.56 $\pm$ 2.26)	9.15 $\pm$ 6.54* ( $<$ 0.01 $\pm$ 0.00)	23.15 $\pm$ 1.30 (27.75 $\pm$ 9.74)	171.75 $\pm$ 3.30 (67.83 $\pm$ 17.66)	Latif et al. (2018), *Banerjee et al. (2010)	
Spinach	36.85 $\pm$ 1.10 (32.94 $\pm$ 4.94)	97.35 $\pm$ 10.6 (125.1 $\pm$ 21.4)	4.65 $\pm$ 0.13* (21.27 $\pm$ 9.1)	bdll $\pm$ 0.00 ( $<$ 0.01 $\pm$ 0.00)	217.20 $\pm$ 9.71 (319.5 $\pm$ 104)	769.88 $\pm$ 44.2 (873.0 $\pm$ 167)	Zakir et al. (2018), *Latif et al. (2018)	
Beef	2.37 $\pm$ 0.10 (5.11 $\pm$ 1.01)	11.01 $\pm$ 0.17 (128.1 $\pm$ 30.4)	0.25 $\pm$ 0.12** (5.96 $\pm$ 2.63)	1.15 $\pm$ 0.29* ( $<$ 0.01 $\pm$ 0.00)	0.98 $\pm$ 0.23 (0.88 $\pm$ 0.54)	19.87 $\pm$ 0.19 (126.98 $\pm$ 52.4)	Inobeme et al. (2018), *Nkansah and Ansah (2014), **Ihedioha et al. (2014)	
Broiler (muscle)	4.60 $\pm$ 0.18 (3.73 $\pm$ 1.00)	33.50 $\pm$ 1.14 (35.65 $\pm$ 5.71)	6.14 $\pm$ 2.59* (1.32 $\pm$ 0.61)	8.77 $\pm$ 0.54 ( $<$ 0.01 $\pm$ 0.00)	25.73 $\pm$ 1.01 (12.09 $\pm$ 3.41)	214.62 $\pm$ 7.32 (312.36 $\pm$ 68.1)	Alturqi and Abedair (2012), *Iwegbue et al. (2008)	
Ruhi (muscle)	1.13 $\pm$ 0.12 (3.09 $\pm$ 0.72)	4.22 $\pm$ 0.21 (54.78 $\pm$ 22.9)	1.27 $\pm$ 0.12* (0.13 $\pm$ 0.03)	7.67 $\pm$ 2.08* ( $<$ 0.01 $\pm$ 0.00)	0.58 $\pm$ 0.18** (2.24 $\pm$ 1.19)	21.59 $\pm$ 0.51 (70.98 $\pm$ 8.66)	Noor and Zutshi (2016), *Shovon et al. (2017), **Paul et al. (2016)	
Tilapia (muscle)	0.45 $\pm$ 0.08 (2.70 $\pm$ 0.69)	4.73 $\pm$ 0.54 (36.78 $\pm$ 1.99)	0.47 $\pm$ 0.05 (0.48 $\pm$ 0.13)	0.55 $\pm$ 0.31 ( $<$ 0.01 $\pm$ 0.00)	1.10 $\pm$ 0.34 (2.42 $\pm$ 1.06)	13.32 $\pm$ 3.83 (67.40 $\pm$ 16.4)	El-Batrawy et al. (2018)	

bdll below detectable limit



**Fig. 2** Amount of nutritional important metals through human consumption of different foodstuffs (fresh weight basis) collected from Chottogram and Mymensingh city of Bangladesh along with the recommended daily allowance (RDA)

lentils' consumption in the Bangladeshi diet is very much lower than the RDA (Fig. 2). On the other hand, the average contents of all heavy metals in lentils obtained by the present study were lower except Cu than previously published results from different parts of the world (Table 2).

Vegetables are ubiquitous in the Bangladeshi diet, which is also an essential part of a healthy diet worldwide. There were 2 (two) types of vegetables selected for this study, viz., brinjal (fruit vegetables), which is consumed throughout the year in our diet, and spinach (leafy vegetables), which is usually incorporated in our diet in the winter season. The contents of Cu, Zn, Ni, and Mn in brinjal collected from the Chottogram city were higher than the same collected from the Mymensingh city, but Fe content in brinjal showed the opposite trend (Fig. 1). On the other hand, in the spinach case, the contents of Cu, Zn, and Mn collected from the Chottogram city were higher than the spinach collected from the Mymensingh city. In contrast, Ni and Fe contents in the same vegetable showed a vice versa trend. However, Pb contents were  $<0.01 \pm 0.00 \mu\text{g g}^{-1}$  in both brinjal and spinach samples collected from both the cities (Fig. 1). It is very much evident from Fig. 1 that the contents of all metals were higher in spinach compared to brinjal collected from both cities. In general, leafy vegetables like spinach

accumulate metals like Fe, Mn, and Ni in higher amounts as some of these metals mediate the photosynthetic reactions in leaves (Ondo et al. 2013). Considering the RDA, per capita per day consumption of brinjal in the Bangladeshi diet was insufficient to supply nutritionally essential metals (Fig. 2).

On the contrary, it can be said from Fig. 2 that the consumption of spinach collected from both the cities provides more than sufficient amounts of Mn per capita per day, which may be health hazardous. But per capita per day consumption of spinach in our daily diet was insufficient to supply other nutritionally essential metals. It can also be inferred from the data presented in Figs. 1 and 2 that spinach is an excellent source of Fe, which may reduce Fe deficiency in humans by incorporating this item in our regular diet chart. However, the average amounts of Ni and Mn in brinjal, and Zn, Ni, Mn, and Fe contents in spinach obtained by the present study were higher than the previously published results from different parts of the world, including Bangladesh (Table 2).

### Heavy Metal Status in Meats and Fish Samples

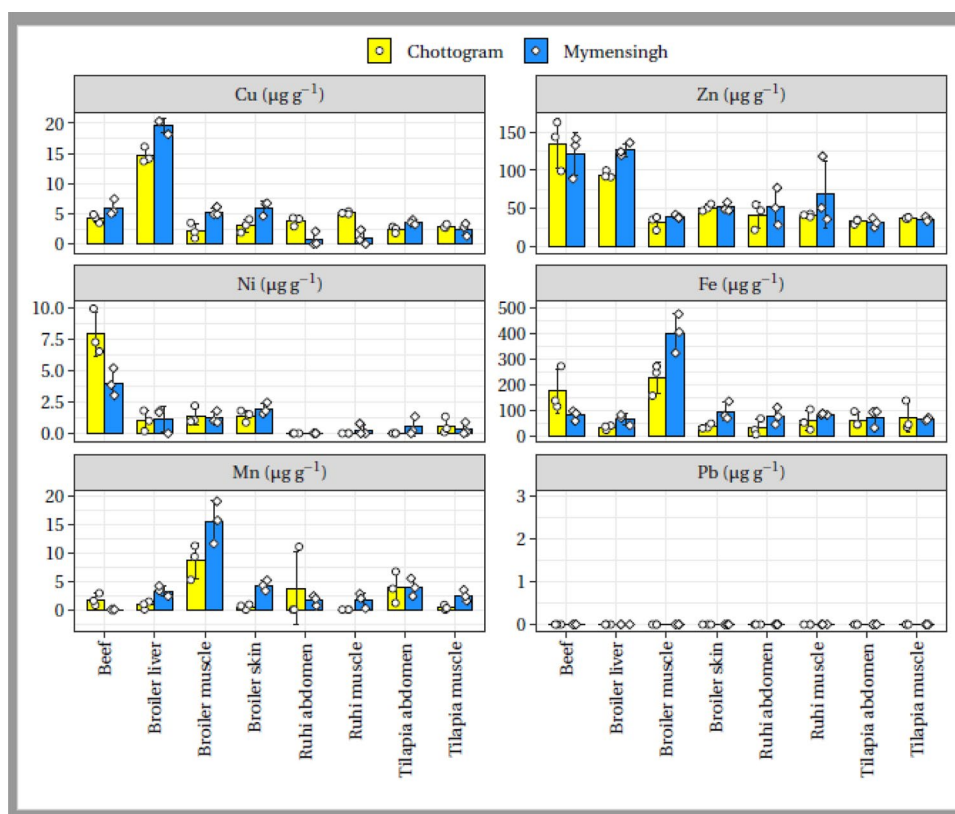
Meat and fish are the most important and common protein sources in the regular diet worldwide. The mean

concentrations of Cu, Zn, Ni, Pb, Mn, and Fe in beef, broiler (liver, muscle, and skin), and fish species (ruhi and tilapia) (muscle and abdomen) are presented in Fig. 3. Among the meat proteins, nowadays, broilers captured the lions share in the Bangladeshi diet due to its price difference with beef, mutton, and others, and according to HIES (2017), in Bangladesh per capita per day consumption rate of beef is 7.54 g and chicken is 17.33 g. The contents of Zn, Ni, Mn, and Fe in beef collected from the Chottogram city were higher compared to the same collected from the Mymensingh city, but Cu content in beef showed the opposite trend (Fig. 3). In broiler, all heavy metal contents in different parts (liver, muscle, and skin) were comparatively higher in samples collected from the Mymensingh city. Such variations of metal contents in meats of other cities may be due to the animal's age and size and feed used to rear those animals. Among the different parts of the broiler, the accumulation pattern of Cu and Zn followed the sequence as liver > skin > muscle; Mn and Fe showed the order as muscle > skin > liver; while Ni followed the sequence as skin  $\geq$  muscle > liver. However, Pb contents were  $<0.01 \pm 0.00 \mu\text{g g}^{-1}$  in beef and different broiler parts collected from both the cities (Fig. 3). Compared to beef and broiler muscle collected from both cities, the contents of Cu, Zn, and Ni in broiler were higher, while Mn and Fe contents were higher in beef (Fig. 3). However, in the context of the RDA, per capita per day consumption of beef and broiler of our regular diet provides an insufficient

amount of nutritionally important (Fig. 2), as well as toxic metals; thus, it can be assumed as safe for human health. So, a higher amount of these items can be included in our diet chart to meet the protein requirements. On the other hand, the average contents of Cu, Zn, Ni, and Fe in beef, and Zn, and Fe contents in broiler muscle obtained by the present study were higher than the previously published results from different parts of the world (Table 2).

Among all population groups, fish plays a vital role in the Bangladeshi diet. In between the 2 (two) selected fish species, tilapia is among the commonly consumed fish species by the lower-income group people. In contrast, the higher-income group people usually choose ruhi. The average contents of Cu and Mn in the ruhi collected from the Chottogram city were higher than the same collected from the Mymensingh city, but Zn, Ni, and Fe contents in the same species showed the opposite trend. In the tilapia, Cu and Zn contents were comparatively higher in samples collected from the Chottogram city. At the same time, Ni, Mn, and Fe were relatively higher in samples collected from the Mymensingh city (Fig. 3). Such variations of different metal contents in the same fish species of other cities are mainly influenced by the quality of feeds and raw materials (Sarker et al. 2017). Among the different parts of fish species studied, the accumulation pattern of Cu, Zn, Ni, and Fe in the ruhi followed the sequence as muscle > abdomen, while Mn contents showed the order as abdomen > muscle.

**Fig. 3** The concentration of heavy metals in different meat [beef, broiler (liver, muscle and skin)] and fish (abdomen and muscle) samples collected from Chottogram and Mymensingh cities of Bangladesh





In the tilapia case, the accumulation pattern of Zn, Ni, Mn, and Fe followed the sequence as muscle > abdomen, while Cu content showed the order as abdomen > muscle. Lead contents were  $<0.01 \pm 0.00 \mu\text{g g}^{-1}$  in different parts (muscle and abdomen) of the ruhi and tilapia collected from both the cities. Similarly, Ni contents were also  $<0.01 \pm 0.00 \mu\text{g g}^{-1}$  in different parts (muscle and abdomen) of the ruhi collected from the Chottogram city (Fig. 3). As regards to fish muscles collected from both the cities, the contents of Cu, Zn, and Fe were higher in the ruhi, while Mn and Ni contents were higher in the tilapia (Fig. 3). Considering the RDA, per capita per day consumption of fish in Bangladeshi regular diet chart supplied comparatively little amount of nutritionally important (Fig. 2), as well as toxic metals; thus, it can be assumed as safe for human health. So, similar to beef and broiler, a higher amount of fish species can also be included in our diet chart to meet the protein requirements. On the other hand, it can also be seen from Figs. 2 and 3 that both fish species are a good source of Fe, which may help to reduce Fe deficiency in peoples of the cities through the consumption of those fish species. Then again, the average contents of Cu, Zn, Mn, and Fe in the ruhi muscle, and Cu, Zn, Ni, Mn, and Fe contents in the tilapia muscle determined in this study were higher than the previously published results from different parts of the world (Table 2).

## Assessment of Human Health Risk

### Daily Metal Intakes (DMI)

Daily metal intake was calculated to measure human health risks associated with different heavy metals via commonly consumed foodstuffs. In the calculation of DMI, different food ingestion rates were taken from a preliminary report on the HIES of Bangladesh. According to this report, per capita per day consumption rates were 367.19 g for rice, 19.83 g for wheat, 15.60 g for lentil, 167.30 g for vegetables, 7.54 g for beef, 17.33 g for chicken, and 62.58 g for fish (HIES 2017). The calculated DMI values for different foodstuffs of both the cities and genders are presented in Table 3, along with the UTIL and oral reference doses (RfD) of heavy metals. It can be seen from Table 3 that the DMI values obtained for Cu, Zn, Mn, and Fe due to consumption of both types of rice in both cities; the values found for Pb due to the consumption of both kinds of rice in Mymensingh city for male and female were higher than that of UTIL.

Moreover, the same values obtained for Ni, Mn, and Fe due to intake of spinach for both genders in both cities were greater than that of UTIL, which indicates that the cities' peoples may have to face adverse effects due to dietary consumption of those foodstuffs. Similarly, DMI values for Ni for both males and females were also higher than that of UTIL due to the dietary intake of coarse rice collected from

the Mymensingh city and brinjal collected from Chottogram city (Table 3). Zakir et al. (2020a) also reported higher DMI values for different heavy metals for both genders due to the consumption of different rice varieties cultivated in wastewater irrigated sites of Bhaluka area, Mymensingh.

### Target Hazard Quotients (THQ)

Target hazard quotients were calculated following the procedure outlined by US EPA (2010) to evaluate both males' and females' possible health risks due to long-term exposure to heavy metals. If the calculated THQ value is less than 1.0, then it is assumed as "safe"; a value in between 1.0 to 5.0 indicates "there exists a possibility of adverse effects," and a value greater than 5.0 specifies "unsafe" when peoples are exposed to heavy metals. The calculated THQ and hazard index (HI) values of heavy metals for different genders due to the consumption of studied foodstuffs of both cities are presented in Table 4. It can be comprehended from the Table 4 that the calculated THQ values for individual metals of different foodstuffs collected from the Mymensingh city decreased in the sequence as-Mn > Pb > Cu > Zn > Fe > Ni. In contrast, Chottogram city, the values decreased in the order as-Mn > Cu > Zn > Fe > Ni > Pb. Alternatively, considering individual foodstuffs, which contributed to a higher THQ value, the most contaminated food item was rice, followed by spinach, brinjal, and lentil. In rice, Mn contributed a major risk for both males and females in both cities. It accounted for 71.28 and 69.78% for coarse and fine rice, respectively of the HI in Chottogram city, followed by Cu (10.70 and 17.36%, respectively), Zn (7.70 and 9.46%, respectively), and Fe (9.88 and 2.87%, respectively). On the other hand, in the case of Mymensingh city, Mn contributed for 44.26 and 36.43% for coarse and fine rice, respectively of the HI, followed by Pb (37.61 and 47.03%, respectively), Cu (8.95 and 9.44%, respectively), Zn (4.10 and 5.30%, respectively), and Fe (4.19 and 1.77%, respectively) (Table 4). However, the calculated THQ values for all metals in grains (rice and lentil) and vegetables were higher than 1.0 for both adult males and females; thus, all of these metals are exceeded the acceptable level of non-carcinogenic harmful health risk. On the contrary, the THQ values for all metals in most of the protein food items were below 1.0 for both adult males and females; thus, metals are within an acceptable level of non-carcinogenic harmful health risk in both cities. The calculated HI values also revealed that the peoples of Mymensingh city are more vulnerable than Chottogram city, considering most of the food items except vegetables and wheat flour. It can also be inferred from this result that females are more vulnerable than men regarding the heavy metal risk associated with consuming different foodstuffs collected from both the cities.

**Table 3** Daily metals intakes (DMI) estimate for grains, vegetables, meat and fish samples collected from two divisional cities of Bangladesh along with the upper tolerable intake level (UTIL) and oral reference doses (RfD)

	Cu	Zn	Ni	Pb		Mn		Fe				
				CHO	MYM	CHO	MYM	CHO	MYM	CHO	MYM	
UTIL (mg day <sup>-1</sup> person <sup>-1</sup> )	10.00 <sup>a</sup>	40.00 <sup>a</sup>	1.00 <sup>a</sup>	0.24 <sup>b</sup>	11.00 <sup>b</sup>	45.00 <sup>a</sup>						
RfD (mg kg <sup>-1</sup> day <sup>-1</sup> )	0.040 <sup>c</sup>	0.300 <sup>c</sup>	0.020 <sup>c</sup>	0.0035 <sup>d</sup>	0.014 <sup>c</sup>	0.700 <sup>c</sup>						
	CHO	MYM	CHO	MYM	CHO	MYM	CHO	MYM	CHO			
Rice (coarse)	15.29	28.45	82.60	97.68	4.60E-02	1.44E+00	4.65E-02	1.05E+01	35.66	4.92E+01	247.07	232.94
Rice (fine)	21.40	39.83	115.64	136.75	6.50E-02	2.01E+00	6.51E-02	1.46E+01	49.93	6.89E+01	345.89	326.12
Lentil (coarse)	21.53	21.78	87.94	91.67	4.70E-02	4.80E-02	4.73E-02	9.50E+00	30.29	2.94E+01	62.35	71.29
Lentil (small)	30.14	30.49	123.12	128.34	6.60E-02	6.70E-02	6.62E-02	1.33E+01	42.40	4.12E+01	87.29	99.81
Wheat flour	2.08	1.67	7.94	8.93	2.00E-03	2.30E-02	2.00E-03	2.10E-03	2.39	3.08E+00	20.17	23.37
Brinjal	2.92	2.34	11.12	12.51	3.00E-03	3.20E-02	2.90E-03	2.90E-03	3.34	4.31E+00	28.24	32.72
Spinach	2.76	2.21	9.81	11.08	2.00E-03	3.00E-02	2.00E-03	2.00E-03	1.73	2.97E+00	23.99	22.48
Beef	3.87	3.09	13.73	15.51	3.00E-03	4.20E-02	2.80E-03	2.80E-03	2.42	4.16E+00	33.59	31.47
Broiler (liver)	1.08	1.13	3.20	3.43	3.00E-03	1.54E+00	2.60E-03	2.60E-03	2.12	2.60E-03	5.13	10.65
Broiler (muscle)	1.51	1.58	4.47	4.80	4.00E-03	2.16E+00	3.60E-03	3.60E-03	2.97	3.60E-03	7.19	14.90
Broiler (skin)	3.43	2.61	5.73	4.10	1.19E+00	6.57E-01	1.60E-03	1.70E-03	5.02	4.26E+00	9.45	13.30
Ruhi (abdomen)	4.80	3.65	8.01	5.74	1.67E+00	9.20E-01	2.30E-03	2.40E-03	7.02	5.96E+00	13.23	18.61
Ruhi (muscle)	3.20	3.02	13.28	10.07	1.14E+00	1.54E+00	9.00E-04	1.10E-03	32.95	2.69E+01	93.09	69.82
Tilapia (abdomen)	4.48	4.23	18.60	14.09	1.59E+00	2.16E+00	1.20E-03	1.50E-03	46.13	3.77E+01	130.33	97.75
	0.12	0.17	3.77	3.53	2.20E-01	1.17E-01	3.00E-04	3.00E-04	0.05	3.00E-04	4.86	2.33
	0.17	0.24	5.28	4.95	3.08E-01	1.64E-01	4.00E-04	4.00E-04	0.07	4.00E-04	6.80	3.26
	0.97	1.29	6.26	8.31	6.70E-02	7.60E-02	7.00E-04	7.00E-04	0.05	2.17E-01	2.14	4.17
	1.36	1.80	8.76	11.64	9.30E-02	1.06E-01	9.00E-04	9.00E-04	0.08	3.04E-01	3.00	5.84
	0.13	0.37	1.98	2.72	8.70E-02	8.50E-02	6.00E-04	7.00E-04	0.54	1.07E+00	13.93	27.57
	0.19	0.51	2.77	3.80	1.22E-01	1.18E-01	9.00E-04	1.00E-03	0.75	1.50E+00	19.50	38.60
	0.32	0.73	5.35	6.24	1.46E-01	2.34E-01	1.00E-03	1.20E-03	0.06	5.17E-01	3.76	11.33
	0.45	1.02	7.49	8.74	2.05E-01	3.28E-01	1.50E-03	1.70E-03	0.08	7.24E-01	5.26	15.86
	0.83	0.17	9.06	12.70	2.00E-03	2.00E-03	2.20E-03	2.40E-03	0.81	4.07E-01	6.84	18.53
	1.16	0.24	12.68	17.78	3.00E-03	3.00E-03	3.10E-03	3.40E-03	1.13	5.69E-01	9.57	25.95
	1.03	0.20	8.21	13.82	2.00E-03	5.30E-02	2.00E-03	2.00E-03	0.56	5.50E-01	11.85	16.67
	1.44	0.29	11.49	19.35	3.00E-03	7.40E-02	2.80E-03	2.80E-03	0.78	7.70E-01	16.59	23.34
	0.69	0.85	7.88	8.50	2.00E-03	1.46E-01	2.30E-03	2.80E-03	0.90	6.89E-01	14.10	17.66
	0.96	1.18	11.03	11.91	3.00E-03	2.04E-01	3.20E-03	3.90E-03	1.26	9.64E-01	19.74	24.73

Table 3 (continued)

	Cu	Zn	Ni	Pb	Mn	Fe
UTTIL (mg day <sup>-1</sup> person <sup>-1</sup> )	10.00 <sup>a</sup>	40.00 <sup>a</sup>	1.00 <sup>a</sup>	0.24 <sup>b</sup>	11.00 <sup>a</sup>	45.00 <sup>a</sup>
RFD (mg kg <sup>-1</sup> day <sup>-1</sup> )	0.040 <sup>c</sup>	0.300 <sup>c</sup>	0.020 <sup>c</sup>	0.0035 <sup>d</sup>	0.014 <sup>c</sup>	0.700 <sup>c</sup>
	CHO MYM	CHO MYM	CHO MYM	CHO MYM	CHO MYM	CHO MYM
Tilapia (muscle)	Male 0.56 0.51	7.11 7.42	1.16E-01 7.42	1.90E-03 6.90E-02	0.45 2.00E-03	13.65 8.03E-01
	Female 0.78 0.71	9.95 10.39	1.62E-01 10.39	2.70E-03 9.60E-02	0.63 2.90E-03	19.11 1.12E+00

CHO Chottogram, MYM Mymensingh

<sup>a</sup>FDA (2001)

<sup>b</sup>Garcia-Rico et al. (2007)

<sup>c</sup>USEPA (2010)

<sup>d</sup>Khan et al. (2008)

## Principal Component Analysis (PCA)

Research in food contamination involves various food substances of diverse origin. Hence, to obtain valid results on heavy metals contamination, it requires modern analytical methods with sensitivity, specificity, and high accuracy; simultaneously, it demands the employment of complex statistical methods that provide the big picture in what they are concerned. Multivariate statistical techniques are the right tool for viewing and analyzing complex data (da Silva Torres et al. 2006). PCA and cluster analysis (CA) are two unsupervised methods that allow us to deduce how certain variables such as heavy metal concentrations, the origin of foodstuffs and other parameters characterize target substances and define their associations (Gergen and Harmanescu 2012). The CA method is used for samples grouping original variables, PCA estimates the correlation structure of the variables by finding hypothetical new variables (principal components-PC) that account for as much as possible of the variance (or correlation) in a multi-dimensional data set. This method helps us to identify groups of variables (i.e., heavy metals concentrations or locations or other parameters) based on the loadings and groups of samples based on the scores (Hammer et al. 2001). This study applied the PCA for different metal contents of various foodstuffs from the two target cities. The loading plot and score plot of the PCA results obtained for the city-wise heavy metal data points and their eigen analysis of data for various food categories are presented in Fig. 4 (A and B) and Table 5.

In Fig. 4A, each eigenvector's length is proportional to the variance in the data for individual food items, and the angle between the eigenvectors signifies the correlations among the different food items. In the figure, the colored groups of food items indicated by (I), (II), and (III) show strong positive correlations with each other. Interestingly, foods of similar type clustered in the same group except for brinjal, ruhi fish and broiler skin. In organisms, homeostasis mechanisms are in place at the cellular level to regulate the transition metals such as Fe, Zn, Cu, etc. (Pilon et al. 2009). Hence, the same sources' food items showed a somewhat similar trend in metal content, likewise showing a strong positive correlation. However, for items like ruhi fish or brinjal, the metal variation was significantly contributed by sampling location. The difference in variety or production practice might play critical roles in this difference.

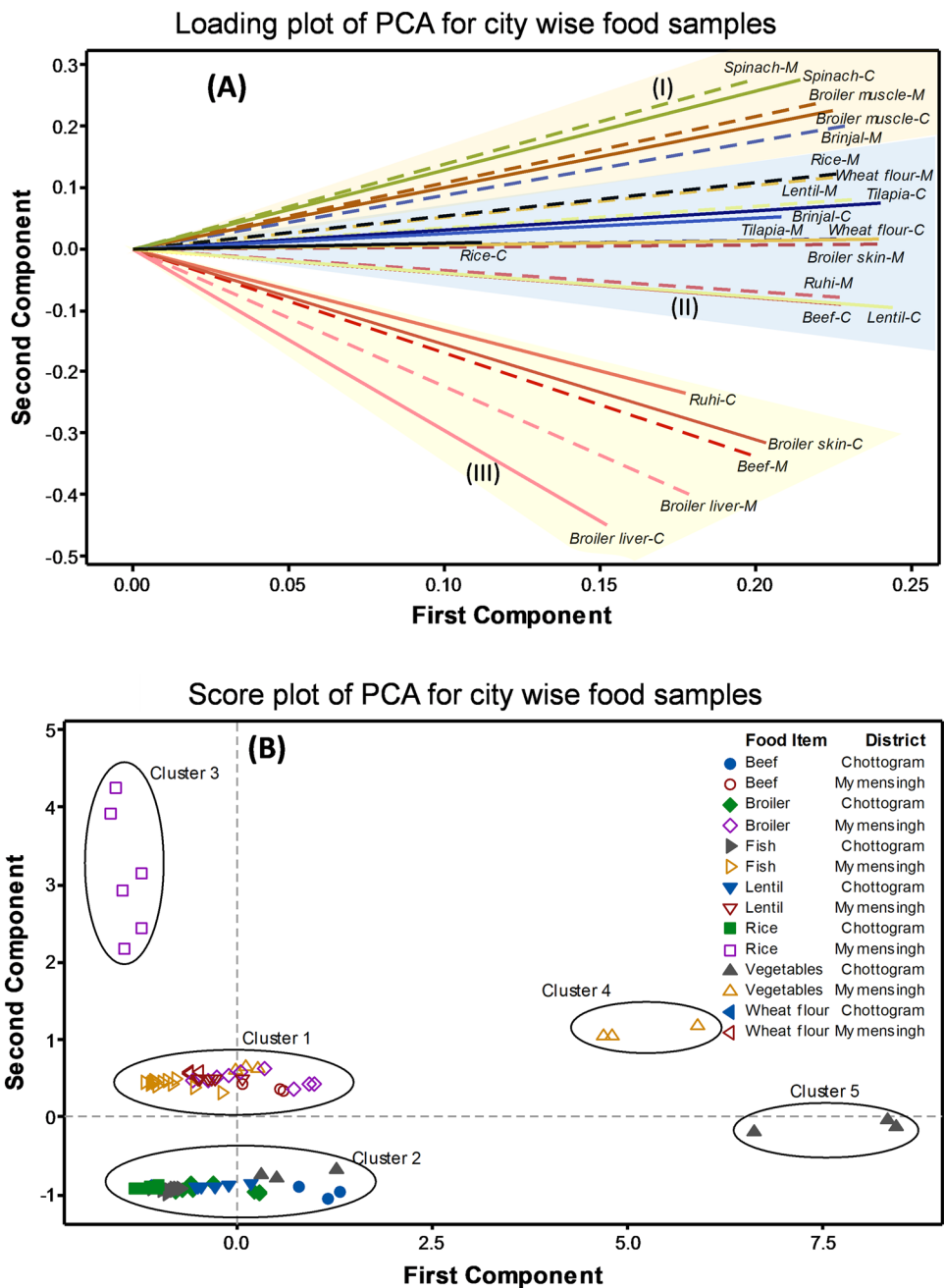
Again, if we consider only the sampling site as a principal variant (shown in Fig. 4B), the food items collected from the two divisional cities cluster separately (closed symbols for Chottogram city and open symbols for Mymensingh city). Based on the heavy metal status of the food items, there were five distinct clusters in the PCA outcome, where rice, wheat flour, brinjal, lentil, fish, beef, and broiler collected from Chottogram city form cluster 1,

**Table 4** Target hazard quotients (THQ) and hazard index (HI) of heavy metals estimate for grains, vegetables, meat, and fish samples collected from two divisional cities of Bangladesh

Name and type of sample	Gender	THQ values of heavy metals														HI values	
		Cu		Zn		Ni		Pb		Mn		Fe		CHO	MYM		
		CHO	MYM	CHO	MYM	CHO	MYM	CHO	MYM	CHO	MYM	CHO	MYM				
Rice (coarse)	Male	5.460	10.16	3.933	4.651	0.033	1.026	0.190	42.70	36.39	50.25	5.042	4.754	51.05	113.54		
	Female	10.70	19.91	7.710	9.117	0.046	2.011	0.266	83.69	71.33	98.50	9.883	9.318	99.93	222.54		
Rice (fine)	Male	7.689	7.779	4.188	4.365	0.034	0.034	0.193	38.76	30.90	30.02	1.272	1.455	44.28	82.41		
	Female	15.07	15.25	8.208	8.556	0.047	0.067	0.270	75.97	60.57	58.83	2.494	2.852	86.66	161.53		
Lentil (coarse)	Male	0.744	0.596	0.378	0.425	0.001	0.017	0.008	0.008	2.434	3.143	0.412	0.477	3.98	4.67		
	Female	1.459	1.168	0.741	0.834	0.002	0.032	0.012	0.017	4.771	6.160	0.807	0.935	7.79	9.15		
Lentil (small)	Male	0.987	0.789	0.467	0.528	0.001	0.021	0.008	0.008	1.760	3.034	0.490	0.459	3.71	4.84		
	Female	1.934	1.547	0.915	1.034	0.002	0.042	0.011	0.016	3.450	5.946	0.960	0.899	7.27	9.48		
Wheat flour	Male	0.386	0.403	0.152	0.163	0.002	1.103	0.010	0.010	2.164	0.003	0.105	0.217	2.82	1.90		
	Female	0.756	0.790	0.298	0.320	0.003	2.161	0.015	0.021	4.242	0.005	0.205	0.426	5.52	3.72		
Brinjal	Male	1.224	0.932	0.273	0.195	0.853	0.469	0.007	0.007	5.118	4.344	0.193	0.271	7.67	6.22		
	Female	2.399	1.826	0.534	0.383	1.672	0.920	0.013	0.014	10.03	8.515	0.378	0.532	15.03	12.19		
Spinach	Male	0.282	0.266	0.156	0.118	0.200	0.272	0.001	0.001	8.291	6.775	0.468	0.351	9.40	7.78		
	Female	0.552	0.521	0.306	0.232	0.392	0.533	0.002	0.002	16.25	13.28	0.918	0.689	18.42	15.26		
Beef	Male	0.042	0.062	0.180	0.168	0.157	0.084	0.001	0.001	0.050	0.000	0.099	0.048	0.53	0.36		
	Female	0.083	0.122	0.352	0.330	0.308	0.164	0.002	0.002	0.098	0.001	0.194	0.093	1.04	0.71		
Broiler (liver)	Male	0.348	0.459	0.298	0.396	0.048	0.054	0.003	0.003	0.054	0.221	0.044	0.085	0.79	1.22		
	Female	0.681	0.900	0.584	0.776	0.093	0.106	0.005	0.005	0.107	0.434	0.086	0.167	1.56	2.39		
Broiler (muscle)	Male	0.048	0.131	0.094	0.129	0.062	0.060	0.003	0.003	0.548	1.092	0.284	0.563	1.04	1.98		
	Female	0.093	0.257	0.185	0.253	0.122	0.118	0.005	0.006	1.075	2.139	0.557	1.103	2.04	3.88		
Broiler (skin)	Male	0.114	0.260	0.255	0.297	0.105	0.167	0.004	0.005	0.056	0.528	0.077	0.231	0.61	1.49		
	Female	0.223	0.510	0.499	0.582	0.205	0.328	0.008	0.010	0.110	1.035	0.150	0.453	1.20	2.92		
Ruhi (abdomen)	Male	0.296	0.062	0.431	0.605	0.002	0.002	0.009	0.010	0.825	0.415	0.140	0.378	1.70	1.47		
	Female	0.580	0.122	0.846	1.185	0.003	0.003	0.017	0.019	1.617	0.813	0.274	0.741	3.34	2.88		
Ruhi (muscle)	Male	0.367	0.073	0.391	0.658	0.001	0.038	0.008	0.008	0.570	0.562	0.242	0.340	1.58	1.68		
	Female	0.720	0.143	0.766	1.290	0.003	0.074	0.016	0.016	1.116	1.101	0.474	0.667	3.10	3.29		
Tilapia (abdomen)	Male	0.245	0.302	0.375	0.405	0.002	0.104	0.009	0.011	0.915	0.703	0.288	0.360	1.84	1.89		
	Female	0.481	0.592	0.735	0.794	0.003	0.204	0.019	0.022	1.794	1.378	0.564	0.706	3.60	3.70		
Tilapia (muscle)	Male	0.199	0.182	0.338	0.353	0.083	0.049	0.008	0.008	0.461	0.819	0.279	0.301	1.37	1.71		
	Female	0.389	0.357	0.663	0.693	0.162	0.096	0.015	0.016	0.904	1.606	0.546	0.590	2.68	3.36		

CHO Chottogram, MYM Mymensingh

**Fig. 4** Loading plot (A) and score plot (B) presentation of the Principle Component Analysis (PCA) results showing the city wise clustering of data points of various food categories. In (A), the length of each eigenvector is proportional to the variance in the data for that food item. The angle between the eigenvectors represents the correlations among the different food items. The colored groups of food items indicated by (I), (II) and (III) show strong positive correlations with each other. In figure A, samples from Chottogram (C) and Mymensingh (M) cities are indicated by continuous and dashed lines, respectively. In (B), closed and open symbols represent the food items collected from Chottogram and Mymensingh divisional cities, respectively.



indicating a more or less similar trend in the heavy metal status. Cluster 2 includes wheat flour, brinjal, lentil, fish, beef, and broiler samples collected from Mymensingh city. On the other hand, rice and spinach collected from Mymensingh city form clusters separately (clusters 3 and 4, respectively), and spinach collected from Chottogram city also creates a cluster alone (cluster 5), indicating different patterns of metal content. Such distinct clustering of the same food items from different locations may suggest that the products reaching in the two locations' city markets were from diverse origins and/or under different production management.

### Conclusions

Recently, food adulteration and contamination in Bangladesh have received significant attention due to implementing a government strategy to provide safe food to the peoples of the country. This study was planned to evaluate heavy metal status in commonly consumed foodstuffs collected from two divisional cities of the country and to assess the possible health risks for both males and females. Based on THQ, the study results identified rice as the most contaminated food item, followed by spinach, brinjal, and lentil. On the contrary, the consumption of

**Table 5** Eigenanalysis of the correlation matrix and the PC values of the eigenvectors up to six variables

	PC1	PC2	PC3	PC4	PC5	PC6
Eigenvalue	15.914	2.943	1.392	1.033	0.375	0.201
Proportion	0.723	0.134	0.063	0.047	0.017	0.009
Cumulative	0.723	0.857	0.920	0.967	0.984	0.994
<i>Variable</i>						
Rice-CHO	0.112	0.012	0.754	-0.070	-0.054	-0.024
Lentil-CHO	0.244	-0.096	0.052	-0.044	0.199	0.029
Wheat flour-CHO	0.240	0.016	-0.051	0.153	0.368	-0.169
Brinjal-CHO	0.208	0.053	0.167	0.390	0.469	0.242
Spinach-CHO	0.214	0.278	0.079	-0.019	0.163	-0.182
Beef-CHO	0.227	-0.091	-0.138	0.248	-0.338	0.241
Broiler liver-CHO	0.152	-0.450	-0.108	0.052	0.089	0.228
Broiler muscle-CHO	0.225	0.227	-0.110	-0.085	-0.210	0.055
Broiler skin-CHO	0.203	-0.317	0.135	0.009	-0.041	0.270
Ruhi-CHO	0.178	-0.235	-0.073	0.470	-0.298	-0.548
Tilapia-CHO	0.240	0.075	-0.200	0.004	-0.145	0.057
Rice-MYM	0.226	0.122	-0.305	-0.020	0.073	0.088
Lentil-MYM	0.231	0.080	0.296	-0.094	-0.043	-0.021
Wheat flour-MYM	0.225	0.118	0.145	0.145	-0.456	0.270
Brinjal-MYM	0.229	0.202	-0.090	-0.016	0.164	-0.090
Spinach-MYM	0.197	0.274	-0.018	0.366	0.063	-0.100
Beef-MYM	0.198	-0.337	0.072	-0.109	-0.048	-0.282
Broiler liver-MYM	0.178	-0.399	0.007	-0.119	0.154	0.071
Broiler muscle-MYM	0.219	0.237	0.094	-0.214	-0.140	0.053
Broiler skin-MYM	0.239	0.009	-0.117	-0.247	0.000	0.195
Ruhi-MYM	0.227	-0.079	0.010	-0.342	-0.054	-0.404
Tilapia-MYM	0.226	0.016	-0.219	-0.333	0.063	-0.032

CHO Chottogram, MYM Mymensingh

different sources of protein foodstuff was found as safe. The calculated hazard index for both adult males and females exceeded the threshold value 1, which may create health risk such as cancer. The study results also revealed that in most cases, the peoples of Mymensingh city are more vulnerable than Chottogram city. Among the individual metals, Mn was the most dominant metal in different foodstuffs collected from both the cities, which contributed significant risks to the peoples of the cities. The PCA results also suggest that the foodstuffs available in the city markets were from diverse origins and/or under different production management. Thus, it can be concluded that Bangladesh's food consumption patterns should be changed. More protein items and higher amounts may be incorporated in our regular diet chart by reducing carbohydrates, especially rice.

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**Data Availability** The data that support the findings of this study are available on request from the corresponding author.

## Compliance with Ethical Standards

**Conflict of interest** Authors have declared that no competing interests exist. The products used for this research are commonly and predominantly use products in our area of research and country. There is absolutely no conflict of interest among the authors and producers of the products because we do not intend to use these products as an avenue for any litigation but for the advancement of knowledge.

**Ethical Approval** All studies were conducted in accordance with principles for international, national, and/or institutional guidelines for the care and use of animals. This article does not contain any studies involving human participants performed by any of the authors. The manuscript in part or in full has not been submitted or published anywhere.

**Consent for Publication** All of the authors have read and approved the paper for submission of publication.

## References

- Akter M, Zakir HM, Sharmin S, Quadir QF, Mehrin S (2020) Heavy metal bioaccumulation pattern in edible tissues of different farmed fishes of Mymensingh area, Bangladesh and health risk assessment. *Adv Res* 21(4):44–55. <https://doi.org/10.9734/air/2020/v21i430200>
- Al-Jassir MS, Shaker A, Khaliq MA (2005) Deposition of heavy metals on green leafy vegetables sold on roadsides of Riyadh City, Saudi Arabia. *Bull Environ Contam Toxicol* 75(5):1020–1027. <https://doi.org/10.1007/s00128-005-0851-4>
- Alturiqi AS, Albedair LA (2012) Evaluation of some heavy metals in certain fish, meat and meat products in Saudi Arabian markets. *Egypt J Aquat Res* 38:45–49. <https://doi.org/10.1016/j.ejar.2012.08.003>
- Ametepey ST, Cobbina SJ, Akpabey FJ, Duwiejuah AB, Abuntori ZN (2018) Health risk assessment and heavy metal contamination levels in vegetables from Tamale Metropolis, Ghana. *Int J Food Contam* 5:5. <https://doi.org/10.1186/s40550-018-0067-0>
- Aysha MIJ, Zakir HM, Haque R, Quadir QF, Choudhury TR, Quraishi SB, Mollah MZI (2017) Health risk assessment for population via consumption of vegetables grown in soils artificially contaminated with arsenic. *Arch Curr Res Int* 10(3):1–12. <https://doi.org/10.9734/acri/2017/37612>
- Banerjee D, Bairagi H, Mukhopadhyay S, Pal A, Bera D, Ray L (2010) Heavy metal contamination in fruits and vegetables in two districts of West Bengal, India. *Electron J Environ Agril Food Chem* 9(9):1423–1432
- Bawiec P, Halabis M, Marzec Z, Kot A, Solski J, Gawel K (2014) Evaluation of chromium, nickel, iron and manganese content in wheat, flour, bran and selected baked products. *Curr Issues Pharm Med Sci* 27(2):71–75. <https://doi.org/10.2478/cipms-2014-0016>
- BBS (2015) Health and morbidity status survey-2014. Bangladesh Bureau of Statistics, Statistics and Informatics Division, Ministry of Planning, Govt. of the People's Republic of Bangladesh. [www.bbs.gov.bd](http://www.bbs.gov.bd)
- Connie WB, Christine SR (2009) Handbook of clinical nutrition and aging, 2nd edn. Humana Press, New York
- da Silva Tores EAF, Garbelotti ML, Neto JMM (2006) The application of hierarchical cluster analysis to the study of the composition of foods. *Food Chem* 99(3):622–629. <https://doi.org/10.1016/j.foodchem.2005.08.032>
- El-Batrawy OA, El-Gammal MI, Mohamadein LI, Darwish DH, El-Moselhy KM (2018) Impact assessment of some heavy metals on tilapia fish, *Oreochromis niloticus*, in Burullus Lake, Egypt. *J Basic Appl Zool* 79:1–12. <https://doi.org/10.1186/s41936-018-0028-4>
- FDA (2001) Dietary reference intakes for vitamin A, vitamin K, arsenic, boron, chromium, copper, iodine, iron, manganese, molybdenum, nickel, silicon, vanadium, and zinc. Report of the Panel on Micronutrients. National Academy Press, Washington, DC
- Garcia-Rico L, Leyva-Perez J, Jara-Marini ME (2007) Content and daily intake of copper, zinc, lead, cadmium, and mercury from dietary supplements in Mexico. *Food Chem Toxicol* 45:1599–1605. <https://doi.org/10.1016/j.fct.2007.02.027>
- Gergen I, Harmanescu M (2012) Application of principal component analysis in the pollution assessment with heavy metals of vegetable food chain in the old mining areas. *Chem Cent J* 6:156. <https://doi.org/10.1186/1752-153X-6-156>
- Hammer Q, Harper DAT, Ryan PD (2001) Past: Paleontological statistics software package for education and data analysis. *Palaeontol Electron* 4(1):4. [http://palaeo-electronica.org/2001\\_1/past/issue1\\_01.htm](http://palaeo-electronica.org/2001_1/past/issue1_01.htm)
- Haque R, Zakir HM, Aysha MIJ, Mallick S, Rahman MS (2018) Heavy metal uptake pattern and potential human health risk through consumption of tomato grown in industrial contaminated soils. *Asian J Adv Agric Res* 5(4):1–11. <https://doi.org/10.9734/AJAAR/2018/40169>
- HIES (2017) Preliminary report on household income and expenditure survey 2016. Bangladesh Bureau of Statistics, Ministry of Planning, Govt. of the People's Republic of Bangladesh, Dhaka-1000, Bangladesh
- Hossain MA, Zakir HM, Kumar D, Alam MS (2017) Quality and metallic pollution level in surface waters of an urban industrialized city: a case study of Chittagong city, Bangladesh. *J Ind Safety Engg* 4(2):9–18. <https://doi.org/10.37591/joise.v4i2.1941>
- Ihedioha JN, Okoye COB, Onyechi UA (2014) Health risk assessment of zinc, chromium, and nickel from cow meat consumption in an urban Nigerian population. *Int J Occup Environ Health* 20(4):281–288. <https://doi.org/10.1179/2049396714Y.0000000075>
- Ikem A, Egiebor NO (2005) Assessment of trace elements in canned fishes (mackerel, tuna, salmon, sardines and herrings) marketed in Georgia and Alabama (United States of America). *J Food Comp Anal* 18(8):771–787. <https://doi.org/10.1016/j.jfca.2004.11.002>
- Inobeme A, Obigwa PA, Olori E, Eziukwu C, Bamigboye O (2018) Heavy metal contents of meats from Auchi, Edo State, Nigeria. *Environ Res J* 12(2–4):19–22
- Islam F, Zakir HM, Rahman A, Sharmin S (2020) Impact of industrial wastewater irrigation on heavy metal deposition in farm soils of Bhalauka area, Bangladesh. *J Geol Environ Earth Sci Int* 24(3):19–31. <https://doi.org/10.9734/JGEEESI/2020/v24i330207>
- Iwegbue CMA, Nwajei GE, Iyoha EH (2008) Heavy metal residues of chicken meat and gizzard and turkey meat consumed in southern Nigeria. *Bulg J Vet Med* 11(4):275–280
- Jafari A, Kamarehie B, Ghaderpoori M, Khoshnamvand N, Birjandi M (2018) The concentration data of heavy metals in Iranian grown and imported rice and human health hazard assessment. *Data Brief* 16:453–459. <https://doi.org/10.1016/j.dib.2017.11.057>
- Khan S, Cao Q, Zheng YM, Huang YZ, Zhu YG (2008) Health risks of heavy metals in contaminated soils and food crops irrigated with wastewater in Beijing, China. *Environ Pollut* 152:686–692. <https://doi.org/10.1016/j.envpol.2007.06.056>
- Lamastra L, Suci NA, Trevisan M (2018) Sewage sludge for sustainable agriculture: contaminants' contents and potential use as fertilizer. *Chem Biol Technol Agric* 5:1–6. <https://doi.org/10.1186/s40538-018-0122-3>
- Latif A, Bilal M, Asghar W, Azeem M, Ahmad MI, Abbas A, Ahmad MZ, Shahzad T (2018) Heavy metal accumulation in vegetables and assessment of their potential health risk. *J Environ Anal Chem* 5(1):1–7. <https://doi.org/10.4172/2380-2391.1000234>
- Leshe S, Tessema M (2014) Determination of levels of essential and toxic heavy metals in lentil (*Lens culinaris* Medik) by flame atomic absorption spectroscopy. *Afr J Chem Edu* 4(4):16–34
- Llobet JM, Falco G, Casas C, Teixido A, Domingo JL (2003) Concentration of arsenic, cadmium, mercury, and lead in common foods and estimated daily intake by children, adolescents, adults, and seniors of Catalonia, Spain. *J Agril Food Chem* 51(3):838–842. <https://doi.org/10.1021/jf020734q>
- Ludajic GI, Pezo LL, Filipovic NK, Filipovic JN (2015) The content of essential and toxic elements in wheat bran and flour. *Hemijiska Ind* 69(4):417–423. <https://doi.org/10.2298/HEMIND140322056L>
- Nkansah MA, Ansah JK (2014) Determination of Cd, Hg, As, Cr and Pb levels in meat from the Kumasi Central Abattoir. *Int J Sci Res Pub* 4(8):1–4
- Noor N, Zutshi B (2016) Bioaccumulation of trace metals in tissues of rohu fish for environmental risk assessment. *J Water Res Protect* 8:472–481. <https://doi.org/10.4236/jwarp.2016.84040>
- Ondo JA, Biyogo RM, Eba F, Prudent P, Fotio D, Ollui-Mboulou M, Omva-Zue J (2013) Accumulation of soil-borne aluminium, iron, manganese and zinc in plants cultivated in the region of Moanda (Gabon) and nutritional characteristics of the edible

- parts harvested. *J Sci Food Agric* 93(10):2549–2555. <https://doi.org/10.1002/jsfa.6074>
- Paul BN, Chanda S, Sridhar N, Saha GS, Giri SS (2016) Proximate, mineral and vitamin contents of Indian major carp. *Indian J Anim Nutr* 33(1):102–107. <https://doi.org/10.5958/2231-6744.2016.00018.9>
- Pilon M, Cohu CM, Ravet K, Abdel-Ghany SE, Gaymard F (2009) Essential transition metal homeostasis in plants. *Curr Opin Plant Biol* 12(3):347–357. <https://doi.org/10.1016/j.pbi.2009.04.011>
- Radwan MA, Salama AK (2006) Market basket survey for some heavy metals in Egyptian fruits and vegetables. *Food Chem Toxicol* 44(8):1273–1278. <https://doi.org/10.1016/j.fct.2006.02.004>
- Rana M, Bakali B, Mia MY, Zakir HM (2014) Physico-chemical properties of effluents discharged from different industries of Gazipur, Bangladesh and its suitability for agricultural land. *J Environ Sci Nat Resour* 7(1):157–167. <https://doi.org/10.3329/jesnr.v7i1.22165>
- Sarbu C, Pop HF (2005) Principal component analysis versus fuzzy principal component analysis. A case study, the quality of Danube water 1985–1996. *Talanta* 65:1215–1220. <https://doi.org/10.1016/j.talanta.2004.08.047>
- Sarker MS, Quadir QF, Zakir HM, Nazneen T, Rahman A (2017) Evaluation of commonly used fertilizers, fish and poultry feeds as potential sources of heavy metals contamination in food. *Asian Australas J Food Safety Security* 1(1):74–81
- Sharmin S, Mia J, Miah MS, Zakir HM (2020) Hydrogeochemistry and heavy metal contamination in groundwaters of Dhaka metropolitan city, Bangladesh: Assessment of human health impact. *HydroRes* 3:106–117. <https://doi.org/10.1016/j.hydres.2020.10.003>
- Shovon MNH, Majumdar BC, Rahman Z (2017) Heavy metals (lead, cadmium and nickel) concentration in different organs of three commonly consumed fishes in Bangladesh. *Fish Aqua J* 8(3):1–5. <https://doi.org/10.4172/2150-3508.1000207>
- Singh D, Chhonkar PK, Pandey RN (1999) Soil, plant and water analysis: a method manual. IARI, New Delhi, India
- US EPA (2010) Human Health Risk Assessment: Risk-Based Concentration Table. [http://www.epa.gov/reg3hwmd/risk/human/rb-concentration\\_table/Generic\\_Tables/html](http://www.epa.gov/reg3hwmd/risk/human/rb-concentration_table/Generic_Tables/html). Accessed 25 Dec 2019
- Zakir HM, Sumi SA, Sharmin S, Mohiuddin KM, Kaysar S (2015) Heavy metal contamination in surface soils of some industrial areas of Gazipur, Bangladesh. *J Chem Biol Phy Sci* 5(2):2191–2206
- Zakir HM, Islam MM, Hossain MS (2016) Impact of urbanization and industrialization on irrigation water quality of a canal—a case study of *Tongi* canal Bangladesh. *Adv Environ Res* 5(2):109–123. <https://doi.org/10.12989/aer.2016.5.2.109>
- Zakir HM, Islam MM, Hossain MS (2017) Heavy metal contents in sediments of an urban industrialized area: a case study of *Tongi* canal, Bangladesh. *Asian J Water Environ Pollut* 14(1):59–68. <https://doi.org/10.3233/AJW-170007>
- Zakir HM, Aysha MIJ, Mallick S, Sharmin S, Quadir QF, Hossain MA (2018) Heavy metals and major nutrients accumulation pattern in spinach grown in farm and industrial contaminated soils and health risk assessment. *Arch Agric Environ Sci* 3(1):95–102. <https://doi.org/10.26832/24566632.2018.0301015>
- Zakir HM, Islam F, Quadir QF, Rahman A (2020a) Metallic health risk through consumption of different rice varieties cultivated in industrial wastewater irrigated farmers' fields of Bhaluka area, Bangladesh. *Curr J App Sci Technol* 39(11):76–91. <https://doi.org/10.9734/CJAST/2020/v39i1130650>
- Zakir HM, Sharmin S, Akter A, Rahman MS (2020b) Assessment of health risk of heavy metals and water quality indices for irrigation and drinking suitability of waters: a case study of Jamalpur Sadar area, Bangladesh. *Environ Adv* 2:100005. <https://doi.org/10.1016/j.envadv.2020.100005>

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