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Presence of heavy metals in fruits and vegetables: Health risk implications in Bangladesh

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HIGHLIGHTS

- Sample collected from 30 different agro-ecological zones for the first time in Bangladesh.
- Lead content in Mango was six times higher than Maximum Allowable Concentration level at production level.
- Inhabitants were exposed to slight carcinogenic risk from Lead.
- Health risks (Hazard Index) from vegetable was higher while fruits were found safe for consumption.

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ABSTRACT

The presence of toxic heavy metals such as As, Cd, Pb, Cr, Mn, Ni, Cu, and Zn in nationally representative samples of highly consumed fruits and vegetables was determined by inductively coupled plasma mass spectrometry (ICP-MS). Their concentrations exceeded the maximum allowable concentration (MAC) set by FAO/WHO for Pb in mango and Cd in tomato among the analyzed fruits and vegetables. Pb content in mango was found to be six times higher than the safe limit at production level. Health risks associated with the intake of these metals were evaluated in terms of estimated daily intake (EDI), and carcinogenic and noncarcinogenic risks by target hazard quotient (THQ) and hazard index (HI). EDI values of all the metals were found to be below the maximum tolerable daily intake (MTDI). The THQs of all metals were <1, suggesting no health hazards for adult population. However, total THQs of Mn and Cu were >1 through consumption of all vegetables, indicating significant health risks. HI was found to be <1 (0.825) for consumption of fruits; however, it was >1 (3.727) for vegetable consumption, suggesting adverse health effects from vegetable consumption only. The total carcinogenic risk (CR) of As was below the threshold level (10^{-6}) and 9.82E-05 for Pb, suggesting no potential CR from As consumption, but indicating the risk of Pb-induced carcinogenesis. The findings of this study reveal the health risks associated with the consumption of heavy metals through the intake of selected fruits and vegetables in adult population of Bangladesh.

1. Introduction

Food safety is a major public health concern worldwide. Because of the increasing risk of contamination of food by pesticides, heavy metals, and/or toxins, the food safety issues have attracted the attention of research recently (D'Mello, 2003). Contamination with heavy metals is important, particularly in agricultural production systems and human health. Factors influencing the concentration of heavy metals in plants include climate, environmental pollution, nature of the soil on which the plant is grown, and the degree of maturity of the plant at the time of harvesting (Lake et al., 1984; Scott et al., 1996; Voutsa et al., 1996). Fertilizers also contain heavy metals, thereby becoming an additional source of metal pollution in vegetables (Yusuf et al., 2003). Fresh fruits, vegetables, and fiber are of significance in the diet because they contain

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vitamins and mineral salts. They are very important and useful components for the maintenance of a better health and the prevention and management of various diseases. However, these plants contain both essential and toxic metals over a wide range of concentrations.

Trace metals have been found to play both positive and negative roles in human health (Adriano, 1984; Divrikli et al., 2003; Dundar and Saglam, 2004; Colak et al., 2005). They can be classified as toxic (arsenic, cadmium, lead, mercury, nickel, etc.), probably essential (vanadium), and essential (copper, zinc, iron, manganese, selenium, and cobalt) metals (Munoz-Olivas and Camara, 2001). However, toxic effects of the last two classes of metals have also been identified when the intake is excessively high (Celik and Oehlenschlager, 2007). Heavy metals have damaging effects on humans and animals, because of their nonbiodegradable nature, long biological half-lives, and potential to accumulate in different body parts as there is inadequate mechanism for their elimination from the body (Jarup, 2003; Reilly, 1980; Davies and White, 1981).

Accumulation of heavy metals has been reported to exhibit carcinogenic, mutagenic, and teratogenic effects (IARC, 1993; Pitot and Dragan, 1996; Radwan and Salama, 2006). Pb and Cd are the most abundant heavy metals, and their excessive intake is associated with cardiovascular, kidney, nervous, and bone diseases (WHO, 1992, 1995; Steenland and Boffeta, 2000; Jarup, 2003).

Average per capita daily intake of nonleafy vegetables and fruits in Bangladesh is 130 and 44.7 g, respectively (HIES, 2011). Different types of vegetables are grown throughout the year, but there is a lack of information on their metal contents (Alam et al., 2003). However, few previous studies on heavy metal contents in fruits and vegetables were conducted sporadically, but they were confined to a specific region.

There is limited information on heavy metal contents in highly consumed fruits and vegetables of Bangladesh. To the best of the authors' knowledge, this is first study of its kind to investigate heavy metal content of nationally representative samples (collected from 30 agro ecological zones of seven divisions) of highly consumed fruits (banana, mango, and jackfruit) and vegetables (brinjal, bean, carrot, green chilli, onion, potato, and tomato) grown in Bangladesh.

2. Materials and methods

2.1. Samples and sampling procedure

Sample selection and prioritization was done according to the key food approach throughout the year. The samples were selected randomly from 30 agro ecological zones across all seven divisions of Bangladesh (Fig. 1 and Table S1) based on the population census model with 17 sites including 14 Haats (village markets) and three city markets to ensure coverage of both urban and rural population. A total of 10 samples of predominant varieties, including seven different vegetable species (*Solanum melongena* (brinjal), *Dolichos lablab* (bean), *Daucus carota* (carrot), *Capsicum frutescens* (green chilli), *Allium cepa* (onion), *Solanum tuberosum* (potato), and *Lycopersicon esculentum* (tomato)) and three fruit species (*Musa paradisiaca* (banana), *Artocarpus heterophyllus* (jackfruit), and *Mangifera indica* (mango)), were collected for heavy metal analysis. From each sampling site, a composite of at least 12 samples for each food item was prepared. First, vegetable and fruit samples were washed with distilled water and cut into small pieces. Then, they were freeze-dried. After drying, the samples were crushed with a porcelain mortar and pestle, sieved through a 2-mm nylon sieve, and stored in airtight Ziploc bags at −20 °C in a laboratory of the Institute of Nutrition and Food Science (INFS), University of Dhaka. The preprocessed samples were then brought to the Laboratory of Environment and Information Sciences, Yokohama National University, Japan, and analyzed for the presence of arsenic (As), cadmium (Cd), lead (Pb), chromium (Cr), manganese (Mn), nickel (Ni), copper (Cu), and zinc (Zn). Metal contents were expressed as milligrams per kilogram fresh weight (fw) of the composite samples.

2.2. Analysis of samples

All chemicals were analytical-grade reagents, and Milli-Q (Elix UV5 and MilliQ, Millipore, USA) water was used for the preparation of solution. For metal analysis, 0.3 g of the freeze-dried samples was digested with 6 mL of 69% HNO_3 and 2 mL of 30% H_2O_2 (Wako Chemical Co., Japan) in a microwave digestion system (Berghof speedwave® Germany). The digested samples were then transferred to a Teflon beaker, whose total volume was made up to 50 mL by adding Milli-Q water. The digested solution was then filtered using a syringe filter (DISMIC®-25HP PTFE, pore size = 0.45 μm, Toyo Roshi Kaisha, Ltd., Japan) and stored in 50-mL polypropylene tubes (Nalgene, New York, USA).

2.3. Instrumental analysis and quality assurance

The samples were analyzed by inductively coupled plasma mass spectrometry (ICP-MS, Santa Clara, CA, USA). The detection limits of ICP-MS were 0.7, 0.6, 0.8, 0.4, 0.06, and 0.09 ng/L for Cr, Ni, Cu, As, Cd, and Pb, respectively. In order to satisfy the defined internal quality controls (IQCs), each sample was made to run, including blank and certified reference materials (CRM), to validate the internal standards. For excluding batch-specific errors, each sample was analyzed in triplicate. Standard stock solutions containing 10 μg/L of each element (Cd, As, Pb, Cr, Ni, Zn, Cu, and Mn) and internal standard solutions containing 1.0 mg/L of indium (In), yttrium (Y), beryllium (Be), tellurium (Te), cobalt (Co), and titanium (Ti) (Spex CertiPrep®, USA) were prepared. The standard curve was established by using multielement standard solution. Relative standard deviation (RSD < 5%) was inspected by a tuning solution purchased from Agilent Company.

2.4. Calculation

2.4.1. Estimated daily intake of heavy metals

Estimated daily intakes (EDIs) of heavy metals were calculated using their respective average concentration in food samples by the weight of food items consumed by an individual (body weight 60 kg for an adult in Bangladesh) (FAO, 2006), which was obtained from the household income and expenditure survey (HIES, 2011) and calculated by the following formula:

$$EDI = (FIR \times C)/BW,$$

where FIR is the food ingestion rate (g/person/day), C is the metal concentration in food samples (mg/kg), and BW is the body weight.

2.4.2. Noncarcinogenic risk

The target hazard quotient (THQ) and total target hazard quotient (TTHQ) can be calculated as (FAO/WHO, 2011)

$$THQ = (Efr \times ED \times FIR \times C)/(RfD \times BW \times AT) \times 10^{-3}$$

$$TTHQ(\text{individual food}) = \text{THQ metal 1} + \text{THQ metal 2} + \ldots\ldots\ldots + \text{THQ metal n.}$$

In order to assess the overall potential for noncarcinogenic effects from more than one heavy metal, a hazard index (HI) has been formulated based on the Guidelines for Health Risk Assessment of

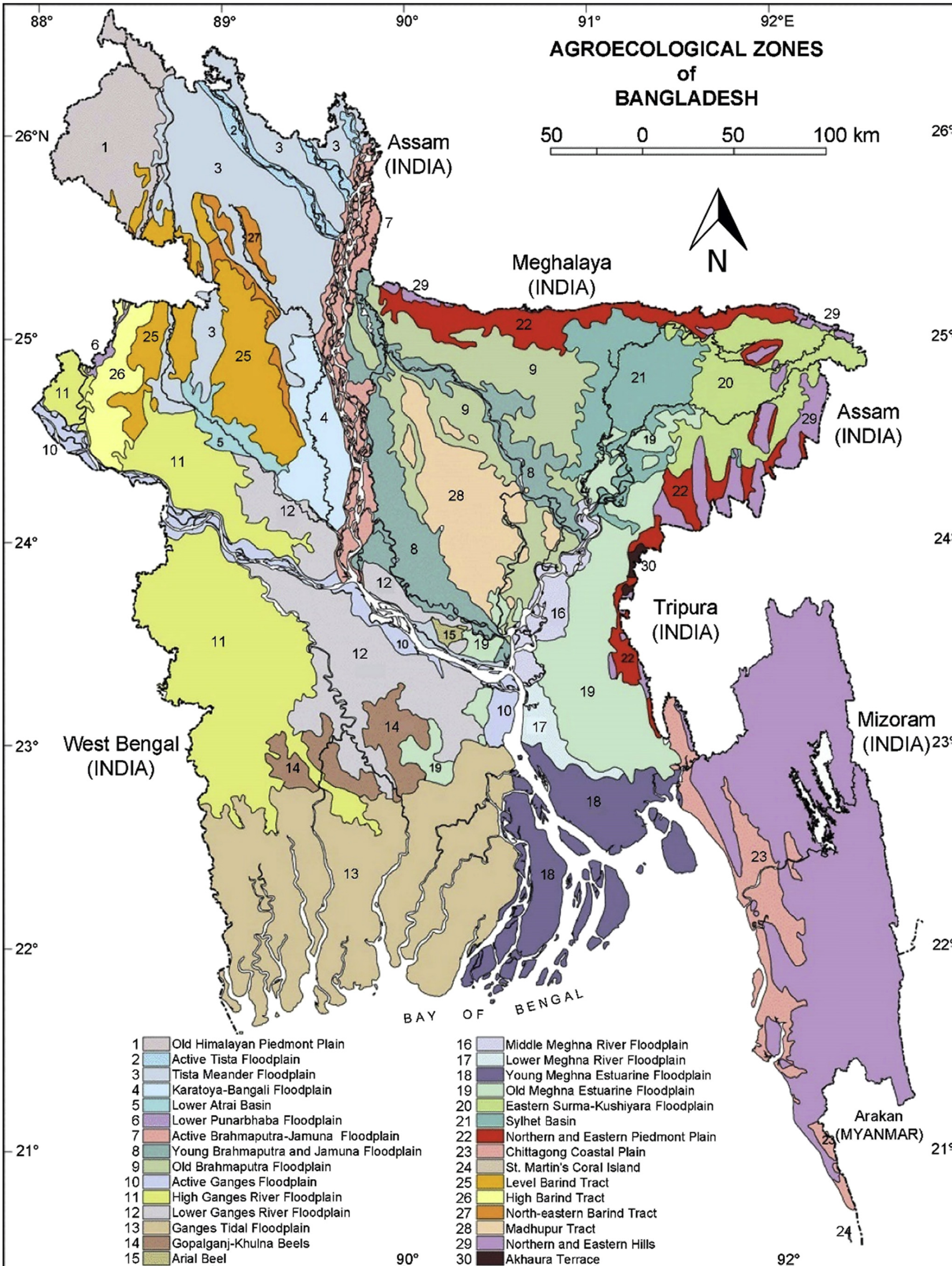

Fig. 1. Agro-ecological zones of Bangladesh (Source: http://www.bpedia.org).

Chemical Mixtures of USEPA as follows (USEPA, 1989):

$$HI = \sum \mathrm{TTHQ} = \mathrm{TTHQ\ food\ 1} + \mathrm{TTHQ\ food\ 2} + \ldots\ldots + \mathrm{TTHQ\ food}\ n,$$

where THQ is the target hazard quotient, EFr is the exposure frequency (365 days/year), ED is the exposure duration (70 years), FIR is the food ingestion rate (g/person/day), C is the metal concentration in foods (mg/kg), RfD is the oral reference dose (mg/kg/day), and AT is the averaging time for noncarcinogens (365 days/year $\times$ number of exposure years). The oral reference doses are 1.5, 0.02, 0.04, 0.3, 0.0003, 0.003, 0.0035, and 0.14 mg/kg/day for Cr, Ni, Cu, Zn, As, Cd, Pb, and Mn, respectively (USEPA, 2015). In order to determine the appropriate RfD for THQ, it is assumed that all chromium ions in the fruits and vegetables are trivalent (noncarcinogenic) and all arsenic ions are inorganic. If THQ < 1, the exposed population is unlikely to experience obvious adverse effects. If THQ $\geq$ 1, there is a potential health risk (Wang et al., 2005), and related interventions and protective measurements are needed to be taken.

2.4.3. Carcinogenic risk

The target carcinogenic risk (CR) factor (lifetime cancer risk) (USEPA, 1989) can be calculated as

$$TR = (Efr \times ED \times FIR \times C \times Csfo)/(BW*AT) \times 10^{-3},$$

where TR represents the target cancer risk or the risk of cancer over a lifetime, AT is the averaging time for carcinogens (365 days/year $\times$ ED), and Csfo is the oral carcinogenic slope factor obtained from the integrated risk information system (USEPA, 2015) database, which was 1.5 and 8.5 $\times$ 10^{-3} $(mg/kg/day)^{-1}$ for As and Pb, respectively.

3. Results and discussion

3.1. Metal concentrations in fruits and vegetables

In this study, concentrations of As, Cd, Pb, Cr, Mn, Ni, Cu, and Zn (mg/kg fw) were determined in the composite Bangladeshian samples of three highly consumed fruits, including banana, (*M. paradisiacal*), jackfruit (*A. heterophyllus*), and mango (*M. indica*), and seven vegetables, including brinjal (*S. melongena*), bean (*D. lablab*), carrot (*D. carota*), green chilli (*C. frutescens*), onion (*A. cepa*), potato (*S. tuberosum*), and tomato (*L. esculentum*). To the best of the authors' knowledge, this is the first study of its kind to use composite samples from Bangladesh. The levels of heavy metals found in the analyzed fruit and vegetable samples are listed in Tables 1 and 2. All metal concentrations were expressed in fw basis.

The lowest and highest As content in fruit samples was found in jackfruit (0.007 mg/kg) and mango (0.013 mg/kg), respectively (Table 1). The lowest and highest As content in vegetable samples was found in green chilli (0.004 mg/kg) and bean (0.018 mg/kg), respectively (Table 2). Mean As concentrations in all fruit and vegetable samples were lower than the safe limit at production level (0.1 mg/kg) (Codex, 2001; Table 1). Previous studies have reported As content in brinjal as 0.2 mg/kg and 0.01 mg/kg in potato on dry weight basis (Alam et al., 2003). In another study, the mean As concentration in all vegetables imported from Bangladesh were found to be 0.0545 mg/kg (Al Rmalli et al., 2005). Two recent studies conducted in Bangladesh reported mean As concentration of vegetables as 0.2 (0.009–7.9) (Islam et al., 2015) and 0.05 (0.01–0.2) mg/kg (Rahman et al., 2013).

The lowest and highest cadmium (Cd) level in fruit samples was found in mango (0.005 mg/kg) and jackfruit (0.037 mg/kg), respectively (Table 1). The lowest and highest Cd content in vegetable samples was found in bean (0.008 mg/kg) and tomato (0.056 mg/kg), respectively (Table 2). Among the fruit and vegetable samples, Cd concentration exceeded the permissible limit (0.05 mg/kg) only in tomato (Table 2). Previous studies found Cd amounts as 0.4 and 0.1 mg/kg in brinjal and potato, respectively (Alam et al., 2003), and 0.02 and 0.001 mg/kg in banana by Radwan and Salama (2006) and Karavoltsos et al. (2002), respectively. Al-Rmalli et al. (2012) reported Cd levels in nonleafy vegetables (0.008 $\pm$ 0.0008 mg/kg) and fruits (0.002 $\pm$ 0.001 mg/kg). Other studies reported Cd level as 0.0032 mg/kg in nonleafy vegetables grown in Mumbai, India (Tripathi et al., 1997), 0.21 mg/kg in selected vegetables grown in Nigeria (Sobukola et al., 2010), 0.008 mg/kg on fw basis in UK (Ysart et al., 1999), and 0.22 mg/kg (dry wt.) with a range of 0.09–0.62 mg/kg (dry wt.) in Nigerian leafy and fruity vegetables (Onianwa et al., 2000). Three recent studies conducted in Bangladesh reported mean Cd concentration in vegetable samples as 0.1 (0.001–1.6) (Islam et al., 2015), 0.06 (0.006–0.3) (Rahman et al., 2013), and 0.6 mg/kg (Ahmad and Goni, 2010).

In the fruit samples, lead (Pb) content was found to be the highest in mango (0.642 $\pm$ 0.556 mg/kg) and lowest in banana (0.003 $\pm$ 0.003 mg/kg) (Table 1). In vegetables, Pb was found to be the highest in bean (0.057 mg/kg) and lowest in tomato (0.005 mg/kg) (Table 2). The maximum limit of Pb in fruits and vegetables at production level is 0.1 mg/kg as recommended by WHO/FAO (Codex, 2001). According to the findings of this study, Pb content in mango was found to be six times higher than the safe limit level, although all other fruits and vegetables samples were below the permissible level value (Tables 1 and 2). Other studies showed that Pb content in banana was 0.118 mg/kg (Sobukola et al., 2010) and 0.02 mg/kg (Parveen et al., 2003). Pb concentration of the vegetable samples ranged from 3E-05 to 7E-04 mg/kg in a study on 24 different types of vegetables grown in the Chapai Nawabganj district of Bangladesh (Islam et al., 2015). Other studies reported the mean Pb content in potato as 0.5 (Alam et al., 2003) and 0.02 mg/kg (Ysart et al., 1999). Recent studies conducted in Bangladesh have found average Pb concentration as 0.5 (0.03–6.3) (Islam et al., 2015), 3.7 (0.7–17) (Rahman et al., 2013), and 3.9 mg/kg (Ahmad and Goni, 2010) in different vegetable species.

In the fruit samples, chromium (Cr) content was found to be the lowest in banana (0.317 mg/kg) and highest in mango (0.893 mg/kg) (Table 1). In the vegetable samples, the highest amount was found in bean (1.110 mg/kg) and the lowest amount in carrot (0.296 mg/kg) (Table 2). Mean Cr concentrations in all the fruit and vegetable samples were below the maximum permissible limit value (2.3 mg/kg) (Codex, 2001). In the literature, Cr contents were reported as 0.6 (Rahman et al., 2013) and 7.90 in vegetable samples (Maleki and Zarasvand, 2008), 0.32 in fruits grown at the roadsides in Turkey (Hamurcu et al., 2010), and 0.00–7.14 mg/kg on dry weight basis in Iran (Jafarian and Alehashem, 2013). Considering the levels of Cr in vegetables, the results of this study were in agreement with those of some previous studies (Islam et al., 2015; Ahmad and Goni, 2010).

In the fruit samples, the minimum and maximum manganese (Mn) levels observed were 6.06 mg/kg in mango and 24.76 mg/kg in jackfruit (Table 1). In the vegetable samples, the lowest and highest amounts of Mn were found in potato (6.928 mg/kg) and bean (28.352 mg/kg), respectively (Table 2). In the literature, median concentration of Mn was found as 65 mg/kg (Rahman et al., 2013) and ranging from 0.18 to 2.8 mg/kg (Singh and Taneja, 2010).

The lowest and highest levels of nickel (Ni) in the fruit samples were found in banana (0.037 mg/kg) and jackfruit (0.882 mg/kg), respectively (Table 1). In the vegetable samples, the minimum and

Table 1
Mean concentrations (mg/kg fw) and standard deviations (means ± SD) of heavy metals in the composite samples of three fruits (banana, jackfruit, and mango) highly consumed by the Bangladeshi population.

Fruits	Heavy metals (mg/kg fw)							
	As	Cd	Pb	Cr	Mn	Ni	Cu	Zn
Banana	ND	ND	0.003 ± 0.003	0.317 ± 0.012	10.747 ± 0.36	0.037 ± 0.001	0.946 ± 0.043	0.235 ± 0.003
Jackfruit	0.007 ± 0.0	0.037 ± 0.0	0.017 ± 0.015	0.863 ± 0.080	24.76 ± 1.01	0.882 ± 0.012	11.78 ± 0.419	1.19 ± 0.01
Mango	0.013 ± 0.0	0.005 ± 0.0	0.642 ± 0.556	0.893 ± 0.149	6.06 ± 0.315	0.293 ± 0.004	7.891 ± 0.307	0.604 ± 0.01
MAC (FAO/WHO, 2002)	1.0	0.05	0.1	1.0		0.8	4.5	

MAC = Maximum allowable concentration; fw = fresh weight.

Table 2
Mean concentrations (mg/kg fw) and standard deviations (means ± SD) of heavy metals in the composite samples of seven vegetables (brinjal, bean, carrot, green chilli, onion, potato, and tomato) highly consumed by the Bangladeshi population.

Vegetables	Heavy metals (mg/kg fw)							
	As	Cd	Pb	Cr	Mn	Ni	Cu	Zn
Brinjal	0.006 ± 0.001	0.041 ± 0.032	0.011 ± 0.011	0.497 ± 0.029	13.752 ± 2.354	0.949 ± 0.074	6.819 ± 0.552	0.567 ± 0.03
Bean	0.018 ± 0.007	0.008 ± 0.001	0.057 ± 0.050	1.110 ± 0.054	28.352 ± 1.96	2.047 ± 0.012	6.036 ± 0.355	4.75 ± 0.12
Carrot	0.006 ± 0.001	0.023 ± 0.003	0.029 ± 0.025	0.296 ± 0.021	6.986 ± 0.656	0.103 ± 0.007	2.254 ± 0.147	0.074 ± 0.02
Green chilli	0.004 ± 0.001	0.023 ± 0.011	0.006 ± 0.005	0.650 ± 0.039	16.95 ± 1.247	2.259 ± 0.133	8.726 ± 0.693	1.967 ± 0.08
Onion	0.008 ± 0.002	0.023 ± 0.017	0.027 ± 0.023	0.542 ± 0.131	12.016 ± 1.478	0.548 ± 0.05	3.632 ± 0.308	3.449 ± 0.13
Potato	0.006 ± 0.001	0.013 ± 0.007	0.007 ± 0.006	0.528 ± 0.051	6.928 ± 1.254	0.643 ± 0.028	4.3 ± 0.514	3.019 ± 3.0
Tomato	0.006 ± 0.002	0.056 ± 0.004	0.005 ± 0.004	0.795 ± 0.059	16.319 ± 0.849	0.69 ± 0.031	9.718 ± 0.555	2.0 ± 0.2
MAC (FAO/WHO, 2011)	0.1	0.05	0.1	2.3		10	40	

MAC = Maximum allowable concentration; fw = fresh weight.

maximum Ni levels were found as 0.103 mg/kg in carrot and 2.259 mg/kg in green chilli, respectively (Table 2). Ni concentrations in all the vegetable samples were found below the maximum permissible value (10 mg/kg) set by WHO/FAO (Codex, 2001). In the literature, average Ni concentration in vegetables was reported as 1.9 (0.02–12) (Islam et al., 2015), 1.4 (0.3–4.7) (Rahman et al., 2013), and 3.0 mg/kg (Ahmad and Goni, 2010) in three different regions of Bangladesh, whereas a Nigerian study reported Ni concentrations in the range of 0.083–0.119 mg/kg in banana and 0.24 ± 0.01 mg/kg in selected different leafy vegetables (Sobukola et al., 2010).

In this study, copper (Cu) concentration varied in the range of 0.946–11.78 mg/kg in fruit samples, and decreased in the following order: jackfruit > mango > banana (Table 1). In the vegetable samples, Cu concentration varied in the range of 2.254–9.718 mg/kg, and decreased in the following order: tomato > green chilli > brinjal > bean > potato > onion > carrot (Table 2). Cu content in all the vegetable samples was found to be below the safe limit (40 mg/kg) set by WHO/FAO (Codex, 2001). In the literature, Cu content has been reported to be in the range of 0.27–0.05 in Turkey (Hamurcu et al., 2010) and 2.4–9.8 mg/kg in vegetable foodstuff (Singh and Taneja, 2010). Average Cu concentration was found to be 0.03 in eight different leafy vegetables from selected markets in Nigeria (Sobukola et al., 2010), 11.50 (Maleki and Zarasvand, 2008), 8.51 in nonleafy vegetables grown in Samta village, Bangladesh (Alam et al., 2003), 1.30 and 0.526 in leafy and nonleafy vegetables grown in Mumbai, India (Tripathi et al., 1997), and 0.651 mg/kg in vegetables from Tianjin, China (Zhou et al., 2000). Three recent studies in Bangladesh reported Cu concentration in vegetables as 2.5 (0.3–32) (Islam et al., 2015), 21 (2.1–86) (Rahman et al., 2013), and 3.9 mg/kg (Ahmad and Goni, 2010).

Mean zinc (Zn) concentration in fruits varied in the range of 0.235–1.19 mg/kg in fruits (Table 1) with the lowest level in banana and highest in jackfruit. In the vegetable samples, mean Zn concentration varied in the range of 0.074–4.75 mg/kg with the lowest value in carrot and highest in bean (Table 2). In the literature, mean Zn concentration in banana was found to be 0.046 (Sobukola et al., 2010), 5.59 (Radwan and Salama, 2006), and 1.50 mg/kg (Onianwa et al., 2000). Median Zn concentration in the vegetable samples was found to be 50 mg/kg in a severely As-contaminated area of Bangladesh (Rahman et al., 2013) and in the range of 19.54–42.06 mg/kg in another study (Ahmad and Goni, 2010).

3.2. EDI of heavy metals

The EDI of eight metals (As, Cd, Pb, Cr, Mn, Ni, Cu, and Zn) was calculated according to the mean concentration of each metal in each food and the respective consumption rates. The EDI and maximum tolerable daily intake (MTDI) of studied metals from consumption of fruits and vegetables are shown in Table 3. Total daily intake of As, Cd, Pb, Cr, Mn, Ni, Cu, and Zn were 1E-04, 3E-04, 7E-04, 0.008, 0.184, 0.01, 0.077, and 0.023 mg/day, respectively. Daily intakes of all the metals were less than the MTDI. In fruit and vegetable samples, mean values of EDI decreased in the following order: Mn > Cu > Zn > Ni > Cr > Pb > Cd > As.

3.3. Noncarcinogenic risk

The health risks from consumption of contaminated fruits and vegetables by adult populations were assessed based on THQ, which is the ratio of determined dose of a pollutant to a reference dose level. If THQ > 1, the exposed population will likely to experience a detrimental effect (Wang et al., 2005). THQs of the eight studied metals are listed in Table 4. It is evident from the table that THQs of all the metals were <1 for all fruit and vegetable species. The ranking orders of TTHQ for fruit and vegetable samples were jackfruit > mango > banana and bean > tomato > brinjal > carrot > potato > onion > green chilli, respectively.

TTHQ was found to be >1 only in bean, indicating the potential health risk of its consumption. TTHQs of Mn and Cu were >1 (Fig. 2) through consumption of all vegetables, suggesting that people would experience significant health risks if they only ingested these two metals from all those vegetables. However, TTHQs through consuming other fruits and vegetables were ≤1, indicating no or little potential significant health risks in the consumption of

Table 3
Comparison of the estimated daily intake (EDI) of heavy metals from highly consumed fruit and vegetable samples with the corresponding maximum tolerable daily intake (MTDI) in the Bangladeshi population.

	Consumption rate (g/day/person)	Estimated daily intake (EDI) toxic trace elements (mg/day)							
		As	Cd	Pb	Cr	Mn	Ni	Cu	Zn
Fruits									
Banana	44.7	ND	ND	2.2E-06	0.0002	0.008	2.8E-05	7E-04	2E-04
Jackfruit	44.7	5.2E-06	2.8E-05	1.3E-05	0.0006	0.018	7E-04	0.009	9E-04
Mango	44.7	9.7E-06	3.7E-06	5E-04	0.0007	1.2E-05	2E-04	0.006	4E-04
EDI from all fruits		1.5E-05	3.1E-05	5E-04	0.002	0.026	9E-04	0.015	0.002
Vegetables									
Brinjal	130	1.3E-05	8.9E-05	2.4E-05	0.001	0.03	0.002	0.015	0.001
Bean	130	3.9E-05	1.7E-05	1E-04	0.002	0.061	0.004	0.013	0.01
Carrot	130	1.3E-05	5E-05	6.3E-05	6E-04	0.015	2E-04	0.005	2E-04
Green chilli	10.5	7E-07	4E-06	1.1E-06	1E-04	0.003	4E-04	0.002	3E-04
Onion	22	2.9E-06	8.4E-06	9.9E-06	2E-04	0.004	2E-04	0.001	0.001
Potato	70.3	7E-06	1.5E-05	8.2E-06	6E-04	0.008	8E-04	0.005	0.004
Tomato	130	1.3E-05	1E-04	1.1E-05	0.002	0.035	0.001	0.021	0.004
EDI from all vegetables		8.9E-05	3E-04	2E-04	0.007	0.157	0.01	0.062	0.021
Total intake from food		1E-04	3E-04	7E-04	0.008	0.184	0.01	0.077	0.023
MTDI		0.13	0.021	0.21	0.2	2–5	0.3	30	60

Table 4
Target hazard quotient (noncarcinogenic risk) of heavy metals from consuming fruits and vegetables.

	Target hazard quotient (THQ)								
	As	Cd	Pb	Cr	Mn	Ni	Cu	Zn	TTHQ
Fruits									
Banana	0	0	6E-04	2E-04	0.057	0.001	0.018	6E-04	0.078
Jackfruit	0.017	0.009	0.004	4E-04	0.132	0.033	0.219	0.003	0.418
Mango	0.032	0.001	0.140	4E-04	9E-05	0.012	0.147	0.001	0.33
TTHQ from fruits	0.05	0.01	0.141	0.001	0.189	0.045	0.384	0.005	**HI** = 0.825
Vegetables									
Brinjal	0.043	0.03	0.007	7E-04	0.213	0.103	0.369	0.004	0.77
Bean	0.130	0.006	0.035	0.002	0.439	0.222	0.327	0.034	1.194
Carrot	0.043	0.017	0.018	4E-04	0.108	0.011	0.122	5E-04	0.320
Green chilli	0.002	0.001	3E-04	8E-05	0.021	0.02	0.038	0.001	0.084
Onion	0.01	0.003	0.003	1E-04	0.031	0.01	0.033	0.004	0.095
Potato	0.023	0.005	0.002	4E-04	0.101	0.038	0.126	0.012	0.307
Tomato	0.043	0.04	0.003	0.001	0.253	0.075	0.526	0.014	0.956
TTHQ from vegetables	0.296	0.102	0.069	0.005	1.166	0.478	1.542	0.071	**HI** = 3.727
TTHQ	0.345	0.112	0.21	0.006	1.355	0.523	1.926	0.076	**HI** = 4.552

TTHQ (Total THQ); HI (Hazard Index).

these food items.

The HI value expresses the combined noncarcinogenic effects of multiple elements. For consumption of selected fruits, HI was <1 (0.825), whereas for consumption of selected vegetables, HI was >1 (3.727) (Table 4), indicating that consumers may experience adverse health effects from vegetable consumption only, while fruits were found to be safer.

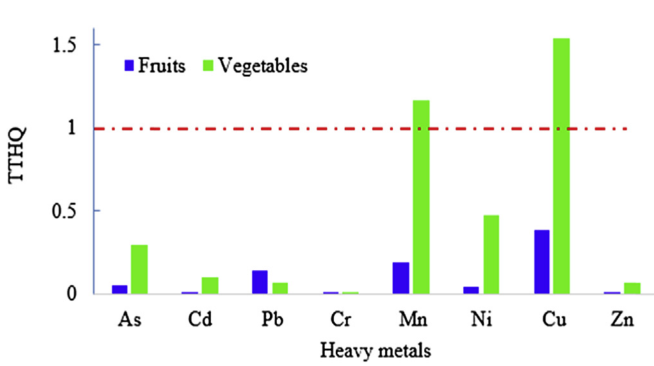

Fig. 2. Total target hazard quotient (TTHQ) of heavy metals from consuming fruits and vegetables. Horizontal dotted red line indicates the threshold value of hazard quotient.

3.4. CRs of As and Pb

The target CRs derived from the intake of As and Pb were calculated as these elements may promote both noncarcinogenic and carcinogenic effects depending on the exposure dose. The CRs of As and Pb due to exposure from different species of fruits and vegetables are listed in Table 5. CRs of As ranged from 0 to 1.5E-08 in fruits and 2.1E-09 to 5.9E-08 in vegetables, whereas 1.9E-08 to 4.1E-06 in fruits and 8.9E-09 to 9.2E-05 in vegetables, for Pb.

Total CR $> 10^{-4}$ is considered to be unacceptable (USEPA, 1989, 2015). In all fruits and vegetables, CRs of As were found to be lower than the negligible range, whereas those of Pb were found within the acceptable range in tomato and mango among the studied samples, indicating no CR from As consumption from fruits and vegetables in the adult population, but a small concern for Pb-induced CR does exist, particularly from consumption of mango and tomato.

4. Conclusion

This study revealed the presence of heavy metal concentration in selected highly consumed fruits and vegetables grown in Bangladesh as well as EDI of metal from those foods by adult Bangladeshi people and their health risk implications from

Table 5
Target carcinogenic risk (CR) of heavy metals due to consumption of fruits and vegetables.

	As	Pb
Fruits		
Banana	0	1.9E-08
Jackfruit	7.8E-09	1.1E-07
Mango	1.5E-08	4.1E-06
Vegetables		
Brinjal	1.9E-08	2.0E-07
Bean	5.9E-08	1.1E-06
Carrot	1.9E-08	5.3E-07
Green chilli	2.1E-09	8.9E-09
Onion	4.4E-09	8.4E-08
Potato	1.1E-08	7.0E-08
Tomato	1.9E-08	9.2E-05
Total CR	1.56E-07	9.82E-05

consuming those foods in terms of THQ and CR. Heavy metal concentration widely varied in fruits and vegetables, and all the metals in the analyzed foods were found to be below the recommended maximum allowable concentration (MAC), except for Pb in mango and Cd in tomato. Pb content in mango was six times higher than the safe or permissible limit. From consumption point of view, EDIs of all the metals through consumption of fruits and vegetables were below the MTDI level. From human health point of view, TTHQ values of Mn and Cu were >1 through consumption of all vegetables, indicating that people would experience significant health risks if they ingest these two vital metals through consuming the studied vegetables. Consumption of fruits would be safe for the consumers as HI value was <1 (0.825), whereas that of the selected vegetables consumption was >1 (3.727), indicating the possible adverse health effects from vegetable consumption only. Concerning the CR, the total CRs of As were found to be below 10^{-6} and 9.82E-05 for Pb, suggesting no potential CR from As consumption, but slight concern for Pb-induced CR through consumption of the studied fruits and vegetables. The findings of this study significantly contribute to the field of food safety, considering the health risk for Bangladeshi population as it represents the composite samples of highly consumed fruits and vegetables, grown and consumed in the country.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2016.02.060.

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