

## Concentrations of heavy metals in vegetables around the industrial area of Dhaka city, Bangladesh and health risk assessment

\*Md Saiful Islam and Hoque, M. F.

Department of Soil Science, Patuakhali Science and Technology University, Dumki Patuakhali-8602, Bangladesh

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### Abstract

The present study evaluates the concentration of heavy metals in eight different vegetables species around industrial area of Dhaka city Bangladesh. Metals were measured by inductively coupled plasma mass spectrometry after microwave digestion system. The levels of Cr, Ni, Cu, Zn, As, Cd and Pb in vegetables were 0.61 to 3.0, 1.6 to 12, 8.3 to 34, 16 to 119, 0.007 to 0.24, 0.009 to 1.0 and 0.06 to 3.5 mg/kg fw, respectively. There were differences in the concentrations of metals between vegetables species. Potential health risks of heavy metals to the local population via vegetables consumption were estimated. The health risks of Cr, Cu, As, Cd and Pb should be of great concern due to vegetables consumption.

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### Introduction

Heavy metal pollution of agricultural soil and vegetables is one of the most severe ecological problems on a world scale and also in Bangladesh. The food chain contamination is the major pathway of heavy metal exposure for humans (Khan *et al.*, 2008). The consumption of vegetables is one of the most important pathways for trace metals that harm human health (Sipter *et al.*, 2008). Industrial or municipal wastewater irrigation is a common reality in three fourth of the cities in Asia, Africa, and Latin America (Gupta *et al.*, 2008). In Bangladesh, more than 90 vegetables and 60 fruits are being grown in the country (Alam *et al.*, 2003). Major vegetable crops include brinjal (egg plant), chilli, lady's finger, potato, tomato, bottle gourd, red amaranth etc. In an average Bengali home, the main meal would consist of boiled rice served with some sort of vegetables. Investigations on the accumulation of trace metals from vegetables grown around the industrial sites have revealed high levels of Ni, Pb and Cd in vegetables (Ahmad and Goni, 2010). In this context, the risks associated with the consumption of contaminated food grown near the industrial area may be a potential health concern. However, description on metal toxicity scarcely found in literature and related data are severely insufficient to accomplish the risk assessment. Health risks have been evaluated by numerous methods but most commonly, risk to the human health is computed in terms of Target hazard quotient (THQ) which is based on the concentration of trace metals in the edible parts of vegetables, in

comparison with the reference dose of the metal intake and body weight of the consumers (Pandey *et al.*, 2012). The present study investigates the levels of Cr, Ni, Cu, Zn, As, Cd and Pb in eight different vegetable species from agricultural fields besides the Turag River in Dhaka city, Bangladesh. The aim of this work is to measure the levels of heavy metals in edible portion of vegetables and to assess the health risk due to vegetables consumption.

### Materials and Methods

#### Study area

For present study, the agriculture fields selected near the industrial area of Dhaka city, located besides the Turag River (Figure 1). Agricultural fields besides the Turag River were selected based on farmer's interview where irrigation with contaminated river water has been a common practice for many years. Numerous industries (leathers, textiles, metals processing, paper mills, electronic goods, power plant, fertilizers, pharmaceuticals, dyeing, battery manufacturing, ink manufacturing, Pb-Zn melting, brick fields, etc.) are situated near the selected fields (Rahman *et al.*, 2012). Most of the treated and untreated industrial effluents are being discharged to this river. Several acres of agricultural land irrigated by contaminated river water and farmers cultivate various types of vegetable crops of economic importance. The greater Dhaka city is one of the most densely populated cities in the world, home to approximately twelve million people of which less than 25% are served by sewage treatment facility

\*Corresponding author.  
Email: [msaifulpstu@yahoo.com](mailto:msaifulpstu@yahoo.com)

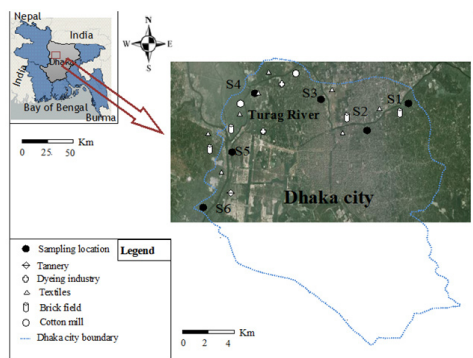


Figure 1. Map showing the study area in Dhaka city, Bangladesh.

(Islam *et al.*, 2014; Ahmad *et al.*, 2010). The Turag River received domestic raw sewage as well as industrial effluents from the surrounding habitation and nearby industrial belt. As per the information given by the local farmers we have identified the above area for the present study.

### Vegetable sampling

The sampling was conducted in February-March, 2010. The samples were collected from six agriculture fields besides the river. At each sampling station, same species of vegetables were collected as sub-samples and were thoroughly mixed to form a composite sample. Forty eight samples of eight different vegetables i.e. tomato (*Solanum lycopersicum*), bottle gourd (*Lagenaria siceraria*), brinjal (*Solanum melongena*), pumpkin (*Cucurbita maxima*), green amaranth (*Amaranthus viridis* L.), red amaranth (*Amaranthus paniculatus* L.), chilli (*Capsicum annum* L.) and banana (*Musa* sp.) were collected from the selected agricultural fields. All samples were put in polythene zip-bags and transported to the laboratory on the day of sampling. Each vegetable sample was carefully washed with distilled water and the edible parts were cut into small pieces and then oven dried at 70–80°C to attain constant weight (Tiwari *et al.*, 2011). The moisture contents were calculated by recording the fresh and dry weights. The dried vegetable samples were crumbled and pulverized with a porcelain mortar and pestle, sieved through 2 mm nylon sieve and stored in polythene bottle in refrigerator. The processed samples were brought to Yokohama National University, Japan for chemical analysis.

### Sample digestion

All chemicals were analytical grade reagents and MilliQ water was used for solution preparation. The Teflon vessel and polypropylene containers were cleaned, soaked in 5% HNO<sub>3</sub> for more than 24 h, then rinsed with Milli-Q water and dried. For vegetable, 0.2 g of dried sample was digested with

6 mL 69% HNO<sub>3</sub> (Kanto Chemical Co, Japan) and 2 mL H<sub>2</sub>O<sub>2</sub> (Wako Chemical Co, Japan) in a Microwave Digestion System. The digested samples were then transferred into a Teflon beaker and total volume was made up to 50 mL with MilliQ water (Elix UV5 and Milli-Q, Millipore, USA). The digested solution was then filtered by using 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). Afterwards, the vessels were cleaned by Milli-Q water and dried with air. Finally, blank digestion with 5 mL HNO<sub>3</sub> following the said digestion procedures were carried out to clean up the digestion vessels (Berghof's product user manual, 2008).

### Instrument analysis and quality check

For trace metals, samples were analysed by using Inductively Coupled Plasma mass spectrometer (ICP-MS, Agilent 7700 series). Multi-element Standard XSTC-13 (Spex CertiPrep® USA) solutions was used to prepare calibration curve. Internal calibration standard solutions containing 1.0 mg/L of Indium (In), Yttrium (Y), Beryllium (Be), Tellurium (Te), Cobalt (Co) and Thallium (TI) were purchased from Spex Certi Prep® USA. A multielement solution (Agilent Technologies, USA) 1.0 μg/L was used as tuning solution covering a wide range of masses of elements. All test batches were evaluated using an internal quality approach and validated if they satisfied the defined Internal Quality Controls (IQCs). For each experiment, a run included blank, certified reference materials (CRM) and samples analysed in duplicate to eliminate any batch-specific error. Plant materials (NIST, 1547 Peach leave) of National Institute and Technology were used as CRM in each sample batch to verify the accuracy and precision of the digestion procedure and for subsequent analyses.

### Data analysis

#### Estimated daily intake of trace metals from vegetables

The estimated daily exposure of trace metals (EDEM) through vegetables was dependent on metal concentrations in vegetables (on fresh weight basis), daily vegetable consumption, as well as body weight, which was calculated with the following formula:

$$EDEM = \frac{DIM}{BW} \quad (1)$$

DIM = Daily intake of metals = daily vegetable consumption × mean metal concentration in vegetable

BW = Body weight

Where, daily vegetable consumption rate for adult residents was an average of 166.08 g (BBS, 2010, Preliminary report on household income and expenditure survey) and the bodyweight of an adult resident was set to 60 kg in the present study.

#### Calculation of health risk

In this study, the health risks associated with the consumption of vegetables by the local inhabitants were assessed based on the target hazard quotients (THQs). This method of estimating risk using THQ was provided in the U.S. EPA Region III risk-based concentration table (USEPA, 2000) and is based on the following equation:

$$THQ = \frac{EF \times ED \times FI \times MC}{RfD \times BW \times AT} \times 10^{-3} \quad (2)$$

Where, EF is the exposure frequency (365 days/year), ED is the exposure duration (70 years), FI is the food ingestion (g/person/day), MC is the metal concentration in vegetables (mg/kg, on fresh weight basis), RfD is the oral reference dose (mg/kg/day), BW is the average body weight (adult, 60 kg), AT is the averaging time for non-carcinogens (365 days/year  $\times$  number of exposure years, assuming 70 years in this study). The oral reference doses were based on 0.003, 0.02, 0.04, 0.3, 0.0003, 0.0005, and 0.004 mg/kg/day for Cr, Ni, Cu, Zn, As, Cd and Pb, respectively (JECFA, 1993; USEPA, 2007). If the THQ is less than 1, the exposed population is unlikely to experience obvious adverse effects. If the THQ is equal to or higher than 1, there is a potential health risk, and related interventions and protective measures should be taken.

#### Statistical analysis

The data were statistically analyzed using the statistical package, SPSS 16.0 (SPSS, USA). The means and standard deviations of the metal concentrations in vegetables were calculated. A Pearson bivariate correlation was used to evaluate the inter-element relationship in vegetables. Multivariate Post Hoc Tukey tests were employed to examine the statistical significance in the differences of mean concentrations of trace metals among vegetables and sites.

## Results and Discussion

#### Heavy metals concentration in vegetables

The heavy metal concentrations (mg/kg fw) in the edible part of vegetables are listed in Table 1. The concentration of metals varied greatly among

plant species and sample locations. The average concentrations of trace metals in all vegetable samples are in the following decreasing order: Zn > Cu > Ni > Cr > Pb > Cd > As. The mean concentrations of heavy metals in various vegetable species collected from the study area were compared with the standards set for vegetables by FAO & WHO, 2011 (Table 1). Zn for most of the vegetables and As, Cd and Pb in some vegetables exceeded the FAO & WHO standards in food (Table 1) (The guideline values are decided based on contaminants, toxins and many substances regarding their occurrence in foods and their significance for human and animal health). On the other hand, significant differences in metal concentrations among the different vegetables implied that different vegetable species had different abilities and capacities to take up and accumulate different metals (Table 1).

The metal contents in the leafy vegetables were slightly higher than the fruit vegetables could probably be ascribed to its high foliar surface area and industrial activities. When smelting and other industrial activities started, heavy smoke containing various kinds of trace metals, metalloids and organic pollutants would have been discharged into the air and the vegetables growing on these sites could well have been the first recipients of these substances (Bi *et al.*, 2009; Luo *et al.*, 2011). A major pathway for Pb may enter the above-ground tissues of plants is through foliar deposition (Xu *et al.*, 2013). Our observations revealed pronounced Pb concentration in vegetable species due to the industrial waste, coal in the brick fields and power station at the study sites, which might result to the deposition of particulate matter (PM) on vegetables. Thus, the vegetables were exposed to fine particles of Pb from PbSO<sub>4</sub>, PbO and PbCO<sub>3</sub>. Uzu *et al.* (2011) showed that PM deposited on plant leaves and penetrate inside the plant tissues. This finding suggests that growing of fruit vegetables in these areas are not safe for human consumption. However, when comparing with results of some other study in Bangladesh and other countries indicated the contamination of vegetables by studied metals (Table 2).

#### Correlation analysis

Statistical analyses were performed in order to elucidate the associations among metals in vegetables. Inter metal interactions may illustrate the sources and pathways of the metals present in vegetables. Pearson's correlation coefficients for the investigated metals are depicted in Table 3. A clear pattern of strong association was found among the metal pairs in vegetables (Parashar and Prasad,

Table 1. Heavy metal content (mg/kg fw) in vegetables from Turag riverside fields of Dhaka City, Bangladesh

English name	Scientific name		Cr	Ni	Cu	Zn	As	Cd	Pb
Tomato (6)	<i>Solanum lycopersicum</i>	Mean ±SD	1.23±0.32 <sup>a</sup>	2.13±0.46 <sup>a</sup>	17.50±2.51 <sup>a</sup>	24.73±2.36 <sup>ab</sup>	0.01±0.00 <sup>a</sup>	0.05±0.01 <sup>a</sup>	0.23±0.05 <sup>ab</sup>
		Range	0.82-1.64	1.61-2.69	13.94-20.44	21.57-28.47	0.01-0.02	0.04-0.06	0.17-0.31
Boottle gourd (6)	<i>Lagenaria siceraria</i>	Mean ±SD	0.91±0.24 <sup>a</sup>	8.05±1.73 <sup>b</sup>	10.42±1.49 <sup>b</sup>	29.29±2.79 <sup>a</sup>	0.02±0.00 <sup>a</sup>	0.04±0.01 <sup>a</sup>	0.69±0.15 <sup>b</sup>
		Range	0.61-1.21	6.08-10.19	8.30-12.17	25.55-33.72	0.01-0.02	0.03-0.05	0.52-0.94
Brinjal (6)	<i>Solanum melongena</i>	Mean ±SD	1.02±0.27 <sup>a</sup>	4.52±0.97 <sup>a</sup>	17.04±2.44 <sup>a</sup>	18.68±1.78 <sup>b</sup>	0.04±0.01 <sup>a</sup>	0.24±0.05 <sup>bc</sup>	0.07±0.02 <sup>a</sup>
		Range	0.68-1.35	3.42-5.73	13.57-19.90	16.29-21.50	0.03-0.04	0.17-0.30	0.06-0.10
Pumpkin (6)	<i>Cucurbita maxima</i>	Mean ±SD	1.45±0.38 <sup>ac</sup>	5.82±1.25 <sup>c</sup>	11.44±1.64 <sup>b</sup>	46.10±4.39 <sup>c</sup>	0.02±0.00 <sup>a</sup>	0.01±0.00 <sup>a</sup>	0.25±0.06 <sup>a</sup>
		Range	0.96-1.93	4.40-7.38	9.12-13.37	40.20-53.06	0.02-0.03	0.01-0.01	0.19-0.34
Green amaranth (6)	<i>Amaranthus viridis</i> L.	Mean ±SD	2.28±0.60 <sup>bc</sup>	2.46±0.53 <sup>a</sup>	15.60±2.23 <sup>a</sup>	78.45±7.48 <sup>d</sup>	0.19±0.04 <sup>c</sup>	0.15±0.03 <sup>b</sup>	2.54±0.56 <sup>d</sup>
		Range	1.52-3.04	1.86-3.12	12.42-18.22	68.42-90.31	0.14-0.24	0.10-0.18	1.93-3.45
Red amaranth (6)	<i>Amaranthus paniculatus</i> L.	Mean ±SD	2.13±0.56 <sup>b</sup>	5.76±1.24 <sup>c</sup>	19.35±2.77 <sup>ac</sup>	103.74±9.89 <sup>c</sup>	0.15±0.03 <sup>b</sup>	0.84±0.17 <sup>d</sup>	1.99±0.44 <sup>c</sup>
		Range	1.42-2.84	4.35-7.30	15.41-22.60	90.47-119.42	0.11-0.19	0.60-1.05	1.51-2.70
Chilli (6)	<i>Capsicum annum</i> L.	Mean ±SD	1.23±0.32 <sup>a</sup>	4.76±1.03 <sup>b</sup>	24.18±3.46 <sup>c</sup>	33.29±3.17 <sup>a</sup>	0.01±0.00 <sup>a</sup>	0.33±0.07 <sup>c</sup>	0.17±0.04 <sup>a</sup>
		Range	0.82-1.64	3.60-6.03	19.26-28.24	29.04-38.33	0.01-0.01	0.23-0.41	0.13-0.23
Banana (6)	<i>Musa</i> sp.	Mean ±SD	1.27±0.34 <sup>a</sup>	9.21±1.98 <sup>b</sup>	29.39±4.21 <sup>d</sup>	75.46±7.19 <sup>d</sup>	0.01±0.00 <sup>a</sup>	0.05±0.01 <sup>a</sup>	0.11±0.02 <sup>a</sup>
		Range	0.85-1.70	6.96-11.67	23.41-34.33	65.81-86.87	0.01-0.02	0.04-0.06	0.08-0.15
Permissible levels as per (FAO & WHO, 2011)			2.3	66.9	40	20	0.1	0.2	0.3

Table 2. Comparison with other reports of metal concentrations (mg/kg fw) in vegetables of the present study in Bangladesh

District (Country)	Sampling site description	Cr	Ni	Cu	Zn	As	Cd	Pb	References
Dhaka (Bangladesh)	Industrial area	1.44 (0.61-3.04)	5.34 (1.61-11.7)	18.1 (8.30-34.3)	51.2 (16.3-119)	0.057 (0.007-0.24)	0.21 (0.009-1.05)	0.76 (0.06-3.45)	This study
Dhaka (Bangladesh)	Industrial area	1.66	2.97	3.85	NA	NA	0.62	3.89	Ahmad and Gani, 2008
Noakhali (Bangladesh)	Arsenic contaminated area	0.64 (0.18-1.91)	1.44 (0.32-4.67)	20.6 (2.1-86.3)	NA	0.05 (0.011-0.145)	0.058 (0.006-0.265)	3.7 (0.67-16.5)	Rahman <i>et al.</i> , 2013
Dabaoshan (China)	Near mine area	NA	NA	1.18 (0.28-3.61)	10.53 (2.34-40.2)	NA	0.19 (0.001-0.71)	0.17 (0.01-0.39)	Zhuang <i>et al.</i> , 2009
Varanasi (India)	Urban area	NA	NA	36.4 (20.5-71.2)	NA	NA	2.08 (1.1-4.5)	1.42 (0.9-2.2)	Sharma <i>et al.</i> , 2007

Table 3. Correlation coefficient of heavy metals in vegetables samples from Turag riverside fields of Dhaka City, Bangladesh

Metal	Cr	Ni	Cu	Zn	As	Cd	Pb
Cr	1						
Ni	-0.23	1					
Cu	-0.009	0.301*	1				
Zn	0.664**	0.208	0.334*	1			
As	0.700**	-0.28	-0.071	0.731**	1		
Cd	0.380**	-0.014	0.245	0.563**	0.525**	1	
Pb	0.802**	-0.242	-0.175	0.697**	0.921**	0.449**	1

Table 4. Estimation of daily intake of metal (mg/kg bw) and target hazard quotient (THQ)

English name		Cr	Ni	Cu	Zn	As	Cd	Pb	Total
Tomato (6)	EDI	0.0034	0.0059	0.048	0.0685	0.00004	0.00014	0.00062	0.13
	THQ	1.14	0.29	1.21	0.23	0.12	0.28	0.16	3.43
Boottle gourd (6)	EDI	0.003	0.022	0.029	0.081	0.00005	0.00012	0.002	0.14
	THQ	0.84	1.11	0.72	0.27	0.18	0.24	0.48	3.84
Brinjal (6)	EDI	0.003	0.013	0.047	0.052	0.0001	0.0007	0.0002	0.12
	THQ	0.94	0.63	1.18	0.17	0.33	1.33	0.05	4.62
Pumpkin (6)	EDI	0.0040	0.02	0.03	0.13	0.00007	0.00003	0.00070	0.18
	THQ	1.33	0.81	0.79	0.43	0.23	0.067	0.18	3.83
Green amaranth (6)	EDI	0.01	0.01	0.04	0.22	0.0005	0.0004	0.0070	0.28
	THQ	2.10	0.34	1.08	0.72	1.73	0.81	1.76	8.55
Red amaranth (6)	EDI	0.01	0.02	0.05	0.29	0.0004	0.0023	0.0055	0.37
	THQ	1.97	0.80	1.34	0.96	1.38	4.64	1.38	12.46
Chilli (6)	EDI	0.0034	0.01	0.07	0.09	0.00002	0.00091	0.00046	0.18
	THQ	1.13	0.66	1.67	0.31	0.08	1.82	0.12	5.79
Banana (6)	EDI	0.0035	0.025	0.08	0.21	0.00004	0.00014	0.00030	0.32
	THQ	1.18	1.27	2.03	0.70	0.14	0.28	0.07	5.67
RfDo (mg/kg/day)		0.003	0.02	0.04	0.3	0.0003	0.0005	0.0035	0.3673

2013). Significant correlation was observed for Cr with most of the metals except Ni and Cu, where as Cu showed correlation with Ni and Zn (Table 3). These strong correlations among metal-metal pair may be an indication of common sources of these metals (Abbasi *et al.*, 2013; Mohamed *et al.*, 2003); similar or nearly identical metal accumulation properties of vegetables (Xu *et al.*, 2013). There are many industries, agricultural farms using herbicides, fungicides and chemical fertilizers besides the Turag River of Dhaka city through which metals can be released.

#### Health risk assessment

The accumulation of metals in the edible parts of vegetables could have a direct impact on the health of nearby inhabitants. Therefore, the metal

contaminated vegetables could be a concern to the local residents. The concentrations of Zn, Cd and Pb in most vegetables exceeded the food safety standards, with the average levels of metals being 2.6, 1.1, and 2.5 times that of the maximum permissible level, respectively (Table 1). The estimated daily intake of metal (EDI) and target hazard quotients (THQ) were used to assess the human health risks associated with trace metal contamination of selected vegetables grown in the study area. For adults, the average EDI values of Cr, Ni, Cu, Zn, As, Cd and Pb were, 0.004, 0.015, 0.05, 0.14, 0.0002, 0.0006 and 0.0021 mg/kg bw, respectively (Table 4). The THQ of each metal through consumption of vegetables in the study area decreased in the order of Cr > Cu > Cd > Ni > As > Pb > Zn. The THQ value of each metal was less than 1, indicating that intake of a single metal through consumption of vegetables does not pose a considerable potential health hazard except Cu. However, total metal THQ value (sum of individual metal THQ) for individual vegetable was higher than 1 (Table 4). Potential health risks from exposure to vegetables are therefore of some concern.

In addition, there are also other sources of metal exposures, such as consumption of other foodstuffs and dust inhalation, which were not included in this study. The result indicates that those living around the study area were probably exposed to some potential health risks through the intake of metals via consuming locally grown vegetables. If the whole intake of metals through dietary means (vegetables) is taken into account, the potential health risks involved in the consumption of local food should not be ignored.

## Conclusion

This study showed that the intensive uncontrolled operation of various industries has resulted in the release of trace metals in the local environment. Vegetables grown in the nearby sites were contaminated by the relevant metals, especially Zn, Cd and Pb, which could be a potential health concern to the local residents. In terms of food safety, although metal present in some vegetables were below the recommended legal limits established by the FAO/WHO, they must be eaten in moderation due to possible hazard and risks derived from metals ingestion. Moreover, different remediation measures should be taken promptly to remove existing metal contamination of the study area.

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