Health risk assessment for carcinogenic and non-carcinogenic heavy metal exposures from vegetables and fruits of Bangladesh

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Abstract: Most popular vegetables and fruits and their corresponding soil from the sub-urban industrial area of Bangladesh were collected and the concentration of carcinogenic (Pb, As, and Cd) and non-carcinogenic (Fe, Co, V, Cu, Cr, Zn, Mn, and Ni) heavy metals was determined. Health risk was evaluated by estimating daily heavy metal intake and computing cancer and non-cancer risks (ILCR and THQ) using probabilistic risk assessment model of US-EPA. Heavy metals in vegetables varied with vegetable species as well as metal types. Higher daily intake of As, Fe, Mn, and Pb was observed from the consumption of root and leafy vegetables. Moreover, the probability of an adult for developing cancer from the consumption of studied vegetables was greater than US-EPA threshold risk limit (>10⁻⁴) for As and Cd. In addition, cumulative cancer risk (∑ILCR) of all the studied vegetables and fruits exceeded the limit for fruit, root, leafy vegetables, and fruits (22, 15, 59, and 4%) with As, Cd, and Pb as 17, 81, and 2%, respectively. Non-cancer risk index also presented Pb, As, Mn, and Fe as the dominant contaminants of root and leafy vegetables that contributed 80–90% of HI. It suggests that the study area is unsuitable for growing leafy and root vegetables due to the risk of higher intakes of heavy metals which affect food safety. Mn, Pb, Fe, and As are the most predominant heavy metals posing non-cancer risk while Cd caused the highest cancer risk.

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PUBLIC INTEREST STATEMENT
The heavy metal content in the various vegetables and fruit species were investigated by collecting it at field condition. Further the toxicity level was determined by comparing the present data with the permissible limit recommended by WHO. Further, the carcinogenic and non-carcinogenic health risks through the dietary intake was computed by using the US-EPA health risk assessment models.

The precise data specify that the cumulative non-cancer risk for Mn, Pb, Fe and As contributed 80–90% through leafy and root vegetables whereas Cd has the highest cancer risk. The outcome of this research will help to encourage the researchers to conduct research on health risk through the food intake. The output of this extensive research will be helpful to the concerned authorities to construct health safety policies to save the public from health risks.
1. Introduction

Vegetables and fruits are the most important components of human diet and it is well known that consumption of these food items on a regular basis is one of the possible health-improving practices. Moreover, recently, people all over the world have been concerned about the higher intake of fresh vegetables and fruits instead of red meat for good health as they significantly reduce the incidence of chronic diseases such as diabetics, cancer, cardiovascular diseases, and other age-related diseases (Prakash, Upadhyay, Gupta, Pushpangadan, & Singh, 2012). However, unsafe food consumption is a severe problem because of heavy metal contamination, which is caused by various anthropogenic activities (Cui et al., 2004). Usually, agricultural foods are adulterated with pollutants, especially heavy metals by direct and indirect industrial activities, automobile exhaust, excess metal-based fertilization, and pesticide application. In contrast, some heavy metals (As, Cd, and Pb) have no known beneficial role in human metabolism and are considered as chemical carcinogens even at very low levels of exposure (Jarup, 2003). Generally, heavy metals present in the environment in minute quantities become part of various food chains through biomagnification and their concentration increases to such a level that may prove to be toxic to both humans and other living organisms. However, except occupational exposures at related industries, diet is the main route of human exposure to heavy metals, which are one of the potential hazards associated with foodstuffs (Martí-Cid, Llobet, Castell, & Domingo, 2008). It has been reported that the dietary intake of lead, copper, and chromium through food is due to plant origin (fruits, vegetables, and cereals) and it is sometimes higher than permissible limits within urban areas (Yebpella et al., 2011).

Further, food safety is a major public health concern and the importance of the association with consumption of foodstuffs contaminated by heavy metals has increased worldwide (DMello, 2003). WHO has expressed its anxiety about the impact of food safety on public health in Bangladesh on its website. It reveals that unsafe food can be a significant reason for many chronic and non-chronic diseases including diarrhea, cancer, heart diseases, various kidney diseases, and birth defects (Akram & Richard, 2013). In Bangladesh, most of the foodstuffs, whether harvested, manufactured, or processed, are unsafe for consumption or adulterated to varying degrees. Rapidly increasing urbanization and industrialization of Bangladesh have been continuously emitting metal-contaminated fumes from industries and vehicles, have contributed to the contamination of agricultural soils, and consequently in food chain by depositing heavy metals on the fruit and vegetable surfaces during their production, transportation, and retailing (Ikeda et al., 2000). Further, application of wastewater to irrigate agricultural land and the use of agrochemicals such as metal-based fertilizers and pesticides play an important role in the contamination of agricultural products, where the use of agrochemicals is not well controlled (Naser, Shil, Mahmud, Rashid, & Hossain, 2009). Regular monitoring and assessment of heavy metal concentration in food crops near industrial and mining areas has been carried out in some developed and developing countries, but limited published data are available on heavy metal concentrations and dietary intake of these pollutants in the vegetables and fruits in the industrial areas of Bangladesh (Ahmad & Goni, 2010; Ikeda et al., 2000; Mahfuza et al., 2014; Narottam, Zaman, & Sofiur, 2012; Naser et al., 2009; Saiful & Hoque, 2014). Further, cancer is predicted to be an increasingly important cause of morbidity and mortality in Bangladesh in the next few decades. The estimated incidence of 12.7 million new cancer cases will rise to 21.4 million by 2030. International Agency for research on cancer (IARC) has estimated cancer-related death rate in Bangladesh to be 7.5% in 2005 and 13% in 2030 (Akram & Richard, 2013).

The uptake of heavy metals from soil to the edible parts of the vegetables and fruits is a significant path to harm human health. It is therefore important to control and limit the accumulation of heavy metals in vegetables from soil. To accomplish this, we need to investigate the transfer
characteristics of heavy metals from soil to vegetables. Further, the dietary intake of heavy metals through vegetables is very important for assessing their risk to human health. The knowledge of dietary intake of toxic elements in Bangladesh is limited. Therefore, this study evaluated the contamination level of carcinogenic (Cd, As, and Pb) and non-carcinogenic (Fe, V, Co, Cr, Cu, Zn, Mn, and Ni) heavy metals in common vegetables and fruits mostly consumed for health nutrition in Bangladesh and grown near industrial areas of sub-urban Dhaka. Further, the plant transfer factor (PTF) and the daily intake of heavy metals were estimated to know the source of human exposure risk. Finally, the probabilistic risk assessment models were used to determine the cancer and non-cancer risks of heavy metals intake through vegetables and fruits of study area to specify the toxic level of heavy metals which cause health risks.

2. Materials and methods

2.1. Geology and geomorphology of the study area

The study area lies within Dhaka-Gazipur terrace, a part of Madhupur Tract, located in the central part of Bangladesh. Its coordinates are 25°15′0″ N and 89°30′0″ E in DMS (Degrees Minutes Seconds) or 25.25 and 89.5 (in decimal degrees). The tract is a structural high that extends from the folded hills in the eastern fringe of Bengal basin. This elevated area is only a few meters above the surrounding rivers such as the Buriganga and Turag on the west and the Balu on the east. It is formed of Madhupur Clay. Due to higher elevation than the surrounding plains, the terrace has become a seat of urban and industrial development (Wikipedia, 2016). The map of the study area is shown in Figure 1.
2.2. Sampling
Sampling was carried out from January to August 2015 in the agricultural fields of two locations around the sub-urban Gazipur industrial area of Dhaka, Bangladesh. These fields have been irrigated with Turag River and Turag Khal water which receive pollution load from several industries such as textile and dyeing, metal processing, battery manufacturing, ink manufacturing, fertilizers, pharmaceuticals, and Pb-Zn melting and also from urban pollution, especially from domestic sewerage and heavy traffic. Several acres of agricultural land of this area are irrigated by contaminated river water and farmers cultivate various types of food crops for economic importance (Saiful & Hoque, 2014). The edible parts of the various kinds of popular and cheap vegetables and fruits from the agricultural fields and corresponding soil were collected randomly in triplicate for the analysis of heavy metals. The details of the vegetable and fruit samples collected from the study area are given in Table 1.

2.3. Analysis of carcinogenic and non-carcinogenic heavy metals
Vegetable and fruit samples were washed thoroughly with tap water and finally with deionized water and the edible parts of the vegetables were cut into small pieces for drying. Further, unwanted portions like plant root in the soil samples were removed and kept in watch glass for drying. Then, the collected vegetable and soil samples were air dried and then oven dried at 70–80°C until constant weight was obtained. The dried samples were sieved and ground to fine powder and preserved in zip polyethylene bags in desiccators for analysis. These processed vegetable, fruit, and soil samples were brought in to the National Institute of Radiological Sciences, NIRS, Chiba, Japan for subsequent analysis.

Hundred milligrams of finely ground soil samples were digested in Teflon PFA decomposition vessels with mineral acids (concentrated HNO₃ and HF) using a microwave pressure digester (CEM, MARS-5). Further, 50 mg of finely ground vegetable samples were digested with mineral acids and evaporated the digested soil and vegetable samples to dryness. Then, the soil and vegetable residues were dissolved in 2% HNO₃ to yield the final sample solutions of 50 ml and 20 ml, respectively. The digestion samples were made in duplicate. All the acids used were ultrapure analytical grade (Tama Chemicals, AA-100). The decomposition and analytical procedures are same as Yoshida, Muramatsu, Tagami, and Uchida (1998). The carcinogenic (As, Cd, and Pd) and non-carcinogenic (Fe, V, Co, Cr, Mn, Ni, Cu, and Zn) heavy metals in vegetable, fruit, and soil samples were measured by ICP-MS (Agilent Technologies 8800 ICP-MS Triple Quad) after diluting the solution to a suitable concentration. The standard solutions were prepared from SPEX Multi Element Plasma standards (XSTC1 and 355) supplied by SPEX industries, Inc. (Meutuchen, NJ, USA) and used to get calibration curves. The elements In, Rh, and Bi were chosen as the internal standards in this analysis. The RSD for the analyzed heavy metals was found to be (0.087–7.281). The standard reference materials such as SRM 1573a (Tomato leaves), GBW 07603 (Bush leaves and branches), and GBW 07604 (Poplar leaves)

| Sample No. | Common name | Scientific name | Vegetable ID | Vegetable type/fruit |
|------------|-------------|-----------------|--------------|----------------------|
| 1          | Papaya      | Carica papaya   | PA           | Fruit vegetable      |
| 2          | Egg plant   | Solanum melongena L | EP         | Fruit vegetable      |
| 3          | Bottle gourd| Cucurbita Pepo  | GD           | Fruit vegetable      |
| 4          | Cauliflower | Brassica oleracea botrytis | CF       | Fruit vegetable      |
| 5          | Radish      | Raphanus sativus | RA           | Root vegetable       |
| 6          | Pui shak    | Basella alba    | PUI          | Leafy vegetable      |
| 7          | Pat shak    | Carchorus Capsularis | PT       | Leafy vegetable      |
| 8          | Kalmi shak  | Ipomoea Aquatica | KL          | Leafy vegetable      |
| 9          | Helencha    | Enhydra fluctuans | HL          | Leafy vegetable      |
| 10         | Guava       | Psidium guajava  | GV           | Fruit                |
| 11         | Banana      | Musa balbisiana  | BA           | Fruit                |
for vegetables and rock sample JB-1b for soil were digested and analyzed with the studied samples for the validity of the analytical method. Good agreement between the certified and measured values was observed in the reference samples analyzed in this study (Table 2).

### 2.4. PTF calculation

PTF that is the uptake of elements by plants from the soil was calculated from the ratio of the heavy metal concentrations in the plant to soil using the following equation (Mahfuza et al., 2014).

\[
PTF = \frac{\text{Heavy metal content, mg/kg, dry weight in plant tissue}}{\text{Heavy metal content, mg/kg, dry weight in corresponding soil}}
\]

#### 2.5. Estimation of daily heavy metal intake

The health risk posed to inhabitants was determined by dietary intake of heavy metals and compared with maximum permissible risk level for human population. The estimated daily intake (EDI) of heavy metals through vegetable and fruit consumption was calculated by the following equation (United States Environmental Protection Agency [US-EPA], 2000).

\[
\text{EDI} = V_{\text{DIR}x} \times C_V \div \text{BW}
\]

where \(V_{\text{DIR}x}\) is the daily vegetable and fruit consumption rate. In this study, the daily vegetable and fruit consumption rate (\(V_{\text{DIR}x}\)) was taken categorically for individual leafy, or root, or fruit vegetables and fruits from the report of WHO/FAO (2003). The daily ingestion of total vegetables and fruits (\(V_{\text{DIRtotal}}\)) for the Bangladeshi people (adult and child) is 211 and 42.2 g person\(^{-1}\) day\(^{-1}\), respectively (FAO/WHO, 2004), while the average adult and child body weights for Bangladesh population were considered to be 49.5 and 16.5 kg, respectively (Mahfuza et al., 2014).

### 2.6. Health risk assessment

Health risk assessment was computed based on the average contents of carcinogenic and non-carcinogenic metals determined in the vegetable and fruit samples using United States Environment Protection Agency human health risk assessment models (NFPCSP Nutrition Fact Sheet, 2011). This study evaluated both cancer and non-cancer risks of heavy metals through vegetable and fruit consumptions to take safety measures.

| GBW 07603 | GBW07604 | SRM 1573a |
|-----------|-----------|-----------|
| Elements (mg/kg) | Obs value | Cer. value | % of recovery | Obs value | Cer. value | % of recovery | Obs value | Cer. value | % of recovery |
| Fe | 1,042 | 1,070 | 97 | 272.66 | 274 | 99 | 428 | 368 | 116 |
| V | 2 | 2 | 90 | 0.47 | 0.5 | 102 | 0.738 | 0.835 | 88 |
| Cr | 2 | 3 | 77 | 0.55 | 0.5 | 100 | 2 | 2 | 100 |
| Co | 0.4 | 0.4 | 100 | 45 | 45 | 100 | 0.6 | 0.6 | 100 |
| Cu | 6 | 7 | 93 | 9 | 9 | 100 | 4.3 | 4.7 | 9 |
| Zn | 55 | 55 | 100 | 37 | 37 | 100 | 30 | 31 | 99 |
| Mn | 63 | 61 | 103.4 | 45 | 45 | 100 | 0.6 | 0.6 | 100 |
| Ni | 1 | 2 | 88 | 1.8 | 1.9 | 96.8 | 1.9 | 1.6 | 116 |
| As | 1 | 1 | 100 | 0.2 | 0.4 | 65 | 0.1 | 0.1 | 100 |
| Cd | 1 | 1 | 100 | 0.4 | 0.3 | 121 | 1.4 | 1.5 | 93 |
| Pb | 53 | 47 | 112 | 1.7 | 1.5 | 113 | 1.7 | 1.5 | 113 |
2.7. Cancer risks

The possibility of cancer risks in the studied vegetables and fruits through intake of carcinogenic heavy metals was estimated using the Incremental Lifetime Cancer Risk (ILCR) (Liu et al., 2013).

\[ \text{ILCR} = \text{CDI} \times \text{CSF} \]

where CDI is chronic daily intake of chemical carcinogen, mg/kg BW/day which represents the lifetime average daily dose of exposure to the chemical carcinogen.

The US EPA ILCR is obtained using the cancer slope factor (CSF), which is the risk produced by a lifetime average dose of 1 mg/kg BW/day and is contaminant specific (Micheal, Patrick, & Vivian, 2015). ILCR value in vegetable or fruit represents the probability of an individual’s lifetime health risks from carcinogenic heavy metals’ exposure (Pepper, Gerba, & Brusseau, 2012). The level of acceptable cancer risk (ILCR) for regulatory purposes is considered within the range of 10\(^{-6}\) to 10\(^{-4}\) (Li & Zhang, 2010). The CDI value was calculated on the basis of the following equation and CSF values for carcinogenic heavy metals were used according to the literature (Liu et al., 2013).

\[ \text{CDI} = (\text{EDI} \times \text{EF}_r \times \text{ED}_{\text{tot}}) / \text{AT} \]

where EDI is the estimated daily intake of metal via consumption of vegetable and fruit; \(\text{EF}_r\) is the exposure frequency (365 days/year); \(\text{ED}_{\text{tot}}\) is the exposure duration 70.7 years, average lifetime for Bangladesh; AT is the period of exposure for non-carcinogenic effects \((\text{EF}_r \times \text{ED}_{\text{tot}})\), and 70 years life time for carcinogenic effect (Micheal et al., 2015). The cumulative cancer risk as a result of exposure to multiple carcinogenic heavy metals due to consumption of a particular type of vegetable or fruit was assumed to be the sum of the individual heavy metal increment risks and calculated by the following equation (Liu et al., 2013).

\[ \sum_{i=1}^{n} \text{ILCR}_i = \text{ILCR}_1 + \text{ILCR}_2 + \cdots + \text{ILCR}_n \]

where \(n = 1, 2, \ldots, \) is the individual carcinogenic heavy metals or vegetable or fruit species.

2.8. Non-cancer risks

Non-carcinogenic risks for individual heavy metal or vegetable were evaluated by computing the target hazard quotient (THQ) using following equation (Micheal et al., 2015).

\[ \text{THQ} = \text{CDI} / \text{RfD} \]

CDI is the chronic daily heavy metal intake (mg/kg/day obtained from the previous section and \(\text{RfD}\) is the oral reference dose (mg/kg/day) which is an estimation of the maximum permissible risk on human population through daily exposure, taking into consideration a sensitive group during a lifetime (Li & Zhang, 2010). EPA-recommended \(\text{RfD}\) values of Fe, V, Cr, Co, Cu, Zn, Mn, Ni, As, Cd, and Pb were used in the above equation (Li et al., 2013; US-EPA, 2002).

To evaluate the potential risk to human health through more than one heavy metal, chronic hazard index (HI) is obtained as the sum of all hazard quotients (THQ) calculated for individual heavy metals for a particular exposure pathway (NFPCSP Nutrition Fact Sheet, 2011). It is calculated as follows:

\[ \text{HI} = \text{THQ}_1 + \text{THQ}_2 + \cdots + \text{THQ}_n \]

where \(1, 2, \ldots, n\) are the individual heavy metals or vegetable and fruit species.

It is assumed that the magnitude of the effect is proportional to the sum of the multiple metal exposures and that similar working mechanism linearly affects the target organ (RAIS, 2007). The calculated HI is compared to standard levels: the population is assumed to be safe when HI < 1 and in a level of concern when 1 < HI < 5 (Guerra, Trevizam, Muraoka, Marcante, & Caniatti-Brazaca, 2012).
3. Results and discussion

3.1. Concentration of carcinogenic and non-carcinogenic heavy metals in soil

The average concentrations of carcinogenic and non-carcinogenic heavy metals in the investigated agricultural soil of Gazipur industrial area are shown in Figures 2a and 2b. A wide variation of heavy metal concentrations was observed in the studied soils where the highest metal content was for Fe with average concentration of 23,474.75 mg/kg and the lowest metal content was for Cd with average concentration of 0.32 mg/kg, respectively. The trend of the heavy metal contents according to average concentration found in the studied soil is followed: Fe > Mn > Zn > V > Cr > Ni > Pb > Cu > As > Co > Cd.

The toxic level of heavy metals in soil was estimated by comparing the data with the safe limit set by NEPA of China 1995 (Kabata-Pendias & Pendias, 1992; National Environmental Protection Agency, 2001; Ogwok, Bamuwamye, Apili, & Musalima, 2014). The trend of the toxicity of heavy metal was as follows: Mn > V > Ni > Cu > Zn > Cr > Fe > Pb > As > Cd > Co. According to the pollution level proposed by Hakanson (1980) and Kabata-Pendias and Pendias (1992), it reveals that the soil of this area is moderately polluted by Mn, V, Ni, Zn, and Cu. Further, compared to the background level reported by Kashem and Singh (1999) and Hakanson (1980), the content of carcinogenic heavy metals in the studied soil increased about two-fold which might be due to the repeated use of industrial waste water as irrigation and for excess use of metal-based fertilizer and pesticides. Similar results were observed for the heavy metal contents in different industrial sites of Bangladesh (Ahmad & Goni, 2010; Mahfuza et al., 2014; Naser et al., 2009).

Therefore, in the present study, Mn, V, Ni, Zn, Cd, Pb, and Cu are identified as potential heavy metal toxicants in the studied soil of sub-urban area of Dhaka.
3.2. Concentration of heavy metals in vegetables and fruits
Carcinogenic and non-carcinogenic heavy metal contents of the vegetables and fruits grown in the industrial area of sub-urban Gazipur industrial area were investigated which are shown in Figures 3a and 3b, respectively.

Average concentration of all the heavy metals was observed to be higher in leafy vegetables as compared with fruit vegetables, root vegetables, and fruits. Usually, heavy metal concentration varied among different vegetables and fruits depending on the different accumulation capacities and variation in the soil properties (NFPCSP Nutrition Fact Sheet, 2011). Further, highest deposition of Fe was observed in all of the vegetables and fruits which varied from 88 to 1,902 mg/kg depending on vegetable or fruit species. Usually, Fe participates in chlorophyll synthesis and photosynthesis. So, the green of the plant contains higher Fe than fruit (Mahfuza et al., 2014). However, excess Fe in the body may play a role in etiology of heart disease and type 2 diabetes (Kashem & Singh, 1999).
Besides this, high content of Zn and Mn was also observed in all vegetables and fruits. Zn and Mn are airborne pollutants and so might accumulate from the atmospheric deposition of the surrounding metal industries as well as vehicle exhaust (Ikeda et al., 2000). Higher deposition of Pb (22 mg/kg) and Cd (1.2 mg/kg) was observed in helencha grown in the studied industrial area. Elevated levels of heavy metals such as Pb, Cd, and Zn in plants near metal smelters had previously been demonstrated in another investigation (Cui et al., 2004).

Moreover, the toxic level in the vegetables for each heavy metal except Mn was evaluated and compared with the permissible limit recommended by WHO/FAO (1984, 2011) (Tidemann-Andersen, Acham, Maage, & Malde, 2011) (Figures 3a and 3b). The permissible limit of Mn for foodstuff was not available. The sequence of heavy metals according to pollution for fruit, root, leafy vegetables, and fruits is Co > Pb > Zn > Cr > Ni > Fe > As > Cu > V > Cd; Pb > Zn > As > V > Fe > Ni = Cr > Cd > Co > Cu; Pb > Zn > Ni > Cd > As > Co > Fe > Cr > V > Cu; and Pb > Zn > Cr = Ni > Fe > Cu > Co > Cd > V > As, respectively. Except Cu, all heavy metals’ content exceeded the permissible limit in root and leafy vegetables while Cr, Pb, Co, and Zn exceeded the permissible limit for fruit vegetables and Zn and Pb in fruits grown in the studied areas. Elevated levels of Mn, Zn, Cd, and Pb were found in leafy vegetables and might be deposited from surrounding metal industries or from vehicle emission which readily get fixed to hairy or waxy cuticles of leaves which transport these metals to foliar cells of the studied vegetables (Mahfuza et al., 2014). The toxic levels of heavy metals like Cd, Cr, Ni, Pb, Zn, Mn, and Fe in the studied vegetables may cause adverse health impact on the surrounding inhabitants. Therefore, vegetables grown in the sub-urban area are greatly polluted with non-carcinogenic and carcinogenic heavy metals which may harm human health. So, regular monitoring and assessment of heavy metal contents in food crops of this area is recommended.

3.3. Heavy metal transfer from soil to food crops

PTF of carcinogenic and non-carcinogenic heavy metals from soil to vegetable and fruits was calculated and provided in Table 3. PTF of studied vegetables and fruits varied with heavy metal types and
also with vegetable or fruits species. It may be attributed to differences in the concentration of metals in soils and different capacities of the heavy metal uptake by different vegetables (Cui et al., 2004).

Moreover, the greater the transfer factors value more than 20%, the greater the chances of vegetables for heavy metal contamination by anthropogenic activities (WHO/FAO, 2011). In this study, higher PTF (>20%) value for Zn, Mn, Cd, Cu, Ni, and Pb was observed in various vegetables and fruits grown in the studied soil indicating they were polluted by various anthropogenic activities.

Therefore, leafy vegetables are high in Zn, Cd, Pb, Cu, Ni, and Mn, root vegetables are high in Zn, Ni, Mn, Pb, and Cd, fruit vegetables and fruits are high in Cu, Cd, and Zn contents. The higher uptake of heavy metals in leafy vegetables may be due to higher transpiration rate to maintain the growth and moisture content of these plants (Khan, Farooq, Sahabaz, Khan, & Sadique, 2009). Similar finding was also reported by Mahfuza et al. (2014), Khan et al. (2009), and WHO/FAO (2011).
3.4. Estimated daily intake of heavy metals

The degree of toxicity of heavy metals to human being depends upon their daily intake. The average fruit and vegetable consumption per person in Bangladesh is still low: 211 g/d compared to desirable intake of WHO, 400 g/d (FAO/WHO, 2004; WHO/FAO, 2003). Further, in sub-urban Dhaka, most of the vegetables and fruits grown in industrial area are adulterated to varying degrees by heavy metals (Ikeda et al., 2000; Naser et al., 2009). The intake of edible parts of these contaminated vegetables and fruits causes human health risks to the surrounding population (Saiful & Hoque, 2014). The EDI of carcinogenic and non-carcinogenic heavy metals for inhabitants of the study area is listed in Table 4. The computed EDI for each element in the studied vegetables was compared with the toxicologically accepted level, oral reference dose, and RfD value (Table 3) established by US-EPA (2002), Li et al. (2013), RAIS (2007), and US-EPA (2002). Our EDI for Pb, Mn, As, and Fe was above the RfD value in few vegetables. The intake of heavy metals from the consumption of studied fruits was below the RfD limit. The higher intake of Mn was found through the consumption of all types of vegetables, the higher intake of Pb was found in the leafy vegetable, whereas higher intake of As and Fe was found in the root vegetable. Therefore, the health risks for heavy metals through the consumption of vegetables could be a great concern for the population of sub-urban Dhaka.

3.5. Human health risks

3.5.1. Cancer risks

As, Cd, and Pb are classified by the IARC as being carcinogenic agents (Tani & Barrington, 2005). Chronic exposure to low doses of As, Cd, and Pb could therefore result into many types of cancers (Jarup, 2003). The computed ILCR and cumulative incremental lifetime cancer risk (ΣILCR) for As, Cd, and Pb through the studied vegetables and fruits are presented in Table 5.

US-EPA recommended the safe limit for cancer risk is below about 1 chance in 1,000,000 lifetime exposure (ILCR < 10⁻⁶) and threshold risk limit (ILCR > 10⁻⁴) for chance of cancer is above 1 in 10,000 exposure where remedial measures are considerable and moderate risk level (ILCR > 10⁻³) is above 1 in 1,000 where public health safety consideration is more important (Pepper et al., 2012; Tchounwou, Yedjou, Patlolla, & Sutton, 2014). ILCR for Cd violated the threshold risk limit (>10⁻⁴) in all the studied vegetables and fruits, As violated the risk in leafy and root vegetables, whereas none of the vegetables or fruits crossed the designated risk limit for Pb. The trend of risk for developing cancer as a result of consuming studied vegetables and fruits showed: Leafy vegetables > Root vegetables > Fruit vegetables > Fruit.
Moreover, cumulative cancer risk (∑ILCR) of all the studied vegetables and fruits exceeded the recommended threshold risk limit (>10⁻⁴) with 22, 15, 59, and 4%, respectively, for fruit vegetables, root vegetables, leafy vegetables, and fruits (Figure 4a). Further, among all the studied vegetables and fruits, halencha (Enhydra fluctuans) has the highest chances of cancer risks (ILCR 4 × 10⁻³) and banana (Musa balbisiana) has the lowest chances of cancer risk (ILCR 1 × 10⁻⁴). These risk values indicate that consumption of halencha (Enhydra fluctuans) would result in an excess of 41 cancer cases per 10,000 people exposure while consumption of banana (Musa balbisiana) would result in an excess of 10 cancer case per 100,000 people exposure (US-EPA, 2001). Among the carcinogenic heavy metals, Cd contributed to risk of about 78, 60, 88, and 91% in fruit vegetables, root vegetables, leafy vegetables, and fruits, respectively (Figure 4b). Therefore, Cd is the most dominant carcinogen in the study area and thus attention should be paid to control its exposure to environment to save the population from cancer risk. Furthermore, the cumulative cancer risk (∑ILCR) for heavy metals in root and leafy vegetables reached the moderate risk limit (>10⁻³) and halencha (Enhydra fluctuans) exceeded this limit. Therefore, consumption of halencha (Enhydra fluctuans) is most risky in this study area which is most susceptible to cancer risk. Metal oxide fumes escaping from Pb–Zn smelters and automobiles of the studied area have highly contaminated the soil and vegetation with chemical carcinogens like Cd and Pb (Naser et al., 2009) which consequently were exposed through the

| Vegetables and fruits | Fe  | V    | Cr   | Co   | Cu   | Zn   | Mn   | Ni   | As   | Cd   | Pb   |
|-----------------------|-----|------|------|------|------|------|------|------|------|------|------|
| PA                    | 0.50| 0.001| 0.02 | 0.002| 0.02 | 0.11 | 0.03 | 0.004| 0.00002| 0.0001| 0.003|
| EP                    | 0.40| 0.001| 0.006| 0.001| 0.03 | 0.06 | 0.03 | 0.006| 0.000063| 0.00012| 0.001|
| GD                    | 0.70| 0.001| 0.02 | 0.1  | 0.032| 0.12 | 0.01 | 0.01 | 0.000084| 0.000063| 0.003|
| CF                    | 0.60| 0.002| 0.002| 0.0003| 0.007| 0.10 | 0.02 | 0.008| 0.00021| 0.0001| 0.001|
| RA                    | 1.00| 0.01 | 0.006| 0.001| 0.005| 0.01 | 0.02 | 0.006| 0.00039| 0.00021| 0.003|
| Pui                   | 0.80| 0.001| 0.0038| 0.001| 0.012| 0.05 | 0.10 | 0.009| 0.00012| 0.00016| 0.008|
| Pat                   | 0.20| 0.001| 0.003 | 0.0002| 0.008| 0.05 | 0.02 | 0.0036| 0.00001| 0.000140| 0.003|
| KL                    | 0.53| 0.0002| 0.002 | 0.003 | 0.009| 0.01 | 0.10 | 0.01 | 0.0001| 0.0001| 0.002|
| HL                    | 0.63| 0.0013| 0.002 | 0.001 | 0.009| 0.11 | 0.14 | 0.0068 | 0.00013| 0.000063| 0.01|
| GU                    | 0.30| 0.0004| 0.002 | 0.0003| 0.015| 0.02 | 0.005| 0.002 | 0.000007| 0.00006| 0.001|
| BA                    | 1.00| 0.0001| 0.001 | 0.00003| 0.007| 0.02 | 0.002| 0.001 | 0.000002| 0.000016| 0.0006|
| RD                    | 0.70| 0.009| 1.5 | 0.03 | 0.04 | 0.30 | 0.014| 0.02 | 0.0003| 0.001| 0.00035|

| Vegetables and fruits | As   | Cd  | Pb  | ∑ILCR |
|-----------------------|------|-----|-----|-------|
| PA                    | 3E-05| 5E-04| 2.00E-05| 6E-04|
| EP                    | 9.00E-05| 7E-04| 1.00E-05| 8.00E-04|
| GD                    | 1E-04| 4.00E-04| 3.80E-04| 5E-04|
| CF                    | 3.00E-04| 6.00E-04| 1E-05| 8.0E-04|
| RA                    | 6.00E-04| 1.00E-03| 3.00E-05| 2.00E-03|
| Pui                   | 2.00E-04| 1.00E-04| 6.00E-05| 1.20E-03|
| PT                    | 1.00E-04| 9.00E-04| 3.00E-05| 1.00E-03|
| KL                    | 1.00E-04| 8.00E-04| 3.00E-05| 2.00E-03|
| HL                    | 2.00E-04| 4.00E-03| 8.00E-05| 4.20E-03|
| GV                    | 1.00E-05| 4.00E-04| 1.00E-05| 4.00E-04|
| BA                    | 3.00E-06| 1.00E-04| 5.00E-06| 1.00E-04|

Moreover, cumulative cancer risk (∑ILCR) of all the studied vegetables and fruits exceeded the recommended threshold risk limit (>10⁻⁴) with 22, 15, 59, and 4%, respectively, for fruit vegetables, root vegetables, leafy vegetables, and fruits (Figure 4a). Further, among all the studied vegetables and fruits, halencha (Enhydra fluctuans) has the highest chances of cancer risks (ILCR 4 × 10⁻³) and banana (Musa balbisiana) has the lowest chances of cancer risk (ILCR 1 × 10⁻³). These risk values indicate that consumption of halencha (Enhydra fluctuans) would result in an excess of 41 cancer cases per 10,000 people exposure while consumption of banana (Musa balbisiana) would result in an excess of 10 cancer case per 100,000 people exposure (US-EPA, 2001). Among the carcinogenic heavy metals, Cd contributed to risk of about 78, 60, 88, and 91% in fruit vegetables, root vegetables, leafy vegetables, and fruits, respectively (Figure 4b). Therefore, Cd is the most dominant carcinogen in the study area and thus attention should be paid to control its exposure to environment to save the population from cancer risk. Furthermore, the cumulative cancer risk (∑ILCR) for heavy metals in root and leafy vegetables reached the moderate risk limit (>10⁻³) and halencha (Enhydra fluctuans) exceeded this limit. Therefore, consumption of halencha (Enhydra fluctuans) is most risky in this study area which is most susceptible to cancer risk.
consumption of root and leafy vegetables to the inhabitants of sub-urban Dhaka for cancer risk. Prompt action should be needed to control the excess use of heavy metal-based fertilizer and pesticides and also emission of heavy metal exhaust from the industries and automobiles should be checked to save the urban population from cancer risk.

3.5.2. Non-cancer risk
The non-cancer risks (THQ) of the investigated heavy metals through the consumption of vegetables and fruits for both adults and children inhabitants of the study area were determined and presented in Tables 6a and 6b.

Risk level of Target Hazard Quotient (THQ > 1) was observed for Pb in leafy vegetables, As in root vegetables, Mn in all vegetables, and Fe in few vegetables for both adults and children. It indicates that intake of these heavy metals through consumption of vegetables poses a considerable non-cancer risk. On the other hand, the intake of individual heavy metals V, Cr, Co, Cd, Cu, Zn, and Ni through consumption of vegetables and fruits in this area is safe (THQ < 1) for the inhabitants. The health risk of Cr was minimum for both adults and children compared to other heavy metals investigated, which is due to its high RfD. The sequence of THQ according to risk level followed a decreasing order of fruit, root, leafy vegetables, and fruits with Mn > Fe > Pb > Cu > Ni > Zn; Fe > Mn > As > Pb > V > Ni > Zn > Cd > Cu > Co > Cr; Mn > Pb > Fe > Ni > As > Cd > Cu > Zn > V > Co > Cr; and Cu > Fe > Mn > Pb > Zn > Ni > Cd > V > As > Co > Cr, respectively. The sequence of risk was the same for both adults and children although the children had higher THQ values in all cases. Similar observations have been reported previously by Micheal et al. (2015) and Liu et al. (2013).
Further, the non-cancer risks for each type of vegetables and fruits were expressed as the cumulative HI, which is the sum of individual metal THQ. All the studied vegetables and fruits except banana showed the risk level (HI > 1) with highest (15.89) in helencha (*Enhydra fluctuans*) and lowest (0.79) in banana (*Musa balbisiana*). All the vegetables and fruits have non-cancer risk (HI > 1) contributing 19, 27, 49, and 5% in fruit vegetables, root vegetables, leafy vegetables, and fruits, respectively (Figure 5a). Further, HI showed Mn, Pb, Fe, and As as the dominant contaminants in root and leafy vegetables, whereas Mn, Pb, Fe, and Cu are in the fruit vegetables and fruits which contributed together 80% to 90% of HI (Figure 5b). It suggests that the inhabitants of sub-urban Dhaka might be exposed to some potential health risk through the intake of heavy metals. Therefore, it is urgent to take proper measures for the reduction of Pb, Mn, Fe, and As pollution in this area to save the sub-urban population from non-cancer risks.

### Table 6a. THQ and HI for adult population through the consumption of different vegetable and fruit species in the study area

| THQ  | Fe  | V    | Cr  | Co  | Cu  | Zn  | Mn  | Ni  | As  | Cd  | Pb  | HI = ΣTHQ |
|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|-----|----------|
| PA   | 0.70| 0.11 | 0.01| 0.01| 0.40| 0.36| 2.87| 0.20| 0.10| 0.10| 0.9  | 5        |
| EP   | 0.51| 0.07 | 0.004| 0.002| 0.75| 0.2  | 1.96| 0.30| 0.02| 0.12| 0.4  | 4        |
| GD   | 1.00| 0.11 | 0.01| 0.20| 0.80| 0.4  | 0.71| 0.50| 0.03| 0.06| 0.9  | 5        |
| CF   | 1.00| 0.17 | 0.001| 0.01| 0.20| 0.3  | 1.8  | 0.04| 0.70| 0.10| 0.4  | 5        |
| RA   | 1.40| 0.43 | 0.004| 0.02| 1.25| 0.26| 1.35| 0.30| 1.30| 0.21| 0.97 | 6        |
| PUI  | 1.12| 0.14 | 0.003| 0.02| 0.30| 0.16| 5   | 0.45| 0.40| 0.21| 2.22 | 10       |
| PT   | 0.30| 0.06 | 0.002| 0.01| 0.20| 0.16| 8.57| 0.20| 0.30| 0.14| 0.01 | 2.22 | 10       |
| KL   | 0.80| 0.02 | 0.002| 0.11| 0.22| 0.04| 6.4  | 0.55| 0.33| 0.12| 0.45 | 9        |
| HL   | 1.00| 0.14 | 0.001| 0.02| 0.22| 0.36| 10   | 0.34| 0.43| 0.63| 2.85 | 16       |
| GV   | 0.41| 0.04 | 0.001| 0.01| 0.04| 0.06| 0.35| 0.10| 0.02| 0.10| 0.28 | 1.7       |
| BA   | 0.12| 0.01 | 0.001| 0.001| 0.17| 0.08| 0.16| 0.05| 0.01| 0.02| 0.18 | 0.8       |

### Table 6b. THQ and HI for children through the consumption of different vegetable and fruit species in the study area

| THQ  | Fe  | V    | Cr  | Co  | Cu  | Zn  | Mn  | Ni  | As  | Cd  | Pb  | HI = ΣTHQ |
|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|-----|----------|
| PA   | 0.96| 0.15 | 0.014| 0.080| 0.60| 0.50| 4.01| 0.25| 0.10| 0.11| 0.120| 8        |
| EP   | 0.71| 0.01 | 0.60 | 0.30| 1.05| 0.030| 2.74| 0.42| 0.003| 0.17| 0.60 | 6        |
| GD   | 1.40| 0.15 | 0.01| 0.28| 1.12| 0.56| 1.00| 0.7  | 0.03| 9.08| 1.30 | 7        |
| CF   | 1.17| 0.24 | 0.001| 0.014| 0.24| 0.042| 2.52| 0.56| 1.00| 0.14| 0.60 | 7        |
| RA   | 2.00| 0.60 | 0.006| 0.022| 0.20| 0.36| 2.00| 0.42| 1.82| 0.29| 1.4  | 8        |
| PUI  | 1.60| 0.20 | 0.0042| 0.028| 0.42| 0.200| 7.00| 0.63| 0.60| 0.22| 3.1  | 14       |
| PT   | 0.40| 0.08 | 0.0028| 0.0098| 0.30| 0.20| 12.00| 0.21| 0.45| 0.20| 1.4  | 15       |
| KL   | 1.05| 0.03 | 0.0028| 0.154| 0.22| 0.04| 6.00| 0.55| 0.33| 0.12| 0.4  | 9        |
| HL   | 1.00| 0.14 | 0.001| 0.02| 0.31| 0.50| 14.00| 0.48| 0.61| 0.10| 4.00 | 21       |
| GV   | 0.60| 0.056| 0.0014| 0.014| 0.52| 0.08| 0.49| 0.14| 0.030| 0.10| 0.40 | 2.4       |
| BA   | 0.20| 0.014| 0.001| 0.001| 0.24| 0.11| 0.22| 0.07| 0.01| 0.02| 0.24 | 1.1       |

Further, the non-cancer risks for each type of vegetables and fruits were expressed as the cumulative HI, which is the sum of individual metal THQ. All the studied vegetables and fruits except banana showed the risk level (HI > 1) with highest (15.89) in helencha (*Enhydra fluctuans*) and lowest (0.79) in banana (*Musa balbisiana*). All the vegetables and fruits have non-cancer risk (HI > 1) contributing 19, 27, 49, and 5% in fruit vegetables, root vegetables, leafy vegetables, and fruits, respectively (Figure 5a). Further, HI showed Mn, Pb, Fe, and As as the dominant contaminants in root and leafy vegetables, whereas Mn, Pb, Fe, and Cu are in the fruit vegetables and fruits which contributed together 80% to 90% of HI (Figure 5b). It suggests that the inhabitants of sub-urban Dhaka might be exposed to some potential health risk through the intake of heavy metals. Therefore, it is urgent to take proper measures for the reduction of Pb, Mn, Fe, and As pollution in this area to save the sub-urban population from non-cancer risks.
4. Conclusions
Agricultural fields near sub-urban industrial areas of Bangladesh have been suffering increasing heavy metal damages from various pollution sources. Average heavy metal content was found higher in leafy vegetables compare to non leafy vegetables and fruits. Risk within the carcinogenic heavy metals in the studied vegetables and fruits showed Cd (81%) > As (17%) > Pb (2%) and among the vegetables and fruits : leafy vegetables (59%) > Fruit vegetables (22%) > root vegetable (15%) > fruit (4%), respectively. Most of the vegetables had non-cancer risk for Mn, Pb, Fe, and As. The inhabitants of this area have no risk (THQ < 1) for V, Cr, Co, Cd, Cu, Zn, and Ni. The sequence of risk was the same for both adults and children although children had higher risk in all cases.

Cd has the highest carcinogenic risk while Fe, Pb, As, Mn, and Cd generate the highest non-cancer risk and consumption of helencha (*Enhydra fluctuans*) has the most cancer and non-cancer risks. Inappropriate agricultural practice, metal oxide fumes, and particulate matter escaping from the industries and automobiles of the study area might contaminate the soil and vegetation heavily with toxic metals at risk level. Therefore, effective measures should be taken to control Cd, Pb, As, Fe, and Mn pollution in the agricultural fields of the sub-urban industrial area of Bangladesh. Extensive research program on the health risk of heavy metals on the agricultural products grown in sub-urban environment of Bangladesh is recommended in order to ensure food safety.
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