Original Research Article

Assessment of trace metals in foodstuffs grown around the vicinity of industries in Bangladesh

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A B S T R A C T

In the present study, we investigated the levels of chromium (Cr), nickel (Ni), copper (Cu), arsenic (As), cadmium (Cd) and lead (Pb) in eight groups of foods, namely, cereals, pulses, vegetables, fruit, fish, meat, eggs and milk. The range of Cr, Ni, Cu, As, Cd and Pb in the foodstuffs was 0.18–4.8, 0.008–10, 0.47–22, 0.003–0.98, 0.0003–0.85 and 0.005–3.7 mg/kgfw, respectively. The daily intakes (EDIs) of Cr, Ni, As, Cd and Pb were higher than the maximum tolerable daily intake (MTDIs), indicating their potential sources from dietary intake. The combined metal hazard quotients (CMHQs) from rice, fruit, vegetables and fish were higher than 1, meaning that metals may pose a considerable risk to local inhabitants due to consumption of these four food items. From the human health point of view, this study showed that the studied foods were not safe for the local inhabitants, and potential risk cannot be neglected for regular or excessive consumers.

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1. Introduction

Metals and metalloids are ubiquitous in the environment, through either natural or anthropogenic intervention, and elevated concentrations in the environment result from waste disposal, smelter stacks, atmospheric deposition, application of fertilizer, pesticide and sewage sludge in the arable land (Cui et al., 2005; Zheng et al., 2007; Islam et al., 2014a). While they are essential for plant growth and/or human nutrition, some micronutrients (e.g. Cu, Cr and Ni) might be toxic at high concentrations (McLaughlin et al., 1999; Rahman et al., 2014). Other toxic trace elements such as As, Cd and Pb might also inadvertently enter the food chain and pose risks to human and animals (Sankar et al., 2006; Sharma et al., 2007; Bundschuh et al., 2012; Ji et al., 2013; Rahman et al., 2014). Trace metals such as Cr, Ni, As, Cd and Pb have been considered as the most toxic elements in the environment by the US Environment Protection Agency (EPA) (Lei et al., 2010; Islam et al., 2014a). Therefore, the risks associated with metal contamination in foodstuffs are of great concern.

Although the relative contribution of trace metals has not yet been clearly established, dietary intake is considered as the vital exposure pathway (Kachenko and Singh, 2006; Sharma et al., 2007). Fig. 1 shows those possible food chain pathways through which the Bangladeshi population is generally being exposed to metal toxicity, “soil–plant–human” and/or “plant–animal–human” and/or “soil–water–animal” could be the potential food chain pathways of metal accumulation in human populations. In this study we tried to trace food chain pathways in the natural ecosystem, by means of which metals may contaminate human foods so that we can assess the potentiality of these pathways in exposing trace metals to human.

Rice is the major staple food in many countries, particularly in Asian countries like Bangladesh, India, Thailand, China and
Vietnam, where soil and ground water pollution with high level of As and trace metals have been reported (Roychowdhury et al., 2003; Duxbury et al., 2003; Meharg and Rahman, 2003; Das et al., 2004; Rahman et al., 2013). Increased levels of trace metals in agricultural soils and their uptake in rice, vegetables and other food crops have become a serious health issue in this region (Meharg and Rahman, 2003; Williams et al., 2006; Islam et al., 2014b). While a significant number of studies have been focused on As in Bangladeshi rice and vegetables (Alam et al., 2003; Das et al., 2004; Karim et al., 2008; Rahman and Hasegawa, 2011), studies on other trace metals in other daily consumable foods are scarce (Alam et al., 2003; Rahman et al., 2013). Therefore, this study aimed to evaluate the levels of trace metals in foodstuffs that are generally consumed by the Bangladeshi population, and to assess health risk associated with dietary intake of metals.

2. Materials and methods

2.1. Study area and sample

Dhaka, with a metropolitan area of 815.8 km², is surrounded by the three major rivers Turag, Buriganga and Shitalakha, which are currently used for industrial waste disposal. Dhaka is one of the most densely populated cities in the world with 12 M people, of which fewer than 25% are served by sewage treatment facilities (Islam et al., 2014c). The basic information of the study areas is presented in Table S1 (Supplementary material). The most consumed foods for Bangladeshi people, i.e. cereals, pulses, vegetables, fruits, fish, meat, eggs and milk, were collected during February–March, 2012 and August–September, 2013. About 173 food samples were collected from three rivers and their adjacent areas around the Dhaka city metropolitan area, Bangladesh (Fig. 2). A composite sample for each

![Fig. 1. Possible food chain pathways through which humans may be exposed to trace metals. Modified after Rahman et al. (2008).](image1)

Fig. 1. Possible food chain pathways through which humans may be exposed to trace metals.

![Fig. 2. Map of the study area around Dhaka City, Bangladesh.](image2)

Fig. 2. Map of the study area around Dhaka City, Bangladesh.
food item was prepared and homogenized in a food processor, and 50 g test portions were stored at −20 °C in the laboratory of the Institute of Nutrition and Food Science (INFS), University of Dhaka, Bangladesh. The prepared samples were brought to Yokohama National University, Japan, for chemical analysis.

2.2. Sample analysis

All chemicals were analytical grade reagents; Milli-Q water (Elix UV5 and MilliQ, Millipore, Boston, MA, USA) was used for the preparation of solutions. The Teflon vessel and polypropylene containers were cleaned, soaked in 5% HNO₃ for more than 24 h, then rinsed with Milli-Q water and dried. For metal analysis, 0.3–0.5 g of the food sample was treated with 6 ml 69% HNO₃ (Kanto Chemical Co, Tokyo, Japan) and 2 ml 30% H₂O₂ (Wako Chemical Co, Tokyo, Japan) in a closed Teflon vessel and was digested in a Microwave Digestion System (Berghof speedwave®, Eningen, Germany). The digested samples were then transferred into a Teflon beaker, and total volume was made up to 25 ml with Milli-Q water. The digested solution was then filtered by using syringe filter (DISMIC®, 25HP PTFE, pore size = 0.45 μm; Toyo Roshi Kaisha, Ltd., Tokyo, Japan) and stored in 50 ml polypropylene tubes (Nalgene, New York, NY, USA).

2.3. Instrumental analysis and quality control

For trace metals, samples were analyzed using inductively coupled plasma mass spectrometer (ICP-MS, Agilent 7700 series, Santa Clara, CA, USA). Instrument operating conditions and parameters for metal analysis are presented in Table S2 (Supplementary material). The detection limits of ICP-MS for the studied metals 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. Multi-element Standard XSTC-13 (Spex CertiPrep®, Metuchen, NJ, USA) solutions were used to prepare calibration curves. Internal calibration standard solutions containing 1.0 mg/L of indium (In), yttrium (Y), beryllium (Be), tellurium (Te), cobalt (Co) and thallium (Tl) were purchased from Spex CertiPrep® (Metuchen, NJ, USA). During the procedure, 10 μg/L internal standard solution was prepared from the primary standard and added to the digested samples. Multi-element solution (purchased from Agilent Technologies) 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). Before starting the analysis sequence, relative standard deviation (RSD, <5%) was checked by using tuning solution purchased from Agilent Technologies. The certified reference materials INCT-CF-3 (corn flour) and DORM-2 (dogfish muscle), both from the National Research Council (Canada), were analyzed to confirm analytical performance and good precision (relative standard deviation bellow 20%) of the applied method (Table S3; Supplementary material).

2.4. Data calculation

2.4.1. Estimated daily intake of metals due to food consumption

The estimated daily intake of trace metals (EDI) through foodstuffs depends on metal concentrations (on fresh weight basis), daily vegetable consumption, as well as body weight, which was calculated with the following formula:

$$ EDI = \frac{\sum \text{FIR}_i \times C_i}{\text{BW}} \times 10^{-3}, $$

where FIR is the daily food consumption rate (on fresh weight basis) for adult residents presented in Table 3 [(questionnaire survey of this study and Report of the household income and expenditure survey 2010 (HIES, 2011); in this study, we conducted 270 food consumption questionnaire surveys from 15 February to 15 March, 2012], C is the metal concentrations (mg/kg fw) in foods, and BW is the body weight of an adult resident, which was set to 60 kg in the present study (Islam et al., 2014a).

2.4.2. Non-carcinogenic and carcinogenic risk

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

$$ HQ = \frac{EDI}{RfD} \times 10^{-3}, $$

where EDI is the estimated daily intake of metal (mg/day) and RfD is the oral reference dose (mg/kg/day). The oral reference doses were based on 0.003, 0.02, 0.04, 0.0003, 0.001 and 0.004 mg/kg/day for Cr, Ni, Cu, As, Cd and Pb, respectively (USEPA, 2010; Islam et al., 2014a). If the HQ is equal to or higher than 1, there is a potential health risk, and related interventions and protective measures should be taken (Islam et al., 2014a).

The target carcinogenic risks (CR) derived from the intake of As and Pb were calculated using the equation provided in USEPA Region III Risk-Based Concentration Table (USEPA, 2006):

$$ CR = \frac{Efr \times ED \times EDI \times CSF_{Co}}{AT} \times 10^{-3}, $$

where Efr is the exposure frequency (350 days/year), ED is the exposure duration (30 years) (USEPA, 2006) and AT is the averaging time for carcinogens (365 days/year × 70 years). CSF is the oral carcinogenic slope factor from the Integrated Risk Information System (USEPA, 2010) database were 1.5 and 8.5 × 10⁻³ (mg/kg/day)⁻¹ for As and Pb.

2.5. Statistical analysis

The data were statistically analyzed using the statistical package SPSS 16.0 (International Business Machines Corporation [IBM] Armonk, NY, USA). The means and standard deviations of the metal concentrations in foodstuffs were calculated. Multivariate Post Hoc Tukey tests were employed to examine the statistical significance of the differences among mean concentrations of trace metals among foodstuffs. Cluster analysis (CA) was used to obtain the detailed information of the dataset and gain insight into the distribution of trace metals by detecting similarities or differences in samples.

3. Results and discussion

3.1. Metal concentrations in foodstuffs

The concentrations of Cr, Ni, Cu, Zn, As, Cd and Pb (mg/kg fw) were determined in the most commonly consumed foodstuffs in Bangladesh (cereals, pulses, vegetables, fruits, fish, meat, eggs and milk) and presented in Table 1 and Fig. 3. Average concentrations of trace metals among the food groups showed the descending order of: vegetables > fish > fruits > pulses > cereals > egg > meat > milk. For all types of foods, a relatively large variability in metal concentrations was observed, even within the same kind of food. The observed variation in metal concentrations in foodstuffs could be due to variation in absorption and accumulation capabilities (Pandey and Pandey, 2009), growth period and stages of food crops (Saha and Zaman, 2013) or climatic differences of the study areas (Santos et al., 2004).

The mean concentration of Cr in foodstuffs followed the descending order of: fish > pulse > cereals > milk > fruit > egg >
Table 1
Trace metal concentrations [mg/kg fw, (mean ± SD)] in most frequently consumed foodstuffs in Dhaka City, Bangladesh.

<table>
<thead>
<tr>
<th>Foodstuff</th>
<th>Scientific name</th>
<th>Cr</th>
<th>Ni</th>
<th>Cu</th>
<th>As</th>
<th>Cd</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cereals</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rice (n = 13)</td>
<td>Oryza sativa</td>
<td>0.91</td>
<td>0.42</td>
<td>1.4</td>
<td>1.6</td>
<td>6.3</td>
<td>3.1</td>
</tr>
<tr>
<td>Maize (n = 8)</td>
<td>Zea mays</td>
<td>1.8</td>
<td>0.33</td>
<td>1.3</td>
<td>0.44</td>
<td>2.3</td>
<td>0.39</td>
</tr>
<tr>
<td>Pulses</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lentil (n = 7)</td>
<td>Lens culinaris</td>
<td>1.5</td>
<td>1.1</td>
<td>0.76</td>
<td>0.46</td>
<td>3.3</td>
<td>2.2</td>
</tr>
<tr>
<td>Mung Bean (n = 3)</td>
<td>Vigna radiata</td>
<td>2.6</td>
<td>1.3</td>
<td>3.0</td>
<td>0.86</td>
<td>4.4</td>
<td>1.6</td>
</tr>
<tr>
<td>Blackgram (n = 5)</td>
<td>Vigna mungo</td>
<td>2.2</td>
<td>1.3</td>
<td>2.8</td>
<td>1.7</td>
<td>4.6</td>
<td>1.4</td>
</tr>
<tr>
<td>Vegetables</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Brinjal (n = 9)</td>
<td>Solanum melongena</td>
<td>0.84</td>
<td>0.41</td>
<td>3.1</td>
<td>1.6</td>
<td>16</td>
<td>5.7</td>
</tr>
<tr>
<td>Bottle gourd (n = 9)</td>
<td>Lagenaria siceraria</td>
<td>0.60</td>
<td>0.28</td>
<td>3.1</td>
<td>1.9</td>
<td>10</td>
<td>3.1</td>
</tr>
<tr>
<td>Pumpkin (n = 9)</td>
<td>Cucurbita maxima</td>
<td>0.62</td>
<td>0.21</td>
<td>4.8</td>
<td>3.9</td>
<td>10</td>
<td>3.3</td>
</tr>
<tr>
<td>Tomato (n = 9)</td>
<td>Solanum lycopersicum</td>
<td>0.68</td>
<td>0.19</td>
<td>1.6</td>
<td>1.0</td>
<td>11</td>
<td>3.2</td>
</tr>
<tr>
<td>Fruits</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Papaya (n = 3)</td>
<td>Carica papaya</td>
<td>1.5</td>
<td>0.41</td>
<td>0.85</td>
<td>0.48</td>
<td>3.7</td>
<td>1.7</td>
</tr>
<tr>
<td>Banana (n = 5)</td>
<td>Musa paradisiaca</td>
<td>1.2</td>
<td>1.3</td>
<td>1.0</td>
<td>0.56</td>
<td>4.9</td>
<td>2.5</td>
</tr>
<tr>
<td>Mango (n = 3)</td>
<td>Mangifera indica</td>
<td>2.1</td>
<td>1.1</td>
<td>0.76</td>
<td>0.76</td>
<td>4.5</td>
<td>3.2</td>
</tr>
<tr>
<td>Jack fruit (n = 3)</td>
<td>Artocarpus heterophyllus</td>
<td>1.0</td>
<td>0.34</td>
<td>2.3</td>
<td>2.3</td>
<td>8.9</td>
<td>2.6</td>
</tr>
<tr>
<td>Fish</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stinging catfish (n = 18)</td>
<td>Channa punctatus</td>
<td>2.0</td>
<td>0.90</td>
<td>1.0</td>
<td>0.96</td>
<td>3.0</td>
<td>1.5</td>
</tr>
<tr>
<td>Banded gourami (n = 18)</td>
<td>Trichogaster fasciata</td>
<td>2.1</td>
<td>0.82</td>
<td>1.5</td>
<td>0.75</td>
<td>4.7</td>
<td>1.2</td>
</tr>
<tr>
<td>Meat</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chicken (n = 5)</td>
<td>Gallus gallus domesticus</td>
<td>1.4</td>
<td>0.31</td>
<td>0.39</td>
<td>0.43</td>
<td>2.5</td>
<td>1.6</td>
</tr>
<tr>
<td>Beef (n = 3)</td>
<td>Bos primigenus</td>
<td>1.3</td>
<td>0.24</td>
<td>0.10</td>
<td>0.08</td>
<td>1.2</td>
<td>0.49</td>
</tr>
<tr>
<td>Mutton (n = 3)</td>
<td>Capra hircus</td>
<td>1.2</td>
<td>0.16</td>
<td>1.5</td>
<td>1.6</td>
<td>2.5</td>
<td>1.6</td>
</tr>
<tr>
<td>Chicken egg (n = 10)</td>
<td>Gallus gallus domesticus</td>
<td>1.4</td>
<td>0.19</td>
<td>1.9</td>
<td>1.8</td>
<td>4.0</td>
<td>2.1</td>
</tr>
<tr>
<td>Milk</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cow milk (n = 12)</td>
<td>Bos primigenus</td>
<td>1.6</td>
<td>0.41</td>
<td>1.5</td>
<td>2.2</td>
<td>2.3</td>
<td>1.2</td>
</tr>
</tbody>
</table>

meat > vegetables. Statistical significant differences (p < 0.05) were observed for Cr concentrations among the food samples (Fig. 3). The highest mean concentration of Cr was observed in fish of Trichogaster fasciata (2.9 mg/kg) and the lowest was found in vegetables of Lagenaria siceraria (0.60 mg/kg fw) (Table 1). The concentrations of Cr in most of the foods were higher than the maximum allowable concentration (MAC) of Cr in foods (Table 2), indicating severe Cr contamination of foodstuffs. The observed elevated concentrations of Cr in the fish samples might be due to the untreated wastewater coming from various industries such as dying and tanning, photography, textile, manufacturing green varnish, paints, and inks run-off from upstream agricultural fields (Kashem and Singh, 1999; Islam et al., 2009, 2014a; Bhiyani et al., 2011; Rahman et al., 2012, 2013).

The mean concentration of Ni in the studied foods followed the descending order of: vegetables > pulse > egg > milk > fish > cereals > fruit > meat. Among the studied vegetables, the highest mean of Ni was observed in Cucurbita maxima (4.8 mg/kg), followed by the Lagenaria siceraria, Solanum melongena and Solanum lycopersicum (Table 1). The concentrations of Ni in vegetables were higher than the recent study by Rahman et al. (2013), where mean Ni concentration was reported as 1.4 mg/kg in non-leafy vegetables from Bangladesh.

Among the food items, the mean Cu concentrations followed the descending order of: vegetables > fruit > egg > pulse > fish > cereals > milk > meat. The highest mean concentration of Cu was found in Solanum melongena: (mean: 16 mg/kg) and the lowest in beef meat (0.10 mg/kg) (Table 1). A recent study by Rahman et al. (2013) reported mean Cu concentrations of 17 and 21 mg/kg in leafy and non-leafy vegetables, respectively, collected from households in the Noakhali district in Bangladesh. Alam et al. (2003) reported Cu concentrations of 8.5 and 15 mg/kg in leafy and non-leafy vegetables, respectively, from Samta village in Bangladesh. The results showed that Cu concentration in vegetables of the present study were comparable with studies conducted for garden vegetables from the other areas in Bangladesh (Rahman et al., 2013; Alam et al., 2003).

The mean As concentrations (mg/kg) followed the descending order of: cereals (0.64) > fruit (0.38) > fish (0.25) > pulse (0.24) > egg (0.087) > vegetables (0.080) > milk (0.056) > meat (0.037). The mean concentration of As in Oryza sativa was 0.17 mg/kg, which was in line with the previous study conducted by Williams et al. (2005), where As concentration in Bangladeshi rice was 0.13 mg/kg. The elevated As concentration in Oryza sativa can be due to the higher transfer of As from soil to grain in the anaerobic paddy soil systems for rice production (Williams et al., 2007; Roberts et al., 2011; Neumann et al., 2010) and uncontrolled application of As enriched fertilizers and pesticides (Renner, 2004).

The second highest concentrations of As were observed in fruits (0.48, 0.41, 0.36 and 0.22 mg/kg for Musa paradisiaca, Mangifera indica, Artocarpus heterophyllus and Carica papaya, respectively) (Table 2). The mean concentration of As in cereals, fruits, fish and pulses were higher than the maximum allowable concentration (MAC) of As (0.1 mg/kg), indicating these foods are contaminated by As and might pose risk to the consumers. The present study explained the causes and sources of the elevated level of As in the foods as due to the effect from both natural and anthropogenic sources, especially use of ground water for irrigation containing As (Neumann et al., 2010, 2011; Hug et al., 2011), as well as uncontrolled application of As-enriched fertilizers and pesticides (Renner, 2004; Islam et al., 2014a).

Cadmium is a metallic element that occurs naturally at low levels in the environment (Rahman et al., 2014). Food, rather than air or water, represents the major source of cadmium exposure (FSANZ, 2003). The mean Cd concentrations (mg/kg) followed the descending order of: vegetables (0.15) > cereals (0.075) > fruit (0.04) > fish (0.035) > milk (0.029) > meat (0.027) > egg (0.022) > pulse (0.018 mg/kg). Statistical significant difference (p = 0.05) was observed for Cd in vegetables compared to the other foodstuffs. Among vegetables, the highest Cd concentration was observed in S. melongena (mean: 0.42, range: 0.22–0.85 mg/kg) followed...
**S. lycopersicum, L. siceraria** and **C. maxima** (Table 1). In the present study, concentrations of Cd in vegetables were slightly higher than in some of the studies on Bangladeshi vegetables by Alam et al. (2003) (range: 0.012–0.22 mg/kg), Rahman et al. (2013) (mean: 0.13 mg/kg, range: 0.006–0.43 mg/kg) and Rahman et al. (2014) (mean: 0.14 mg/kg, range: 0.009–0.43 mg/kg). Among the foodstuffs, Cd in vegetables exceeded the MAC, indicating that vegetables are contaminated by Cd.

**Table 2**

Maximum allowable concentration (MAC) (mg/kg fw) of trace metals in the foodstuffs collected from the vicinity industries around Dhaka City, Bangladesh.

<table>
<thead>
<tr>
<th>Foodstuffs</th>
<th>Cra</th>
<th>Nib</th>
<th>Cuc</th>
<th>Asd</th>
<th>Cad</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rice</td>
<td>1.0</td>
<td>0.5</td>
<td>20c</td>
<td>0.1</td>
<td>0.4</td>
<td>0.2</td>
</tr>
<tr>
<td>Maize</td>
<td>1.0</td>
<td>0.5</td>
<td>–</td>
<td>0.1</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>Pulses</td>
<td>1.0</td>
<td>0.5</td>
<td>–</td>
<td>0.1</td>
<td>0.1</td>
<td>0.2</td>
</tr>
<tr>
<td>Vegetables</td>
<td>1.0</td>
<td>1.5c</td>
<td>10c</td>
<td>0.1</td>
<td>0.05</td>
<td>0.1</td>
</tr>
<tr>
<td>Fruits</td>
<td>1.0</td>
<td>1.0c</td>
<td>–</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Fish</td>
<td>1.0</td>
<td>0.9c</td>
<td>4.5c</td>
<td>1c</td>
<td>2.0</td>
<td>0.3</td>
</tr>
<tr>
<td>Meat</td>
<td>1.0</td>
<td>0.5</td>
<td>–</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Egg</td>
<td>1.0</td>
<td>–</td>
<td>–</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Milk</td>
<td>1.0</td>
<td>0.1</td>
<td>–</td>
<td>0.1</td>
<td>0.1</td>
<td>0.02</td>
</tr>
</tbody>
</table>

d JECFA (2005).

Lead is a toxic metal that enters into the body system through air, water and food and cannot be removed by washing and cooking of foods (Sharma et al., 2007). The mean Pb concentrations (mg/kg) followed the descending order of: fish (1.0) > vegetables (0.84) > fruit (0.53) > pulse (0.35) > meat (0.25) > egg (0.24) > milk (0.20) > cereals (0.17). The highest mean concentration of Pb was found in *T. fasciata* (1.3 mg/kg) followed by *H. fossilis* (1.1 mg/kg) and *C. punctatus* (0.63 mg/kg) (Table 1). Statistically significant differences (*p* < 0.05) were observed for Pb in fish with other foods. The elevated concentration of Pb in fish and vegetables could probably be due to the lead smelting activity in the study area. The concentrations of Pb in foodstuffs were higher than the recommended permissible levels in foods, indicating severe contamination.

Furthermore, using the overall trace metal concentrations in eight different food composites, cluster analysis (CA) with dendrogram using Ward’s Method was adopted to divide the foods into several groups as shown in Fig. 4. Different clusters were formed between different selected foods, where the foods in each group were of similar nature. Moreover, on the basis of trace metal concentrations in some food groups showed strong significant correlations by forming primary groups/clusters with each other (Fig. 4). The primary clusters such as egg–milk–meat, cereals–fruit–pulse in selected food stuffs were formed within a distance of five on the scale (Fig. 4).
3.2. Health risk assessment

3.2.1. Daily intake of metals from foodstuffs

The consumption rates and estimated daily intakes (EDIs) of trace metals in adult inhabitants from different food groups are listed in Table 3. To evaluate the daily intake, the median concentrations of metals in each food category were calculated and then multiplied by the respective consumption rate. Total daily intake of Cr, Ni, Cu, As, Cd and Pb were 0.97, 1.2, 5.6, 0.15, 0.047 and 0.28 mg/day, respectively (Table 3). The maximum contribution of dietary intake of metals came from rice and vegetables, due to these being the most highly consumed foods (445 and 191 g/person/day, respectively). Metal specific EDIs revealed that EDI of Cr, Ni, As, Cd and Pb from consumption of all examined foodstuffs were higher than the maximum tolerable daily intake (MTDI) (Table 3). Based on these data, we conclude that Cr, Ni, As, Cd and Pb were the major components contributing to the potential health risk via consumption of the studied food items around the industrial area in Bangladesh.

The contributions of trace metals from different food items to the dietary intake are summarized in Fig. 5. Rice and vegetables contributed 53%, 77%, 84%, 59%, 85% and 55% for Cr, Ni, Cu, As, Cd and Pb, respectively. The dietary intake of trace metals in Dhaka city, Bangladesh indicated that rice and vegetables were the main sources of trace metals through the diet. Rice contributed to the largest proportion (52%) of total As intake to the exposed population, which could be due to the higher consumption rate than other foods. Previous studies (Ma et al., 2008; Xu et al., 2008) revealed an elevated accumulation of arsenic in rice from arsenic-contaminated crop fields as well as excessive use of ground water as irrigation. Fish contributed 21%, 15% and 31% for Cr, As and Pb, respectively.

Table 3: Food consumption rates and estimated daily intakes of trace metals from commonly consumed foodstuff by Bangladeshi adult population.

<table>
<thead>
<tr>
<th>Foodstuff</th>
<th>Consumption rate (g/day)</th>
<th>Estimated daily intake (EDI) (mg/day)</th>
<th>Cr</th>
<th>Ni</th>
<th>Cu</th>
<th>As</th>
<th>Cd</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rice</td>
<td>445(^a)</td>
<td>0.38</td>
<td>0.41</td>
<td>2.6</td>
<td>0.076</td>
<td>0.023</td>
<td>0.028</td>
<td></td>
</tr>
<tr>
<td>Maize</td>
<td>26.09(^b)</td>
<td>0.047</td>
<td>0.032</td>
<td>0.060</td>
<td>0.015</td>
<td>0.0019</td>
<td>0.004</td>
<td></td>
</tr>
<tr>
<td>Pulse</td>
<td>14.3(^c)</td>
<td>0.033</td>
<td>0.022</td>
<td>0.054</td>
<td>0.0013</td>
<td>0.0001</td>
<td>0.005</td>
<td></td>
</tr>
<tr>
<td>Vegetable</td>
<td>191(^d)</td>
<td>0.13</td>
<td>0.49</td>
<td>2.2</td>
<td>0.010</td>
<td>0.018</td>
<td>0.126</td>
<td></td>
</tr>
<tr>
<td>Fruit</td>
<td>50.59(^d)</td>
<td>0.063</td>
<td>0.049</td>
<td>0.28</td>
<td>0.018</td>
<td>0.0011</td>
<td>0.024</td>
<td></td>
</tr>
<tr>
<td>Fish</td>
<td>87.16(^e)</td>
<td>0.20</td>
<td>0.12</td>
<td>0.33</td>
<td>0.022</td>
<td>0.0024</td>
<td>0.086</td>
<td></td>
</tr>
<tr>
<td>Meat</td>
<td>17.4(^f)</td>
<td>0.021</td>
<td>0.0033</td>
<td>0.030</td>
<td>0.0006</td>
<td>0.0002</td>
<td>0.0022</td>
<td></td>
</tr>
<tr>
<td>Egg</td>
<td>11.3(^g)</td>
<td>0.016</td>
<td>0.020</td>
<td>0.036</td>
<td>0.0007</td>
<td>0.0002</td>
<td>0.0015</td>
<td></td>
</tr>
<tr>
<td>Milk</td>
<td>39.2(^h)</td>
<td>0.066</td>
<td>0.023</td>
<td>0.096</td>
<td>0.0026</td>
<td>0.0012</td>
<td>0.00044</td>
<td></td>
</tr>
<tr>
<td>Daily intake via food consumption</td>
<td>21%</td>
<td>0.2(^h)</td>
<td>0.3(^h)</td>
<td>30(^h)</td>
<td>0.126(^h)</td>
<td>0.046(^h)</td>
<td>0.21(^h)</td>
<td></td>
</tr>
</tbody>
</table>

Maximum tolerable daily intake (MTDI)

\(^a\) Meharg et al. (2009).
\(^b\) HIES (2011).
\(^c\) Food consumption survey of the present study.
\(^d\) RDA (1989).
\(^e\) WHO (1996).
\(^f\) JECFA (2003).

Fig. 5. The contribution of individual foodstuff to the total daily intake of metals in Dhaka City, Bangladesh.
suggesting that people may be exposed these metals due to the consumption of fish from the study area.

3.2.2. Non-carcinogenic and carcinogenic risk

Risk assessment is the process that evaluates the potential health effects from doses to humans of one contaminant received through one or more exposure pathways. The non-carcinogenic risks from consumption of foodstuffs by the adult inhabitants were assessed based on the hazard quotients (HQs). The estimated HQ values of metals are shown in Table 4. The HQ of each metal through consumption of foods in the study area decreased in the order of: As > Cr > Cu > Pb > Ni > Cd. Among the studied metals, the HQ values of Cr, Cu and As exceeded the threshold value of 1 suggested that there are some obvious health risks related to these three metals associated with the consumption of rice, fruit and fish. The data in Table 4 show that cumulative risk of the studied metals (∑HQs) are higher than 1 for rice, vegetables, fruit and fish, indicating that people would be subjected to considerable on-carcinogenic risks from trace metals if they ingest even just these four items of food.

The carcinogenic risks of As and Pb from consumption of foodstuffs by the adult inhabitants were assessed based on the target carcinogenic risk (CR). The estimated CR values of metals are shown in Table 4. The CR values for the studied food items were 6.0 × 10⁻⁶–7.9 × 10⁻⁴ for As, and 1.0 × 10⁻⁸–8.6 × 10⁻⁷ for Pb (Table 4). Carcinogenic risk of Pb from the foodstuffs was less than 10⁻⁶ and regarded as negligible. On the other hand, the risk for As from most of the foods was higher than the acceptable level of 10⁻⁴. Therefore, the potential carcinogenic risks for the urban residents through food ingestion should not be overlooked. The present study has thus clearly revealed that consumption of these food items definitely poses cancer risks to the Bangladeshi population.

4. Conclusions

The concentrations of trace metals varied widely among foodstuffs in Bangladesh and some metals in foods were higher than the maximum allowable concentration (MAC) in foods. Rice contributes the highest intake of Cr, Cu, As, and Cd, whereas vegetables contribute high Ni and Pb. Most of the metals from dietary intake were higher than the maximum tolerable daily intake (MTDI), suggesting a considerable risk. The cumulative risks of studied metals through consumption of rice, fruits, vegetables and fish exceeded unity (HQs > 1), indicating that people would experience significant risks if they ingested these food items. Human health risks associated with food consumption were not negligible, and the sources of metal pollution in foodstuffs should be controlled. Furthermore, this study recommended that urgent attention is required to evaluate the trace metal concentrations in both human beings (blood and urine) and animals of the study area in order to evaluate if any potential health risks from trace metal exposure do exist.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jfca.2014.12.031.

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