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

Nazma Shaheen [a], [*], Nafis Md. Irfan [a], Ishrat Nourin Khan [a], Saiful Islam [a],
Md. Saiful Islam [b], Md. Kawser Ahmed [c]

[a] *Institute of Nutrition and Food Science (INFS), University of Dhaka, Dhaka 1000, Bangladesh*
[b] *Department of Soil Science, Patuakhali Science and Technology University, Dumki, Patuakhali, Bangladesh*
[c] *Department of Oceanography, University of Dhaka, Dhaka 1000, Bangladesh*

HIGHLIGHTS

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

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ABSTRACT

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

1. Introduction

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

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[*] Corresponding author. Institute of Nutrition and Food Science (INFS), University of Dhaka, Dhaka 1000, Bangladesh.
E-mail address: nzmshaheen58@gmail.com (N. Shaheen).

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

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

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

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

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

2. Materials and methods

2.1. Samples and sampling procedure

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

2.2. Analysis of samples

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

2.3. Instrumental analysis and quality assurance

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

2.4. Calculation

2.4.1. Estimated daily intake of heavy metals

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

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

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

2.4.2. Noncarcinogenic risk

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

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

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

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