Public Health Risk Assessment of Heavy Metal Uptake by Vegetables Grown at a Waste-water-Irrigated Site in Dhaka, Bangladesh

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Introduction

Industrial or municipal wastewater is mostly used for the irrigation of crops due to its easy accessibility, difficulty of disposal and scarcity of fresh water. Irrigation with wastewater is known to contribute significantly to the heavy metals content of soil.1 Wastewater may contain various heavy metals, including zinc (Zn), copper (Cu), lead (Pb), arsenic (As), nickel (Ni), chromium (Cr), and cadmium (Cd), depending upon the type of activities associated with it. Continuous irrigation of agricultural land with sewage and industrial wastewater may cause heavy metal accumulation in the soil and vegetables.2-4

Background. Wastewater is often used in developing countries for irrigation of crops. This wastewater often contains a number of heavy metals which are harmful to human health due to their non-biodegradable nature, long biological half-lives and their potential to accumulate in different body parts. There are many potential risks to human health from heavy metal contamination of vegetables grown at waste-water-irrigated sites.

Objectives. This study was carried out to assess the concentration of heavy metals and the risk to human health by heavy metals (lead (Pb), nickel (Ni) and arsenic (As)) through the intake of locally grown vegetables collected from wastewater irrigated agricultural fields. The objectives of the present study were to determine concentrations of heavy metals in vegetables collected from the waste-water-irrigated fields, to calculate daily intake of heavy metals from the consumption of vegetables for both adults and children, and to evaluate their potential health risk.

Methods. Twenty-seven samples of nine different types of vegetables were analyzed by an Atomic Absorption Spectrometer (AAS) (Varian AAS 240 F S).

Results. The range of various metals in waste-water irrigated vegetables were not detected (ND)-0.188, 0.072-1.069 and ND-0.076 mg/kg for Pb, Ni and As, respectively. The highest mean concentration of all metals was detected in jute leaf, except for Pb which was found in the stem amaranth leaf. The mean concentration of all metals in all vegetables was within the safe limits of the World Health Organization/Food and Agricultural Organization (WHO/FAO) and China’s national standards. The health risk index was more than 1 for As in jute leaf for both adults and children. The metal pollution index was highest (0.16 mg/kg) in jute leaf, whereas green papaya showed the lowest metal pollution index (MPI) value (0.005 mg/kg). Among all vegetables tested, the highest intake values of Ni and As were from consumption of jute leaf and the highest intake values of Pb from consumption of stem amaranth leaf for both adults and children.

Conclusions. Higher values in the metal pollution index and health risk index indicate heavy metal contamination in wastewater-irrigated soils that present the potential for a significant negative impact on human health.

Competing Interests. The authors declare no competing financial interests.

Keywords. Heavy metal, vegetable, metal pollution index, daily intake, health risk index, health risk assessment, Bangladesh

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mechanism for their elimination from the body.\textsuperscript{1} Intake of heavy metals through the food chain by human populations has been widely reported throughout the world.\textsuperscript{3} Individual metals exhibit specific signs of toxicity. The nature of their effects can be toxic (acute, chronic or sub-chronic), neurotoxic, carcinogenic, mutagenic or teratogenic.\textsuperscript{6}

Vegetables are very important because they contain essential components of protein, vitamins, iron, calcium and other nutrients.\textsuperscript{7} However, their nutritional value and consumer acceptance as food must be taken into consideration, because vegetables can contain both essential and nonessential elements over a wide range of concentrations.\textsuperscript{5,9} It is well established that the daily intake of heavy metal-contaminated vegetables may pose a risk to human health. This is because heavy metals can accumulate in living organisms and at elevated levels, they can be toxic. It has been reported that prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of metals in the kidney and liver of humans, causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney, and bone diseases.\textsuperscript{10}

Therefore, the aim of present study was: (1) to determine concentrations of heavy metals in vegetables collected from a waste water-irrigated agricultural field, (2) to calculate daily intake of heavy metals through the consumption of vegetables for both adults and children, and (3) to evaluate the potential health risks to local consumers.

**Methods**

**Study Area and Sampling**

Twenty-seven samples (3 samples for each vegetable) of nine different types of vegetables (e.g., red amaranth, radish, bottle gourd, green Indian spinach, green papaya, pointed gourd, stem amaranth leaf, jute leaf and coriander) were collected during February to April, 2015 at randomly selected areas from an agricultural field beside a large canal into which most local industries discharge their wastewater with little or no treatment, located in Savar Upazila, Dhaka District, Bangladesh. The details of the different vegetables sampled during the study are shown in Table 1. The edible portions of test vegetables were collected from the sampling site. The sample size was at least 1 kg. The samples were marked and labelled and kept in sterile polyethylene bags, put into ice boxes and immediately transferred to the laboratory (Bangladesh Council of Scientific and Industrial Research (BCSIR)) and stored at 4°C until analysis within 24 hours.

**Chemical Analysis**

All the collected samples were washed with double distilled water to remove airborne pollutants. The edible parts of the vegetable samples were weighed and air-dried for 24 hours to reduce water content. All the samples were then oven-dried in a hot air oven at 70–80°C for 24 hours to remove all moisture. Dried samples were
powdered using a pestle and mortar and sieved through muslin cloth.

For each vegetable sample, approximately 0.5 g dried samples were digested in a microwave digestion system (Berghof, Speed Wave) using nitric acid: hydrogen peroxide (9:1) to prepare a clear solution. Nitric acid and hydrogen peroxide were collected from Merck, Germany. This clear solution was then brought up to volume with 2 moles (M) nitric acid (400 ml deionized water + 100 ml nitric acid) for heavy metal analysis. The working standard solutions for each metal were prepared before each analysis. The samples were analyzed at the Analytical Research Division, BCSIR Laboratories, Dhaka, using an atomic absorption spectrophotometer (AAS) (Varian AAS 240 F S). Arsenic was analyzed using hydride vapor generation AAS (APHA 3114.C, Varian 220), Ni was measured by air acetylene flame AAS (APHA 3110.B, Varian 240 Z), and Pb by graphite furnace AAS (APHA 3110.B, Varian 240 Z).

**Quality Control Analysis**

Analytical reagent blanks were prepared with each batch of digestion set and then analyzed for the same element in each of the samples. The analytical procedures were verified with National Institute of Standards and Technology (NIST) traceable certified reference standards. Quality control measures were taken to assess contamination and reliability of data. Accuracy of the analytical method was performed as percent recoveries for each of the elements.

**Data Analysis**

**Metal Pollution Index (MPI)**

To examine the overall heavy metal concentrations of vegetables, the metal pollution index (MPI) was computed. This index was obtained by calculating the geometrical mean of concentrations of all the metals in the vegetables, cereals and milk.

\[
MPI \ (\text{mgkg}^{-1}) = \left( C_{f1} \times C_{f2} \times \ldots \times C_{fn} \right)^{1/n}
\]

Where, \( C_{fn} \) = concentration of metal \( n \) in the sample

**Daily Intake of Metals (DIM)**

The daily intake of metals (DIM) was determined by the following equation:

\[
\text{Daily intake of metal (DIM)} = \frac{(\text{Concentration of metal (mg/kg)} \times \text{Daily food intake})}{(\text{Average body weight})}
\]

The average adult and child body weights were considered to be 55.9 and 32.7 kg respectively, while average daily vegetable intakes for adults and children were considered to be 0.345 and 0.232 kg/person/day, respectively.\(^{12-13}\)

**Health Risk Index (HRI)**

The health risk index (HRI) was calculated from the ratio of estimated exposure of test vegetable and oral reference doses.\(^{14}\) Oral reference doses were 3 \(\times\) \(10^{-4}\) mg kg\(^{-1}\) day\(^{-1}\) for As, and 0.004 and 0.02 mg kg\(^{-1}\) day\(^{-1}\) for Pb and Ni, respectively.\(^{15,16}\) The oral reference dose (RfD) for As has been determined based on critical health effects like hyperpigmentation, keratosis and possible vascular complications.\(^{15}\) The RfD for Pb has been determined based on its particular health effects such as neurotoxicity, developmental delay, hypertension, impaired hearing acuity, impaired hemoglobin synthesis, and male reproductive impairment, and the RfD for Ni has been determined based on critical health effects such as decreased body and organ weights.\(^{16}\) Estimated exposure is obtained by dividing the daily intake of heavy metals by their safe limits. An index of more than 1 is considered to be unsafe.

| Edible part of vegetable | Common name       | Botanical name            | Family       |
|--------------------------|------------------|---------------------------|--------------|
| Leaf                     | Red amaranth     | *Amaranthus gangeticus*   | *Amaranthaceae* |
| Root and Leaf            | Radish           | *Raphanus sativus L.*     | *Brassicaceae* |
| Fruit and Leaf           | Bottle gourd     | *Lagenaria siceraria Mol.*| *Cucurbitaceae* |
| Leaf                     | Indian spinach (green) | *Basella alba*          | *Baselaceae* |
| Fruit                    | Green papaya     | *Carica papaya*          | *Caricaceae*  |
| Fruit                    | Pointed gourd    | *Trichosanthas dioica*   | *Cucurbitaceae* |
| Leaf                     | Stem Amaranth Leaf | *Amaranthus lividus*     | *Amaranthaceae* |
| Leaf                     | Jute leaf        | *Corchorus capsularis*   | *Malvaceae*   |
| Leaf                     | Coriander        | *Coriandrum sativum L.*  | *Apiaceae*    |

**Table 1 — Vegetable Samples Collected from the Experimental Sites**
### Health risk index (HRI) = DIM/RfD

Where DIM is daily intake of metal and RfD is the reference oral dose for each metal.

### Statistical Analysis

All analyses were performed in triplicate. Results were expressed as means with ± (standard deviation) SD. The statistical software package Statistical Package for Social Sciences Version 16.0 (SPSS 16.0) was used for the data analysis.

### Results

#### Heavy Metal Concentrations of Vegetables

The mean concentrations of heavy metals in edible parts of various vegetables collected from wastewater-irrigated agricultural fields in Savar Upazila of Dhaka District, Bangladesh are shown in Table 2. The results reveal significant levels (P < 0.05) of identified metals in different vegetable samples. The application of wastewater leads to changes in the physicochemical characteristics of soil and consequently leads to heavy metal uptake by vegetables.1

In general, leafy vegetables (red amaranth, radish, Indian spinach (green), stem amaranth leaf, jute leaf and coriander) accumulated much higher concentrations of Pb, Ni and As compared to non-leaf vegetables (bottle gourd, green papaya and pointed gourd) grown on wastewater-irrigated soils at the same site. Leafy vegetables generally accumulate metals at a higher rate than roots/tuberous vegetables, which is due to the fact that leafy vegetables have high translocation, high transpiration and fast growth rates.5,17,18

The mean concentration of metals in radish, jute leaf and stem amaranth leaf are shown in Table 2. Metals were detected in radish, jute leaf and stem amaranth leaf in the following decreasing order: Ni > Pb > As, Ni > As > Pb and Ni > Pb > As, respectively. For the rest of the vegetables, the order of metal ion concentration was Ni > Pb > As. Islam and Hoquere reported that the order of metal ion concentration in vegetables was Ni > Pb > As with reported total metals concentrations much higher compared to the present study for bottle gourd and red amaranth.19 Singh et al. also reported that the order of metal ion concentration was Ni > Pb for bottle gourd and Ni > Pb for radish in their study.20

Generally, the mean concentration range of Pb in all vegetables analyzed was not detected (ND) to 0.188 mg/kg, with the highest concentration (0.188 mg/kg) in stem amaranth leaf and lowest concentration (ND less than 0.001 mg/kg) in red amaranth, green papaya, pointed gourd and coriander. On the

### Table 2 — Mean Concentration of Heavy Metals (mean ± standard deviation, mg/kg⁻¹) in Vegetable Samples (fresh weight basis)

| Vegetable               | Pb     | Ni     | As     | Total  |
|-------------------------|--------|--------|--------|--------|
| Red amaranth             | ND     | 0.273 ±0.1118 | ND     | 0.273  |
| Radish                  | 0.075 ±0.008 | 0.816 ±0.1051 | 0.022 ±0.0763 | 0.913  |
| Bottle gourd            | 0.026 ±0.0092 | 0.072 ±0.544  | ND     | 0.098  |
| Indian spinach (green)  | 0.049 ±0.011 | 0.575 ±0.0189 | ND     | 0.624  |
| Green papaya            | ND     | 0.098 ±0.1014 | ND     | 0.098  |
| Pointed gourd           | ND     | 0.198 ±0.0712 | 0.003 ±0.0015 | 0.201  |
| Stem amaranth leaf      | 0.188 ±0.0091 | 0.679 ±0.0873 | 0.002 ±0.0019 | 0.869  |
| Jute leaf               | 0.05 ±0.360 | 1.069 ±0.0763 | 0.076 ±0.041 | 1.195  |
| Coriander               | ND     | 0.977 ±0.0721 | 0.021 ±0.0081 | 0.998  |
| FAO/WHO permissible levels | 0.3    | 66.9    | 0.1    | 0.1    |
| Permissible levels in China | 0.3    | —       | 0.5    | 0.5    |
other hand, Ni was detected in the range of 0.072 to 1.069 mg/kg, with the highest concentration (1.069 mg/kg) in jute leaf and lowest concentration (0.072 mg/kg) in bottle gourd. The mean concentration range of As in all vegetables analyzed was ND to 0.076 mg/kg, with the highest concentration (0.076 mg/kg) in jute leaf and lowest concentration (ND less than 0.001 mg/kg) in Indian spinach (green), green papaya and bottle gourd. In the present study, the mean concentrations of all studied heavy metals in all vegetables were much lower than the concentrations reported in a previous study due to less wastewater contamination, less frequency of irrigation with wastewater, and thus lower duration of exposure to wastewater.

**Discussion**

The mean concentrations of metals were compared with the safe limits given by the World Health Organization/Food and Agricultural Organization (WHO/FAO) (Table 2) and all metal concentrations were within the safe limits for all vegetables.\(^{24}\)

Metal concentrations were within the permissible limits for all vegetables.\(^{24}\)

The results of the present investigation agrees with the findings of previous studies with regard to heavy metal contamination in the edible parts of vegetables produced in wastewater-irrigated sites.\(^{25,26}\) Table 3 shows a comparison of the results of previous studies in Bangladesh and other countries indicating the contamination of vegetables by the studied metals. Studies conducted by Muchuweti et al., Sharma et al., and Liu et al. also demonstrated that plants grown on wastewater-irrigated soils are generally contaminated with heavy metals, which pose a major health concern.\(^{5,21,26}\)

| **District (Country)** | **Sampling site description** | **Ni** | **As** | **Pb** | **References** |
|------------------------|-----------------------------|--------|--------|--------|----------------|
| Dhaka (Bangladesh)     | Industrial area             | 5.34   | 0.057  | 0.76   | Islam and Hoque\(^{20}\) |
| Dhaka (Bangladesh)     | Industrial area             | 2.97   | NA     | 3.89   | Ahmad and Ghani\(^{20}\) |
| Noakhali (Bangladesh)  | Arsenic contaminated area    | 1.44   | 0.05   | 3.7    | Rahman et al\(^{20}\) |
| Dabaoshan (China)      | Near mine area              | NA     | NA     | 0.17   | Zhuang et al\(^{20}\) |
| Varanasi (India)       | Urban area                  | NA     | NA     | 1.42   | Sharma et al\(^{3}\) |

**Table 3 — Comparison of Studies of Metal Concentrations in Vegetables**

*ND—Not detected. All values are presented as (mg/kg fw)*

To appraise the health risk associated with heavy metal contamination of vegetables, the daily intake of metals and risk index were calculated. There are several possible pathways of exposure to humans, but the food chain is the most important. The daily intake of metals was calculated according to the average vegetable consumption for both adults and children (Table 4). Among all vegetables tested, the highest intake values of Ni and As were from consumption of jute leaf and the highest intake of Pb came from consumption of stem amaranth leaf for both adults and children. The daily intake of metals in present study is similar to the provisional tolerable daily intake for adults (PTDI) set by the WHO/FAO.\(^{27}\)

The health risk index of metals with regard to noncarcinogenic effects from...
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The consumption of vegetables for both adults and children is presented in Table 5. The results revealed that As contamination in jute leaf posed the greatest health risk to both adults and children. There is growing evidence that chronic exposure to inorganic arsenic (iAs) may increase the risk of keratosis, hyperpigmentation, and cardiometabolic (CM) disorders, including diabetes mellitus (DM) and cardiovascular diseases (CVD).15, 31-33

The health risk index was less than 1 for Pb, As and Ni in all vegetables, except for As in jute leaf for both adults and children. Singh et al. found that the health risk index was less than 1 for Pb and Ni in bottle gourd, radish and pointed gourd, which is similar to the results in the present study.20

In the present study, Pb and Ni were not found to present any risk to the local population. In contrast, Cui et al. reported that local residents of an area near a smelter in Nanning, China were exposed to Pb through consumption of vegetables and there was a potential health risk to the local population.14

Table 4 — Daily Intake of Metals for Individual Heavy Metals in Different Vegetables for Adults and Children
*ND—Not detected. Units of daily intake are expressed as mg/kg/day

| Vegetable          | Pb  | Ni    | As    | Pb  | Ni    | As    |
|--------------------|-----|-------|-------|-----|-------|-------|
| Red amaranth       | ND  | 0.0017| ND    | ND  | 0.0019| ND    |
| Radish             | 0.0005| 0.0050| 0.001 | ND  | 0.0005| 0.0058|
| Bottle gourd       | 0.0002| 0.0004| ND    | ND  | 0.0002| 0.0005|
| Indian spinach (green) | 0.0003| 0.0035| ND    | 0.0003| 0.0041| ND    |
| Green papaya       | ND  | 0.0006| ND    | ND  | 0.0007| ND    |
| Pointed gourd      | ND  | 0.0012| 0.00002| ND  | 0.0014| 0.00002|
| Stem amaranth leaf | 0.0012| 0.0042| 0.00001| 0.0013| 0.0048| 0.00001|
| Jute leaf          | 0.0003| 0.0066| 0.0005| 0.0004| 0.0076| 0.0005|
| Coriander          | ND  | 0.0061| 0.0001| ND  | 0.0069| 0.0001|

Conclusion

The build-up of heavy metals due to continuous irrigation with wastewater has led to the contamination of vegetables in the study area. The highest mean concentration of all metals was detected in jute leaf, except for Pb, which was found in the highest concentrations in stem amaranth leaf.

The mean concentrations of all metals in all vegetables were within the safe limits of the WHO/FAO and China’s national standards. The health risk index was greater than 1 for As in jute leaf for both adults and children. Higher values of metal pollution and health risk indexes indicate that heavy metal contamination in wastewater-irrigated soils present the potential
for a significant negative impact on human health. Therefore, regular monitoring of these toxic heavy metals from industrial effluents in vegetables and other food materials is essential to prevent their excessive build-up in the food chain.

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