

Research article

Levels of three heavy metals in ground water and cereals in Chattogram Division, Bangladesh

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ABSTRACT

Heavy metals determination in cereals and vegetables is one of the vital interests, because of their essential or toxic nature. In order to assess the levels and the impact of different toxic metals on human health, a study was carried out to determine the concentration of different toxic metals present in ground water and edible cereals (rice, wheat, and maize), which are grown in Chattogram area of Bangladesh. The concentrations of three heavy metals, Lead (Pb), Chromium (Cr) and Copper (Cu) in ground water and three varieties of cereals were determined by using Atomic Absorption Spectroscopy (AAS). Results on ground water showed that the mean concentrations of Pb content were within the permissible limits in terms of Bangladesh Drinking water standards, but exceeds the WHO Drinking Water Standards (WHODWS), US Environmental Protection Agency Drinking Water Standards (USEPADWS) values. However, Cr and Cu content were within the permissible limits in terms of Bangladesh Drinking water standards, WHODWS and USEPADWS values, respectively. The concentrations of Pb, Cr and Cu in rice ranged from 0.853 to 9.265mg/L, 0.096 to 0.475µg/L and 0.096 to 0.562mg/L; in wheat, from 0.363 to 7.523 mg/L, 0.056 to 0.284 µg/L and 0.065 to 0.326 mg/L; in maize, from 0.739 to 8.372 mg/L, 0.058 to 0.248 µg/L and 0.036 to 0.478 mg/L, respectively. Thus, awareness should be raised among the people about the adverse health effects of these metals. The carcinogenic and non-carcinogenic risks of Pb, Cr and Cu for human health is a matter of concern.

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1. INTRODUCTION

Toxic heavy metals such as lead (Pb), chromium (Cr), and copper (Cu) are the major contaminants in food supplies (Radwan and Salama, 2006). Their presence in the atmosphere, soil, water, and in various agricultural products such as cereals, even in trace amounts, can cause serious health

problems, especially cardiovascular, kidney, nervous as well as bone diseases. Furthermore, bioaccumulation of toxic heavy metals in the food chain can be highly dangerous to human health due to their persistent nature and potential toxicity (Das et al., 1997 and Melamed et al., 2003). Thus information on the intake of toxic heavy metals through the food chain is important in assessing risk to human health.

Heavy metals enter the human body through inhalation and ingestion, with ingestion being the main route (Tripathi et al., 1997). The elements such as Cd, Cr, and Arsenic are considered as carcinogenic (cancer-producing elements), while Fe, Cu, Zn, Ni, and Mn are considered as essential elements, but, these elements are found in higher than their permit limits may create a toxic effect in human (Edem et al., 2009).

The People's Republic of Bangladesh is a developing country in South Asia, and is overburdened with an enormous population, poverty, illiteracy and frequent natural disasters (Frisbie et al., 2002). Bangladesh is an agriculture-based country with the vast majority of its people involved in food production. Rice is grown during the rainy season and is used primarily for domestic consumption. In the irrigated areas, a second rice crop is possible, followed by wheat, maize, and vegetables in the short, dry winter from November to February.

Cereals are a major component of the human diet, and a source of essential nutrients, antioxidants and metabolites (Singh et al., 2010). However, intake of toxic metal-contaminated cereals may pose a risk to human health. Toxic metal contamination of foodstuff is one of the most important aspects of food quality assurance (Wang et al., 2005 and Khan et al., 2008). Agricultural activities have been identified as contributors to increasing toxic metal contamination through the application of various types of pesticides and fertilizers (Khairiah et al., 2009). Water is one of the most valuable natural resources on earth and contains minerals, which are extremely important in human nutrition (Bulut et al., 2010).

Groundwater is highly valued because of certain properties not possessed by surface water. Thus, it is used for different purposes, viz. drinking, domestic, irrigation and industrial, depending upon its intrinsic quality. Therefore, it is of prime importance to have prior information on the quantity and quality of water resources available in the region (Goel, 2006). People around the world have used groundwater as a source of drinking water, and even today more than half of the world's population depends on groundwater for survival (Behera et al., 2012). The supply of safe potable water has a

significant impact on the prevention of water-transmissible diseases (Lerda and Proserpi, 1996; Altun et al., 2009). The abundance of organic compounds, toxic metals, radionuclides, nitrites, and nitrates in potable water may cause adverse effects on human health (Ikem et al., 2002). Therefore, the assessment of water quality is important for knowing its suitability for various purposes. Thus, the purpose of this study was to determine the concentrations of toxic metals, including Pb, Cr, and Cu in groundwater and in locally collected cereals, such as, rice, wheat, and maize in the Chattogram area of Bangladesh.

2. MATERIALS AND METHODS

Sample collection

Collection of groundwater

A total of 20 samples of groundwater was collected from 10 locations during March to April 2015 in the Chattogram area of Bangladesh. All the samples were divided in three (n=3) portion. All samples were collected directly into 1 litre polyethylene bottles without filtering. Samples were analyzed for pH immediately, after collection by glass electrode, preserved by acidification to pH < 2 with 18.6% (w/w) HNO₃, and stored in ice-packed coolers. The temperature of all the stored samples was maintained at 0–4°C until analysis.

Collection of cereals (rice, wheat and maize)

Samples of rice (*Oriza sativa*), wheat (*Triticum aestivum*) and maize (*Zea mays*) were collected from the field, wrapped in polyethylene bags and transported to the laboratory. Total 30 samples of cereals (10 rice, 10 wheat and 10 maize samples) were collected from 10 locations in the Chattogram area of Bangladesh. Each sample was divided in three (n=3) portion. Each portion of the samples were then dried properly and labeled it in the laboratory. The dried samples were finally preserved at 4°C until analysis.

Sample preparation

Groundwater samples were prepared for analysis by adopting the method described by Gregg (Gregg, 1989). A hundred milliliters of each representative water sample were

transferred into the Pyrex beakers containing 10 ml of concentrated HNO_3 . The samples were boiled slowly and then evaporated on a hotplate to the lowest possible volume (about 20 ml). The beakers were allowed to cool and another 5 ml of conc. HNO_3 was added. Heating was continued with the addition of conc. HNO_3 as necessary until digestion was complete. The samples were evaporated again to dryness and the beakers were cooled, followed by the addition of 5 ml of HCl solution (1:1 v/v). The solutions were then warmed and 5 ml of 5M NaOH was added, and then filtered through Whatman No. 42 filter paper. The filtrates were transferred to 100 ml volumetric flasks and diluted up to the mark with distilled water. These solutions were then used for the elemental analysis. Solid samples (rice, wheat, and maize) were prepared by the dry ashing method, which is described in literature (Bader, 2011). Samples were washed with deionized water and allowed to dry at room temperature. Dry ashing or oxidation is usually performed by placing 0.5g dried sample in an open vessel and destroying the combustible (organic) portion of the sample by thermal decomposition, normally in the presence of an ashing aid, using a muffle furnace. Typical ashing temperatures are 450 to 550°C and times are 4 to 6 hours at atmospheric pressure, and the ash residues were dissolved in 20 ml 2N nitric acid in combination with 2N sulfuric acid (3:1). Finally, the samples were filtered using Whatman No. 42 filter paper and then analyzed for toxic elements.

Analytical methods and instrumentation

Lead, chromium and copper in groundwater and cereals were determined according to previously described methods (Al-Saleh and Shinwari, 2001; Llobet et al., 2003). The samples were analyzed in a Quality Control Laboratory, Department of Applied Chemistry and Chemical Technology, CVASU, with quality assurance schemes by using “Analytikjena Atomic Absorption Spectrophotometer, model: ZEEnit300P, Germany”. For the determination of lead, chromium and copper by atomic absorption spectrophotometer (AAS), calibration curves were prepared by respective standard solution. Standards with the concentration of lead at 0.40 ppm, 0.80 ppm, and 1.20 ppm, respectively, were set for the

calibration of the AAS. The AAS was also calibrated with copper at the same concentration. Standards with the concentration of chromium at 5 ppb, 10 ppb and 15 ppb, respectively, were set for the calibration of the AAS. The calibration curve of well-prepared standards and an accurate AAS should present as a linear curve. The data for the calibration of lead, chromium and copper are shown in Figures 1, 2 and 3, respectively.

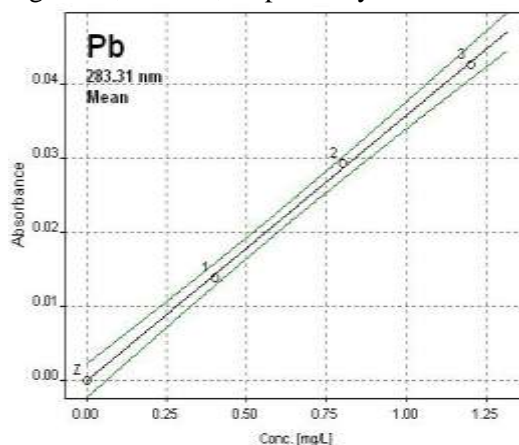


Figure 1. Lead calibration curve.

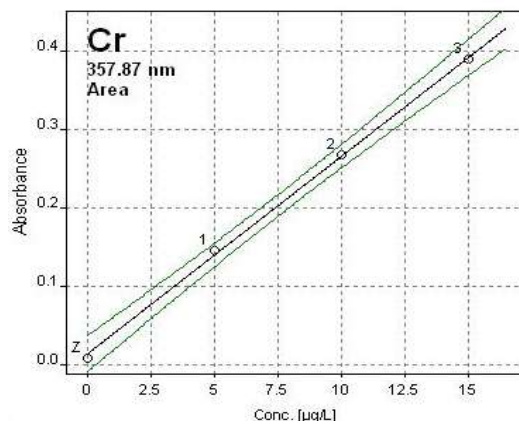


Figure 2. Chromium calibration curve.

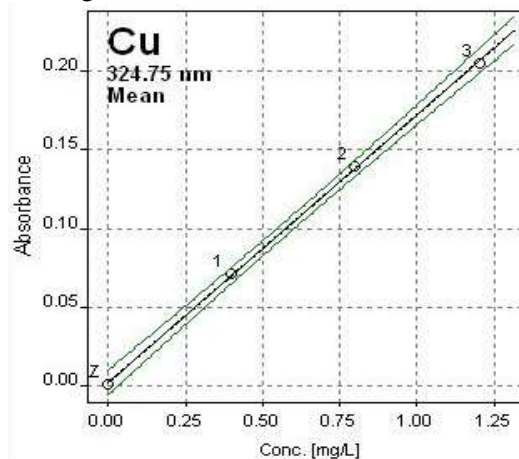


Figure 3. Copper calibration curve.

Data analysis

The concentration of lead, chromium and copper in groundwater and cereals were determined by using ASpect LS 1.2.0.0, Analytik Jena AG 2011-2012 system software. Statistical analysis was performed using SPSS statistical software, version 18. All values were expressed as mean \pm standard deviation (SD).

3. RESULTS

The concentration of lead, chromium and copper in groundwater samples are given in the table 1. In this study, the concentration of lead, chromium, and copper in groundwater samples were found in the range from 0.006 mg/L to 0.073 mg/L, 0.002 μ g/L to 0.024 μ g/L, and 0.003 mg/L to 0.083 mg/L, respectively. The highest concentration of lead, chromium, and copper in groundwater samples were found 0.073 mg/L (GW-6), 0.024 μ g/L (GW-13), and 0.083 mg/L (GW-7), respectively.

The level of lead, chromium and copper in rice samples are given in the table 2. The lead, chromium, and copper concentration in rice samples were found in the range from 0.853 mg/L to 9.265 mg/L, 0.096 μ g/L to 0.475 μ g/L, and 0.096 mg/L to 0.562 mg/L, respectively. The highest level of lead chromium, and copper were found 9.265 mg/L (R-4), 0.475 μ g/L (R-2), and 0.562 mg/L (R-8), respectively.

The content of lead, chromium, and copper in wheat samples are given in the table 3. The lead, chromium, and copper concentration in wheat samples were found in the range from 0.363 mg/L to 7.523 mg/L, 0.056 μ g/L to 0.284 μ g/L, and 0.065 mg/L to 0.326 mg/L, respectively. The highest concentration of lead, chromium, and copper were found 7.523 mg/L (W-3), 0.284 μ g/L (W-8), and 0.326 mg/L (W-5), respectively.

The concentration of lead, chromium, and copper in maize samples are given in the table 4. The concentrations of lead, chromium, and copper in maize samples were found in the range from 0.739 mg/L to 8.372 mg/L, 0.058 μ g/L to 0.248 μ g/L, and 0.036 mg/L to 0.478 mg/L, respectively. The maximum concentration of lead, chromium, and copper in maize samples were found 8.372 mg/L (M-7), 0.248

μ g/L (M-2), and 0.478 mg/L (M-8), respectively.

4. DISCUSSION

Although the sample sizes for the four kinds of samples were less, however, the findings of the study raises serious concerns relating to environmental health issues caused by multi-metal effects. In this study, the lead content in groundwater samples were exceed maximum permeable limit as set by WHO (WHO, 1993; WHO, 1996) but below the maximum permeable limit as set by Bangladesh drinking water standards (BDWS, 1997; Islam et al., 2012). The chromium content in the groundwater samples were found under maximum permeable limit as set by WHO. WHO considered the level of copper is 2 mg/L in groundwater. The highest concentration of copper in groundwater was found 0.083 mg/L (table 1), which is below the safe limit for copper in groundwater and health-related certain substances. Lead, chromium and copper in the groundwater of the studied area are associated with known health risks. Lead is a 'possible human carcinogen' because of inconclusive evidence of human and sufficient evidence of animal carcinogenicity (Frisbie et al., 2002). The 0.01 mg/L WHO drinking water guideline for lead was calculated using the lowest measurable retention of lead in the blood and tissues of human infants (Frisbie et al., 2002). The International Agency for Research on Cancer categorizes Cr(VI) as 'carcinogenic to humans' and Cr(III) as 'not classifiable (IARC, 1987). However, the USEPA lists total Cr in drinking water as having 'inadequate or no human and animal evidence of carcinogenicity (USEPA, 1996). WHO states that 0.05 mg/L drinking water guideline for total Cr is unlikely to cause significant health risks (Frisbie et al., 2002).

The Joint FAO/WHO Food Standards Programmed recommended maximum permissible limit of lead in foodstuff is 7.2 mg/Kg (FAO/WHO, 2011). From the results (table 2 - 4), lead content in cereals samples were found below the maximum permissible limit of lead as prescribed by WHO. The chromium concentration in the investigated cereal samples were in the range from 0.056 to

Table 1. Concentration of lead, chromium and copper in groundwater

Sample no.	Concentration		
	Lead(mg/L) (mean \pm SD)	Chromium(μ g/L) (mean \pm SD)	Copper(mg/L) (mean \pm SD)
GW-1	0.008 \pm 0.004	0.009 \pm 0.0005	0.019 \pm 0.0064
GW-2	0.012 \pm 0.001	0.007 \pm 0.0003	0.008 \pm 0.0002
GW-3	0.038 \pm 0.008	0.006 \pm 0.0008	0.003 \pm 0.0007
GW-4	0.020 \pm 0.005	ND	0.016 \pm 0.0052
GW-5	ND	ND	0.043 \pm 0.0035
GW-6	0.073 \pm 0.004	0.002 \pm 0.0004	ND
GW-7	0.042 \pm 0.008	0.014 \pm 0.0022	0.083 \pm 0.0012
GW-8	0.014 \pm 0.005	ND	0.013 \pm 0.0009
GW-9	0.036 \pm 0.009	0.018 \pm 0.0005	ND
GW-10	0.014 \pm 0.004	ND	0.052 \pm 0.0035
GW-11	0.062 \pm 0.008	ND	0.074 \pm 0.0042
GW-12	0.045 \pm 0.007	0.018 \pm 0.0011	ND
GW-13	0.016 \pm 0.004	0.024 \pm 0.0020	0.023 \pm 0.0041
GW-14	0.053 \pm 0.006	ND	0.026 \pm 0.0052
GW-15	0.042 \pm 0.008	0.006 \pm 0.0009	0.061 \pm 0.0084
GW-16	0.010 \pm 0.004	ND	0.052 \pm 0.0032
GW-17	0.006 \pm 0.003	0.010 \pm 0.0003	0.038 \pm 0.0012
GW-18	ND	0.008 \pm 0.0004	0.056 \pm 0.0040
GW-19	0.048 \pm 0.005	0.020 \pm 0.0011	0.017 \pm 0.0016
GW-20	0.045 \pm 0.008	0.012 \pm 0.0008	0.025 \pm 0.0014
WHODWS ^a	0.01	0.05	2.0
USEPADWS ^b	0.015	0.1	1.3
BDWS ^c	0.05	0.05	1.0

ND = Not detectable, Number of population of each sample is 3 (n=3)

^aWHO Drinking Water Standards (WHO,1993; WHO, 1996). ^bUS Environmental Protection Agency Drinking Water Standards (USEPA, 1996). ^cBangladesh Drinking Water Standards (BDWS, 1997; Islam et al., 2012).

Table 2. Concentration of lead, chromium and copper in rice

Sample no.	Concentration		
	Lead(mg/L) (mean \pm SD)	Chromium(μ g/L) (mean \pm SD)	Copper(mg/L) (mean \pm SD)
R-1	5.657 \pm 0.24	0.142 \pm 0.001	0.284 \pm 0.014
R-2	2.156 \pm 0.08	0.475 \pm 0.084	0.467 \pm 0.083
R-3	2.335 \pm 0.12	0.218 \pm 0.013	ND
R-4	9.265 \pm 0.31	0.284 \pm 0.046	0.096 \pm 0.008
R-5	1.538 \pm 0.09	0.312 \pm 0.065	ND
R-6	5.956 \pm 0.16	0.096 \pm 0.003	0.386 \pm 0.052
R-7	ND	0.279 \pm 0.064	0.169 \pm 0.012
R-8	4.673 \pm 0.26	0.368 \pm 0.042	0.562 \pm 0.068
R-9	0.853 \pm 0.02	ND	0.428 \pm 0.066
R-10	6.154 \pm 0.25	0.185 \pm 0.016	0.263 \pm 0.044

ND = Not detectable, Number of populations of each sample is 3 (n=3)

Table 3. Concentration of lead, chromium and copper in wheat

Sample no.	Concentration		
	Lead(mg/L) (mean ± SD)	Chromium(µg/L) (mean ± SD)	Copper(mg/L) (mean ± SD)
W-1	4.362 ± 0.16	0.104 ± 0.014	ND
W-2	5.831 ± 0.32	ND	0.065 ± 0.003
W-3	7.523 ± 0.62	0.056 ± 0.005	0.106 ± 0.011
W-4	2.368 ± 0.46	0.083 ± 0.011	ND
W-5	0.363 ± 0.08	ND	0.326 ± 0.085
W-6	2.543 ± 0.08	0.074 ± 0.009	0.148 ± 0.025
W-7	4.674 ± 0.55	0.106 ± 0.014	ND
W-8	1.739 ± 0.12	0.284 ± 0.086	0.213 ± 0.046
W-9	2.648 ± 0.18	0.162 ± 0.042	0.192 ± 0.018
W-10	3.674 ± 0.36	0.068 ± 0.008	0.096 ± 0.007

ND = Not detectable, Number of populations of each sample is 3 (n=3)

Table 4. Concentration of lead, chromium and copper in maize

Sample no.	Concentration		
	Lead(mg/L) (mean ± SD)	Chromium(µg/L) (mean ± SD)	Copper(mg/L) (mean ± SD)
M-1	4.874 ± 0.32	0.078 ± 0.008	0.154 ± 0.012
M-2	2.658 ± 0.18	0.248 ± 0.025	0.183 ± 0.016
M-3	0.739 ± 0.06	0.096 ± 0.006	0.036 ± 0.006
M-4	2.768 ± 0.36	0.107 ± 0.010	ND
M-5	4.643 ± 0.28	0.058 ± 0.003	0.278 ± 0.013
M-6	4.968 ± 0.52	0.158 ± 0.012	ND
M-7	8.372 ± 0.52	0.092 ± 0.007	0.175 ± 0.022
M-8	1.695 ± 0.12	0.113 ± 0.008	0.478 ± 0.093
M-9	2.538 ± 0.18	ND	0.274 ± 0.016
M-10	5.832 ± 0.45	0.186 ± 0.014	0.306 ± 0.028

ND = Not detectable, Number of populations of each sample is 3 (n=3)

0.475 µg/L. These values are lower than the maximum permissible limit of 2.3 mg/kg (FAO/WHO, 2001; Tegegne, 2015). The FAO/WHO recommended maximum permissible limit of copper in various foodstuff is between the limit from 0.1 mg/Kg to 0.4 mg/Kg (FAO/WHO, 2011). Copper as cupric sulfate has been evaluated by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 1966, 1970, and 1982. The Provisional Maximum Tolerable Daily Intake (PMTDI) of copper was established by this committee as 0.05-0.5 mg/Kg body weight (FAO/WHO, 2011). In this study, most of the samples were found within the maximum permissible limit.

In several countries, similar studies were previously reported concerning heavy metals as

is the case in the current study. Tegegne (2015) showed that high concentration of Pb was obtained in sorghum, 0.08 mg/kg, followed by wheat, 0.05 mg kg, and barley 0.03 mg kg and not detected in maize. The results were in agreement with that obtained by Sharma et al., (2006), who reported Pb concentration (17.54–25.00 mg kg⁻¹) in vegetables grown in industrial areas. Muchuwetiet et al. (2006) reported the level of Pb (6.77 mg kg⁻¹) in vegetables irrigated with mixtures of waste water and sewage from Zimbabwe to be higher than WHO safe limit (2 mg kg⁻¹). Al Jassir et al. (2005) studied six washed and unwashed green leafy vegetables from Saudi Arabia and noted the highest concentrations of Pb in coriander and purslane. Demirezen and Aksoy (2006) reported that levels of Cu (22.19–76.50 mg kg⁻¹) were higher in the leafy species than the non-leafy

vegetable species from Turkey. Sharma et al. (2006) reported the concentration of Cu ($2.25\text{--}5.42\text{ mg kg}^{-1}$) in vegetables grown in waste water areas of Varanasi, India to be within the safe limit. Furthermore, Radwan and Salama (2006) carried out a survey of various fruits and vegetables in Egypt and noted that the highest levels of Pb, Cd, Cu and zinc were present in strawberries, cucumber, dates and spinach, respectively. Tegegne reported the concentration of copper in wheat, barley, sorghum and maize ranged from 0.13 to 1.72 mg/Kg in the cereals (Tegegne, 2015). Szymczak et al. (1993) reported copper as 1.14 to 11.68 mg/Kg in a total of 117 cereal samples examined. Ali and Al-Qahtani (2012) determined the concentration of some heavy metals including Pb and Cu in various cereals and legumes grown in four major industrial and urban cities in the Kingdom of Saudi Arabia. The obtained results indicated that concentrations of major metals were exceeding the recommended maximum acceptable levels proposed by the Joint FAO/WHO Expert Committee on Food Additives.

Tegegne (2015) reported the levels of chromium in sorghum, maize, wheat and barley were found to be 0.95, 0.52, 0.43 and 0.29 mg/kg, respectively. Mateos et al. (2003) reported that breakfast cereals were rich in Cr, with contents ranging between 0.09 ± 0.04 and $0.55\pm 0.08\text{ }\mu\text{g}\cdot\text{g}^{-1}$ and a mean content of $0.23\pm 0.12\text{ }\mu\text{g}\cdot\text{g}^{-1}$ in Spain. Cr (III) is an essential element required for normal sugar and fat metabolism. It is effective to the management of diabetes and it is a cofactor with insulin. Cr (III) and its compounds are not considered a health hazard, while the toxicity and carcinogenic properties of Cr (VI) have been known for a long time (Ihesinachi and Eresiya, 2014). Accumulation of chromium in the body can cause damage to the liver, kidney, nose, lungs; and possible asthma attack (Kleefstra et al., 2004).

4. CONCLUSION

The results of this study showed relatively higher concentrations of three heavy metals in groundwater and studied cereals. The concentration of lead, chromium and copper in most of the samples were observed within the maximum permissible limit. But in some

samples these values exceeded the recommended maximum acceptable levels. Due to poor sample sizes of the items investigated in this study, further studies containing large number of samples truly representing them in an area such as, Chattogram are required to assess whether any health hazards being posed from them through the drinking water and staple food source contamination.

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