Concentration of selected toxic metals in groundwater and some cereals grown in Shibganj area of Chapai Nawabganj, Rajshahi, Bangladesh

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This study is an attempt to assess the extent of toxic metals, including Pb, Cd, Cr, Mn and As in groundwater and some cereals collected from Shibganj area of Chapai Nawabganj, Rajshahi, Bangladesh. The chemical results of groundwater show that the mean concentrations of Pb, Cd and Mn exceed the values of WHO, USEPA as well as Bangladesh Drinking Water Standards. However, As content is within the permissible limits in terms of Bangladesh Drinking Water Standards, but exceeds the WHO and USEPA values. Cr is in negligible quantity in the examined groundwater samples. Results reveal wide variation in toxic metal content among the three cereal samples. In general, the mean toxic metal concentrations in cereals (rice, wheat and urid bean) are lower than the respective established safe limits for these elements, except for lead concentration in all samples.

Keywords: Cereals, groundwater, health hazards, safety limits, toxic metals.

TOXIC heavy metals such as cadmium, chromium, lead, manganese and arsenic are among the major contaminants in food supplies[1]. 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, and bioaccumulation of toxic heavy metals in the food chain can be highly dangerous to human health due to their persistent nature and potential toxicity[2,3]. 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[4].

The People's Republic of Bangladesh is a developing country in South Asia, and is overburdened with an enormous population, severe poverty, illiteracy and frequent natural disasters[5]. It 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 irrigated areas, a second rice crop is possible, followed by wheat, urid bean 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[6]. 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[7,8]. Agricultural activities have been identified as contributors to increasing toxic metal contamination through the application of various types of pesticides and fertilizers[9].

Water is one of the most valuable natural resources on earth and contains minerals which are extremely important in human nutrition[10]. 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[11]. People around the world have used groundwater as a source of drinking water, and even today more than half the world's population depends on groundwater for survival[12].

The supply of safe potable water has a significant impact on the prevention of water-transmissible diseases[13,14]. The abundance of organic compounds, toxic metals, radionuclides, nitrites and nitrates in potable water may cause adverse effects on human health[15]. Therefore, assessment of water quality is important for knowing its suitability for various purposes.

The purpose of this study is to quantify the concentrations of toxic metals, including Pb, Cd, Cr, Mn and As in groundwater and in locally grown cereals, viz. rice, wheat, and urid bean in Chapai Nawabganj District of Bangladesh.

Chapai Nawabganj District is situated on the north-western part of Bangladesh, bordering the state of West Bengal, India in the west, Rajshahi District in the south-east and Noagaon District towards the northeast. Two major rivers, Ganges and Mahananda, along with tributaries like the Pagla, Purnabha and Tangan flow through the district of Chapai Nawabganj. The district has 5 Upazilas (sub-districts): Nawabganj Sadar, Shibganj, Nachole, Bholarhat and Gomastapur.

The present study was carried out in Shibganj Upazila of Nawabganj District in the Division of Rajshahi, Bangladesh. Shibganj is located at 24.6833°N, 88.1667°E. The sub-district is 525.43 sq. km in area, and is bounded by Bholarhat and West Bengal, India in the north, Nawabganj Sadar and West Bengal, India in the south, Bholarhat, Gomastapur, Nachole and Nawabganj Sadar Upazilas in the east, and West Bengal, India in the west.

The main source of drinking and irrigation water in Shibganj area of Nawabganj is the groundwater and majority of the land is being irrigated by shallow and deep tubewells.

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Figure 1. Map of Shibganj Upazila showing locations where groundwater samples were collected from tubewells.

Groundwater samples were collected from 10 tubewells in 10 locations (Figure 1) during July to August 2010. All sampled tubewells were purged by vigorous pumping for 10 min just prior to sample collection and then collected directly into 1 litre polyethylene bottles without filtering. Samples were analysed for pH immediately after collection by glass electrode, preserved by acidification to pH < 2 with 18.6% (weight/weight) HNO_3, and stored in ice-packed coolers. The temperature of all the stored samples was maintained at 0–4°C until analysis.

Samples of rice (*Oriza sativa*), wheat (*Triticum aestivum*) and urid bean (*Vigna mungo*) were collected from the field, wrapped in polyethylene bags and transported to the laboratory. The samples were then dried properly in the laboratory. The dried samples were finely powdered in a mortar cell, screened through a 2 mm sieve (100 meshes) to have a homogeneous powder, and preserved at 4°C.

Groundwater samples were processed adopting the method described by Gregg[16]. Hundred millilitres of each representative water sample was transferred into Pyrex beakers containing 10 ml of concentrated HNO_3. The samples were boiled slowly and then evaporated on a hot plate 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 5 M NaOH was added, and then filtered through Whatman No. 42 filter paper. The filtrates were transferred to 100 ml volumetric flasks and diluted to the mark with distilled water. These solutions were then used for the elemental analysis.

Open wet digestions of each cereal sample with four different acids (250 ml, 2 N) were evaluated: 2 N HNO_3; 2 N $HClO_4$; 2 N H_2SO_4; 2 N HCl. For each digestion, 0.15 g of dried powdered sample was taken into a 500 ml digestion vessel and 250 ml of each acid was added to it. Condensing system was added to the digestion vessel and heated at 100 ± 10°C, until a clear solution was obtained. The digested samples were filtered using Whatman No. 42 filter paper and then analysed for toxic elements.

All samples were analysed for Pb, Cd, Cr, Mn and As at the Central Science Laboratory of Rajshahi University, Bangladesh. The samples were analysed using an atomic absorption spectrophotometer (AAS; Model AA68, Shimadzu Corporation, Japan). The instrument calibration standards were made by diluting the standard (1000 ppm) supplied by Kanto Chemical Co Inc., Japan.

The amount of toxic metals in cereals was calculated using the following expression: Total metal concentration (mg/kg) = $(d/c) \times v$, where d is the reading of the AAS in mg/l (ppm), c the sample weight taken and v the volume of the sample (after digestion).

The mean values of toxic metals quantified in the groundwater samples in the Shibganj area were compared with those of the World Health Organization Drinking Water Standards (WHODWS), US Environmental Protection Agency Drinking Water Standards (USEPADWS) and Bangladesh Drinking Water Standards (BDWS; Table 1).

Table 1 reveals that mean Pb concentration varies between maximum and minimum of 0.06 (GW_3, GW_7) and 0.16 (GW_2) mg/l respectively. These values are higher than the permissible limits of Bangladesh, WHO and USEPA standards of 0.05, 0.01 and 0.015 mg/l respectively. The Cd levels range from 0.006 (GW_6) to 0.013 (GW_3) mg/l, which indicates that in all samples the cadmium content is in excess of the recommended limit prescribed by the Bangladesh, WHO and USEPA standards of 0.005, 0.003 and 0.005 mg/l respectively. Mn concentration varies from 0.89 (GW_9) to 2.25 (GW_2) mg/l. The maximum permissible limit of Mn in drinking water is 0.1 and 0.5 mg/l as recommended by the Bangladesh and WHO standards respectively. Hence, all water samples have Mn in excess of the upper limit given by the Bangladesh or WHO standards. However, Cr concentration in all samples is lower than the recommended values in drinking water (0.05, 0.05 and 0.1 by the Bangladesh, WHO and USEPA standards respectively) for human consumption.

The above findings raise serious concerns relating to environmental health issues caused by multimetal effects. Pb, Cd and Mn in the groundwater of the study 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. The 0.01 mg/l WHO drinking water guideline for Pb was calculated using the lowest measurable retention of Pb in

Table 1. Toxic content of metals (mean ± SD) in groundwater samples from the sampling sites (mg/l)

Location	Location reference no.	Pb	Cd	Cr	Mn	As
Kansat ($n = 3$)	GW_1	0.08 ± 0.00	0.007 ± 0.00	0.002 ± 0.00	1.07 ± 0.01	0.02 ± 0.01
Mobarakpur ($n = 3$)	GW_2	0.16 ± 0.01	0.011 ± 0.00	Nil	2.25 ± 0.09	0.02 ± 0.00
Chak kirti ($n = 3$)	GW_3	0.06 ± 0.01	0.013 ± 0.00	0.001 ± 0.00	1.63 ± 0.05	0.01 ± 0.00
Satrujitpur ($n = 3$)	GW_4	0.09 ± 0.00	0.007 ± 0.00	Nil	1.01 ± 0.06	0.03 ± 0.01
Binodpur ($n = 3$)	GW_5	0.13 ± 0.01	0.010 ± 0.00	Nil	2.12 ± 0.01	0.03 ± 0.00
Manakosa ($n = 3$)	GW_6	0.10 ± 0.00	0.006 ± 0.00	Nil	1.17 ± 0.01	0.02 ± 0.00
Shyampur ($n = 3$)	GW_7	0.06 ± 0.00	0.012 ± 0.00	Nil	2.11 ± 0.02	0.04 ± 0.01
Dhainagar ($n = 3$)	GW_8	0.12 ± 0.01	0.010 ± 0.00	0.001 ± 0.00	1.05 ±0.03	0.02 ± 0.01
Shahbajpur ($n = 3$)	GW_9	0.07 ± 0.00	0.011 ± 0.00	Nil	0.89 ± 0.00	0.03 ± 0.00
Naya-Naobhanga ($n = 3$)	GW_{10}	0.09 ± 0.01	0.009 ± 0.00	Nil	1.11 ± 0.01	0.01 ± 0.00
WHODWS[a]		0.01	0.003	0.05	0.5	0.01
USEPADWS[b]		0.015	0.005	0.1	None	0.01
BDWS[c]		0.05	0.005	0.05	0.1	0.05

[a]WHO Drinking Water Standards[23,24]. [b]US Environmental Protection Agency Drinking Water Standards[20]. [c]Bangladesh Drinking Water Standards[30].

the blood and tissues of human infants[17]. Cadmium is an environmental pollutant ranked eighth in the top 20 hazardous substances priority list. It is a nonessential element for humans, which can cause various acute and chronic adverse effects, such as renal, nervous, bone and osseous diseases and is identified as carcinogenic to humans[18].

The International Agency for Research on Cancer categorizes Cr(VI) as 'carcinogenic to humans' and Cr(III) as 'not classifiable'[19]. However, the USEPA lists total Cr in drinking water as having 'inadequate or no human and animal evidence of carcinogenicity'[20]. WHO states that 0.05 mg/l drinking water guideline for total Cr is unlikely to cause significant health risks[17].

Manganese is a known mutagen. The accumulation of Mn may cause hepatic encephalopathy[21]. Moreover, the chronic ingestion of Mn in drinking water is associated with neurologic damage[22]. The 0.5 mg/l WHO drinking water guideline for Mn was calculated using human exposures in Japan and Greece, and studies on various laboratory animals where neurotoxic and other effects were observed[17].

Arsenic is a mobile element in the environment and may circulate in various forms through the atmosphere, water and soil. From the chemical results of the present study, it can be seen that arsenic concentration varies from 0.01 to 0.04 mg/kg at different sampling locations (Table 1). The results indicate that 8 out of 10 samples are contaminated with arsenic level above the WHO/USEPA guideline value (0.01 mg/l), of which all samples are below the Bangladesh standard (0.05 mg/l). Arsenic is classified as a 'human carcinogen' based on sufficient epidemiologic evidence. It is important to note that the 0.01 mg/l WHO drinking water guideline for As is based on a 6×10^{-4} excess skin cancer risk for human males in Taiwan[17], which is 60 times higher than the 1×10^{-5} factor that is typically used to protect public health. WHO states that the health-based drinking water guideline for As should be 0.00017 mg/l. However, the detection limit for most laboratories is 0.01 mg/l, which is why the less protective guideline was adopted[23,24]. There is sufficient evidence from human epidemiologic studies linking increased mortality from liver, kidney, bladder and lung cancers to drinking As-contaminated water. Furthermore, a thorough review of As and public health recommends a zero exposure level for As in drinking water[25].

The mean values of toxic metals analysed in the cereal samples collected from the field at Bagantoli in Shibganj area are presented in Table 2.

Toxic metal concentration in cereals shows variations depending upon the acid digestion procedure (Table 2). WHO recommended maximum permissible limit of Pb in foodstuff is 7.2 mg/kg (ref. 26). Table 2 indicates that Pb content in the studied cereals ranges from 7.31 to 12.33 mg/kg. Samples digested with four different acids, all exceed the maximum permissible limit of lead prescribed by WHO. The maximum permissible level of Cd in all foods in Bangladesh is 6 mg/kg (dry wt basis)[27]. Many countries have now set legal or at least provisional guideline levels for maximum permissible concentration (MPC) for one or more toxic metals. A critical examination of Table 2 reveals that the cadmium concentration in cereals of the studied area is within the recommended value. From these results, it could safely be said that the cereals of the area are suitable for human consumption in terms of cadmium contamination. The total Cr concentration in the studied cereals is in the 0.03–0.17 mg/kg range for four acid digestions. None of the cereals studied is burdened with Cr in terms of safe limit, i.e. 0.36 mg Cr/kg (ref. 17). From WHO recommendation the normal limit of Mn in foodstuff varies from 15 to 100 mg/kg. The present results show that the concentration of Mn in cereal samples is within the normal limit and one can infer that the cereals are free from Mn contamination. Al Rmalli *et al.*[28] reported that the concentration of arsenic

Table 2. Toxic metals content (mean ± SD) in cereal samples from the study site (mg/kg) (dry wt basis) with different acid digestions

Metal	Digestion solution	Rice	Wheat	Urid bean
Pb	HNO_3 (2N)	12.33 ± 0.31	7.61 ± 0.06	10.32 ± 0.16
	H_2SO_4 (2N)	11.22 ± 0.52	7.56 ± 0.03	9.84 ± 0.04
	HCl (2N)	10.20 ± 0.05	7.31 ± 0.02	8.20 ± 0.02
	$HClO_4$ (2N)	10.56 ± 0.06	7.39 ± 0.01	9.12 ± 0.01
Cd	HNO_3 (2N)	0.24 ± 0.00	0.17 ± 0.00	0.23 ± 0.01
	H_2SO_4 (2N)	0.23 ± 0.00	0.18 ± 0.00	0.19 ± 0.00
	HCl (2N)	0.21 ± 0.00	0.16 ± 0.00	0.29 ± 0.00
	$HClO_4$ (2N)	0.29 ± 0.00	0.15 ± 0.00	0.20 ± 0.00
Cr	HNO_3 (2N)	0.14 ± 0.00	0.13 ± 0.00	0.08 ± 0.00
	H_2SO_4 (2N)	0.11 ± 0.00	0.17 ± 0.01	0.07 ± 0.00
	HCl (2N)	0.12 ± 0.00	0.14 ± 0.00	0.03 ± 0.00
	$HClO_4$ (2N)	0.15 ± 0.01	0.11 ± 0.01	0.10 ± 0.00
Mn	HNO_3 (2N)	1.35 ± 0.02	1.34 ± 0.00	1.36 ± 0.01
	H_2SO_4 (2N)	1.32 ± 0.01	1.36 ± 0.01	1.39 ± 0.00
	HCl (2N)	1.65 ± 0.01	2.29 ± 0.00	2.47 ± 0.01
	$HClO_4$ (2N)	1.52 ± 0.00	2.03 ± 0.00	1.31 ± 0.02
As	HNO_3 (2N)	0.14 ± 0.00	0.15 ±0.00	0.17 ± 0.01
	H_2SO_4 (2N)	0.12 ± 0.01	0.08 ± 0.00	0.37 ± 0.00
	HCl (2N)	0.12 ± 0.00	0.14 ± 0.00	0.40 ± 0.00
	$HClO_4$ (2N)	0.13 ± 0.00	0.14 ± 0.00	0.08 ± 0.00

in vegetables from Bangladesh ranged from 5 to 540 μg/kg. The recommended tolerance limits of As in foodstuff is 1.0 mg/kg (ref. 29). Thus the examined samples are free from As contamination.

In the developing countries more reliance is placed on local groundwater supplies. This is the case of Shibganj where a majority of the people rely on shallow and deep tubewells to meet their water demands. Based on the results obtained here, it can be concluded that the level of some of the toxic metals in groundwater in the study area is in excess of the WHO guideline values. Lead concentration in Shibganj cereals is greater than the threshold, whereas other toxic metals are within permissible limits.

Thus the build-up of toxic metals in cereals and groundwater needs to be monitored periodically in view of their significant accumulation.

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Occurrence of pseudotachylites in the vicinity of South Almora Thrust zone, Kumaun Lesser Himalaya

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Thin veins of pseudotachylites are observed within the South Almora Thrust (SAT) zone of the Almora crystallines, Kumaun Lesser Himalaya. The SAT zone presents a relatively wide variety of mylonite types. Within this zone thin, dark-coloured veins of pseudotachylites are found. Folded veins of irregular thickness are also observed. The pseudotachylites are formed by rapid crystallization of melts. Intense deformation and friction-related heating is generated during the thrust sheet movement, which is responsible for producing the melts. Small amount of melts generated during the frictional heating cool rapidly in association of the wall rock and form thin veins, which have sharp boundaries (or folded) within the wall rock. Quartz, K-feldspar and plagioclase occur as porphyroclasts in the host rocks.

Keywords: Frictional heating, melts, mylonite, pseudotachylites, veins.

THE Almora Crystalline Zone (ACZ) is one of the largest and better-studied crystalline outcrops of the Lesser Himalaya, extending almost WNW-ESE and tectonically bounded on both the sides by sedimentary rocks. These are believed to be the remnants of a large thrust sheet that has moved from the Higher Himalayan Crystalline Zone along the Main Central Thrust (MCT) to rest over the Lesser Himalayan Sedimentary Belt. ACZ is basically characterized by asymmetrical synformal structure. The unit is delineated along the two flanks by the South Almora Thrust (SAT) and the North Almora Thrust (NAT)[1–7] (Figure 1). ACZ is characterized by the typical occurrence of a variety of mylonite types along its southern boundary (SAT) and is marked by a high degree of shearing, pulverization and recrystallization of rocks[8,9].

Mylonites are a special kind of metamorphic rocks that form in both ductile and ductile–brittle shear zones, and accommodate intense strain dominantly through ductile processes[10]. Mylonites have two types of constituents, i.e. matrix and porphyroclasts[11]; the matrix is composed of the more ductile elements of the rock. A true mylonite is composed of 10–50% porphyroclasts[12]. The heat transfer during thrusting has left signatures in rocks deformed during this process and show changes in the textural and chemical composition in the rocks of ACZ, which exhibit intense shearing and deformation along the northern and southern margins and are highly mylonitized[8]. Though mylonites from ACZ have earlier been described by many workers[5,6,13,14], a detailed study of them in the vicinity of the SAT zone (both the footwall block rocks as well as the hanging wall block rocks) has revealed interesting occurrence of pseudotachylites, which are reported here and would help in understanding the tectonic evolutionary history of ACZ.

In the present study the mylonite zone has been traced all along the southern boundary of the Almora unit, i.e. SAT. The hanging wall along the SAT has typically mylonitic rocks exposed, whereas to the south the footwall block has massive (north-dipping) Nagthat quartzites with their constituents showing prominent deformation (mylonitization), with a well-developed stretching lineation. The mylonitic rocks, in general, show a prominent schistosity as defined by the alignment of ellipsoidal quartz and sometimes by feldspar grains and their aggregates (Figure 2).

A zone of intense deformation was observed during the course of the present study (Figure 3) and the samples were subjected to detailed petrographic studies. It is interesting to note that the samples were collected from within the ultramylonites[6]. This zone shows occurrences of pseudotachylites[15,16] in the form of veins, which have either produced within the host rock. They are formed by rapid crystallization, and are chemically and mineralogically layered. The veins of pseudotachylite also occur as pairs of parallel surfaces, connected by injection veins (Figure 4 *c*). Injection veins are more conspicuous and allow recognition of pseudotachylites within the outcrop. The veins of pseudotachylites have distinct, sharp, straight boundaries with the wall rock. K-feldspar and

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