

Lake and reservoir water quality affected by metals leaching from tropical soils, Bangladesh

Md.R. Islam · P. Lahermo · R. Salminen · S. Rojstaczer · V. Peuraniemi

Abstract The release of metals during weathering has been studied in order to assess its geochemical controls and possible effects on environmental health in Bangladesh. A total of 27 soil samples and 7 surface water samples were collected from four locations covering three major regions in the country. Results show that weathering effects are a strong function of climatic conditions. Surface waters are typically enriched in Al, Mg, Ca, Na, K, As, Ba, Cr, Cu, Ni, Pb and Zn. The solubility of metal ions, organometallic complexes, co-precipitation or co-existence with the colloidal clay fraction are the main processes that lead to metal enrichment in lake and reservoir water. Aluminium concentrations exceed World Health Organization (WHO) drinking-water standards in all samples, and in two regions, arsenic concentrations also significantly exceed WHO standards. The elevated levels of As indicate that arsenic contamination of water supplies in Bangladesh is not confined to groundwater.

Key words Soil · Metals · Leaching · Surface water quality · Bangladesh

Introduction

Due to their potential environmental impact, the distribution and fate of metals in lake environments can be of significant societal importance. The geochemical behaviour of metals in natural tropical lake environments (Paul and Pillali 1983; Rivera and others 1986; Olade 1987), however, has received relatively little attention. Yet in developing tropical countries like Bangladesh, metal contamination can have a significant impact on environmental health. Recently, attention has been focused on extensive arsenic contamination of groundwater in Bangladesh (Ratan and others 1997), but surface water bodies throughout Bangladesh are also subject to potential water-quality hazards associated with metals due to both intense chemical weathering and anthropogenic activities. The issue of surface water quality is very crucial since the government has been suggesting that inhabitants use surface water as drinking water from areas where groundwater has been severely affected by arsenic. No systematic work has been carried out to date to assess the possibility of metal contamination of surface waters in Bangladesh. The main objectives of this study are to evaluate dissolved inorganic materials transported mainly by rainwater into natural lakes and reservoirs, and to identify the environmental factors that control their hydrogeochemistry.

Study area

Bangladesh, located between latitude 20°43' and 26°36' N and longitude 88°3' to 92°40' E (Fig. 1), has a tropical climate characterized by three main seasons, (1) a dry season from November to March and a (2) rainy (monsoon) season from June to October. In addition to the summer monsoon, winter depressions originating from the Mediterranean and the so-called (3) "Nor'Westers" in April and May contribute to annual precipitation. The climatic features of Bangladesh vary from region to region and season to season. During the monsoon, the average rainfall varies from about 890 mm in the western districts (three of the study areas belong to this region) to more than 2030 mm in the south-eastern (one study area belongs to this region) and north-eastern parts of the country (Reimann 1993).

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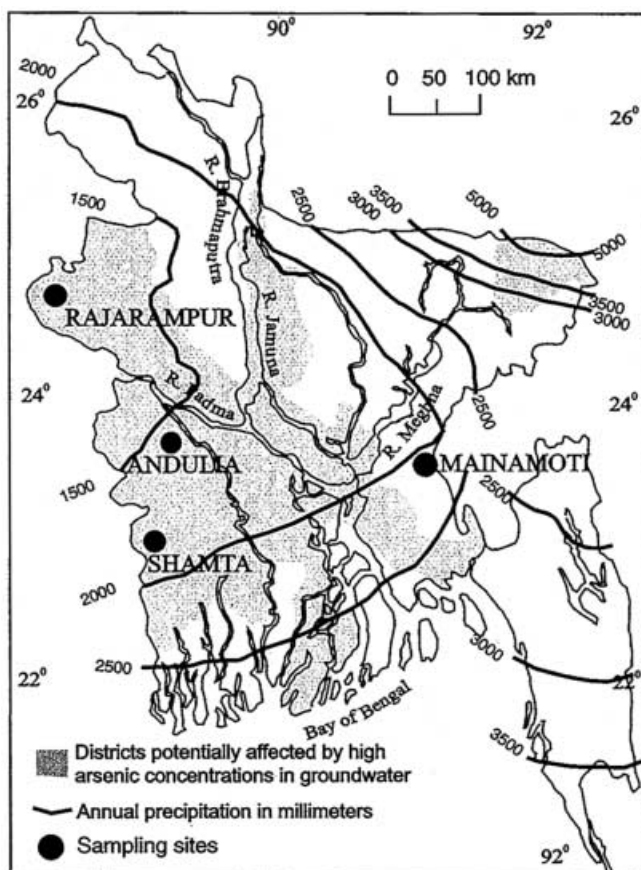


Fig. 1

Location map of the sampling sites in Bangladesh showing Rajarampur, Chapai Nawabgonj; Shamta area, Jessore; Mainamoti area, Comilla and Andulia area, Jhenidah district. Arsenic affected areas are according to Ratan and others (1997) and annual precipitation modified after Reimann (1993)

Bangladesh experiences two distinct temperature conditions, a cool and a warm season, with the former lasting from November to January and the latter from March to October. The average January temperature is about 19°C in the south and 17°C in the north. The average July temperature ranges country-wide from 27 to 29°C. The maximum temperatures recorded from the western districts, which reach 36°C in April, occasionally exceed 38°C (Islam and Miah 1981). The average annual humidity in the northeastern, eastern and southeastern districts is 78.66, 80.74 and 76.91% respectively (Statistical year-book of Bangladesh 1991). The humidity in the western, northwestern and southwestern districts is generally slightly lower.

The natural lakes in the locations studied vary from 2 to 4 km² in area and their mean depth is 4 m. The lakes receive water from both precipitation, river inflow, and runoff from surrounding agricultural lands and densely populated villages. Some of the lakes have outlets during the rainy season. Artificial reservoirs are typically 100 m in length and 70 m in width and have an average depth of around 7 m.

Bangladesh occupies the greater part of the Bengal basin and covers part of the Himalayan piedmont plain and the eastern and southeastern hill ranges of the Sylhet, Chittagong and Chittagong Hill tracts (Paul and Lian 1975). The stratigraphy of Bangladesh is somewhat problematic because most of the country is covered by thick alluvium and almost all the strata are devoid of faunal fossils. The thickest stratigraphic succession is composed of Tertiary sediments, occasionally covered by Quaternary overburden. Sandstone, siltstone, shale and claystones are the main rock types existing all over the country. The weathered sandstones, siltstones, shales and claystones in the geosynclinal foredeep region (east and southeastern part of Bangladesh) contain mainly quartz together with minor amounts of muscovite, plagioclase, K-feldspar, biotite, carbonate, epidote and clay minerals such as chlorite and illite (Islam 1996). The geochemistry of the geosynclinal foredeep region is described in detailed by Islam (1996).

Methodology

The sampling sites are marked on the location map (Fig. 1). From Rajarampur in the northwestern Chapai Nawabgonj district (alluvial sand), 3 surface water and 11 soil samples were collected. Two surface water and six soil samples were collected from Shamta village, Sharsha in the southwestern Jessore district (deltaic silt). One surface water and six soil samples were collected from the Mainamoti area in the eastern district of Comilla (Pleistocene Madhupur clay residue) and one surface water and four soil samples were collected from Andulia, Hari-nakundu in the southwestern Jhenidah district (deltaic silt). The sites were chosen to include a broad coverage and to examine surface water quality in regions impacted by high levels of arsenic in groundwater.

Both non-filtered and filtered (<0.45 µm) surface water samples were taken in 500-ml polyethylene bottles from artificial reservoirs and lakes. Filtered samples were acidified by concentrated HNO₃. Soil samples were collected in the vicinity of the surface water-sampling sites. The soil samples were taken from pits up to 2 m deep and put into polyethylene bags. The samples were dried at 70°C and sieved to the less than a 2-mm fraction. Non-filtered and filtered-acidified surface water samples were analyzed for Al, Mg, Ca, Na, K, As, Ba, Cr, Cu, Ni, Pb and Zn with an ICP-MS at the chemical laboratory of Geological Survey of Finland (GSF), Espoo.

Soil samples were analyzed according to the following procedures

1. Aqua regia-leachable elements: 12 ml of aqua regia (1 HNO₃:3 HCl) and 60 ml of water were added to dried soil samples weighing 1.2 g. Al, Mg, Ca, Na, K, As, Ba, Cr, Cu, Ni, Pb and Zn concentrations in the soil were determined after digestion in 90°C aqua regia for 1 h by ICP-AES (ISO/DIS 1146).
2. Total analyses: crushed samples were used to determine the major elemental composition by X-ray fluo-

rescence (XRF). Analyses were carried out with a Siemens SRS-X-Ray 303 As Spectrometer with standard curves based on International Rock Standards at the Institute of Electron Optics, University of Oulu.

3. Determination of pH: the pH of soil samples was determined in unbuffered 0.025 M NH_4NO_3 solution, which characterizes the type of binding capacity of the cations better than pH measured in water suspension (Räisänen and Lahermo unpublished report from 1985).

Results and discussion

The results of the chemical analyses of soil and surface water are given in Tables 1, 2 and 3 respectively.

Soil depletion and enrichment

The elemental concentrations in soil have been compared with worldwide soil analyses published by Wedepohl (1969–1979) and with unweathered core samples (sandstone, siltstone and shale) by Islam (1996). Because the core samples are not taken from the exact locations as

the soil samples, the presence of vertical variability in composition differences in concentration between the cores and soil samples less than 10% are considered insignificant. Overall, depletion of elements in soil relative to bedrock is a strong function of leaching (Fig. 2). The Mainamoti region receives the greatest amount of precipitation of the four locations and is heavily depleted in all elements except aluminium. In contrast, the Rajarampur region receives the least amount of precipitation and, in general, its soils are relatively undepleted with respect to elements. The difference in depletion suggests that the differences in precipitation are long-term and that climatic trends over Bangladesh have been persistent. In comparison with bedrock (fresh core samples of sandstone, siltstone, shale and claystone), aluminium is at most only slightly depleted in the soils of Shamta and Mainamoti. In the soils of Rajarampur and Andulia, enrichment of Al is significant. The absence of Al enrichment in the Shamta soil, the product of intense weathering and, to a lesser extent, the Mainamoti soils, is somewhat problematic. Solubility and interactions of aluminium and minerals depend on speciation, which is strongly controlled by pH (Nriagu and others 1987). In

Table 1

Selected elemental compositions of soil samples by different methods: Aqua regia leached; < *n* below the detection limit

		A l(g/kg)	Mg (mg/kg)	Ca (mg/kg)	Na (mg/kg)	K (mg/kg)	As (mg/kg)	Ba (g/kg)	Cr (mg/kg)	Cu (mg/kg)	Ni (mg/kg)	Pb (mg/kg)	Zn (mg/kg)
Rajarampur (<i>n</i> = 11)	Mean	21.33	9.93	14.52	0.14	3.85	< 10	98.87	33.54	40	18.33	16.18	57.72
Shamta (<i>n</i> = 6)	Mean	12.91	6.41	16.2	0.1	3.75	< 10	61.41	21.5	29.66	15.5	11.16	39.5
Mainamoti (<i>n</i> = 6)	Mean	17.81	0.83	0.43	< 0.05	0.8	< 10	51.71	21.33	19.83	35.25	15.3	23
Andulia (<i>n</i> = 4)	Mean	25.72	10.17	11.17	0.11	4.58	< 10	114.7	40.75	47.5	16.7	23.2	69
Typical value ^a		13	0.90%	1.40%	1%	1.40%	2	500	27.9	21.1	20	10	32

^a Typical value is according to Wedepohl (1969–1979)

Table 2

Selected elemental compositions of soil samples by different methods: total analyses by XRF; – value not available

		Al ₂ O ₃ (wt%)	MgO (wt%)	CaO (wt%)	Na ₂ O (wt%)	K ₂ O (wt%)	As (mg/kg)	Ba (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	Ni (mg/kg)	Pb (mg/kg)	Zn (mg/kg)	pH
Rajarampur (<i>n</i> = 11)	Mean	14	2.55	2.89	1.23	2.89	15.5	565.45	81.7	37.2	39.4	32.2	78.4	8.73
Shamta (<i>n</i> = 6)	Mean	10.4	1.83	3.13	1.3	2.56	13.3	440.83	59.3	30	25.7	28	64.3	8.57
Mainamoti (<i>n</i> = 6).	Mean	10.3	0.46	0.24	0.44	1.78	18.8	379.5	61.5	16	34	35.5	40.3	5.52
Andulia (<i>n</i> = 4)	Mean	14.8	2.68	2.06	1.03	3.36	20.7	588.75	96.2	47.5	48.5	37	92.5	8.23
Typical value ^a		15.1	0.90%	1.40%	1%	1.40%		500	80	21.1	42.8	17	70	–

^a Typical value is according to Islam (1996)

Table 3The average elemental compositions of surface water ($\mu\text{g/l}$, if not otherwise mentioned)

Elements	Typical value ^a	Rajarampur		Shamta		Mainamoti		Andulia	
		Filtered	Non-filtered	Filtered	Non-filtered	Filtered	Non-filtered	Filtered	Non-filtered
Al	300	–	825.82	519.5	615	517	738	1060	2050
Mg (mg/l)	4.1 ppm	–	13.45	13.71	7.98	4.98	–	5.93	5.99
Ca (mg/l)	18 ppm	–	76.93	89.3	36.45	22.8	–	29.5	29.5
Na (mg/l)	6.1 ppm	–	23.05	–	27.55	29.4	–	23.8	22.9
K (mg/l)	2.3 ppm	–	25.93	29.8	34.4	28.4	27.5	13.6	14.2
As	4	–	97.05	13.71	15.36	5.74	5.23	7.79	14
Ba	20	–	71.06	89.3	78.5	66.6	74.7	187	234
Cr	0.7	–	1.41	<0.02	5.65	0.63	9.3	0.69	2.39
Cu	3	–	7.71	5.4	18.3	4.33	18.9	8.3	11.9
Ni	0.3	–	4	5	6	3.3	5.3	3.55	6.3
Pb	3	–	8.03	4.32	16.56	5.39	9.53	8.06	13.4
Zn	15	–	18	10.6	73	19	67.8	14	20.8
No. of samples			3	1	2	1	1	1	1

^a Typical values taken from Wedepohl (1969–1979) for surface water

the study areas in Bangladesh, pH of the soil profile varies from 4.85 to 9.24, levels not favourable for Al dissolution. On the other hand, Al concentrations in surface water in the areas studied is notably higher than typical values worldwide. Dissolution may be enhanced by organic acids released due to decomposition of soil organic matter or secreted from living organisms into surface water during the rainy season (Stevenson 1967; Bruckert 1970). Magnesium (Mg) in soils is similar in its relative amount of depletion to that of Al. The depletion of the Mg from the soil profiles is due to the degradation of biotite (Islam 1996). Enrichment of Mg is due to the formation of illite. Ca has been enriched in Raja Rampur and Shamta, while it is depleted in Mainamoti and Andulia. The apparent enrichment is most likely an artifact of the difference in location between the core samples and the soil samples. Moderate to major alteration of Na-feldspar has depleted sodium in all locations. Potassium has been enriched in the three sections, with the exception of the Mainamoti area where it has been depleted due to major alteration of K-feldspar. The enrichment of potassium in the soil profile is due to the formation of illite (Islam 1996).

Arsenic (As) has been depleted from all soils with respect to fresh rock (cores of sandstone, siltstone, shale and claystone; 42.36 ppm), as has Ba. Chromium (Cr) and Ni have been depleted in all locations except in Andulia. The absence of Cr depletion at this location is due to the formation of clay minerals. Copper (Cu) has been enriched in soils from all areas due to the formation of clay minerals with the exception of Mainamoti where leaching is apparently so intense that all elements, including Cu, have been depleted. Copper availability is drastically reduced at a soil pH above 7. It is most available especially below pH 5 (Lucas and Knezek 1972). In the Mainamoti area, pH was 4.85.

The high concentrations of Cu in soil in the other three study areas are controlled by the strong affinity of this

metal to organic matter, oxides of Fe, Al and Mn, and clay minerals. In general, copper is the most strongly complexed member of the Cu, Zn, Cd, Pb group and is very commonly influenced by organic complexation. Lead is next, followed by zinc. All members of the group are potentially affected by adsorption on iron and manganese oxyhydroxides. The expected behavior of these metals in the environment can be summarized as follows: under oxidizing conditions at low pH, they are all soluble and mobile (Drever 1997). As might be expected, Pb is strongly depleted in all four study areas with respect to fresh rock.

When soil pH exceeds 7.0, the availability of Zn becomes very low. Zinc is most soluble and phyto-available under acidic conditions (Adriano 1986). The pH of soils in the study areas is generally over 8 (except in Mainamoti), suggesting that Zn is retained strongly in the soil profile. The slight apparent depletion in the Shamta area is considered within the range of error of comparison.

Water chemistry

As might be expected in a tropical region with alluvium, concentrations of all elements analyzed are higher than worldwide averages (Fig. 3). Water chemistry is clearly affected by most weathering reactions. As Bricker and Garrels (1967) emphasized, the concentration of Ca, HCO_3 and Mg in natural water systems may be controlled by dissolution of carbonate rocks. Silicate minerals are a likely major source of Na, K, H_4SiO_4 and even Ca and Mg in both ground and surface waters.

Clays in the colloidal size range in water have an effect on water quality out of proportion to their concentration (Angino 1983). Of the common clays – montmorillonite, illite and kaolinite – montmorillonite has the greatest exchange capacity. From this context, it can be assumed that the high levels of Mg, Ca, Na and K in the surface water of the study areas is the result of co-precipitation with colloidal clays formed during tropical weathering.

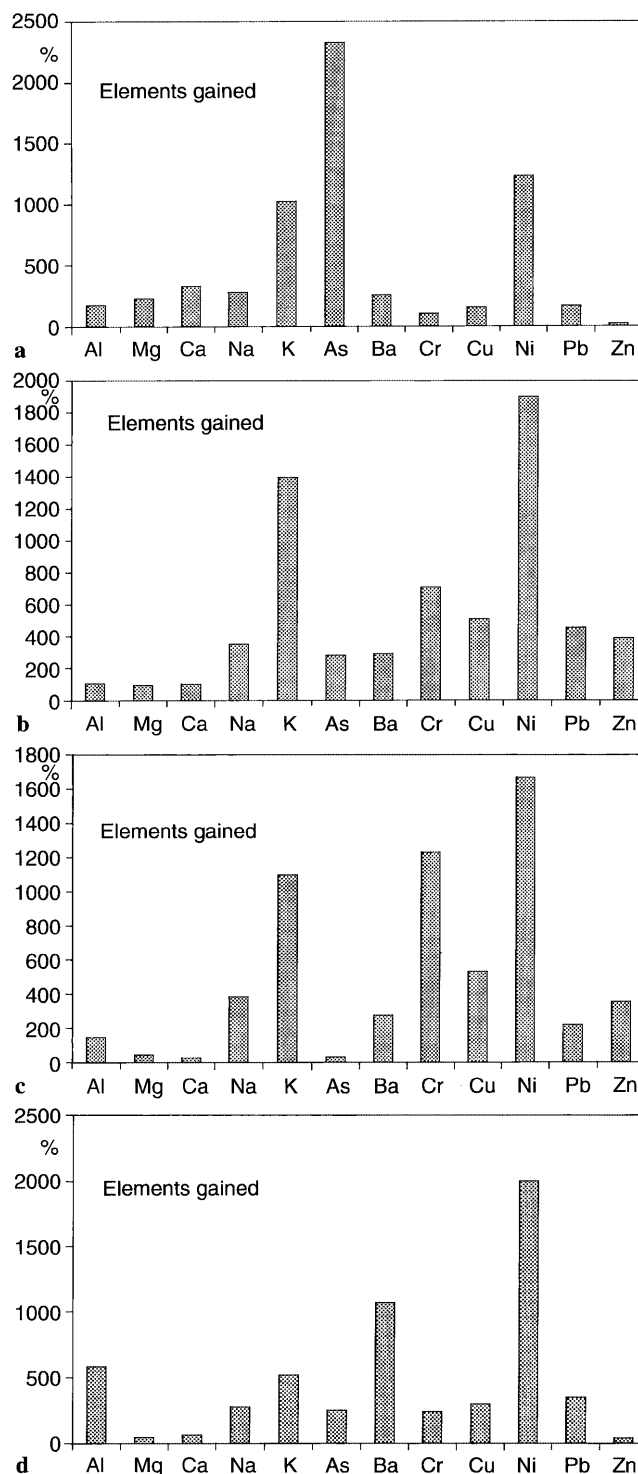
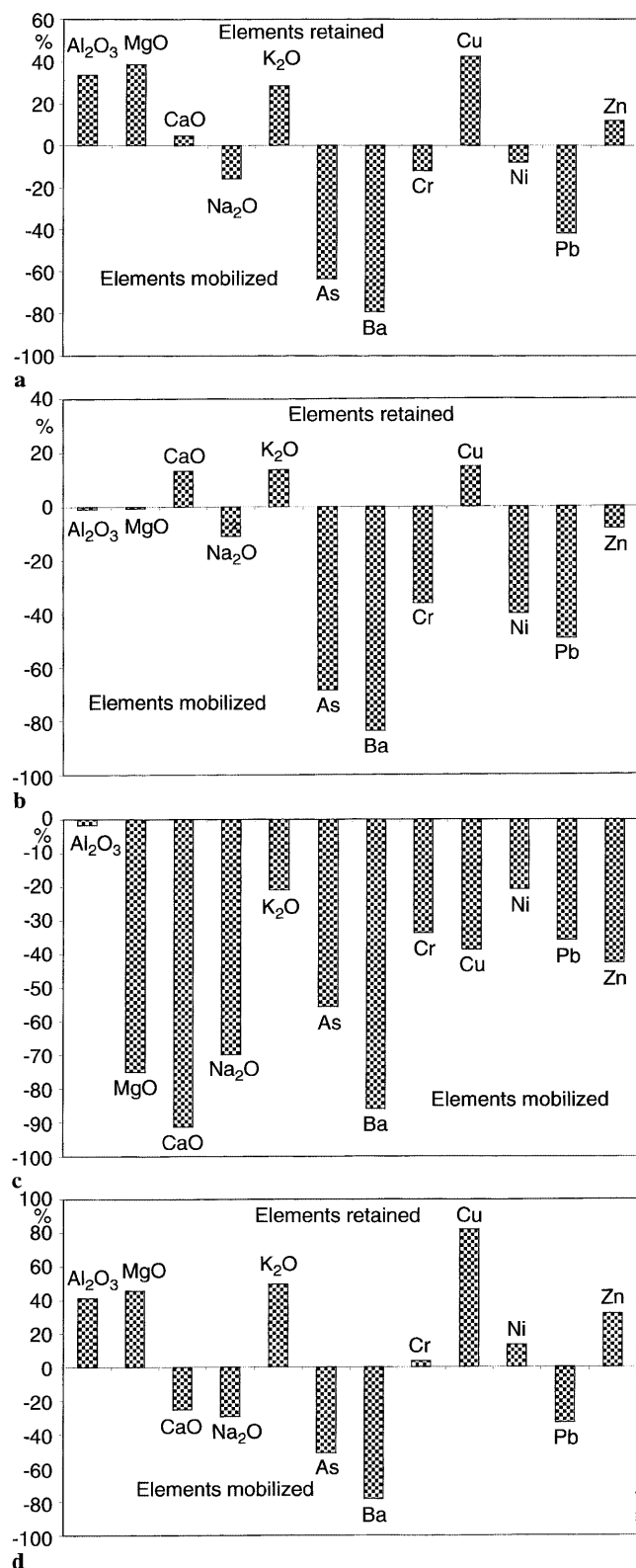


Fig. 3
Percent increase in dissolved elements in water in comparison to worldwide typical values: *a* Rajarampur, *b* Shamta, *c* Mainamoti, *d* Andulia

The dominant clay minerals in the northeastern, eastern and southeastern parts of Bangladesh are chlorite, kaolinite, illite, gibbsite and goethite (Islam 1996), and it is likely that these clay minerals are also dominant in all other

study areas. High levels of Al in surface water are slightly problematic because the soils show no evidence of depletion and may be due to the presence of suspended clay minerals that manage to pass through the water sampling filter.

With regard to trace elements, it should be emphasized that the largest proportion of the dissolved trace element content in natural water systems is usually tied up in the suspended load, typically dominated by clays. The trace metals are tied up primarily in two forms – as weathered solids of precipitates or adsorbed on the surfaces of particulate material –, such as organic debris or clays. Many metals are also present in water as hydroxo- (Angino and others 1971), carbonato- (Long and Angino 1977) and sulfato-complexes. These complexes tend to be absorbed more strongly at clay surfaces than free metal ions.

Arsenic concentrations in these surface waters are significantly higher than worldwide averages in all areas, with the exception of Mainamoti, and is particularly elevated in Rajarampur. The absence of high As in Mainamoti (which has soil with relatively high concentrations of As) is most likely due to the diluting effect of high precipitation. Solubilization of arsenic and colloidal clays in surface water are the natural sources of As.

In addition to depletion from bedrock, runoff derived from groundwater based irrigation can be a source of highly elevated As in lake water. This is particularly true for Rajarampur. The largest lake sampled contained by far the highest levels of arsenic and is surrounded by agricultural lands that are irrigated with groundwater. The correspondence suggests that most arsenic concentrations that significantly exceed drinking water standards reflect the influence of contamination by groundwater.

Chromium in surface water exceeds the worldwide average value in all of the study areas. The pH value of the soil is around 8, at which, however, chromium is not supposed to be mobile and available in surface water. Other factors affecting the mobility and availability of Cr include redox potential, oxidation state, availability of organic matter and hydrous oxides of Fe and Mn. The availability of organic matter is most likely to be the main reason for the relatively high Cr concentrations as there are manyfold higher concentrations of Cr in the non-filtered surface water as compared with the filtered water in all the areas.

Nickel in these surface waters exceeds the average worldwide value several fold. Nickel is known to complex readily with a variety of inorganic and organic ligands. The relatively high concentration of Ni is due to co-precipitation with colloidal clay fractions, and the relatively high concentration of Zn in surface water is due to the complexing effect of organic acids.

Potential health hazards

In Bangladesh, health hazards associated with arsenic in groundwater and pathogens in surface water are pervasive and well known. In the regions sampled here, surface waters also contain potential health hazards due to arsenic and aluminium. Of these, the arsenic hazard is best

understood. There are many clinical manifestations associated with arsenic poisoning, among which the most common are conjunctivitis, melanosis and hyperkeratosis. In severe cases, gangrene in the limbs and malignant neoplasm have also been observed (Das and others 1996). Epidemiological studies undertaken in southwest Taiwan in the 1960s clearly showed a relationship between high concentrations of arsenic in drinking water and skin cancer, keratosis and blackfoot disease (a type of gangrene; Tseng and others 1968; Tseng 1977). Further investigations in Taiwan have established relationships between high arsenic exposure and cancers of the bladder and other internal organs (Chen and others 1988, 1992). In the regions studied, surface waters are below or are just above WHO (1996) drinking-water standards (10 µg/l), except in the Rajarampur lakes that receive irrigation-derived groundwater. This suggests that surface waters that receive little irrigation water will be relatively free of arsenic in comparison to groundwater.

Aluminium concentrations in all of the surface waters greatly exceed that of WHO (1996) drinking-water standards (200 µg/l) in all areas studied. The health impact of such high concentrations is not well understood and, indeed, even average worldwide values of surface waters exceed WHO drinking-water standards. Increased aluminium ingestion is believed to relate to a CNS syndrome termed “dialysis dementia” (Alfrey and others 1976). It has been hypothesized (Crapper and others 1973; Caster and Wang 1981) that Alzheimer’s disease may be related to accumulation of Al in critical parts of the brain. The greater bioavailability of Al in drinking water may render it a relatively more harmful source than in food (Martyn and others 1989).

Conclusion

While leaching of soils is intense throughout the country of Bangladesh, there is variability in climate that causes variability in weathering rates. The current climatic conditions, i.e. significant increases in precipitation from west to east, are reflected in the rates of weathering and this suggests that the country’s current climatic patterns are long term. The major affected minerals during the weathering processes are Na-feldspar, Ca-feldspar, K-feldspar, biotite and muscovite. Spatial variability exists in the rate of mobilization/retention of both major and trace elements that is independent of climate and these differences suggest that local controls are also important. Lake and reservoir waters have been generally enriched in all measured elements, sometimes independently of the degree of depletion or enrichment in the soil profile. The reasons for the high degree of dissolved solids include: (1) leaching of the elements and transportation into surface water; and (2) dissolution of the elements in ionic form and co-precipitation or co-existence with the suspended colloidal clay fraction.

Arsenic and aluminum in the surface waters are at concentrations that approach and commonly exceed World Health Organization (WHO) drinking-water recommendations. The key environmental concern is arsenic, which is particularly high in reservoirs and lakes that receive significant amounts of irrigation drainage water of sub-surface origin. Groundwater in Bangladesh has natural arsenic levels that exceed WHO recommendations throughout much of the country. Recently, the WHO and the Bangladesh government have recommended that citizens avoid contaminated groundwater and use surface water as a substitute. Our studies show that (in addition to potential exposure to pathogens such as cholera) the use of surface water will not always allow the public to avoid arsenic exposure. The high rates of weathering and contamination of surface waters by arsenic laden irrigation water can cause reservoirs and lakes to have dangerous concentrations of arsenic. Public use of reservoirs and lakes for drinking should not be allowed unless those waters have been tested and found to be relatively free of arsenic.

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