For drinking water, the people of Bangladesh used to rely on surface water, which was often contaminated with bacteria causing diarrhea, cholera, typhoid, and other life-threatening diseases. To reduce the incidences of these diseases, millions of tubewells were installed in Bangladesh since independence in 1971. This recent transition from surface water to groundwater has significantly reduced deaths from waterborne pathogens; however, new evidence suggests disease and death from arsenic (As) and other toxic elements in groundwater are affecting large areas of Bangladesh. In this evaluation, the areal and vertical distribution of As and 29 other inorganic chemicals in groundwater were determined throughout Bangladesh. This study of 30 analytes per sample and 112 samples suggests that the most significant health risk from drinking Bangladesh’s tubewell water is chronic As poisoning. The As concentration ranged from < 0.0007 to 0.64 mg/L, with 48% of samples above the 0.01 mg/L World Health Organization drinking water guideline. Furthermore, this study reveals unsafe levels of manganese (Mn), lead (Pb), nickel (Ni), and chromium (Cr). Our survey also suggests that groundwater with unsafe levels of As, Mn, Pb, Ni, and Cr may extend beyond Bangladesh’s border into the four adjacent and densely populated states in India. In addition to the health risks from individual toxins, possible multimetal synergistic and inhibitory effects are discussed. Antimony was detected in 98% of the samples from this study and magnifies the toxic effects of As. In contrast, Se and Zn were below our detection limits in large parts of Bangladesh and prevent the toxic effects of As. Key words: arsenic, arsenic contamination, Bangladesh arsenic, drinking water, environmental toxicity, health risks, metal carcinogenicity, multimetal analysis, multimetal effect, toxic metals. Environ Health Perspect 110:1147–1153 (2002). [Online 20 September 2002] http://ehpnet1.nih.gov/docs/2002/110p1147-1153/frisbie/abstract.html

Geographic, Demographic, and Economic Overview of Bangladesh

The People’s Republic of Bangladesh is a developing country overburdened with an enormous population, severe poverty, common illiteracy, and frequent natural disasters. It is located at one of the largest river deltas in the world: The Ganges, Brahmaputra, and Meghna rivers flow through Bangladesh to the Bay of Bengal. Very little of the country is more than 12 m (40 feet) above sea level, and in a normal monsoon season one-third of its cultivated land is flooded (I). Bangladesh has 127 million people (2) living on 144,000 km² (I); this would be equivalent to one-half the population of the United States living in an area the size of Wisconsin. The infant mortality rate is 58 per 1,000 live births (2). There is one doctor per 5,200 people; by comparison, the United Kingdom has one doctor per 650 people (I). The adult literacy rate is 63% for men and 48% for women. The average annual income is equivalent to US$370 per capita (2). The life expectancy is 55 years (I).

Bangladesh is an agricultural 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 and vegetables in the short, dry winter from November to February. Bangladesh is the world’s leading producer of jute, a strong natural fiber used in the carpet and sacking industries. The principal exports of Bangladesh from smallest to largest are garments, jute and its products, shellfish, tea, and leather (I).

Project Overview

Much of Bangladesh’s surface water is microbially unsafe to drink. Since independence in 1971, between 8 million and 12 million tubewells have been installed to supply microbially safe drinking water to the people of Bangladesh. Today, 97% of Bangladeshis drink well water (3,4). Unfortunately, vast areas of this 127 million-person country contain groundwater with arsenic (As) concentrations above the World Health Organization (WHO) drinking water guideline of 0.01 mg/L (5,6). Chronic As poisoning attributed to groundwater ingestion was first diagnosed in 1993. By 1999, a total of 2,953 cases of chronic As poisoning were identified in Bangladesh (7); however, most of this country remains unsurveyed, and the actual number of cases is expected to be in the tens or hundreds of thousands (8). These diagnoses include melanosis, leukomelanosis, keratosis, hyperkeratosis, nonpitting edema, gangrene, and skin cancer (9).

The 1997 U.S. Agency for International Development (USAID) field program produced the first national-scale map of As concentration in Bangladesh’s tubewell water (10,11). This map indicates that approximately 45% of Bangladesh’s area contains groundwater with As concentrations greater than the 0.05 mg/L Bangladesh national drinking water standard (10). The principal source of As in Bangladesh’s groundwater is geologically deposited sediments. In particular, the major sources might be the reductive dissolution of nonpyrite iron (Fe) or nonpyrite phosphate minerals and the anion exchange of sorbed arsenite or arsenate (10,11).

In addition, the 1997 USAID field program discovered that many of the 127 million people in Bangladesh may be drinking unsafe levels of toxic metals other than As (10,11). At least 27% of the samples contained an analytical interference to the 1,10-phenanthroline methods for measuring Fe(II) and total Fe. This interference was observed from suppressed matrix spike recovery (34%) during the measurement of Fe(II) and from improper color development during the measurement of total Fe. This interference could not be further characterized during the 1997 USAID field program; however, the literature suggested that it resulted from one or more toxic nonarsenic metals in these drinking water samples (12,13). In response to this discovery, we assessed the hypothesis that Bangladeshi
are exposed to toxic metals other than As in their drinking water during our 1998–1999 field program. In this assessment, the concentrations of several analytes (Ag, Al, As, Ba, Bi, Ca, Cd, Co, Cr, Cs, Cu, F, Fe, H+, K, Mg, Mn, Mo, Ni, Pb, Rb, S, Sb, Si, Se, Sr, Ti, V, W, and Zn) in tubewell water were mapped on a national scale (Figure 1). These analytes were selected based on their toxicity and potential to be the analytical interference observed during the 1997 USAID field program. This exposure assessment of As and other toxic metals in Bangladesh’s drinking water is reported here for the first time.

Furthermore, the contention that Bangladeshis are exposed to toxic metals other than As was strengthened by the finding of severe melanosis, keratosis, skin cancer, and other symptoms of chronic As poisoning especially among children (14,15). This observation was the first indication that multimetal health effects might be involved. Therefore, we assessed the hypothesis that Bangladeshis are exposed to antimony (Sb), a metal that magnifies chronic As poisoning (16), during our 1998–1999 field program. Conversely, we also assessed the hypotheses that Bangladeshis are not exposed to selenium (Se) or zinc (Zn), metals that inhibit chronic As poisoning (17,18). This exposure assessment of metals that affect As toxicity is reported here for the first time.

Methods
Groundwater samples were collected from 112 tubewells throughout Bangladesh during 20 December 1998 to 18 January 1999. One sample was collected from each tubewell. All of these samples were analyzed for Ag, Al, As, Ba, Bi, Ca, Cd, Co, Cr, Cs, Cu, F, Fe, H+, K, Mg, Mn, Mo, Ni, Pb, Rb, S, Sb, Si, Se, Sr, Ti, V, W, and Zn.

The sampled tubewells were distributed as evenly throughout Bangladesh as possible, given limited access because of the country’s extensive river delta and developing network of roads (Figure 1). Random samples were typically collected by traveling on roads and across rivers for 20–30 km, stopping at a random location, and collecting groundwater from the first tubewell found. The latitude and longitude of these tubewells were determined using a Garmin Global Positioning System 12-channel Personal Navigator (Garmin International, Inc., Olathe, KS, USA). The accuracy of this instrument was approximately 15 m. The district, thana (a collection of villages or section of a city under the jurisdiction of a single police station), and village of all sampled tubewells were documented. The owner and the owner’s reported depth of each sampled tubewell were recorded.

Established collection, preservation, and storage methodologies were used to ensure that each sample was representative of groundwater quality (12,19). Accordingly, all sampled tubewells were purged by vigorous pumping for 10 min immediately before sample collection. All samples were collected directly into polyethylene bottles and were not filtered. Samples were analyzed for pH immediately after collection by glass electrode or pH paper, preserved by acidification to pH < 2 with 18.6% (weight/weight) HNO₃ and stored in ice-packed coolers. The temperature of all stored samples was maintained at 0°C to 4°C until immediately before analysis.

All samples were analyzed for Mg, Al, Ca, V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Mo, Ag, Cd, Sb, Cs, Ba, W, Ti, Pb, and Bi at the Laboratoire Pierre Süe, Centre National de la Recherche Scientifique in Gif-sur-Yvette, France. These samples were analyzed by inductively coupled plasma mass spectrometry (ICP/MS) with a Fisons PlasmaQuad PQ+ spectrometer (Fisons/VG Analytical, Manchester, UK). Multielement standard solutions were prepared from solutions certified by SPEX CertiPrep, Inc. (Metuchen, NJ). Monoelemental SPEX CertiPrep-certified solutions of Be, In, and Re were used as internal standards. All samples were analyzed twice by ICP/MS. First, undiluted samples were analyzed for trace elements. Then samples were diluted 10 times using ultrapure water and acidified to pH 2 with Prolabo Normalat I grade nitric acid (Prolabo, Paris, France) for the determination of major elements (20,21).

All samples were analyzed for Si, S, K, and Fe at the Université de Bordeaux 1, Laboratoire de Chimie Nucléaire Analytique et Bioenvironnementale, in Gradignan, France. These samples were analyzed by particle-induced X-ray emission Rutherford backscattering spectrometry with the 4 MV Van de Graaff accelerator and nuclear microprobe beamline (22,23). Multielement standard solutions were prepared from Sigma-certified solutions (Sigma Aldrich Chimie, Lyon, France).

Figure 1. Map of Bangladesh showing the locations where groundwater samples were collected from tubewells during the 1998/1999 field program. The Padma, Bramaputra, Jamuna, and Meghana Rivers are outlined.
in Bangladesh’s groundwater (8,9). In addition, the relatively small and negative correlation between As and depth \( (r = -0.13\); Table 1) supports the 1997 hypothesis that drilling deeper tubewells can access drinking water with significantly lower As concentrations approximately 20% of the time (10,11).

Of these analytes, the concentrations of As, manganese (Mn), lead (Pb), nickel (Ni), and chromium (Cr) exceeded WHO \((5,6)\) or U.S. Environmental Protection Agency \((U.S. EPA) \) \((24,25)\) health-based drinking water criteria (Table 2). Maps showing the extent of As, Mn, Pb, Ni, and Cr in groundwater were drawn using kriging (Figures 2–6), a standard geostatistical technique (26).

This map of As concentration (Figure 2) agrees with all three other national-scale surveys of randomly selected tubewells in Bangladesh \((10,11,27,28)\). This agreement suggests that our national-scale maps of Mn, Pb, Ni, and Cr (Figures 3–6) are valid as well. Our map of As concentration (Figure 2) agrees with that produced by the 1997 USAID field program \((10,11)\); however, the 1997 map was based on a 0.03 mg/L detection limit, which was not sensitive enough to delineate the WHO drinking water guideline. In contrast, Figure 2 allows delineation of the 0.01 mg/L WHO and 0.05 mg/L Bangladesh criteria for As in drinking water because the detection limit for As by ICP/MS was 0.0007 mg/L. Figure 2 indicates that approximately 49% of Bangladesh’s area contains ground water with As concentrations greater than the WHO drinking water guideline. These results agree with the estimated 44% of Bangladesh’s area having unsafe levels of As reported by Karim et al. in 1997 \((27)\). In addition, our results agree with an unreviewed national-scale study reported on the Internet by the British Geological Survey/Government of Bangladesh Department of Public Health Engineering (BGS/DPHE) team \((28)\). The BGS/DPHE survey used kriging based on 3,534 samples to estimate that 57 million Bangladeshis are drinking water with As concentrations above the WHO drinking water guideline, a number similar to our survey estimate of 60 million.

It is very important to recognize that the 0.01 mg/L WHO drinking water guideline for As is based on a \(6 \times 10^{-6}\) excess skin cancer risk-based drinking water criteria for Ag, Al, Bi, Ca, Co, Cs, Fe, K, Mg, Pb, total Si, Sr, V, W, and Zn.

| Table 1. Correlation coefficient matrix for tubewell water parameters. |
|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| pH   | F    | Mg   | Al   | Si   | S    | K    | Ca   | V    | Mn   | Fe   | Co   | Ni   | Cu   | Cr   | As   | Se   | Sr   | Rb   | Rs   | Mo   | Cd   | Sb   | Ba   | W    | Ti   | Depth |
| 1.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 | 0.00 | 1.00 |

| Table 2. Risk-based drinking water criteria and the percentage of area exceeding these criteria. |
|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| Element   | Risk-based drinking water criteria (mg/L) | Percentage of Bangladesh’s area exceeding criteria |
|-----------|---------------------------------------------|---------------------------------------------|
| As        | 0.010 0.010 49 49                           |                                              |
| Ba        | 0.003 0.050 0.00 1.00                       |                                              |
| Cd        | 0.003 0.050 0.00 1.00                       |                                              |
| Cr        | 0.005 0.100 < 1 0                           |                                              |
| Cu        | 2.000 1.300 0.00 1.00                       |                                              |
| F         | 1.500 4.000 0.00 1.00                       |                                              |
| Mo        | 0.500 None 50 NA                            |                                              |
| Pb        | 0.020 0.100 < 1 0                           |                                              |
| Sn        | 0.005 0.006 0.00 1.00                       |                                              |
| Se        | 0.010 0.050 0.00 1.00                       |                                              |
| Ti        | None 0.002 NA                               |                                              |

NA, not applicable.

*The WHO \((5,8)\) and U.S. EPA \((24,25)\) have not established risk-based drinking water criteria for Ag, Al, Bi, Ca, Co, Cs, Fe, K, Mg, Pb, total Si, Sr, V, W, and Zn. 
risk for human males in Taiwan (29), which is 60 times higher than the $1 \times 10^{-5}$ 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 (5,6). There is sufficient evidence from human epidemiologic studies linking increased mortality from liver, kidney, bladder, and lung cancers to drinking As-contaminated water; however, this relatively new discovery is not used to calculate the drinking water standard for As due to a lack of dose–response data (30,31). Furthermore, a thorough review of As and public health recommends a zero exposure level for As in drinking water (31). In our study, the As concentration ranged from < 0.0007 to 0.64 mg/L. Arsenic was measured at or above its 0.0007 mg/L detection limit in 84% of the samples. Arsenic exceeded the 0.01 mg/L WHO drinking water guideline in 48% of the samples.

The most important finding of our national-scale study is that approximately 50% of Bangladesh’s area may contain groundwater with Mn concentrations greater than the WHO health-based drinking water guideline (5,6). Our study also indicates that Pb (3% of Bangladesh’s area), Ni (< 1% of Bangladesh’s area), and Cr (< 1% of Bangladesh’s area) concentrations exceed WHO health-based guidelines (5,6). These results are supported by the BGS/DPHE’s national-scale study based on between 20 and 3,530 samples (28), which suggests that 35%, < 1%, 0%, and 0% of Bangladesh’s tubewells exceed the WHO health-based drinking water guidelines for Mn, Pb, Ni, and Cr, respectively. In addition, the BGS/DPHE study suggests that 5.3%, 0.3%, and an unspecified percentage of Bangladesh’s tubewells exceed the WHO health-based drinking water guidelines for boron, barium, and molybdenum, respectively. Moreover, the BGS/DPHE study suggests that 12–50% of Bangladesh’s tubewells exceed the WHO health-based drinking water guideline for uranium.

In our study, Mn exceeded the 0.5 mg/L WHO drinking water guideline in 37% of the samples. The maximum concentration of Mn was 2.0 mg/L. Despite the relatively poor –0.13 correlation coefficient between As and Mn (Table 1), 35% of the samples that exceeded the WHO drinking water guideline for As also exceeded the WHO drinking water guideline for Mn. The areas where the WHO drinking water guidelines were exceeded for both As and Mn can be estimated by superimposing Figures 2 and 3. Similarly, 2% of the samples that exceeded the WHO drinking water guideline for As also exceeded the WHO drinking water guideline for Pb ($r = 0.01$; Table 1). Likewise, 2% of the samples that exceeded the WHO drinking water guideline for As also exceeded the WHO drinking water guideline for Ni ($r = -0.02$; Table 1). Correspondingly, 2% of the samples that exceeded the WHO drinking water guideline for As also exceeded the WHO drinking water guideline for Cr ($r = 0.92$; Table 1).

The above findings raise serious concerns relating to environmental health issues caused by multimetal effects. The As, Mn, Pb, Ni, and Cr in Bangladesh’s drinking water are associated with known health risks. Arsenic is classified as a “human carcinogen” based on sufficient epidemiologic evidence (30). Manganese is a known mutagen (32). The accumulation of Mn may cause hepatic encephalopathy (33). Moreover, the chronic ingestion of Mn in drinking water is associated with neurologic damage (34). The 0.5 mg/L WHO drinking water guideline for Mn was calculated using human exposures in Japan and Greece and studies of various laboratory animals where neurotoxic and other effects were observed (29). Lead is a “possible human carcinogen” because of inconclusive evidence of human and sufficient evidence of animal carcinogenicity (29). In addition, Pb

**Figure 2.** Contour map of As concentration (mg/L) in tubewell water from the 1998/1999 field program. The WHO health-based drinking water guideline is 0.01 mg/L (5,6).

**Figure 3.** Contour map of Mn concentration (mg/L) in tubewell water from the 1998/1999 field program. The WHO health-based drinking water guideline is 0.5 mg/L (5,6).
also causes many noncancerous disorders in humans (35). The 0.01 mg/L WHO drinking water guideline for Pb was calculated using the lowest measurable retention of Pb in the blood and tissues of human infants (29). Nickel is a “probable human carcinogen” (36). The 0.02 mg/L WHO drinking water guideline for Ni was calculated using no observed adverse effects level (NOAEL) and lowest observed adverse effects level (LOAEL) in studies of laboratory rats (37). The International Agency for Research on Cancer categorizes Cr(VI) as “carcinogenic to humans” and Cr(III) as “not classifiable” (38); however, the U.S. EPA lists total Cr in drinking water as having “inadequate or no human and animal evidence of carcinogenicity” (24). The WHO states that 0.05 mg/L drinking water guideline for total Cr is unlikely to cause significant health risks (29).

Figures 2–6 also suggest that groundwater with unsafe levels of As, Mn, Pb, Ni, and Cr extend beyond Bangladesh’s borders into the four adjacent and densely populated Indian states of West Bengal, Assam, Meghalaya, and Tripura. West Bengal has over two million Indians drinking from tubewells with unsafe levels of As and 200,000 suffering from chronic As poisoning from groundwater in West Bengal and neighboring Bangladesh should not be repeated (40). Therefore, the groundwater used for drinking in the adjacent and densely populated Indian states of West Bengal, Assam, Meghalaya, and Tripura should be immediately tested to determine if it is safe.

The severity of chronic As poisoning in Bangladesh might be magnified by exposure to Sb. Antimony in drinking water has been reported to modulate the toxicity of As (16). Antimony was measured at or above its 0.0015 µg/L detection limit in 98% of the samples from this study. Arsenic was measured at or above its 0.7 µg/L detection limit in 84% of the samples from this study. Despite the relatively poor −0.05 correlation coefficient between As and Sb (Table 1), 97% of the samples with detectable concentrations of As had detectable concentrations of Sb. The concentration of Sb ranged from 0.0015 to 1.8 µg/L and did not exceed its 5 µg/L WHO health-based drinking water guideline. However, this guideline is based on the toxicity of exclusively ingesting Sb, not the influence of Sb on chronic As poisoning. The 5 µg/L WHO drinking water guideline for Sb was calculated using the LOAEL for decreased longevity, altered blood glucose levels, and altered blood cholesterol levels in laboratory rats (29). It is possible that these otherwise safe levels of Sb may cause a magnification of As toxicity. Humic substances might also magnify As toxicity (18) and were measured at relatively high concentrations in tubewells from Faridpur, one of Bangladesh’s most severely affected districts (41).

The WHO and U.S. EPA have not established health-based drinking water guidelines for Fe; however, approximately 69% of Bangladesh’s area may exceed the WHO and U.S. EPA’s 0.3 mg/L secondary criteria (24,25) (Figure 7). In addition, As and Fe have a positive 0.25 correlation coefficient (Table 1). Moreover, the As contaminated water has relatively high Fe concentrations; for example, drinking water samples exceeding 0.05 mg/L As have an average of 8.0 mg/L Fe. The potential health effects of these high Fe concentrations on chronic As poisoning are unknown. However, there are reports suggesting high body Fe stores and dietary intakes of Fe are associated with hepatocellular carcinoma in humans (42) and mammary carcinogenesis in female Sprague-Dawley rats (43). In addition, As causes the release of Fe from ferritin, the generation of activated oxygen species, and DNA damage (44).

In contrast, Se is an essential element that prevents the cytotoxic effects of As (17). Se...
was not found above its 3 µg/L detection limit in 93% of the drinking water samples from this study. Significantly, 92% of the samples with detectable concentrations of As did not have detectable concentrations of Se. This general absence of Se and presence of As in drinking water is supported by the relatively poor 0.06 correlation coefficient for these elements (Table 1). The maximum concentration of Se was 5.4 µg/L. Additionally, Zn is an essential element that promotes the repair of tissues damaged by As (18). Zinc was not found above its 0.7 µg/L detection limit in 21% of the drinking water samples from this study. Importantly, 18% of the samples with detectable concentrations of As did not have detectable concentrations of Zn ($r = 0.17$; Table 1). If the sample did not have a detectable concentration of Zn, then the sample did not have a detectable concentration of Se ($r = -0.02$; Table 1). Furthermore, Bangladesh’s agricultural soils might be Se deficient and are often Zn deficient (45); therefore, it is possible that the apparent absence of these essential nutritive elements in drinking water and possibly food may cause a magnification of As toxicity.

**Conclusions**

The catastrophic health crisis caused by drinking metal-contaminated groundwater in Bangladesh affects tens of millions of people and requires urgent attention. Our study suggests that 49% of Bangladesh’s area has As concentrations above WHO guidelines. Similarly, 50%, 3%, < 1%, and < 1% of Bangladesh’s area exceeds WHO guidelines for Mn, Pb, Ni, and Cr, respectively. Our estimate that 60 million Bangladeshi are drinking water with As concentrations above the WHO health-based guideline agrees with the BGS/DPHE’s 57 million-person estimate. In addition, our estimate that 50% of Bangladesh’s area exceeds the WHO health-based guideline for Mn is comparable with the BGS/DPHE’s estimate. Similarly, B, Ba, Cr, Mo, Ni, Pb, and U were discovered at concentrations above WHO health-based guidelines in relatively small areas of Bangladesh by our team, the BGS/DPHE team, or both teams (28). Considering the population of this country and that 97% of its people drink from wells (2,4), these data suggest that tens of millions of Bangladeshis are drinking water with unsafe levels of As, Mn, B, Ba, Cr, Mo, Ni, Pb, or U. Arsenic in Bangladesh’s tubewell water was found to be the most significant health risk. Drinking water with safe levels of As could be supplied to tens of millions by the integrated use of groundwater monitoring, drilling deeper tubewells, and appropriate treatment systems (10,11,39). However, mitigation efforts should not be limited to As; the health risks from other toxins in this region’s drinking water must also be addressed. Figures 2–6 will allow scientists, policy makers, and aid workers to initiate a rapid action program to focus in more detail on the areas with the highest concentrations of As, Mn, Pb, Ni, and Cr as we have documented in these maps. Strategies to supply this region with drinking water that has safe levels of As, Mn, Pb, Ni, Cr, other toxic elements, and agents that magnify chronic As poisoning must be studied, developed, and quickly implemented.

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