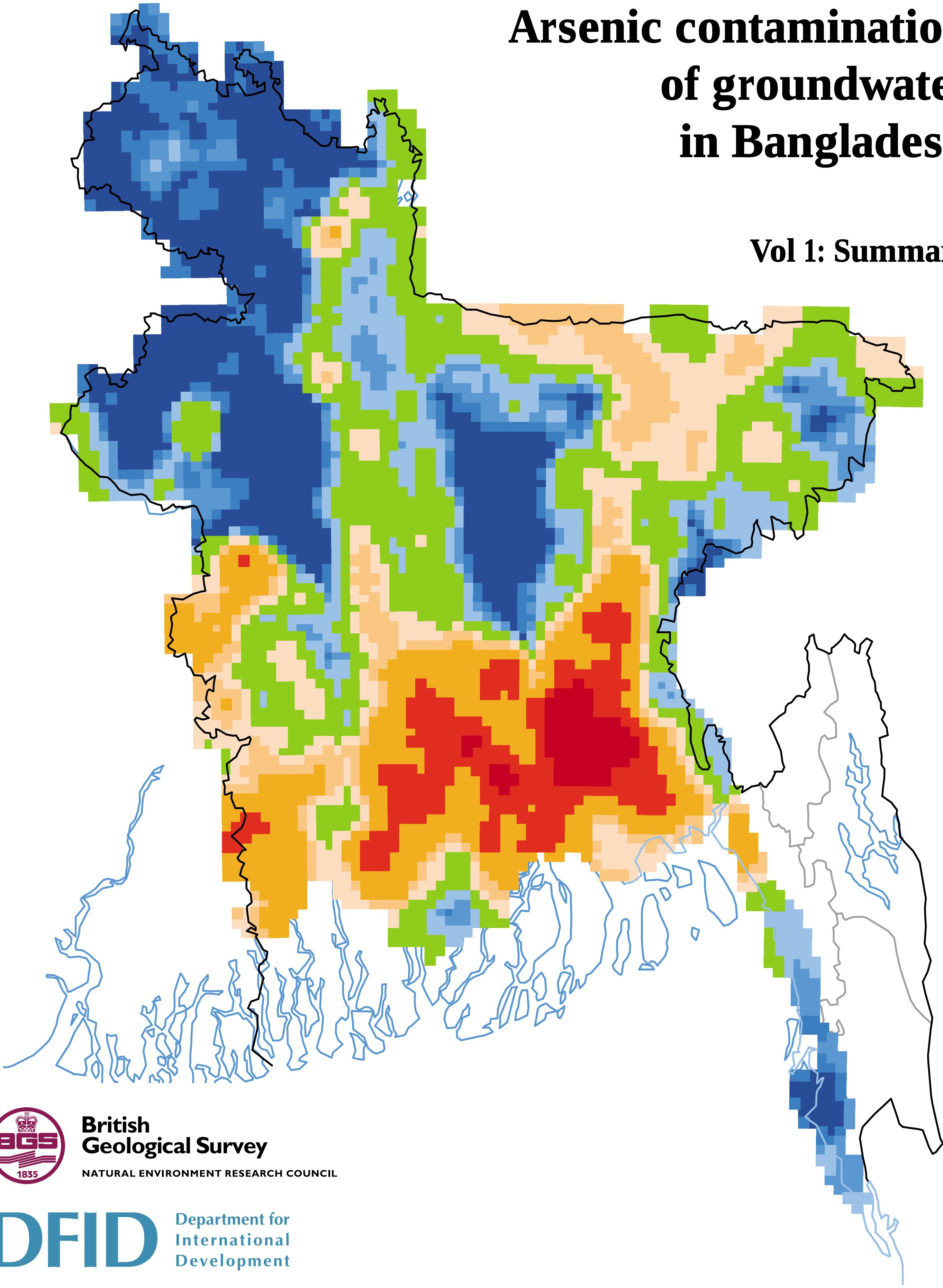
Arsenic contamination of groundwater in Bangladesh

Vol 1: Summary



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D G Kinniburgh and P L Smedley (Editors)

February 2001

The full report comprises four volumes:

- Volume 1. Summary
- Volume 2. Final report
- Volume 3. Hydrochemical atlas
- Volume 4. Data compilation

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Cover Illustration
Map of Bangladesh showing the regional distribution of arsenic in groundwater found during the National Hydrochemical Survey

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Executive summary

A survey of well waters (n=3534) from throughout Bangladesh, excluding the Chittagong Hill Tracts, has shown that water from 27% of the 'shallow' tubewells, that is wells less than 150 m deep, exceeded the Bangladesh standard for arsenic in drinking water (50 µg L^{-1}). 46% exceeded the WHO guideline value of 10 µg L^{-1}. Figures for 'deep' wells (greater than 150 m deep) were 1% and 5%, respectively. Since it is believed that there are a total of some 6–11 million tubewells in Bangladesh, mostly exploiting the depth range 10–50 m, some 1.5–2.5 million wells are estimated to be contaminated with arsenic according to the Bangladesh standard. 35 million people are believed to be exposed to an arsenic concentration in drinking water exceeding 50 µg L^{-1} and 57 million people exposed to a concentration exceeding 10 µg L^{-1}.

There is a distinct regional pattern of arsenic contamination with the greatest contamination in the south and south-east of the country and the least contamination in the north-west and in the uplifted areas of north-central Bangladesh. However, there are occasional arsenic 'hot spots' in the generally low-arsenic regions of northern Bangladesh. In arsenic-contaminated areas, the large degree of well-to-well variation within a village means that it is difficult to predict whether a given well will be contaminated from tests carried out on neighbouring wells.

The young (Holocene) alluvial and deltaic deposits are most affected whereas the older alluvial sediments in the north-west and the Pleistocene sediments of the uplifted Madhupur and Barind Tracts normally provide low-arsenic water. Water from dug wells from a highly contaminated hot spot in northern Bangladesh was also found to normally comply with the WHO guideline value for arsenic and so could be a possible source of low-arsenic water, given the appropriate sanitary precautions.

The arsenic is of natural origin and is believed to be released to groundwater as a result of a number of mechanisms which are poorly understood. This release appears to be associated with the burial of fresh sediment and the generation of anaerobic (oxygen-deficient) groundwater conditions. It probably occurred thousands of years ago. The arsenic is thought to be desorbed and dissolved from iron oxides which had earlier scavenged the arsenic from river water during their transport as part of the normal river sediment load. We call this the iron oxide reduction hypothesis. Natural variations in the amount of iron oxide at the time of sediment burial may be a key factor in controlling the distribution of high arsenic groundwaters. Limited evidence suggests that the isolated arsenic hot spots found in northern Bangladesh occur in areas containing sediments particularly rich in iron oxides, and their accompanying adsorbed arsenic load.

While there is evidence for sulphide minerals in some of the sediments, and in some cases indirect evidence for their oxidation, there is no support for the 'pyrite oxidation' hypothesis in which pyrite oxidation in the zone of water table fluctuation is assumed to release arsenic and ultimately to be responsible for the groundwater arsenic problem. There is no evidence to support the proposition that the groundwater arsenic problem is caused by the recent seasonal drawdown of the water table due to a recent increase in irrigation abstraction.

Monitoring of groundwaters at two-weekly intervals at a number of sites, and at different depths, has shown some variation with time but there is as yet no convincing evidence for seasonal changes. Dramatic changes in contamination are not expected within such a short timescale. A monitoring programme should be undertaken at a range of sites to monitor possible long-term changes. In the three contaminated areas studied in most detail, the arsenic concentration increases most rapidly between 10–20 m below ground level.

While arsenic is the single greatest problem in Bangladesh groundwaters, other elements of concern from a health point of view, are manganese, boron and uranium. Some 35% of the groundwaters sampled exceeded the WHO guideline value for manganese (0.5 mg L^{-1}). The spatial pattern of the arsenic and manganese problem areas was significantly different and only 33% of shallow well waters complied with the WHO guideline values for both arsenic and manganese.

It is unlikely that the regional pattern of arsenic contamination revealed by this, and other studies, will substantially change as more testing refines the picture. There is therefore an urgent need for the arsenic mitigation programme to provide, as a priority, a safe source of drinking water in the worst-affected areas which have now been clearly identified.

Deep groundwaters, where available, appear to offer a long-term source of safe drinking water. Experience gained so far indicates that the great majority of these would not only pass the current Bangladesh standard for arsenic but would pass all other existing national and international standards and guidelines for arsenic. The likelihood of a manganese exceedance is also much lower in these deep groundwaters. Most of the deep groundwaters tested in our surveys were from the southern coastal region where the shallow groundwaters are affected by salinity and these deep groundwaters may not be typical of those from elsewhere in Bangladesh. Therefore the nationwide availability and sustainability of this resource needs to be established in terms of quality, quantity and sustainability. The possible impact of the large-scale abstraction of irrigation water on the deep aquifer also needs to be considered.

From a worldwide perspective, drinking water derived from aquifers showing similar characteristics to those of the Bengal Basin should be considered 'at risk' and need to be systematically tested for arsenic.

Main findings

1. BACKGROUND TO THE STUDY

At the time of the inception of this project (mid 1996), the scale of the groundwater arsenic problem in Bangladesh was largely unknown although the first indications of a problem were apparent from a small number of well water analyses from western Bangladesh. These had been undertaken in response to the well-publicised finding of an extensive groundwater arsenic problem in neighbouring West Bengal. In view of the seriousness of the potential problem and the rapidly-developing awareness of its likely scale, the project was split into two Phases: Phase I, a Rapid Investigation Phase (6 months) was designed to make a rapid assessment of the scale and nature of the problem by reviewing existing data and undertaking a survey of what were then believed to be the worst-affected parts of the country. During Phase I, Mott MacDonald Ltd (UK) led the Bangladesh input to the project. Phase II (21 months) followed with continued sampling and assessment including a groundwater monitoring programme.

The project began in January 1998 and has been funded throughout by the UK Department for International Development (DFID). The Department of Public Health Engineering (DPHE) of the Ministry of Local Government, Rural Development and Cooperatives has acted as the lead agency for the Government of Bangladesh (GoB) but other GoB Departments have been closely involved, principally the Bangladesh Water Development Board (BWDB).

A report on the findings of the Rapid Investigation Phase was published in January 1999 (DPHE/BGS/MML, 1999). During the course of this project, the seriousness and scale of the groundwater arsenic problem in Bangladesh has become apparent. Many GoB agencies, NGOs, international organisations and donors have now become involved and the GoB and the World Bank have begun a large-scale arsenic mitigation programme. A number of other surveys have also been undertaken, principally relating to the immediate needs of the mitigation programme. These have involved the identification of patients and the monitoring of health impacts, and the testing of various mitigation options.

The results of this project are being disseminated in various ways – through reports, presentations and the internet (www.bgs.ac.uk/arsenic). The hydrochemical database created during the project is publicly available.

2. DATA ACQUISITION

The acquisition of a substantial body of reliable water-quality and sedimentological data was a key objective of the project and was achieved through surveys at various scales. The locations of nearly all sample sites were established by hand-held Global Positioning System (GPS) devices which at the time of sampling (1998/99) were accurate to within about 50–100 m. The aim of the surveys was to establish the basic hydrochemistry of Bangladesh aquifers and of course to establish the extent of arsenic contamination. This was achieved by surveys at the national, upazila and mouza (village) scale. While it was not possible to achieve a similar national coverage of the sediments, a variety of sediments was also examined.

The *DPHE/BGS National Hydrochemical Survey* of tubewells attempted to apply a form of stratified random sampling over the whole of Bangladesh (excluding the Chittagong Hill Tracts) with the stratification designed to ensure a reasonably uniform spatial distribution of sampling sites. Such an ideal sampling scheme was difficult to realise for practical reasons, e.g. flooded areas, lack of roads for vehicular access, and the local lack of familiarity with randomised sampling schemes. Specifically, the selection of the sampled tubewells took no account of any prior information about their possible arsenic concentration – this was in any case largely unknown at the time.

The final data set for this survey consisted of samples from 3534 tubewells from 61 of the 64 districts of Bangladesh and from 433 of the 496 upazilas. The sampled area was approximately 129,000 km^2, compared with a total area for Bangladesh of about 152,000 km^2. The sample density was about 8 samples per upazila, or approximately one sample per 37 km^2. This is perhaps 0.03–0.05% of all Bangladesh tubewells. The majority of the sites sampled were Government (DPHE)-installed wells. These are believed to be representative of all wells. Sample collection was undertaken by project staff in close collaboration with local DPHE staff. Arsenic was measured in the BGS laboratories in most cases by atomic fluorescence spectrometry with hydride generation (HG-AFS).

All but four of the 3534 samples were also analysed for a wide variety of other elements by inductively-coupled plasma-atomic emission spectrometry (ICP-AES). A small subset of these samples was also analysed for a range of trace elements using inductively-coupled plasma-mass spectrometry (ICP-MS) to see if there were any other trace elements that were a cause for concern.

A survey of 113 *BWDB Water-Quality Monitoring Network sites* was carried out for arsenic and a wide range of other determinands including anions and trace elements. These sites were distributed across the whole of Bangladesh and were part of a regularly (bi-annually) sampled water-quality monitoring network. The wells included in this network are not a representative subset of all wells in Bangladesh and so any statistics derived from them have to be treated with caution – the network contained a greater proportion of ‘deep’ wells than found in Bangladesh as a whole, for example.

Three *Special Study Areas* were established in the three sadar (headquarter) upazilas of Nawabganj, Faridpur and

Lakshmipur districts in order to undertake sampling at a greater sample density and with a greater range of determinands than was possible in the national survey. Additional determinands included field parameters such as pH, redox potential and dissolved oxygen, a wide range of trace elements and the stable isotopes of oxygen, hydrogen, carbon and in a few cases, sulphur. These study areas were also where the *water-quality monitoring piezometers* were installed and where a regular water-level and water-quality monitoring programme was undertaken. Where possible, sampling piezometers were installed at 10 m intervals down to 50 m and sampled every two-weeks for up to 12 months. Deep boreholes were also drilled at Faridpur and Lakshmipur and included in the monitoring programme (there is no deep aquifer at Chapai Nawabganj, at least not within 150 m). Where possible, each of the piezometers was sampled on one occasion for tritium and ^{14}C as well as for parameters such as dissolved oxygen and redox potential. The drilling programme involved in installing these piezometers also provided core material for detailed logging and mineralogical and chemical analysis.

A Village survey was undertaken in the mouza of Mandari in Lakshmipur sadar upazila. This was known to be in a high-arsenic region of Bangladesh. The wells sampled were selected randomly. Mandari consists of a number of *para* or family settlements spread fairly uniformly over an area of approximately 6 km^2. The population of Mandari is thought to be about 2500. The aim of this survey was to examine the variation of arsenic at the village scale, the scale at which the actual compliance testing would have to take place. We aimed to make maps as accurately as possible within a short timescale and to measure the arsenic in the field as accurately as possible. For this, an 'Arsenator' was used. This was operated by Professor Walter Kosmus (Karl-Franzens University, Graz, Austria), the instrument's designer.

A map of the sample locations in Mandari was prepared using a combination of GPS, visual observation and a SPOT satellite photographic image. A total of 239 tubewells was sampled, analysed and mapped in six days by a team of 6 project staff and 4 local assistants. Additional analysis of the samples by ICP-AES was subsequently undertaken in the BGS laboratories in order to be able to relate the chemistry of arsenic to other water-quality parameters and to establish the nature of the spatial variation for a broad range of elements.

Other samples analysed for arsenic and a broad range of elements included seven river water samples from across Bangladesh and seven pumped public-supply water samples from the city of Dhaka. These were all from Dhaka WASA wells, and included one from each of the main supply divisions of the city

Sediment samples from 10 boreholes were analysed by a variety of techniques including total analysis by X-ray fluorescence spectrometry as well as selective dissolution with acid ammonium oxalate. A subset of 21 sediment samples from the three Special Study Areas were selected for detailed chemical and mineralogical analysis including particle size, magnetic and heavy-liquid separation, SEM observation and detailed magnetic characterisation.

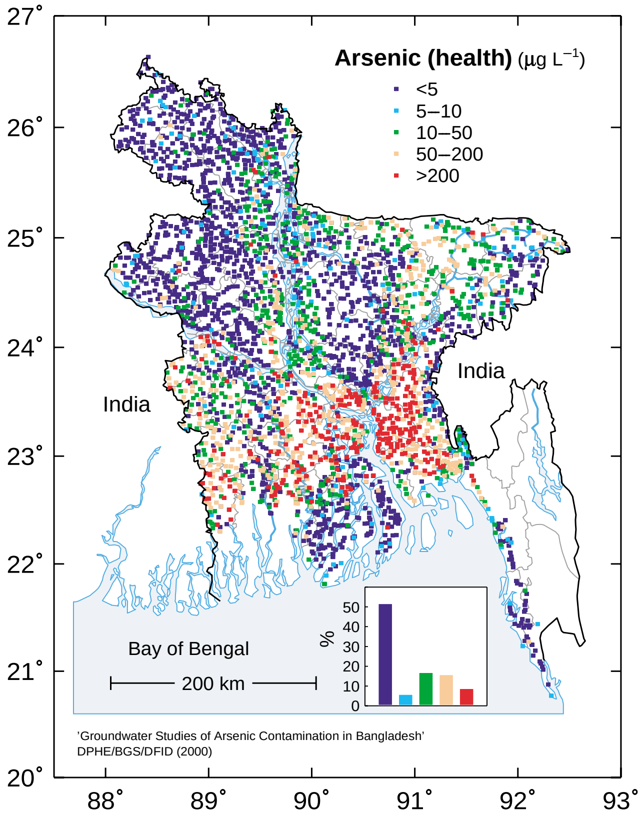

Figure 1. Map showing the concentration of arsenic in groundwaters based on the DPHE/BGS National Hydrochemical Survey. Class divisions are chosen on the basis of health criteria.

3. Scale of the groundwater arsenic problem

The distribution of sample sites from the DPHE/BGS National Hydrochemical Survey and the arsenic concentrations are shown in Figure 1. Arsenic concentrations ranged from less than 0.25 μg L^{-1} to more than 1600 μg L^{-1}. The map shows clear differences in arsenic concentrations in different parts of Bangladesh with the greatest number of high-arsenic wells in the south and south-east of the country. However, superimposed on this regional pattern, there is considerable well-to-well variability over the scale of a few kilometres.

The regional differences can be seen more clearly when the point-source data shown in Figure 1 data are smoothed (Figure 2). This smoothing was carried out by a statistical technique (disjunctive kriging). The high-arsenic region in the south and east of Bangladesh is clear from this map. The DPHE/BGS survey showed that 25% of all the tubewells sampled contain in excess of 50 μg L^{-1} arsenic, the Bangladesh drinking-water standard. In addition, 9% of the tubewells exceeded 200 μg L^{-1}, 1.8% (64) exceeded 500 μg L^{-1} and 0.1% (3) exceeded 1000 μg L^{-1}. Few shallow groundwaters from the south of the country were 'arsenic-free' (i.e. contained less than 3 μg L^{-1}).

On the other hand, 24% of samples fell below the instrumental detection limit for arsenic, normally 0.25 or 0.5 μg L^{-1}. The minimum arsenic concentration is likely to be in the ng L^{-1} range. Concentrations of less than 1 μg L^{-1} are common in northern Bangladesh. They are

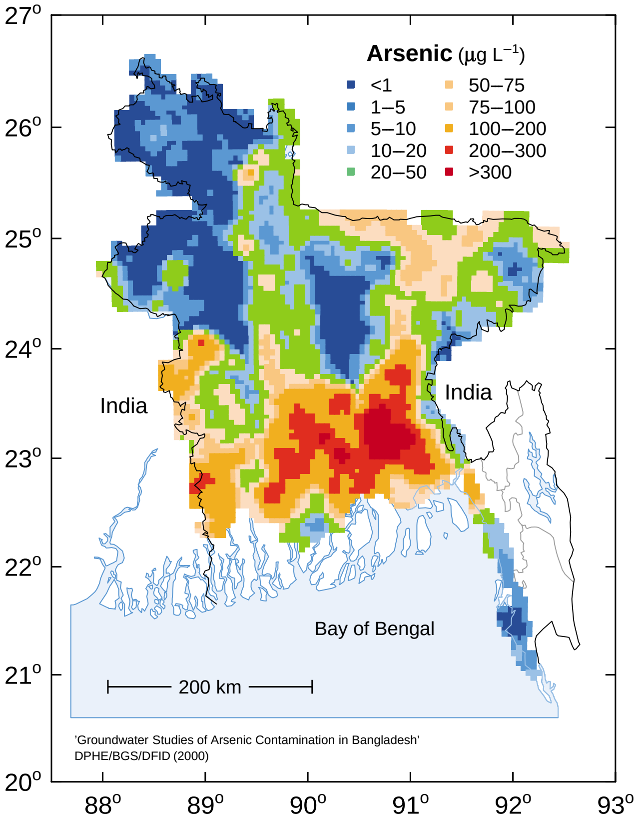

Figure 2. Smoothed map showing the regional trends in groundwater arsenic concentrations in shallow wells.

also typical of the deep aquifer (including water from the city of Dhaka) and the water derived from aquifers in the older sediments of the Madhupur and Barind tracts.

The median arsenic concentration found in all of the tubewells sampled was 4 µg L^{-1} and the maximum concentration found was 1670 µg L^{-1}. The mean concentration was about 55 µg L^{-1}. This value depends to some extent on the concentration of arsenic assumed in the large number of wells containing less than the detection limit. The concentration of arsenic in these less-than-detection limit samples was assumed for statistical purposes to be half the detection limit.

There were important differences between 'shallow' wells and 'deep' wells (defined here as greater than or equal to 150 m depth), as well as between samples from recent (Holocene) alluvium and older (Plio-Pleistocene) alluvium. Arsenic contamination was essentially confined to groundwaters from the shallow aquifer (Figure 3).

Of the wells sampled in the DPHE/BGS National Hydrochemical Survey, 9% were 'deep'. Most of these deep wells were either from the southern coastal belt or from the Sylhet area. There are very few deep wells in the rest of the country. Of the shallow tubewells, 27% contained in excess of 50 µg L^{-1} and 46% in excess of the WHO guideline value for arsenic (10 µg L^{-1}). Only 3 out of the 327 (1%) deep well samples exceeded 50 µg L^{-1} and 16 (5%) exceeded 10 µg L^{-1}. Eight of the 61 sampled districts had no samples exceeding the Bangladesh standard for arsenic (50 µg L^{-1}) and all districts except Thakurgaon had at least one well exceeding the WHO guideline value for arsenic.

The worst-affected districts were (percentage of sampled wells with greater than 50 µg L^{-1} in parentheses): Chandpur (90%), Munshiganj (83%), Gopalganj (79%), Madaripur (69%), Noakhali (69%), Satkhira (67%), Comilla (65%), Faridpur (65%), Shariatpur (65%), Meherpur (60%), Bagerhat (60%) and Lakshmipur (56%). Percentages are the percentage of all wells sampled.

The least-affected districts were: Thakurgaon, Barguna, Jaipurhat, Lalmonirhat, Natore, Nilphamari, Panchagarh, Patuakhali (all 0%), Rangpur (1%), Dinajpur (2%), Naogaon (2%), Gazipur (2%), Cox's Bazar (2%), Bhola (4%), Nawabganj (4%), Jhalakati (6%), Rajshahi (6%), Gaibandha (7%), Tangail (9%) and Kurigram (9%). Again, percentages are the percentage of all wells sampled.

The district-wise average arsenic concentration varies from 1 µg L^{-1} in Thakurgaon to 366 µg L^{-1} in Chandpur (Figure 4). This reflects a very large difference in the average arsenic dose likely to be taken in from drinking water by the people of these two districts.

There is a great deal of short-range variation in the arsenic concentration from well to well which makes predicting the concentration of arsenic in groundwater from unsampled wells in arsenic-contaminated areas difficult even when the concentrations in adjacent wells are known. This points to the necessity for an extensive testing programme. Even in areas of generally low arsenic concentrations, there are occasionally 'hot spots' where a cluster of wells with unusually high concentrations of arsenic are found. Such hot spots are most noticeable in northern Bangladesh. The Chapai Nawabganj hot spot in north western Bangladesh was estimated to be about 5 km by 3 km in extent. The sample density in the DPHE/BGS national survey was insufficient to identify all such hot spots.

Analysis of seven deep well waters from the city of

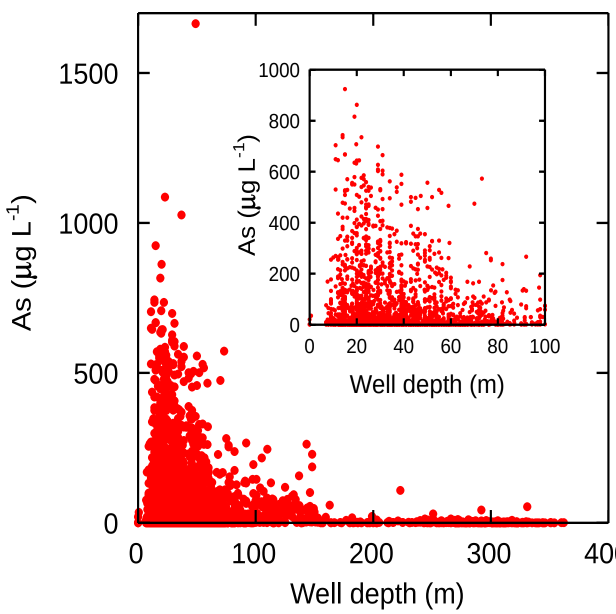

Figure 3. Arsenic concentration of groundwater in wells from the DPHE/BGS National Hydrochemical Survey plotted as a function of well depth.

Figure 4. District-wise average arsenic concentration (in µg L^{-1}) found from the DPHE/BGS National Hydrochemical Survey

Dhaka showed that all contained less than 0.5 µg L^{-1} arsenic.

The population exposed to drinking water in which arsenic exceeds the Bangladesh standard was estimated in two different ways based on slightly different assumptions. These two methods gave estimates of 28 million people (upazila-averaged) and 35 million people (kriging to a 5 km grid). These estimates are based on an estimated 1999 population of 125.5 million for the whole of Bangladesh. If the WHO guideline value is used instead of the Bangladesh standard, these figures increase to 46 and 57 million people, respectively. In the absence of any data to the contrary, we assume that the kriged estimates (larger figures) are more reliable. The problem is clearly very large.

Other findings from the National Hydrochemical Survey

Many groundwaters also contained high concentrations of phosphorus (median 0.3 mg P L^{-1}, n=3530). This phosphorus was probably in part derived from the same source as the arsenic and while the phosphorus-arsenic correlation was not good enough to provide a reliable (or useful) prediction of the concentration of arsenic in a particular well, the two maps do show some correlation when viewed on a regional scale.

The majority of the groundwaters show characteristics that are typical of reducing groundwaters, notably high iron (median 1.1 mg L^{-1}, maximum 61 mg L^{-1}), high manganese (median 0.3 mg L^{-1}, maximum 10 mg L^{-1}) and low sulphate (median 1 mg L^{-1}, minimum less than 0.4 mg L^{-1}) concentrations.

High iron concentrations were widespread but were particularly common in the groundwaters from the Brahmaputra valley in northern Bangladesh (Figure 5). There was a poor overall correlation between arsenic concentrations, although locally significant positive correlations existed.

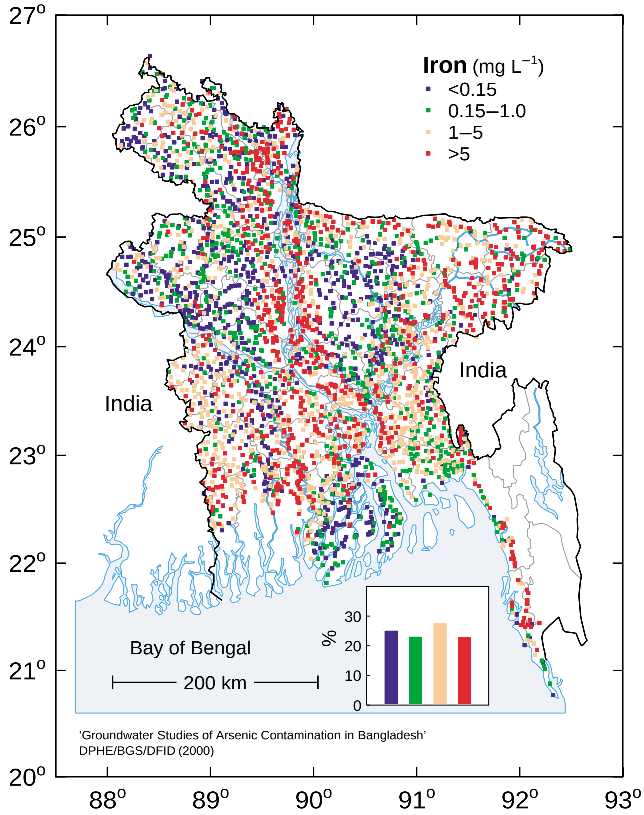

Figure 5. Map showing the concentration of iron found in Bangladesh groundwaters from the DPHE/BGS National Hydrochemical Survey

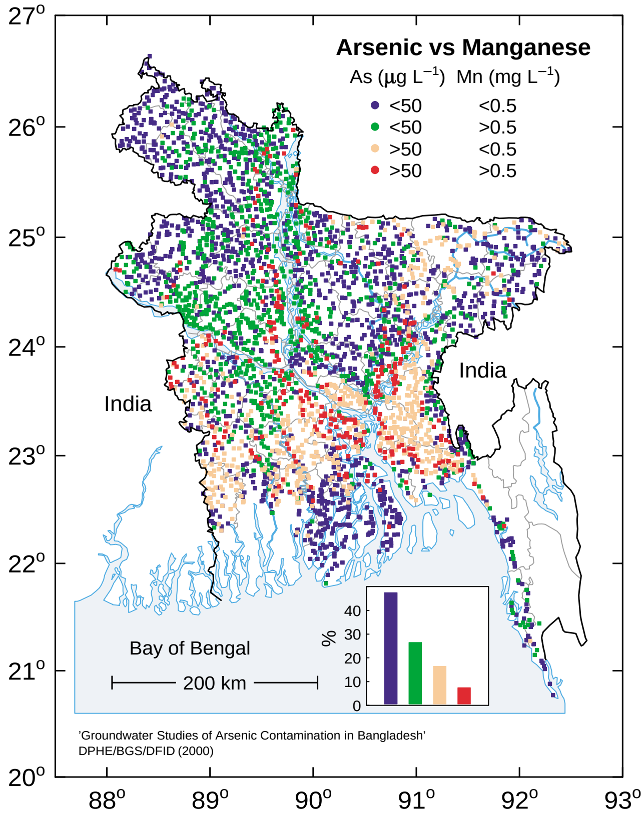

Figure 6. Map showing the joint distributions of arsenic and manganese in Bangladesh groundwaters based on data from the DPHE/BGS National Hydrochemical Survey. Class divisions include the Bangladesh standard for arsenic (50 µg L^{-1}) and the WHO guideline value for manganese (0.5 mg L^{-1}).

Excessive salinity in the coastal regions and particularly high iron concentrations (say more than 5–10 mg L^{-1}) in other areas including northern Bangladesh are important factors restricting the potability of groundwaters in Bangladesh. From our national survey, 23% of tubewell waters contained greater than 5 mg L^{-1} iron and nearly 10% contained more than 10 mg L^{-1}.

Well waters exceeded the WHO guideline value of 0.5 mg L^{-1} for manganese in 35% of the samples from the National Survey. Arsenic and manganese showed distinctly different regional patterns (Figure 6). Only 33% of shallow wells fell below both the WHO arsenic and manganese guideline values. 93% of deep wells did.

Boron exceeded the revised WHO guideline value of 0.5 mg L^{-1} in 5.3% of samples and 9.1% exceeded the former guideline value of 0.3 mg L^{-1}. These high-boron samples are mostly found in the southern coastal region and in the low-lying region around Netrokona-Kishorganj. Boron is a residual component from seawater and high concentrations are usually associated with relatively high salinities.

4. BWDB Water-Quality Monitoring Network

The survey of 113 wells from the BWDB Water-Quality Monitoring Network complemented the DPHE/BGS National Hydrochemical Survey and the results generally showed the same regional trends. The number of samples in the BWDB survey was too small to provide reliable district-wise statistics. A comprehensive analysis of anions was undertaken which indicated that fluoride concentrations were normally low (median 0.2 mg L^{-1}). All were less than 1 mg L^{-1}. Indeed in north-western Bangladesh, they were lower than desirable in drinking water for dental health (Figure 7).

The iodide content of some of the waters from northern Bangladesh was also lower than desirable (less than 3 µg L^{-1}) and could lead to the development of iodine-deficiency disorders without supplementation with dietary iodine.

5. Special Study Areas

The chemistry of the groundwaters from the 1998 and 1999 surveys of the three Special Study Areas showed a high degree of spatial variability on a local scale, as well as with depth. Of the 243 samples collected, arsenic concentrations varied over four orders of magnitude, with the ranges in Lakshmipur, Faridpur and Chapai Nawabganj being respectively <3 to 986 µg L^{-1}, <3 to 1460 µg L^{-1} and <3 to 2342 µg L^{-1}. In Lakshmipur, 55% of all groundwaters sampled exceeded the Bangladesh standard of 50 µg L^{-1}, 41% exceeded this value in Faridpur and 25% in Chapai Nawabganj. Exceedances of the WHO guideline value of 10 µg L^{-1} were 70%, 69%, and 35% in Lakshmi-

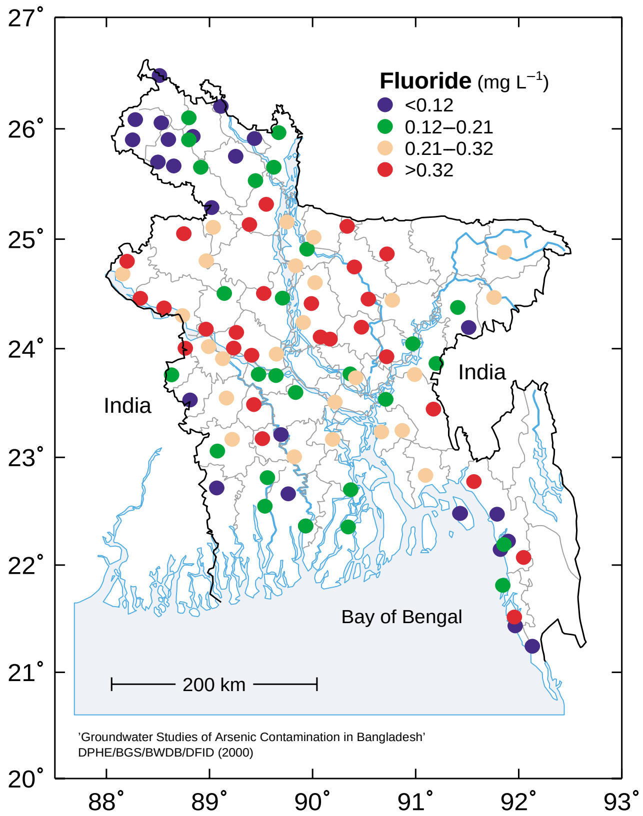

Figure 7. Map showing the concentration of fluoride in groundwaters from the BWDB Water-Quality Monitoring Network.

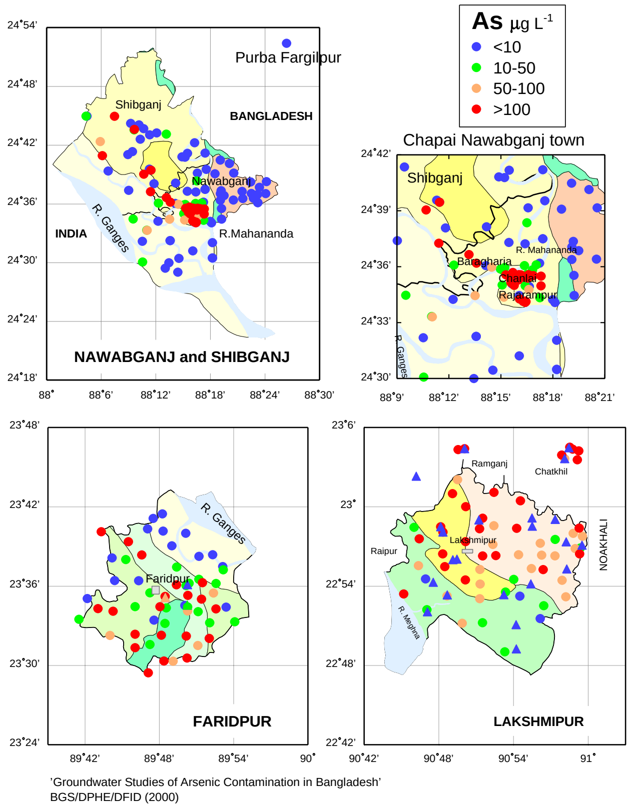

Figure 8. Arsenic concentrations in well waters from the three Special Study Areas. The Chapai Nawabganj map includes data from an earlier 1997 survey. Triangles denote 'deep' wells which were present in Faridpur (below 100 m) and Lakshmipur (below 150 m) but not in Chapai Nawabganj. The colour shading represents different geological units.

pur, Faridpur and Chapai Nawabganj, respectively. Although the greatest proportion of high As concentrations occurred in Lakshmipur, the highest observed concentrations were found in a relatively small number of samples from the Chapai Nawabganj hot spot area. Several of these groundwaters exceeded 1 mg As L^{-1}. Distinct patterns were related in part to geology (Figure 8).

As observed from the National Hydrochemical Survey, groundwaters with high As concentrations occur mostly in the shallow Holocene alluvial aquifer. Concentrations in groundwaters from the deep aquifer in Lakshmipur and Faridpur (which may be Dupi Tila sediments) and from the Dupi Tila aquifer that occurs at shallower depths beneath the Barind Tract in Chapai Nawabganj have mostly low arsenic concentrations. The highest observed value in groundwater from the deep aquifer in the three Special Study Areas was 52 µg L^{-1}, just above the Bangladesh standard for drinking water. This concentration from Faridpur was atypical of the deep aquifer elsewhere. Hand-dug wells sampled from Chapai Nawabganj consistently gave low-arsenic water, even in highly contaminated parts of the study area.

The groundwaters from the Special Study Areas were almost universally reducing, with characteristically high concentrations of iron, manganese and ammonium as well arsenic, and low concentrations of nitrate, dissolved oxygen, copper and selenium. Low sulphate concentrations in most samples point to sulphate reduction as an important process. The highest arsenic concentrations were found in some of the most reducing groundwaters and suggest that the development of reducing conditions is a major factor controlling the mobilisation of arsenic.

Arsenic occurs in the groundwaters in both reduced (As(III)) and oxidised (As(V)) forms. The proportions vary significantly, with As(III)/As_T ratios ranging from less than 0.1 to greater than 0.9 in all areas. Groundwaters from Lakshmipur had a higher proportion of samples with high ratios, i.e. As(III) was a relatively important component there. This suggests that, on average, the Lakshmipur groundwaters are the most reducing. Groundwaters with high concentrations (total arsenic concentrations greater than 400 µg L^{-1}) in Chapai Nawabganj also have relatively high As(III)/As_T ratios, usually greater than 0.6. These results suggest that arsenic needs to be substantially present as reduced As(III) to allow significant mobilisation. However, since not all of the arsenic is present in reduced form, simple reduction of As(V) to As(III) is probably not sufficient in itself to generate the high As concentrations found in the groundwaters. Other factors are likely to be involved in the release of arsenic from the sediments.

Although the oxidation of pyrite has been put forward by other workers as the key process controlling the release of As in the groundwater, this process is not supported by the geochemical evidence. Arsenic concentrations show a broad negative correlation with sulphate concentrations.

This suggests that sulphate reduction rather than sulphide oxidation accompanies arsenic release to groundwater. Sulphur-isotopic compositions of the dissolved sulphate in selected groundwater samples also suggest that these represent residual sulphur resulting from biogenic reduction of sulphate to sulphide.

Stable-isotopic data ($\delta^{18}O$, $\delta^{2}H$) for the groundwaters were variable both within and between aquifers. The greatest variation is seen in Lakshmipur where marine intrusion at some time over the past few thousand years is thought to have been responsible for the most enriched compositions observed. Distinct isotopic differences are seen in each area between the shallow Holocene alluvial aquifer and the deeper aquifer, which in the case of Chapai Nawabganj is the Dupi Tila aquifer. The distinctions suggest that variations exist in the residence times of the groundwaters (i.e. groundwater 'ages') in each aquifer. However, the differences are not consistent between areas and hence do not give an unambiguous indication of relative groundwater age or recharge history.

Tritium, an indicator of modern groundwater, was usually detectable at a few TU in the shallowest groundwaters (a few metres below the water table) from each study area but deeper groundwaters had lower concentrations, typically <0.4 TU. Such low concentrations are indicative of older groundwater, with a large proportion having been recharged prior to the 1960s. A greater number of tritiated waters were detected in the shallow aquifer in Chapai Nawabganj than in Lakshmipur and Faridpur. This observation suggests that Chapai Nawabganj groundwaters have a higher proportion of modern recharge water at shallow depths. This may reflect the greater unsaturated-zone thickness and greater seasonal fluctuations in water level there.

Twenty samples of groundwater from the three sets of piezometers installed were collected for ^{14}C determination. The ^{14}C data suggest distinctive groundwater ages for each of the three sites. Groundwater from 10–40 m depth at Chapai Nawabganj was 'modern' (83 pmc or greater) indicating an age of the order of decades, a conclusion supported by the presence of tritium in many of the samples. Shallow groundwaters from Faridpur were also modern (78 pmc or greater) despite lower tritium concentrations. Groundwater from 150 m at Faripur was notably older (51 pmc) with a model age of about 2000 years. Deep groundwaters from Lakshmipur were even older with ^{14}C activities of 28 pmc or less suggesting the presence of palaeowaters with ages of 2,000–12,000 years.

6. VILLAGE SURVEY: MANDARI, LAKSHMIPUR DISTRICT

Mandari is situated in south-eastern Bangladesh near the boundary between alluvial and deltaic deposits. Salinity is a problem at intermediate depths and the wells tend to be either quite shallow – at a depth of less than 15 m – or deep (below 250 m).

The sample density of the Mandari survey, at nearly 40 samples per km^2, was some 1400 times greater than in the DPHE/BGS National Hydrochemical Survey. The results showed a great deal of short-range spatial variability in the concentration of arsenic and other elements. It would be difficult to predict arsenic concentrations in unsampled wells from adjacent wells with an acceptable degree of accuracy (Figure 9). Arsenic concentrations in shallow wells ranged from 6 µg L^{-1} to 707 µg L^{-1}. 23% of the shallow tubewells (less than 150 m deep and mostly less than 40 m deep) sampled were below the Bangladesh limit for As in drinking water but only 5% were below the WHO guideline value. The ten recently-installed deep wells all contained less than 2.5 µg As L^{-1} and mostly less than 0.25 µg L^{-1}. 14% of the shallow wells exceeded 200 µg As L^{-1}.

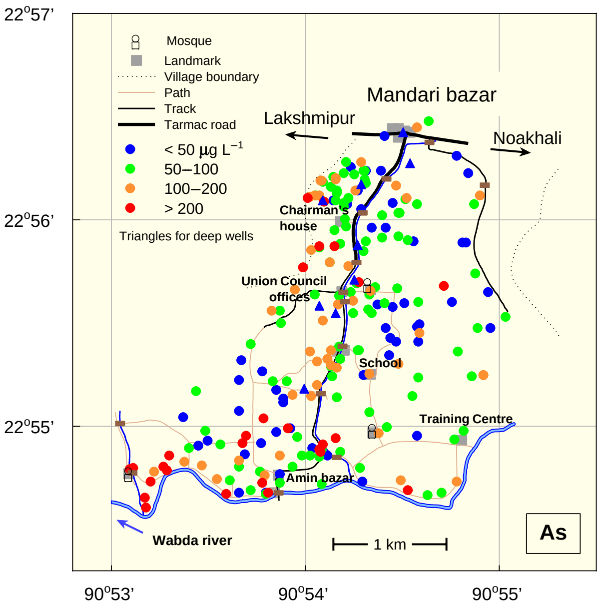

Figure 9. A map of arsenic concentrations in well waters from Mandari village, Lakshmipur. Deep wells (greater than 150 m) are shown by triangles.

High concentrations of arsenic were found on the southern boundary of Mandari adjacent to the Wabda river, particularly in the extreme south-west where many of the wells exceeded 300 µg L^{-1}. The villagers were unaware of the high concentration of arsenic in their well water. An area of predominantly 'low' arsenic concentrations was found in the centre of Mandari.

There were few significant correlations between arsenic and other elements. The most interesting was the inverse relationship between sulphate and arsenic, as also found in the Special Study Areas, probably reflecting the importance of strongly reducing conditions in promoting high arsenic groundwaters.

7. ARSENIC MONITORING

During the course of the project, piezometers installed in each of the Special Study Areas and some existing dug wells and hand-pump tubewells were monitored for arsenic, water levels and a selected number of other elements. Fortnightly monitoring began in April 1999 (June in Lakshmipur) after piezometer completion and has been carried giving a record of slightly less than one year in most cases. The piezometers were purged before sampling but were not pumped between sampling. Other hand-

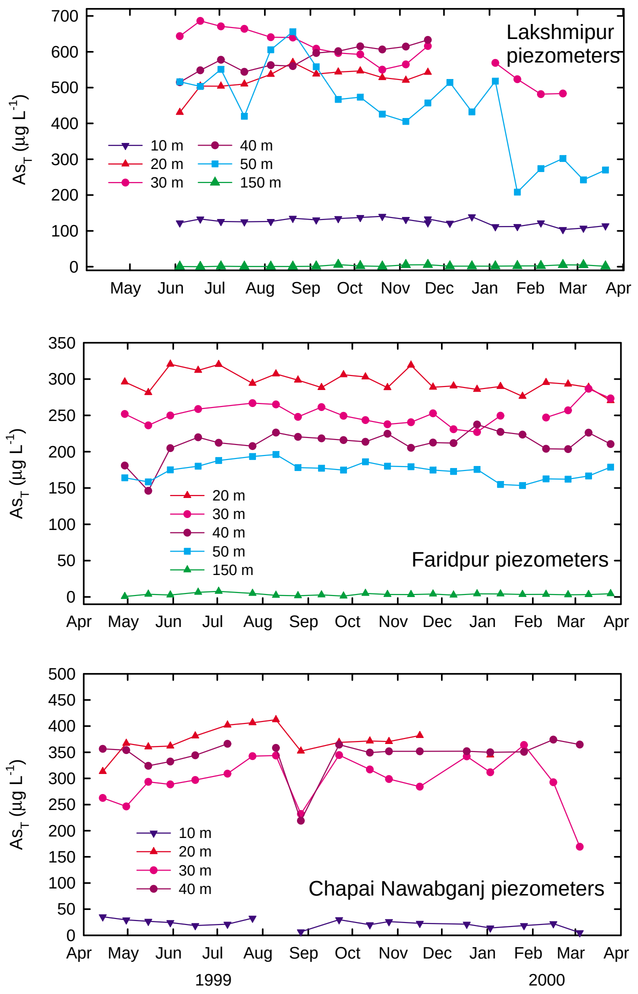

Figure 10. Temporal variations in arsenic concentration in piezometers at various depths from the three Special Study Areas.

pump tubewells sampled in the monitoring were used at other times. Arsenic in the piezometer waters (Figure 10) and other nearby wells had variable concentrations but over the short period of monitoring, did not appear to show significant increases or decreases with time. Variations were greatest at the shallowest depths (10–20 m) where the greatest water movement is expected, but dampened at greater depths. Fluctuating arsenic concentrations were apparent at the start of monitoring of some piezometers, which may have related to initial disturbances in the groundwater following drilling.

Monitored wells included three from the deep aquifer (piezometers at 150 m at both Lakshmipur and Faridpur; Figure 10, and one deep well at 286 m from Lakshmipur). During the monitored period, none of these sites showed increases in arsenic concentration with time and all remained well below 10 µg L^{-1}.

Three shallow dug wells from Chapai Nawabganj also had low arsenic concentrations. None of these showed increasing trends with time, although three individual analyses exceeded 10 µg L^{-1} marginally during the monitored period. Concentrations of some other constituents (chloride, sodium, sulphate) in these samples were typically high and variable, suggesting that pollution has been a significant input to these sources.

No clear or consistent changes in arsenic were detected in the aquifers during the short monitoring interval. However, longer-term monitoring of the wells is required to establish whether there will be significant seasonal and long-term trends in water chemistry. The relatively small variation in arsenic concentrations observed in many of the wells emphasises the need for very careful sampling and high-precision analysis if seasonal or long-term trends are to be detected reliably.

8 SEDIMENT HISTORY

The Bengal Basin is a tectonically active subsiding depression formed at the junction of the Asian, Burmese and Indian plates, and is infilled with more than 15 km of marine and alluvial sediments of Cretaceous to Recent age. Throughout the Quaternary, the combined Ganges, Brahmaputra and Meghna (GBM) river system of Bangladesh has deposited a thick sequence of mixed alluvial and deltaic deposits in response to changes in sea-level rise and fall brought about by glacial cycles.

Within the Basin, there are areas of recent uplift (Madhupur and Barind Tracts) and subsidence (the Sylhet Basin) and major changes in the course of the Tista and Brahmaputra rivers can be seen in the sediments. Patterns of sediment deposition during the Upper Pleistocene were controlled by a fall in sea level to about 150 m below the present day sea level. This decline occurred between the last interglacial 120,000 years ago and the last glacial maximum some 21,000 years ago. Sea level recovered during the Holocene to the present-day level.

Various facies of deposition can be seen within the alluvial fan deltas, fluvial flood plains and deltaic environments. Lithostratigraphic correlation of these sediments has been attempted using palaeosol and peat horizons. Fine-grained deposits were laid down during periods with a relatively high sea level and correspondingly low-energy environment (so-called 'highstand' deposits) whereas coarse-grained deposits characteristic of a high-energy environment formed during periods of glacial maximum or 'lowstand' times.

Core sediments obtained from the two 'deep' boreholes drilled in the Special Study Areas provided the first detailed and relatively undisturbed samples of upper Pleistocene sediments recovered in Bangladesh. These boreholes were logged and correlated with DANIDA logs from the Lakshmipur area and with BGS logs from the Dhamrai-Manikganj area to produce conceptual models of past sediment deposition in the delta area and in the lower fluvial environment of the Brahmaputra valley.

Geological logs of deep boreholes drilled by BWDB were collated to provide an understanding of the possible patterns of sediment deposition during the last highstand and lowstand periods as well as during previous glacial-interglacial cycles. Using these data, tentative maps of the distribution of major incised channels at the last glacial maximum and before the last interglacial period were constructed. Seismic refraction survey data from the western part of the Ganges delta in Bangladesh showed the existence of a series of stacked channels containing coarse-grained sediments at depths greater than 500 m. These could form future sources of deep groundwater within that area.

9. SEDIMENT CHARACTERISTICS

Sediment samples, principally from the Special Study Areas, were analysed by a wide variety of techniques including total dissolution, selective dissolution with ammonium oxalate, magnetic separation, magnetic susceptibility, scanning electron microscopy (SEM) and X-ray diffraction.

In the 21 samples studied in detail, the average total arsenic concentration was 4 mg kg^{-1} and ranged from 0.4 mg kg^{-1} to 10 mg kg^{-1}. Arsenic concentrations were greatest in the fine-grained sediments and were highly correlated with iron content. The principal iron mineral observed was low-titanium magnetite but there was also some oxidised 'rust' material present. There was some evidence that this may have been derived from rust contamination from the drilling rig particularly in the Faridpur and Lakshmipur boreholes. The sediments also showed evidence of partial oxidation during storage.

Other rarer iron-containing minerals observed included titanomagnetite, ilmenite-magnetite composite grains, and ilmenite-hematite intergrowths. There was also abundant mica, particularly in the samples from Lakshmipur. Scanning electron microscopy (SEM) showed the sediments were in all respects typical alluvial and deltaic sediments.

Dissolution of the poorly-ordered metal oxides and desorption of elements by acid ammonium oxalate extractions of sediments from the cored piezometer holes in the Special Studies Areas showed that the sediments from Lakshmipur contained the greatest amounts of extractable arsenic, iron, manganese, aluminium and potassium. Average concentrations of oxalate-extractable As from the three sediment profiles drilled in the project were (in mg kg^{-1}, n=number of samples): Chapai Nawabganj (1.8, n=22); Faridpur (0.8, n=49) and Lakshmipur (2.1, n=48). All three of these are areas of high-arsenic groundwaters. There was also a high correlation between extracted iron and arsenic and in many cases also between iron and aluminium, and between iron and manganese. Average concentrations of extractable iron were (in mg kg^{-1}): Chapai Nawabganj (3000), Faridpur (2300) and Lakshmipur (4600). The fine-grained silts and clays normally contained a greater concentration of iron and many minor elements, including arsenic, compared with coarser-grained sediments which are characteristic of the exploitable parts of the aquifer.

The iron oxides may be derived in part from the weathering of the abundant biotite mica present in the sediments. Mica appears to be particularly abundant in sediments from the distal (lower) part of the delta where it may have been concentrated by natural re-suspension and sedimentation processes.

There was also more extractable sulphur (and sodium) in the Lakshmipur sediments than in the others, reflecting a greater marine contribution. Overall, the Lakshmipur sediments were much more variable than the sediments from Chapai Nawabganj and especially compared with those from Faridpur. This is consistent with the location of Lakshmipur being close to the boundary between the alluvial and deltaic sediments. This was also reflected in the large variability in salinity in the well waters from that area.

Ammonium-oxalate extracts were made on a limited number of sediments from low-arsenic areas including the Barind Tract region of northern-western Bangladesh, coarse-grained sediments from Thakurgaon, and older sediments from the Dupi Tila aquifer beneath Dhaka. These sediments all gave significantly lower average concentrations of extractable arsenic (normally less than 0.2 mg As kg^{-1}) and iron (normally less than 500 mg Fe kg^{-1}) compared with those from arsenic-contaminated areas.

The general conclusions from the sediment studies are that the sediments are typical of alluvial and deltaic sediments with normal amounts of arsenic, mainly in the 1–10 mg kg^{-1} range for total arsenic. However, even normal amounts of arsenic are sufficient to give excessive arsenic in the groundwater if dissolved or desorbed in sufficient quantity. Arsenic-rich groundwaters tended to be found in areas with sediments containing relatively high concentrations of oxalate-extractable iron and arsenic. This is consistent with the iron oxides being a principal source of arsenic in the arsenic-rich groundwaters.

While the role of iron oxides is undoubtedly important, other oxides such as manganese and aluminium oxides may also be important. It is difficult to separate the individual role of the various oxides from extraction data alone because of the non-selectivity of most extractants and the high correlations between the amounts extracted. Everything observed in the sediments is consistent with the desorption and dissolution of arsenic from oxide minerals being the key process controlling the release of arsenic to groundwater.

10. CAUSES OF THE ARSENIC PROBLEM

In order to understand the development of high-arsenic groundwaters in Bangladesh, we have to rely on our scientific knowledge of the likely processes involved, the inferred past history of the aquifers and the present-day evolution of groundwater quality in comparable environments. There is as yet no consensus amongst scientists about the precise cause of the arsenic problem in Bangladesh but below we indicate what we believe to be a plausible scenario that is consistent with most of the known facts. There is still a great deal of uncertainty about the timescale over which the events may have occurred.

Everything points to the arsenic being of natural origin although it is not yet possible to exclude the possibility that modern agricultural practices (groundwater abstraction from shallow wells, irrigation and fertilisation) will have no influence on the groundwater arsenic concentrations whatsoever. It is believed that the arsenic has been in the groundwater for many years, certainly since before the recent extensive abstraction of groundwater.

Both the arsenic content of Bangladesh sediments (0.4–10 mg kg^{-1}) and their mineralogy are typical of young alluvial and deltaic sediments that contain a wide variety of minerals reflecting their diverse source rocks. There is no need to speculate on a unique geological source of high arsenic rocks somewhere upstream of Bangladesh.

However, certain types of sedimentological processes

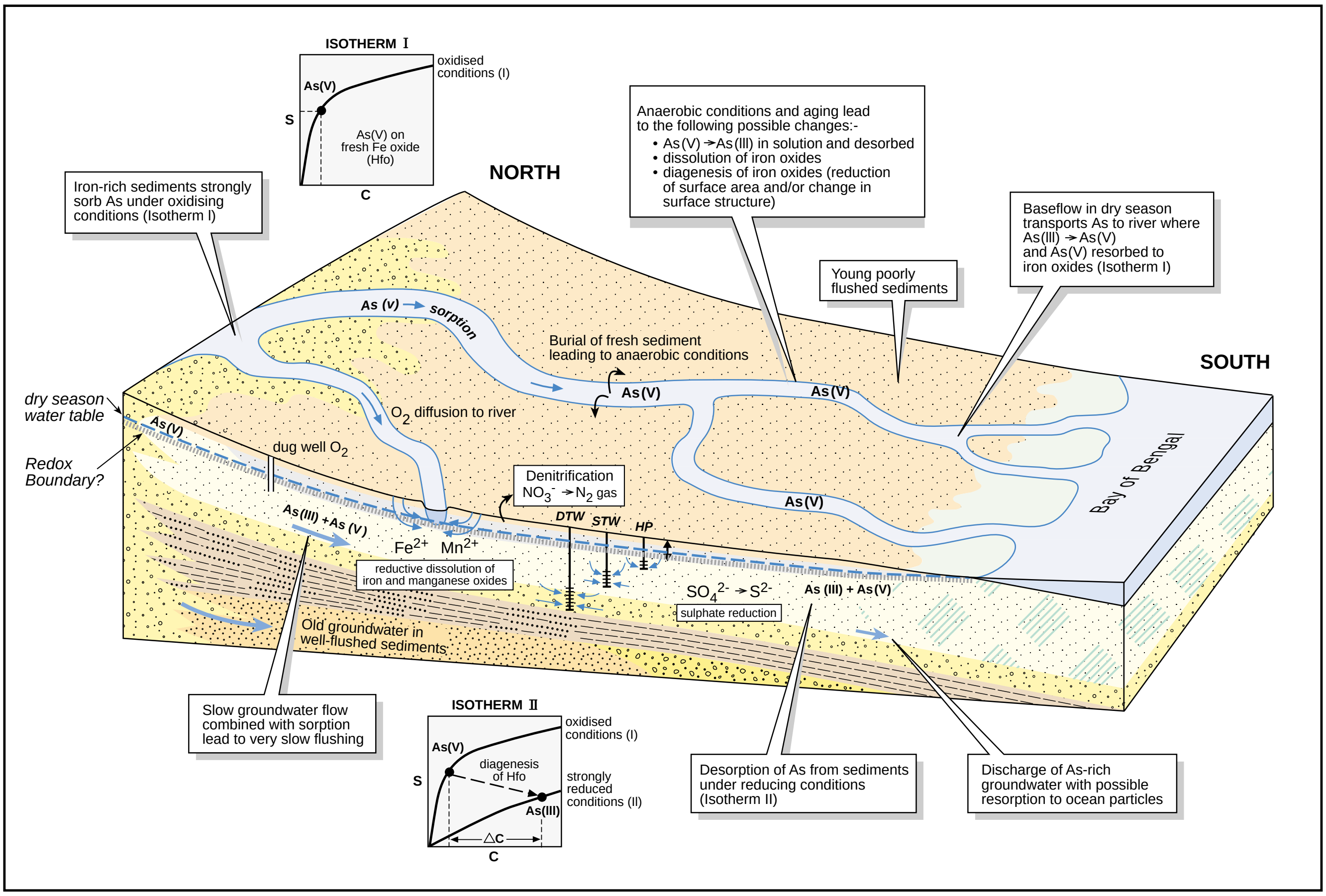

Figure 11. Schematic diagram showing the processes leading to the generation of high-arsenic groundwaters in the Bengal Basin.

have probably been more important in concentrating arsenic in some parts of the delta system than in others. In particular, colloidal-sized iron oxides with their strongly sorbed arsenic and platey, easily suspended, micaceous minerals appear to have been concentrated in the lower part of the delta.

Even 'ordinary' sediments such as these contain sufficient arsenic to give rise to the groundwater arsenic problem observed because of the very low drinking-water guideline value for arsenic and the high solid/solution ratios found in aquifers.

The development of strongly reducing conditions is believed to be the trigger that has been responsible for the release of naturally-occurring arsenic from the sediment into the groundwater. This arises from the rapid consumption of dissolved oxygen by the oxidation of fresh organic matter in the recently-buried sediments. Once strongly reducing conditions are achieved, arsenic is released from the sediments. The mechanism for this release is poorly understood quantitatively but is believed to involve the desorption from, and dissolution of, arsenic from various oxides, especially iron oxides.

This reaction is probably rapid (on geological timescales) and is essentially a diagenetic response of the sediments to a change from an oxidising environment to a reducing environment following burial. Possible changes of significance are a reduction in surface area of the iron oxides following their partial recrystallisation (ageing) and a change in their surface chemistry as a result of the formation of a mixed-oxidation state (Fe(II)–Fe(III)) surface. These changes, as well as the reduction of the strongly sorbed As(V) to the less strongly sorbed As(III), could lead to the release of arsenic from the sediments. Geochemical modelling has confirmed that such changes could account for the high arsenic concentrations observed in Bangladesh groundwaters. Phosphate is believed to be released by the same desorption and dissolution mechanisms (except that the oxidation state of phosphorus is not sensitive to redox conditions). These processes are shown schematically in Figure 11.

The release of arsenic (and phosphate) has also frequently been observed in recently-buried and reducing freshwater and marine sediments, and in flooded soils from many parts of the world. This release is magnified by a number of factors in Bangladesh, especially the large size of the delta and the unusually large depth of recently-deposited sediments, i.e. sediments deposited over the last few thousand years.

The flow of water in the aquifer is also important since this is the normal natural mechanism for flushing away the arsenic so released. The large flat delta region of Bangladesh leads to extremely low hydraulic gradients and correspondingly low rates of flushing of the aquifer. This means that the arsenic released will accumulate, as observed.

Where groundwater flushing is more active, as in parts of northern Bangladesh, or has existed for longer periods as in the deep aquifer, then arsenic concentrations are lower. It is likely that the high concentrations of arsenic found in Bangladesh groundwater will eventually disappear as fresh groundwater flushes through the aquifer, albeit very slowly. The rate of groundwater flow is poorly understood at present but this flushing will probably take thousands or tens of thousands of years. Significant falls in sea level have occurred in the recent past which will have greatly accelerated the flushing of the deeper aquifer. Such changes could occur again if the earth goes through another ice age.

The concept that the present groundwater arsenic problem results from a relatively rapid change in response to recent burial, and that the desorption of arsenic from oxides is an important part of this process, is encouraging in the sense that once the initial release of arsenic has been flushed away, it should not continue to be released unless conditions once again change for the worse, e.g. become even more reducing. This is generally unlikely in the deep aquifer but could occur in those parts of the shallow aquifer that are not yet very strongly reducing. For example, certain changes at the land surface could lead to a reduced rate of diffusion of oxygen to the underlying aquifer. The establishment of more extensive flooding and the puddling of soils associated with paddy fields are the most obvious mechanisms for achieving this. However, the redox buffering by the large volume of sediments involved is large and so any such changes are likely to be slow.

Therefore we believe that the deep aquifers which are currently predominantly arsenic-free in Bangladesh are likely to remain so, at least under natural flow conditions. However, we stress that this is only an initial observation based on limited evidence and that the precautionary principle suggests that this should not be relied on until more solid evidence is established in its favour. In particular, more detailed studies are required on the influence of pumping in both the shallow and deep aquifer to see how this might change the situation. There is conflicting anecdotal evidence on this at present. The connectivity of the shallow and deep aquifers is an important factor.

The careful monitoring of water quality in the aquifers at different depths and over various timescales is essential. A better understanding of the ages of the sediments and groundwaters and of the regional distribution of aquifers and aquicludes would also be very useful.

11. GROUNDWATER FLOW

The groundwater gradient and rate of groundwater flow are controlled by the distance between rivers and the balance between recharge and evaporation. This varies seasonally. In Bangladesh, hydraulic gradients are very low because of the limited relief. Hand-pump tubewells are unlikely to have a major effect on groundwater flow. Irrigation wells with their larger volumes of abstraction will tend to draw water from groundwater rather than from river recharge and may thereby change the local hydraulic gradients significantly. However, groundwater movement and hence aquifer flushing, is inherently very slow in Bangladesh. Under typical groundwater gradients, the timescale to replace the groundwater within an aquifer is of the order of tens of thousands of years. This is revealed by the old 'ages' of groundwater and the large degree of stratification of water quality in the aquifers. It is also reflected in the considerable degree of spatial variation observed in groundwater quality even within a given village.

The magnitude of vertical groundwater flow is important for determining the extent to which arsenic might be transmitted from the shallow contaminated zone to the deeper uncontaminated aquifer. The magnitude depends on the presence, or otherwise, of layers of low hydraulic conductivity (aquicludes) which will restrict vertical flow as well as the presence of thick layers of more permeable material at depth which will enhance vertical flow.

The distribution, nature and size of present-day rivers also has an important effect on groundwater velocities and as such, rivers may play a significant role in controlling the short-range variability of groundwater arsenic concentrations through their effect on local hydraulic gradients. Particularly low groundwater velocities are found in areas surrounded on two or three sides by a river, as for example in the inside of meanders. This may account, in part at least, for localised arsenic-rich 'hot spots' especially where the river system is stable for a long period of time.

Modelling estimates were made of groundwater travel times from the water table to both shallow and deep tubewells based on the aquifer conditions at Faridpur. For the shallow aquifer, assuming well screens at 65–75 m below the water table, it was estimated that 50% of the flow took less than 50 years to reach the well. However, this is highly dependent on the recharge rate; the higher the rate, the shorter the travel time. The approximate lateral distance of flow from the water table to the wells was estimated to be around 50–125 m.

For the deep aquifer, assuming a well screen at 110–135 m below water table, the travel time under pumped conditions was estimated to be in excess of 200 years from a lateral distance of approximately 500 m. Under natural (unpumped) conditions, flow to the same depths was estimated to be in excess of 300 years, with a lateral movement of 1000 m. These travel-time estimates are consistent with the observed presence of tritium in the upper part of the shallow aquifer and its absence from the deep aquifer.

Groundwater modelling has demonstrated that the distribution of vertical flows is highly dependent on the assumed lithological profile. Lithology therefore has to be known in detail and included in the models, before reliable predictions of vertical flows can be made. This is especially important for considering flow to the deep aquifer. Modelling of the Faridpur aquifer indicated that percentages of flow to the deep aquifer (taken to be greater than 130 m depth) can vary by as much as three times (4–12%) depending on the distribution of hydraulic properties of the sediment profile. Lithological information from the borehole log obtained for Faridpur suggests that, unlike elsewhere in Bangladesh, there is not an extensive, well-defined aquitard layer between the shallow and deep aquifer. Groundwater flow to the deep aquifer based on the Faridpur model is therefore likely to represent a worst-case estimate.

12. Other health-related water quality problems

A wide range of inorganic constituents was measured in groundwaters derived from the various surveys undertaken within this project. There were limitations to what could be achieved particularly in the determination of unstable determinands such as ammonium, nitrate, nitrite, bicarbonate and pH. These parameters were not measured in the DPHE/BGS National Hydrochemical Survey because it was not possible to guarantee their preservation before analysis. However, they were measured on most of the samples from the three Special Study Areas. Ammonium, as well as bacteriological quality were also measured in a parallel set of samples collected during Phase I of the national survey by Dr Bilqis Amin Hoque, formerly of the International Center for Diarrhoeal Disease of Bangladesh (ICDDR,B).

Of the inorganic constituents considered in Bangladesh groundwaters, arsenic represents by far the most serious health risk. However, potential problems also arise from a number of other constituents. From the DPHE/BGS National Hydrochemical Survey, 35% of samples exceeded the WHO guideline value (0.5 mg L^{-1}) for manganese in drinking water, and some significantly so (maximum 10 mg L^{-1}). Of the DPHE/BGS National Hydrochemical Survey samples, 8% exceeded both 50 µg L^{-1} arsenic and 0.5 mg L^{-1} manganese, while 48% of samples were below both the Bangladesh arsenic standard and the WHO guideline value for manganese. Wells in parts of western Bangladesh (e.g. the Rajshahi area) are relatively high in manganese but low in arsenic (Figure 6). The reverse is true in much of southern Bangladesh. Altogether, 36% or about one third of samples that were below the Bangladesh standard for arsenic exceeded the WHO manganese guideline value. Groundwater from the older sediments (Barind and Madhupur Tracts), the deep aquifer in the southern coastal region (and in Dhaka), and from the coarse-grained sediments of north-western Bangladesh tended to comply with the WHO guideline values for both arsenic and manganese. Only 2% of the deep wells sampled in the National survey exceeded 0.5 mg Mn L^{-1}.

Five percent of samples from the DPHE/BGS National Hydrochemical Survey also exceeded the WHO guideline value of 0.5 mg L^{-1} for boron. These sites were concentrated in the southern coastal region and in a small region of north-eastern Bangladesh. Boron is a residual component from sea water and therefore tends to be greatest in areas affected by salinity. The sodium concentration in the affected groundwaters usually exceeded 200 mg L^{-1}.

The lack of a high spatial correlation between arsenic, manganese and boron means that there will be some groundwaters that conform to the arsenic drinking water standard but fail on one of the other standards, and *vice versa*.

A much smaller number of samples (0.3%) exceeded the WHO guideline value of 0.7 mg L^{-1} for barium. These were mostly located in the south-west coastal region.

Nitrate and nitrite were not measured in the DPHE/BGS National Hydrochemical Survey but results from the three Special Study Areas indicate that occasional exceedances of guideline values for these determinands will be found but that they are unlikely to be widespread. The strongly reducing conditions found in many Bangladesh aquifers provide a favourable geochemical environment for denitrification and therefore low nitrate concentrations are to be expected and are normally found. Isolated high nitrate concentrations were occasionally found in the shallow groundwaters and are indicative of pollution (e.g. from latrines) and are likely to be accompanied by high concentrations of sulphate, chloride, bromide and often nitrite as well as bacteriological contamination.

Like nitrate, concentrations of nitrite in the Special Study Areas were also mostly low, and usually less than the WHO guideline value of 0.91 mg L^{-1} as NO_2-N. 4% of sampled wells had concentrations in excess of the guideline value. These were mainly but not always in samples which were thought to be polluted.

A wide range of trace elements was measured in the 272 samples collected in total from the Special Study Areas. Results indicated no exceedances above WHO guideline values for antimony, cadmium, chromium, molybdenum or nickel, and most were well below the guidelines. Lead was found in excess of the WHO guideline value (10 µg L^{-1}) in just one sample (concentration 29 µg L^{-1}). Similar concentration ranges, with no exceedances, were found for these trace elements in samples from the BWDB water-quality monitoring network. In these samples, the maximum lead concentration found was 8 µg L^{-1}.

Considering these trace elements, a few exceedances were observed in the subset of 20 samples selected for analysis from the national survey. However, these were in most cases not significantly above the WHO guidelines and are therefore not considered a major problem. Molybdenum was the only exception, with two samples from the national survey having unusually high concentrations of 410 µg L^{-1} and 800 µg L^{-1}.

One element of potential health concern highlighted by the analytical data was uranium. This has been assigned a provisional guideline value of 2 µg L^{-1} by WHO. Concentrations in excess of this were found in a large number of samples – 50% of the subset of samples selected for trace-element analysis from the national survey, 12% of the BWDB survey samples, and 28% of the samples from the three Special Study Areas. Uranium concentrations show quite large differences both within and between the three Special Study Areas (Figure 12).

The maximum uranium concentration observed was 47 µg L^{-1} from the Chapai Nawabganj Special Study Area, although the median concentration was less than 2 µg L^{-1} (0.42 µg L^{-1}). Concentrations were particularly high in dug-well waters from Chapai Nawabganj. Uranium generally showed a negative correlation with arsenic, largely as a result of the variations in redox conditions. Highly reducing conditions favour arsenic mobilisation, whereas more oxidising conditions favour uranium mobilisation.

Constituents considered troublesome on aesthetic grounds include high salinity, iron and ammonium. Salinity is highest in groundwaters from the southern part of Bangladesh where seawater influences have been greatest, as evidenced by high sodium and chloride concentrations and by a high specific electrical conductance. Iron and

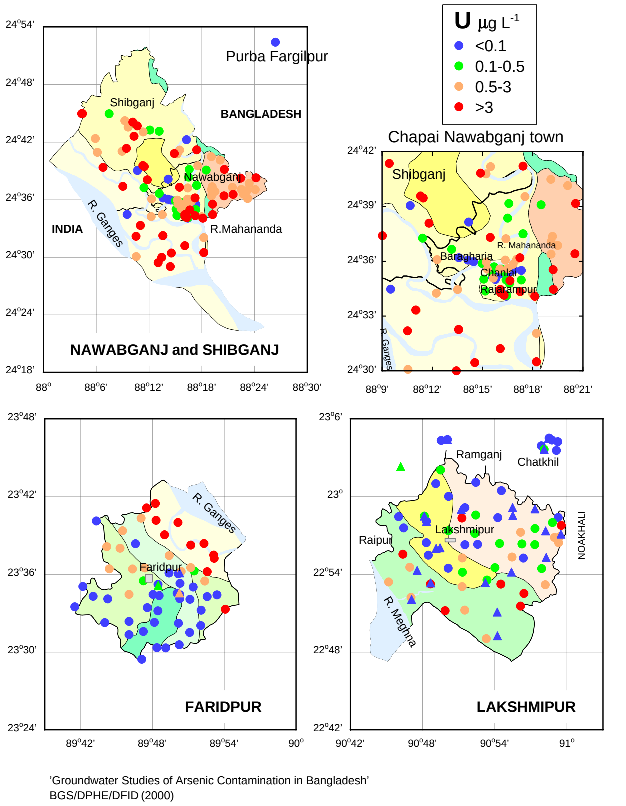

Figure 12. Uranium concentrations in groundwaters from the three Special Study Areas.

ammonium are often present in very high concentrations (up to 11 mg L^{-1} and 17.8 mg L^{-1} respectively) reflecting the reducing conditions of the aquifers. The parallel ICDDR,B survey in Phase I found a median ammonium-N concentration of 1.0 mg N L^{-1}.

High aluminium concentrations are found in a few groundwaters but these are believed to be derived from colloidal material and are not thought to be problematic.

Of the seven deep tubewells analysed from Dhaka, concentrations of the trace metals were usually very low. Concentrations of arsenic, antimony, boron, cadmium, chromium, lead, molybdenum, nickel and uranium were all well below WHO guideline values. Manganese exceeded 0.5 mg L^{-1} in only one sample (0.67 mg L^{-1}).

Only inorganic constituents have been considered in this study and hence no indication can be given of the bacteriological quality of the groundwaters or of potential contamination with pesticides. Occasional high concentrations of nitrate together with nitrite, chloride, sulphate and elevated SEC values, suggest that pollution of some tubewells and dug wells may be severe, especially for those at very shallow depths. Hence bacterial quality of some of these groundwaters is expected to be impaired.

In conclusion, the constituents of greatest concern in Bangladesh groundwaters are arsenic, manganese and possibly uranium, and to a lesser extent boron. With the exception of manganese, these constituents are present in groundwater as neutral species or as anions. Trace metals such as nickel, copper and lead which are mostly present in the groundwater as cations are rarely a cause for concern.

13. IMPLICATIONS FOR ARSENIC MITIGATION

While national surveys such as the one undertaken in this project can identify the worst- and least-affected areas and even provide estimates of the percentage of wells likely to be affected, they cannot identify the individual wells that are affected, e.g. those greater than than 50 µg As L^{-1}.

The large amount of short-range spatial (well-to-well) variation in arsenic concentrations means that ultimately all shallow wells in recent alluvium in Bangladesh need to be tested for arsenic if they are to be used for drinking water. Since timeliness is a crucial factor in any arsenic mitigation programme, the aim should be to tackle the worst-affected areas first as part of a priority programme.

Priority in the surveying and mitigation should therefore be given to the badly-affected areas identified in the south and east of Bangladesh. The resources of such a priority programme should be allocated according to the severity of the problem, based for example on the percentage of wells in a district that are affected, or on the average arsenic concentration in an area, or on the probability that the water quality standard in an area will be exceeded. Our national survey of arsenic provides one set of estimates for setting such priorities and broadly agrees with other studies of the regional distribution of arsenic.

There are areas in north-western Bangladesh that should receive low priority in terms of resources allocated because the extent of contamination there is much lower than elsewhere and, in some places, is not significant.

Our survey has highlighted some quite extensive arsenic-rich areas in northern Bangladesh, particularly in the Netrokona-Sunamganj area. Apparent arsenic hot spots in northern Bangladesh are probably best located by a combination of nationwide public awareness campaigns, backed up by rapid deployment of medical technicians and doctors trained to recognise the early symptoms of arsenic poisoning. This could be accompanied by a rapid low-density survey of every mouza (some 65,000 in all of Bangladesh) in order to identify all sizable hot spots. This could be achieved by sampling just a few wells in each mouza.

Professional statistical expertise should be sought when making estimates of arsenic distribution as many difficult technical decisions need to be taken in arriving at the best estimates, and in understanding the errors involved in these estimates. Reliable statistics enable resources to be allocated most efficiently and the most appropriate action to be taken at an early stage. The large number of well waters with arsenic concentrations at, or close to, the detection limit of our survey (below 1 µg L^{-1}) and the extreme range in concentrations found gives problems in arriving at rigorous statistical estimates of many parameters.

Other arsenic survey data collected and analysed by other agencies under comparable conditions will add to the picture and enable our survey estimates to be updated. Although it is difficult to judge the accuracy of our district-based estimates for the percentage of wells affected, the basic north-south divide appears to be confirmed by all other surveys. While the detailed picture will be refined as more testing is undertaken, it is likely that the broad regional patterns so far identified will be confirmed. Certainly the present knowledge is sufficient to define broad areas for a priority mitigation effort.

There are no long-term monitoring data for arsenic in tubewells anywhere in Bangladesh and so the probable future changes in groundwater quality are largely unknown. The best assumption for planning purposes is probably to assume that the situation will not change appreciably in the short term, and probably not also in the medium term. In the mean time, the careful monitoring of a network of a wells over a broad range of timescales is important. There are very few data for this at present, even for short timescales. Such monitoring is difficult to do because of the relatively small changes expected. There appears to be some short-term variation (over weeks and less) in the arsenic concentration of tubewells. The reasons for this are uncertain but probably reflect changes in the flow paths of the pumped water as the water table or pumping regime changes, combined with a strong stratification of water quality within the aquifer itself.

It is difficult to judge how rapidly the shallow aquifer will change in response to natural groundwater flushing. The overall long-term trend should be downwards, although in the short term, some wells might increase as a pulse of high arsenic groundwater passes through. In practice, such natural flushing is unlikely to be significant on the timescale of the tubewells, e.g. a few decades.

Increased groundwater mixing as a result of pumping is likely to accelerate and to some extent alter these natural changes. This mixing will tend to increase the concentration of arsenic in wells which presently have low arsenic concentrations and reduce it in high-arsenic wells. Because of the extremely heterogeneous nature of the aquifer (spatially), the highly skewed distribution of arsenic concentrations found and the low acceptable concentration of arsenic in drinking water, it is likely that this short-term mixing will lead to an increase in the number of wells exceeding a given water quality standard. But again, the timescale of such changes is at present very uncertain.

Our limited monitoring in three areas over approximately nine months has not identified any significant and consistent changes in water quality during that time. Rather it has highlighted the difficulties of carrying out such exercises in terms of sampling and analytical procedures. In practice, it will probably be difficult to detect changes in chemical concentrations of less than 10%.

Groundwater from the deep aquifer was sampled predominantly from the Barisal, Lakshmipur, Faridpur and Sylhet areas and was found to be essentially arsenic-free (less than 3 $\mu g\ L^{-1}$). These samples may or may not be representative of the deep aquifer in other areas. There is no reason to believe that the deep aquifer will become seriously contaminated, certainly in the short-term, providing that care is taken during borehole construction to isolate the upper and lower aquifers and so prevent direct leakage of contaminated water to the deep aquifer. Therefore deep wells provide a possible option for the long term supply of safe drinking water. Ideally the development of deep wells should be combined with a basic water distribution network that makes it economically feasible as well as being attractive and convenient to use. Existing wells could still be used for non-potable uses.

Experience gained so far indicates that water from the great majority of carefully-constructed deep wells would not only pass the current Bangladesh standard for arsenic but would pass all other existing national and international standards and guidelines for arsenic. The likelihood of a manganese exceedance is also much lower in these deep groundwaters. Most of the deep groundwaters tested in our surveys were from the southern coastal region where the shallow groundwaters are affected by salinity and these deep groundwaters may not be typical of those from elsewhere in Bangladesh. Therefore the nationwide availability and sustainability of this resource needs to be established both in terms of quality and quantity. In some areas, a stony layer at depth makes deep drilling difficult given the drilling rigs currently available in Bangladesh. The possible impact of the large-scale abstraction of irrigation water on the deep aquifer also needs to be considered.

Aside from the mitigation technology *per se*, probably the single greatest contribution that science and technology can make to solving the arsenic problem would be the development of a reliable, sensitive and affordable field-test kit for arsenic analysis at the microgramme-per-litre level. Combining any unavoidable short-term variability from the aquifer itself with the inevitable sampling and analytical errors means that samples close to the adopted arsenic standard will have quite a high probability of changing from ‘acceptable’ to ‘unacceptable’, or *vice versa*, with time. A three-tier classification would therefore be preferable: ‘definitely acceptable’, ‘maybe acceptable’ and ‘definitely unacceptable’. Several improved field-test kits are currently being developed which offer semi-quantitative visual measurements, e.g. 0, 10, 30, 50, 70, 300, and 500 $\mu g\ L^{-1}$. This will greatly improve their value.

A great premium should be placed on obtaining reliable analytical results for arsenic measurements since this will avoid the need for replication, save time wasted in tracking down errors and will give the survey the necessary credence amongst the population at large.

Attempts to reverse the geochemical processes that have given rise to the arsenic-rich groundwaters offer one approach to mitigation. In principle, this could be achieved by either (i) the in situ (re)oxidation of the groundwater and sediment to enhance arsenic sorption processes, or (ii) by inducing the in situ precipitation of pyrite and coprecipitation of arsenic with the addition of injected sulphate (as calcium sulphate). While such approaches have some attractions, there are many practical problems to be overcome before they could become viable – the large quantities of chemicals required, clogging of the aquifer and the need for a reliable electricity supply for the pumps, for example. Perhaps more importantly, Bangladesh has little track record of the successful long-term implementation of such technical solutions in rural areas, particularly where there is a need for substantial recurrent expenditure in terms of maintenance, chemicals and electricity. The absence of a distributed water supply in the rural parts of Bangladesh also significantly hinders all technological solutions to the problem.

14. Reference

DPHE/BGS/MML (1999) Groundwater Studies for Arsenic Contamination in Bangladesh. Phase I: Rapid Investigation Phase. Main Report and five supplementary volumes. Department of Public Health Engineering, Government of Bangladesh, British Geological Survey and Mott MacDonald Ltd (UK).