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# Arsenic pollution of groundwater in Bangladesh†

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**Arsenic concentrations in groundwater around the village of Samta, Jessore District, Bangladesh were measured. Distribution patterns of arsenic in groundwater were determined. Arsenic concentrations in drinking water tubewells mostly exceeded WHO guidelines. Copyright © 2001 John Wiley & Sons, Ltd.**

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## 1. INTRODUCTION

Prior to World War II, pond water was used for drinking without treatment in Bengal. After the war, many wells were installed by UNICEF aid and the population began to use well water. It is believed that, owing to arsenic contamination of the well water, 40 million people are currently exposed to the risk of arsenic poisoning.[1] Although it was only in 1994 that the first group of arsenicosis patients was found in Bangladesh, arsenic contamination of groundwater had already been reported in 1978 in the state of West Bengal in India, the neighboring country on the western border.[2] Around this time, rice growing during the dry season started in Bangladesh under the ‘green revolution’ to increase food production. For this purpose, deep tubewells for irrigation were dug and, accordingly, a large volume of groundwater has been withdrawn. It is assumed that this withdrawal of groundwater has brought about geo-chemical and physical changes underground, causing arsenic contamination of groundwater in turn.

In general, arsenic in groundwater is released from minerals and organic matter under the ground. It is thought that arsenic leaches out from those materials due to changes in the adsorption mechanism. The causes of natural leaching-out of arsenic to groundwater are considered to be due to oxidation[3,4] or reduction.[5] The oxidation theory suggests that groundwater becomes oxidized, causing oxidization of minerals, including pyrite, and finally arsenic leaches out. Bangladesh has a monsoon climate with a clear distinction between the rainy season (June through to early October) and the dry season. For this reason, there is a difference of 3–5 m in the underground water level between the rainy season and the dry season.[6] When the water table lowers in the dry season, air enters into the soil; thus, it is possible that a part of the upper sand layer turns into an oxidation zone. It is also suggested that the large number of deep tubewells (depth of about 100 m) drilled for irrigation allow atmospheric oxygen to enter into the aquifer sediments, causing oxidation zones there.

The groundwater aquifer is normally in a reduction state, and it is thought that arsenic is freed when iron oxyhydroxides ($FeO(OH)$), which are known to scavenge arsenic, are reduced in the aquifer sediments and change to iron hydroxide ($Fe(OH)_2$).[5] It is considered[7] that the withdrawal of groundwater in the dry season causes vigorous circulation of groundwater, which in turn transports arsenic. However, the full reason why arsenic leaches out into groundwater is not yet known. In

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**Table 1** Distribution of arsenic concentration in groundwater in Samta village (May 1997)

| Concentration (mg l$^{-1}$) | Number of tubewell | As (%) |
|---|---|---|
| As $\leq$ 0.01 | 10 | 3.5 |
| 0.01 < As $\leq$ 0.05 | 20 | 7.1 |
| 0.05 < As $\leq$ 0.1 | 57 | 20.2 |
| 0.1 < As $\leq$ 0.3 | 114 | 40.4 |
| 0.3 < As $\leq$ 0.5 | 39 | 13.8 |
| As > 0.5 | 42 | 14.9 |

fact, neither the mechanism of arsenic elution nor its distribution in strata are clearly identified yet.

In March 1997, we analyzed the water of all the tubewells (282) used for drinking in the village of Samta in the Jessore District, Bangladesh, where many patients of arsenicosis have been found. The result of the analysis is shown in Table 1 and Fig. 1.[8] The following facts have been confirmed from the survey: tubewells with arsenic concentrations of less than 0.01 mg l$^{-1}$ (the guideline set by the World Health Organization (WHO) and the limit set for drinking water in Japan) were only 3.5% of the total, and those within the limit (0.05 mg l$^{-1}$) in Bangladesh were approximately 10%. Moreover, highly contaminated tubewells with arsenic concentrations exceeding 0.50 mg l$^{-1}$ amounted to almost 15% of the total, with the highest concentration of 1.40 mg l$^{-1}$. It was assumed, therefore, that arsenic contamination in Samta may be one of the worst in the Ganges basin, including also West Bengal in India.[9] Also, high contaminations of arsenic were found in the south part of the village, and the concentration tended to be lower towards the north.

To clarify the distribution characteristics of the arsenic concentration, we have since carried out surveys of water quality at fixed points in Samta and observed the changes of arsenic concentration with the passage of time and the relationship between arsenic concentrations and other factors. We also analyzed arsenic concentrations of groundwater in the area surrounding Samta. This paper discusses qualitatively the characteristics of arsenic contamination of groundwater in Bangladesh and the mechanism by which arsenic leaches out.

# 2 RESEARCH AREAS AND METHOD OF ANALYSIS

## 2.1 Research areas

Research was carried out in Samta, Sarsha Thana, Jessore District and the southern part of Sarsha Thana (Fig. 2). Water sampling was done from shallow tubewells (30–50 m) in Samta in March 1997, October 1997, May 1998 and May 1999. In

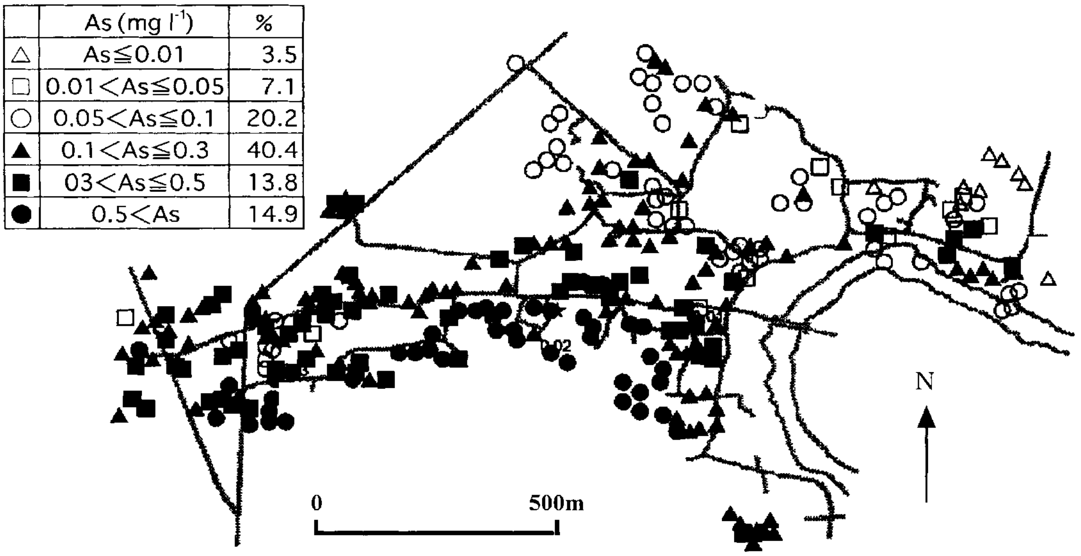

**Figure 1** Distribution of arsenic concentration in groundwater in Samta village (March 1997).

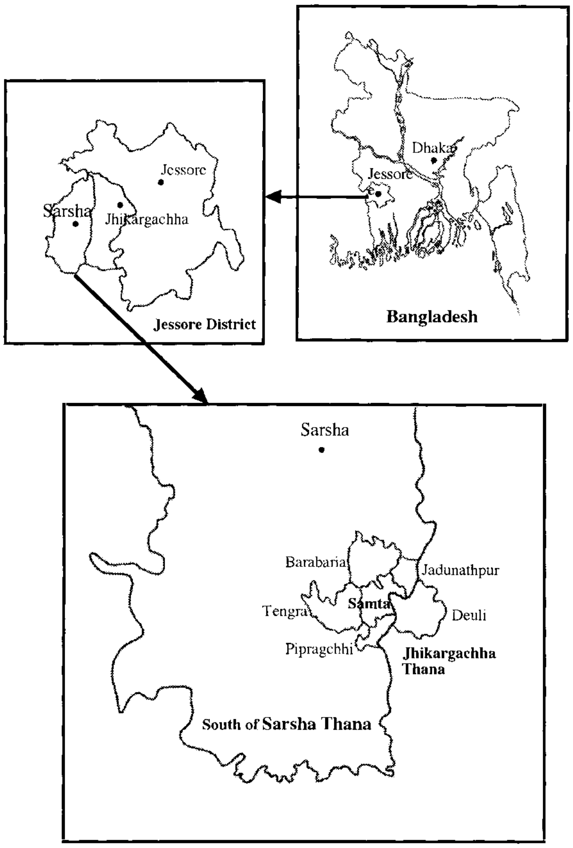

**Figure 2** Location of Samta in Sarsha Thana in Jessore District, Bangladesh.

April–May 1999, water was also collected in Tengra, Pipragchhi, Barabaria, and Jadunathpur, villages in Sarsha Thana, west, south, north and northeast of Samta respectively, and in Deuli of Jhikargachha Thana, a village east of Samta. In Samta, water was collected from deep tubewells for drinking (approximately 200 m) and irrigation wells at the same time.

## 2.2 Analysis

The following items were analyzed:

- pH and electric conductivity (EC), using a U-10 Water Quality Checker (Horiba Seisakusho);
- oxidation–reduction potential (ORP), using a TRX-90 Personal pH/ORP meter (Toko Kagaku Kenkyusho);
- arsenic(III) and arsenic(V) by colorimetry using the Gutzeit method modified by Hironaka and by graphite furnace atomic absorption spectrometry (AAS; Hitachi, Z-8000).
- iron(II) and iron(III) by phenanthroline absorption spectrophotometry[10] — color was formed at the spot and measured later by UV spectrophotomets (Beckman, DU-650);
- $HCO_3^-$, using a separation titration method;[11]
- calcium(I) and magnesium(II) using a flame atomic absorption method (Hitachi, Z-800);
- $Na^+$, $NH_4^+$, $K^+$, $F^-$, $Cl^-$, $Br^-$, $NO_3^-$ and $SO_4^{2-}$, using ion chromatography (Dionex QIC analyzer);
- elution test of drilling core samples, using the method prescribed by the Japanese Government Environment Agency.[12]

## 2.3 Analytical methods for arsenic

The analysis of arsenic was carried out by the Gutzeit method modified by Hironaka at the site and later by graphite furnace AAS (Hitachi, Z-8000). In the Gutzeit method to arsenic compound is reduced by potassium iodide and stannous chloride in acid solution, and then liberated as arsine by hydrogen. The arsine generated produces a yellow–brown stain on mercury bromide disk paper, and the arsenic concentration is measured by the color. In the Hironaka method the reagents other than hydrochloric acid are powdered and color is obtained on a filter paper soaked with mercury bromide, so that analysis can be done easily at the site. The time required for analysis is about 15 min per sample, but five or six samples are measured at one time. A color chart is prepared with divisions varying by 0.01 mg l$^{-1}$ for concentrations between 0 and 0.10 mg l$^{-1}$, and by 0.1 mg l$^{-1}$ for concentrations between 0.1 and 1.5 mg l$^{-1}$. This is suitable for rapid analysis at the site.

Since the Hironaka method uses a colorimetric judgement by eye, there is concern that there may be differences by individuals in judging colors. Figure 3 shows the result of a sensory test amongst 77 students of Miyazaki University in July 1997.[13] Judgement of 0.08 mg l$^{-1}$ on the color chart ranged from 0.05 to 0.10 mg l$^{-1}$, and the percentage of those who made the correct distinction (0.08 mg l$^{-1}$) was 35%. Among the rest, 48% of the students distinguished it lower than 0.08 mg l$^{-1}$ and 17% of them higher. As for a color sample of 0.1 mg l$^{-1}$, although the judgement was in the range of 0.06–0.15 mg l$^{-1}$, 83% of the students judged it as 0.1 mg l$^{-1}$. For a color sample of 0.2 mg l$^{-1}$, judgement was scattered in the range

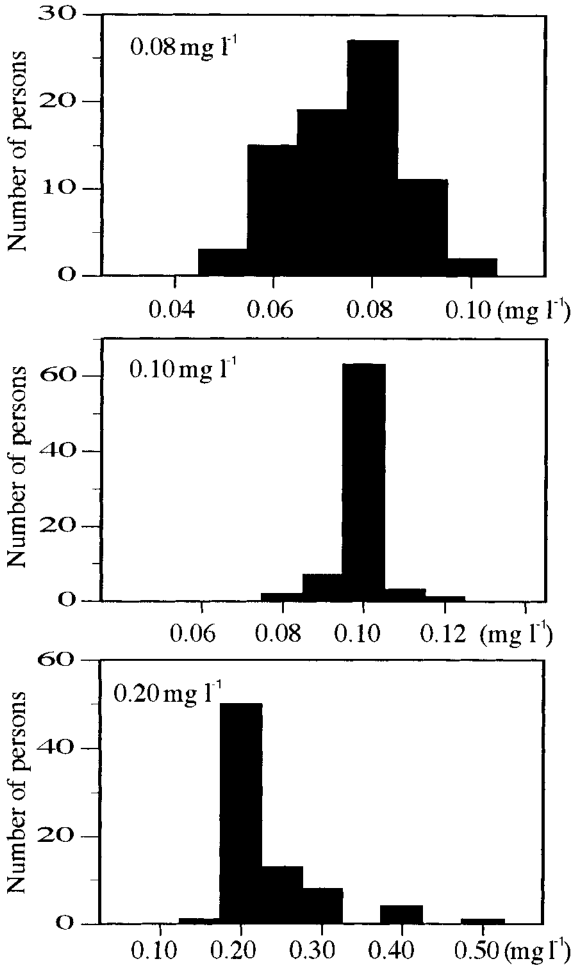

**Figure 3** Sensory test by panelists: simple colorimetry of arsenic by mercury bromide paper disk method modified by Hironaka.

0.15–0.4 mg l$^{-1}$; 64% of the students judged it as 0.2 mg l$^{-1}$, whereas most of the rest judged it higher than 0.2 mg l$^{-1}$. It is assumed that with the Hironaka method people tend to judge arsenic concentrations lower than the actual concentration in case of less than 0.1 mg l$^{-1}$, correctly in the case of 0.1 mg l$^{-1}$, and higher in the case of 0.2 mg l$^{-1}$.

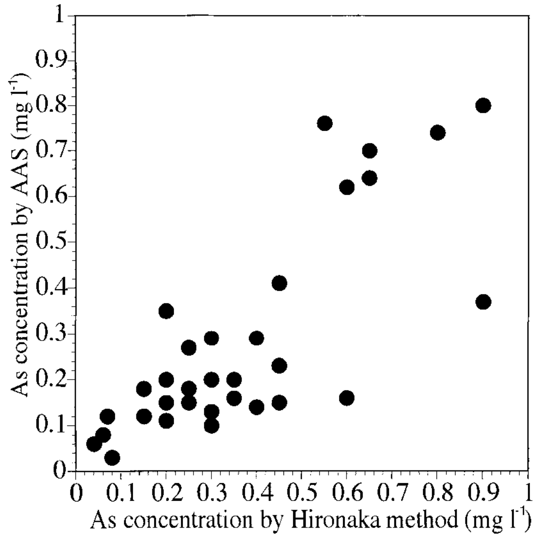

**Figure 4** Comparison of arsenic concentrations between Hironaka method and graphite furnace AAS.

Figure 4 shows the difference in arsenic concentrations of 33 tubewell water samples collected in Samta in October 1997 as measured by the Hironaka method and AAS. From Fig. 4 it can be said that the Hironaka method measured lower in cases below 0.1 mg l$^{-1}$, higher in cases of 0.1–0.6 mg l$^{-1}$, and was in conformity with measurements by AAS in the case of concentrations above 0.6 mg l$^{-1}$. This result also accords with that of the sensory test mentioned above.

Measurement on the spot is easy and simple, compared with analysis at a laboratory, since neither treatment at the time of water collection nor care for preservation are necessary. It is considered, therefore, that the Hironaka method is applicable for measurement of arsenic in the field if considered with an understanding of the above-mentioned tendency of measured concentrations. In this report, figures obtained by the Hironaka method are utilized unless specifically mentioned otherwise.

## 3 RESULTS

### 3.1 Change of arsenic concentrations of groundwater with time

Figure 5 shows the change of arsenic concentrations in Samta with the passage of time, obtained by measuring arsenic concentrations in water from 12 tubewells selected as fixed point observations. Measurement was carried out in March and October 1997, April 1998 and May 1999. The concentrations in October 1997 were those in the rainy season and the rest were in the dry season. Arsenic concentrations were higher in October 1997 (the

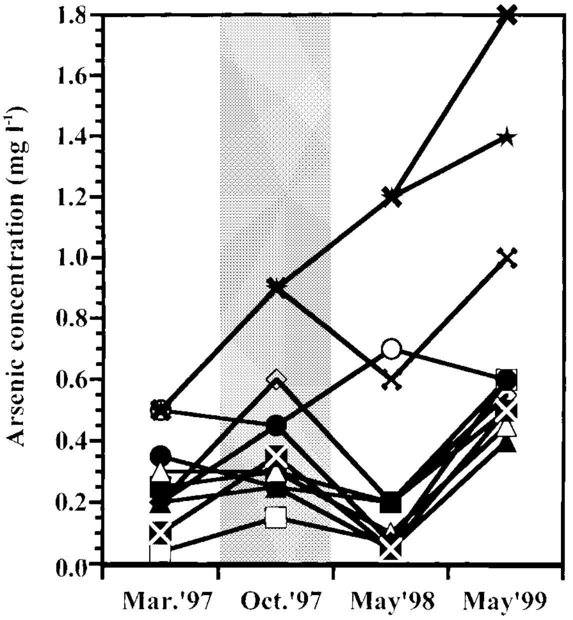

**Figure 5** Change of arsenic concentration of groundwater in Samta with time. Each symbol shows an individual tubewell.

rainy season) than March 1997 (the dry season) and became lower again in April 1998 (the dry season), from which it can be assumed that arsenic concentrations tend to become higher in the rainy season.

As a result of drilling (see Section 4), it was known that the top stratum in Samta consists of a clay layer about 10 m deep with a sand layer following and that the clay layer is arsenic rich. It is possible, therefore, to assume that arsenic concentrations in groundwater in the rainy season become higher than in the dry season, since the arsenic in the clay layer easily leaches out in the rainy season when the water table rises. Moreover, it was also confirmed that the flow of groundwater becomes strong in some areas,[14] and it is very possible that arsenic is transported by the flow of groundwater. From Fig. 5, it is also noted that arsenic concentrations in groundwater seem to increase with the passage of time and that arsenic concentrations of most of the sample wells became high in May 1999. The cause is currently under consideration.

## 3.2 Water quality and arsenic concentration in Samta

The results of the water quality analysis for May 1998 are shown in Table 2. Some features of note are that the concentrations of $HCO_3^-$ and iron were 179–432 mg $l^{-1}$ and 3.9–10.5 mg $l^{-1}$ respectively, much higher than those of normal groundwater, whereas the concentrations of $SO_4^{2-}$ were within the range 0–7.9 mg $l^{-1}$, with most of the samples below 2.0 mg $l^{-1}$, which can be considered very low. Further, the pH was between 7.1 and 7.6, showing alkalescence, and the ORP values were negative, showing a reduction state.

Figures 6 to 8 illustrate the distribution of

**Table 2** Water quality in groundwater in Samta village (May 1998)

| | Tubewell for drinking | Deep tubewell for irrigation | Deep tubewell for drinking |
|---|---|---|---|
| pH | 7.1–7.6 | 7.2–7.3 | 7.5–7.6 |
| EC (μs $cm^{-1}$) | 574–1192 | 806–1228 | 968–1373 |
| ORP (mv) | −88 to −6 | −52 to −3 | −50 to 3 |
| $As^{3+,5+}$ (mg $l^{-1}$) | 0.03–1.2 | 0.03–0.2 | 0.05–0.08 |
| $Fe^{2+}$ (mg $l^{-1}$) | 3.9–10.5 | 4.2–6.3 | 1.1–1.4 |
| $Fe^{3+}$ (mg $l^{-1}$) | 0–1.0 | 0–0.6 | 0 |
| $HCO_3^-$ (mg $l^{-1}$) | 179–432 | 340–358 | 381–392 |
| $NH_4^+$ (mg $l^{-1}$) | 0.1–8.4 | 3.0–8.1 | 0.5–2.0 |
| $K^+$ (mg $l^{-1}$) | 2.1–14.7 | 3.1–10.9 | 5.1–15.8 |
| $Mg^{2+}$ (mg $l^{-1}$) | 13–48 | 27–34 | 36–47 |
| $Ca^{2+}$ (mg $l^{-1}$) | 51–100 | 49–57 | 40–43 |
| $F^-$ (mg $l^{-1}$) | 0–4.4 | 0–1.3 | 1.0–2.4 |
| $Cl^-$ (mg $l^{-1}$) | 0–58.3 | 4.0–14.3 | 26.5–48.4 |
| $Br^-$ (mg $l^{-1}$) | 0–0.10 | 0–0.02 | 0.05–0.15 |
| $NO_3^-$ (mg $l^{-1}$) | 0–0.40 | 0.01–0.08 | 0.03–0.05 |
| $SO_4^{2-}$ (mg $l^{-1}$) | 0–7.9 | 0.1–0.3 | 0.2–0.5 |

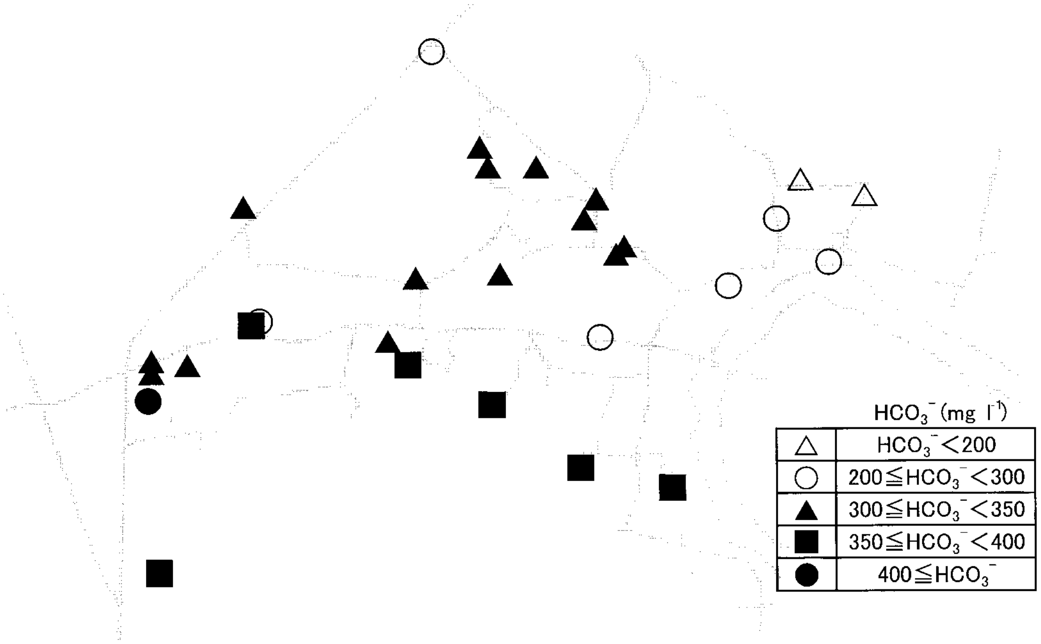

Figure 6 Distribution of $HCO_3^-$ in groundwater in Samta village (May 1998).

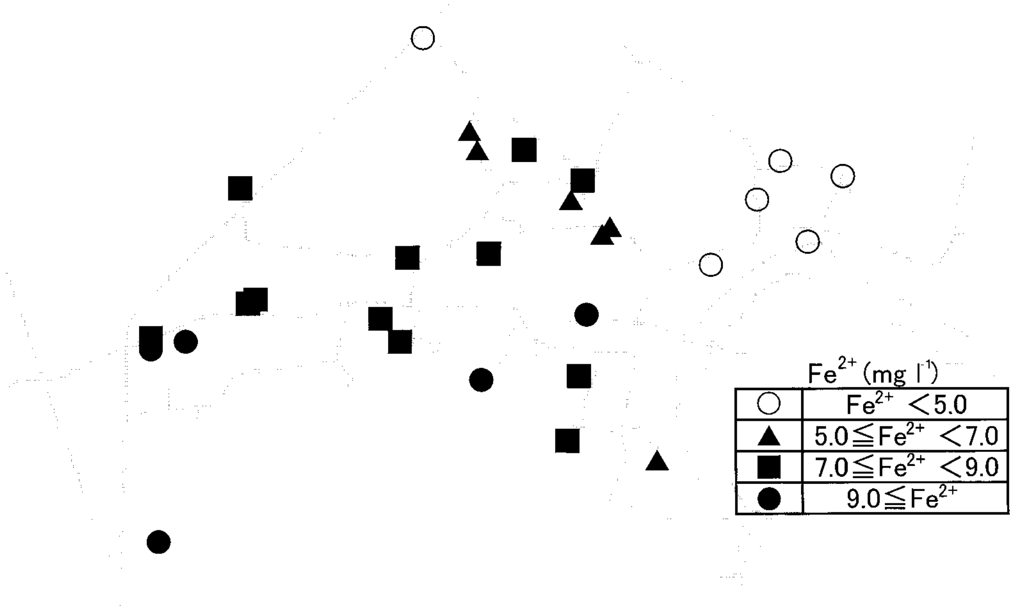

Figure 7 Distribution of iron(II) in groundwater in Samta village (May 1998).

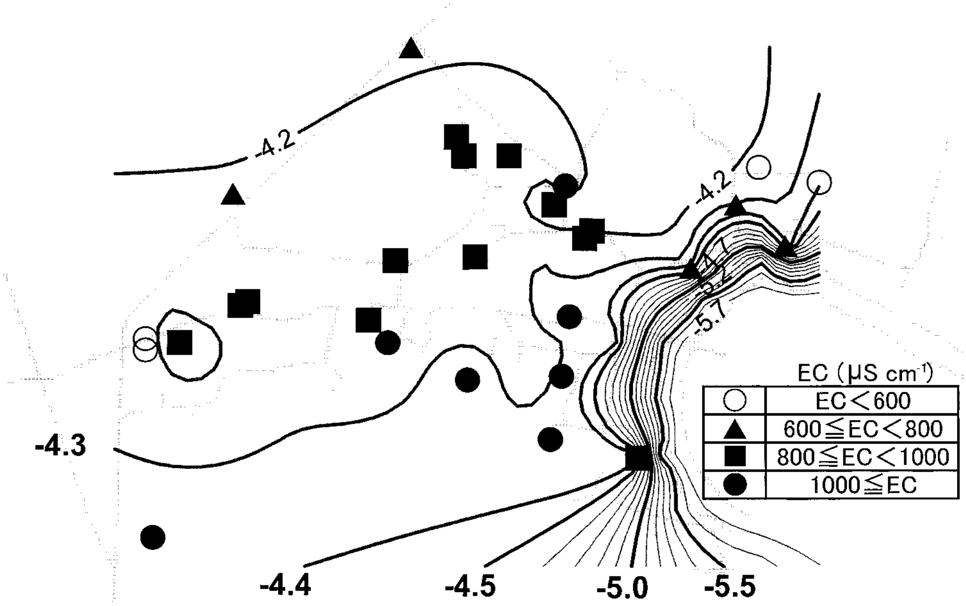

**Figure 8** Distribution of EC of groundwater in Samta village (March 1998).

$HCO_3^-$, iron(II) and EC respectively. When comparing Figs 6 to 8 with Fig. 1, it can be observed that well water with high arsenic concentrations contains high amounts of $HCO_3^-$ and iron(II) and has high EC, whereas in wells with low arsenic concentrations their amounts were low.

$HCO_3^-$ is formed in various ways, such as dissolution of $CO_2$ from atmosphere, decomposition of organic matter, or reduction of $SO_4^{2-}$ by a reducing bacterium. Here it is considered in relation to the decomposition of carbonate. $HCO_3^-$ was in the range of 2.9–7.8 meq $l^{-1}$ whereas $Ca^{2+}$ and $Mg^{2+}$ were 40–130 mg $l^{-1}$ and 13–52 mg $l^{-1}$ respectively. The total content of $Ca^{2+}$ and $Mg^{2+}$ becomes 4.3–10.7 meq $l^{-1}$ and almost accords with the content of $HCO_3^-$. From the analysis of drilling samples in Samta, Akai and coworkers confirmed the presence of calcite ($CaCO_3$) and dolomite ($CaMg(CO_3)_2$) in various alluvial sediments.[15,16] From these findings it is assumed that $HCO_3^-$ was formed by decomposition of calcite and dolomite.

In the theory suggesting oxidation to be the cause of arsenic release, hydrogen is produced in response to oxidation of pyrite and the pH should be low. However, the pH value was as high as 7.1 to 7.6, as mentioned previously, which opposes the oxidation theory. If, however, calcite and dolomite are decomposed as stated above, the pH value does not reduce since hydrogen is consumed during the decomposition.[17] Considering such a reaction, the agreement in the distribution of arsenic concentrations and those of $HCO_3^-$ content would be self-evident. Furthermore, it is observed from Fig. 9 that pH tends to be low in well water with high arsenic concentrations and high in those with low arsenic concentrations. Although the presence of high $HCO_3^-$ content thus supports the theory of oxidation of pyrite in part, it is not consistent with the low content of $SO_4^{2-}$ explained above, even considering that $SO_4^{2-}$ formed during oxidation of pyrite is consumed for decomposition of dolomite. Further studies are required to clarify the decrease of $SO_4^{2-}$ by observing other mechanisms, such as reduction to $H_2S$ in a reduction environment.

On the other hand, the iron(II) content was high in groundwater with high arsenic concentrations, and the fact that groundwater generally showed negative ORP values supports a reduction theory. In addition, as shown in Fig. 10, most iron is present in the form of iron(II), and the relationship between iron(II) and arsenic concentration is strong. With regard to the high amounts of $HCO_3^-$, it can be suggested that this was formed by reduction of $SO_4^{2-}$ and $NO_3^-$.

From the contour lines (m) of the water levels depicted in Fig. 8, it is considered that groundwater flows from the northern towards the southern part of the village, and then into the river to the east. Since

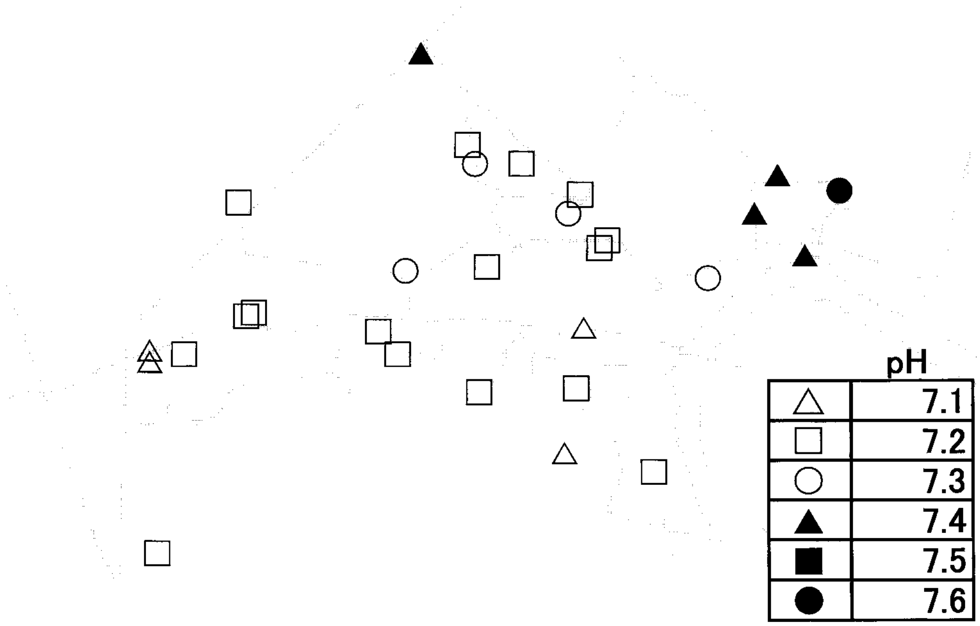

**Figure 9** Distribution of pH of groundwater in Samta village (May 1998).

the EC value is high in the south, where the arsenic concentration is also high, it is assumed that the amount of dissolved ion is large and groundwater stays temporarily, in the south. The distribution in the EC is, therefore, in accordance with the flow of groundwater.

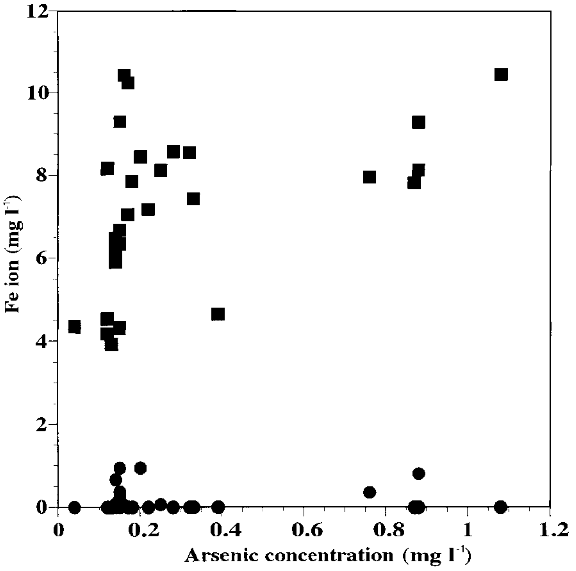

**Figure 10** Relationship between concentration of iron ions and concentration of arsenic ions in well water in Samta: ■, iron(II); ●, iron(III).

Other dissolved components, such as $NH_4^+$ (0.1–8.0 mg $l^{-1}$), $Na^+$ (11–108 mg $l^{-1}$), $K^+$ (2–16 mg $l^{-1}$), $Cl^-$ (0–65.5 mg $l^{-1}$), $Br^-$ (0–0.2 mg $l^{-1}$), $F^-$ (0–4.5 mg $l^{-1}$) and $NO_3^-$ (0–0.4 mg $l^{-1}$), were also detected in groundwater in Samta. Iron and $F^-$ were above the limits set for drinking water in Japan. As seen in Table 2, iron exceeded the limit (below 0.3 mg $l^{-1}$) in all but one deep tubewell, which was newly drilled for drinking purposes, and $F^-$ was above the limit (below 0.8 mg $l^{-1}$) in 74% of the tubewells examined. A high $NH_4^+$ content may indicate direct inflow of excreta to well water. It is necessary, therefore, to assess the whole water quality, including other dissolved components and bacteria, for the supply of safe drinking water.

## 3.3 Arsenic concentration in villages neighboring Samta

In Samta, as illustrated in Fig. 1, high arsenic concentrations were found in the south of the village and the concentration tended to be lower towards the north. To evaluate arsenic distribution in perspective, arsenic was measured over a wider area with the emphasis on the southern part of Sarsha Thana to which Samta belongs. The distribution of arsenic (May 1999) in this wider

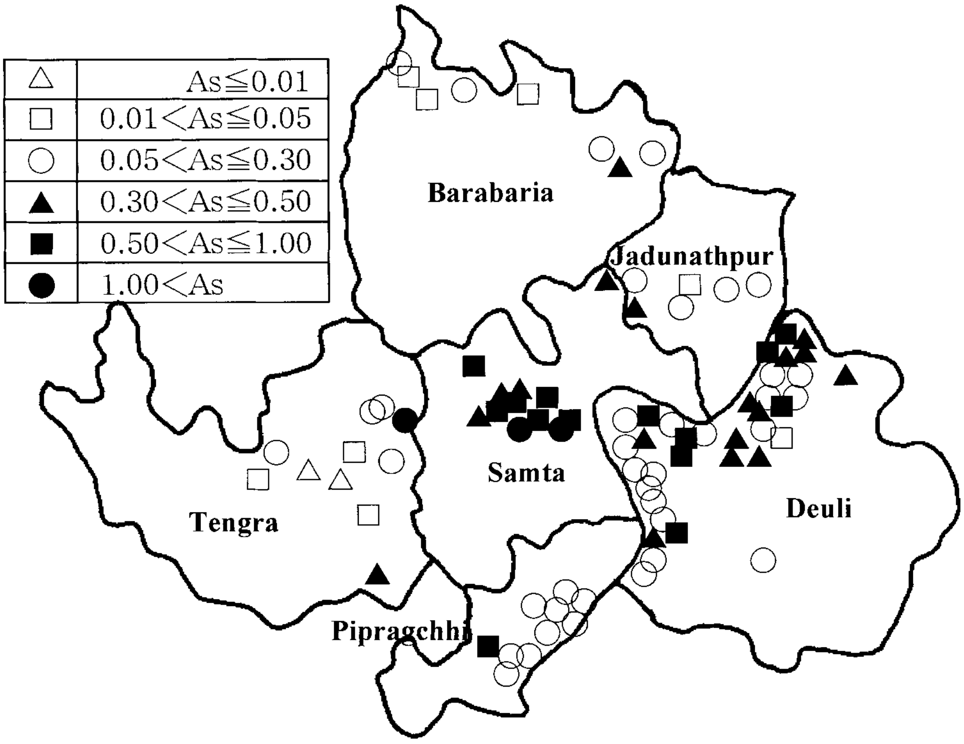

**Figure 11** Distribution of arsenic concentration in groundwater in villages surrounding Samta (March 1999).

area is shown in Fig. 11. Concentrations ranged from 0.40 to 1.80 mg $l^{-1}$ in Samta (12 locations), in Tengra (11 locations) from 0.01 to 0.40 mg $l^{-1}$, in Pipragchhi (nine locations) from 0.10 to 0.90 mg $l^{-1}$, Barabaria (nine locations) from 0.03 to 0.40 mg $l^{-1}$, and in Jadunathpur (eight locations) from 0.05 to 0.45 mg $l^{-1}$. In Deuli of Jhikargachha Thana, a village east of Samta, well water was analyzed at 39 locations and arsenic concentration was in the range of 0.01 to 0.90 mg $l^{-1}$. In Samta, water was collected from deep tubewells used for drinking water (approximately 200 m) and irrigation wells at the same time.

These results show that arsenic concentration is high in the south of Samta and in the east (Deuli), with the highest concentration of 1.80 mg $l^{-1}$ in Samta. The results also revealed a condition of arsenic contamination there is no tubewell that meets the WHO guideline (0.01 mg $l^{-1}$) and only 20% of the tubewells examined were within the limit in Bangladesh (0.05 mg $l^{-1}$). It is also noted that arsenic contamination of groundwater in the area centers around Samta. However, the relationship between arsenic distribution in Samta (Fig. 1) and that of the wider area (Fig. 11) is not fully understood. Further research by increasing measurement points is required in this regard.

## 4 ARSENIC CONTENT AND ELUTION TESTS OF SAMPLES FROM DRILLING CORES

The Research Group for Applied Geology conducted a survey in Samta in May 1998, drilling the ground to a depth of 64 m.[18] Using core samples from the drilling, an elution test was carried out according to the Bottom Sediment Test Method set by the Environment Agency of Japan. The soil sample was mixed with the solvent, which was prepared by adding hydrochloric acid to pure water to adjust the pH to 5.8–6.3 in advance, with a weight/volume ratio of 1:10. The test sample was then shaken for 6 h continuosly in a vibratory apparatus at a frequency of 200 times/min, and the elution sample liquid was filtered after centrifugation for measurement by AAS. The results of the elution test are shown in Fig. 12 together with those

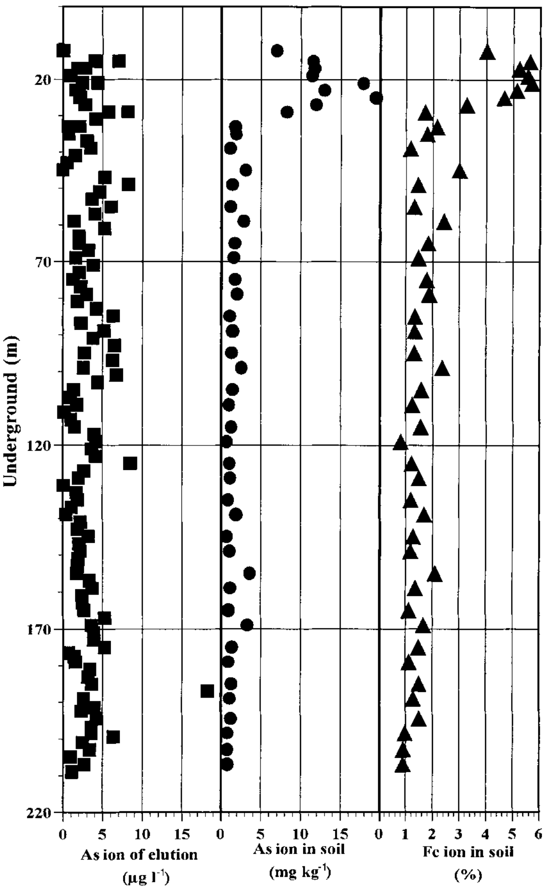

**Figure 12** Arsenic elution, content and iron content of bore core samples in Samta. Left: relation between depth underground and arsenic ions from elution test. Center: relation between depth underground and arsenic ions in soil. Right: relation between depth underground and iron ions in soil.

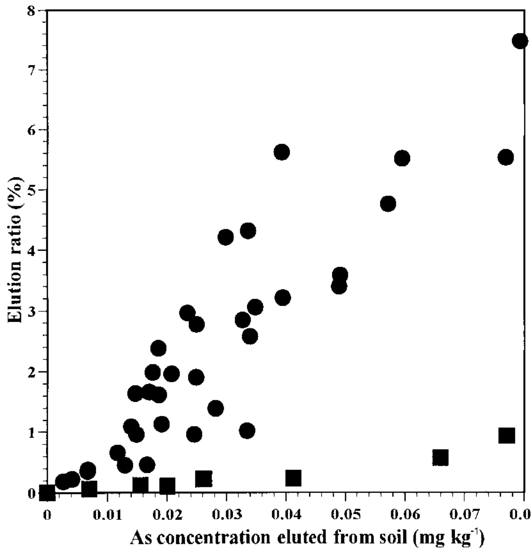

**Figure 13** The ratio of arsenic elution concentration (mg/kg$^{-1}$) to arsenic content of samples (mg/kg$^{-1}$): ■, clay layer; ●, sand layer.

of arsenic and iron content tests conducted by Kubota et al.[19] using the same core samples.

From the tests it was found that upper clay layers (2.5–8.0 m deep) contain arsenic in the range 7–20 mg kg$^{-1}$ and sand layers (8.0–64.0 m deep) in the range 0.07–3.6 mg kg$^{-1}$, which correlates with the result of the iron content test carried out at the same time. In the elution test, the maximum values for arsenic elution were 8 μg l$^{-1}$ in the clay layer and 0.6 μg l$^{-1}$ in the sand layer. Figure 13 shows the elution ratio based on both the arsenic elution test and the content test. The figures on the abscissa were obtained by converting the result of the elution test to 1 kg of soil, and those on the ordinate were obtained by dividing them by the arsenic content in the soil. In the clay layer the elution ratio was below 1%, whereas in the sand layer most of the samples showed a value of several percent, with one exceeding 7%.[20]

The results of the content tests indicate arsenic elution from the upper clay layer, but, from the elution ratio shown in Fig. 13, elution from the sand layer cannot be denied. However, arsenic elution in the ground is related to the values of pH and ORP, and other conditions under which more arsenic leaches out from the clay layer may exist underground. Assessment of elution needs further consideration.

## 5 CONCLUSIONS

As a result of our surveys of water quality at fixed points in Samta since March 1997 and analysis of arsenic concentrations of groundwater in the area surrounding Samta, we have found the following to be the characteristics of arsenic contamination of groundwater and the mechanisms of arsenic elution.

(1) Arsenic concentration of groundwater in Samta is higher in the rainy season than in the dry season, and seems to increase with time.
(2) Arsenic contamination is observed in the

wider surrounding area, although not so high as in Samta, and the distribution of arsenic contamination appears to spread with Samta as the center point.

(3) It was characteristic that $HCO_3^-$ and iron(II) contents of groundwater were high and their distribution was in accordance with that of arsenic contamination. Although it was not possible to identify fully the mechanism of arsenic elution from water analyses, the distribution of dissolved components can be explained by both oxidation and reduction theories.

The mechanism of most groundwater contamination worldwide due to naturally occurring arsenic is yet to be clarified. At present, arsenic contamination is increasing with time. However, many people use arsenic-contaminated water for drinking and cooking. It is hoped that the causes of arsenic contamination will be urgently identified and that a supply system of safe water can be established quickly.

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