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Mechanism of arsenic release to groundwater, Bangladesh and West Bengal

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Abstract

In some areas of Bangladesh and West Bengal, concentrations of As in groundwater exceed guide concentrations, set internationally and nationally at 10 to 50 μg l^{-1} and may reach levels in the mg l^{-1} range. The As derives from reductive dissolution of Fe oxyhydroxide and release of its sorbed As. The Fe oxyhydroxide exists in the aquifer as dispersed phases, such as coatings on sedimentary grains. Recalculated to pure FeOOH, As concentrations in this phase reach 517 ppm. Reduction of the Fe is driven by microbial metabolism of sedimentary organic matter, which is present in concentrations as high as 6% C. Arsenic released by oxidation of pyrite, as water levels are drawn down and air enters the aquifer, contributes negligibly to the problem of As pollution. Identification of the mechanism of As release to groundwater helps to provide a framework to guide the placement of new water wells so that they will have acceptable concentrations of As. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

Following independence, the governments of Bangladesh, assisted by aid agencies, have provided most of the population with bacteriologically-safe drinking water by providing tubewells that abstract water from subsurface alluvial aquifers. This achievement has reduced the incidence of waterborne disease only to replace it with another problem: water from many of the tubewells is contaminated with naturally-occurring As (Saha and Chakrabarti, 1995; Dhar et al., 1997; Bhattacharaya et al., 1997, 1998a, 1998b; Nickson et al., 1998). Concentrations of As in water from tubewells can reach mg l^{-1} levels (Badal et al., 1996) and frequently exceed both the provisional guideline concentration for drinking water set by the World Health Organisation (10 μg l^{-1} WHO, 1994) and the Bangladesh limit for As in drinking water (50 μg l^{-1}; Department of the Environment, Bangladesh, 1991). The problem seems likely to affect a significant proportion of the 3–4 million tubewells in Bangladesh (Arsenic Crisis Information Centre; http://bicn.com.acic/, 15/05/99).

Whilst the calamity may be alleviated by using water from other sources for public supply (e.g. rain or surface water), the attendant storage and bacteriological problems make this difficult. The authors believe that by identifying the chemical and geological processes that give rise to As contamination, it might be possible to use that knowledge in a predictive manner to site

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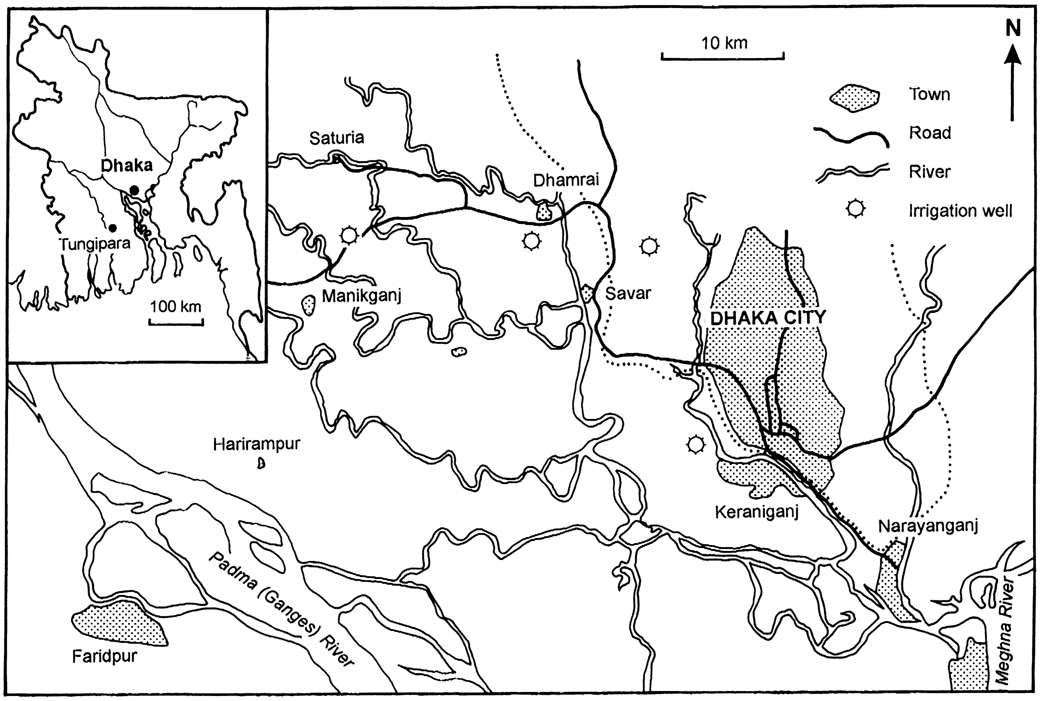

Fig. 1. Conurbations in Bangladesh that were sampled for this study; the scale does not permit individual wells to be differentiated, excepting for 4 irrigation wells outside of town sites. Tungipara, in the district of Gopalganj, is 100 km SW of Dhaka (inset). The area within the dotted line marks the border of the Madhupur Tract.

new tubewells and possibly to remediate existing tubewells, so as to continue the development of a groundwater resource that is bacteriologically safe. As a contribution to this end, it is shown here that the As present in Bangladesh groundwater cannot derive from the presently accepted mechanism, whereby water-level drawdown from abstraction allows atmospheric O_2 into the aquifer and so allows the oxidation of As-bearing pyrite, with a concomitant release of As to groundwater (Das et al., 1995, 1996; Roy Chowdhury et al., 1998). Such a mechanism is incompatible with the redox chemistry of the waters. Arsenic produced this way would be adsorbed to FeOOH, the product of oxidation (Mok and Wai, 1994; Thornton, 1996; references therein), rather than be released to groundwater. The As in the groundwater derives from reductive dissolution of As-rich Fe oxyhydroxide that exists as a dispersed phase (e.g. as a coating) on sedimentary grains. The reduction is driven by microbial degradation of sedimentary organic matter and is the redox process that occurs after microbial oxidation of organic matter has consumed dissolved-O_2 and NO_3.

2. Sedimentological setting

Fluvial and deltaic sediments up to 10km in thickness underlie much of Bangladesh (Khan, 1991). Upwards fining sequences from braided river deposits to meander deposits and ultimately to floodplain deposits are common (Ghosh and De, 1995). The nature of fluvial deposits, however, makes difficult the definition of laterally continuous or contiguous sedimentary layers.

The evolution of the most recent parts of the sedimentary sequence in the Ganges Alluvial Plain have been discussed by Davies (1989, 1994) and Umitsu (1985, 1993). During the last glacial maximum (18 ka BP), the base-level of the rivers was some 100 m lower than in interglacial times. During this low-stand of sea-level, the sediments were flushed and oxidised, thereby giving rise to their characteristic red/brown colour. The Madhupur Tract (underlying Dhaka city) and the Barind Tract are two areas of Plio-Pleistocene sediment that survived this period of erosion. As sea level rose, late Pleistocene-Holocene sediment infilled the valleys with fluvial sands, silts and clays.

3. Material and methods

During May and June, 1997, groundwaters were sampled from 17 wells in Dhaka City that tap the Plio-Pleistocene Dupi Tila aquifer of the Madhupur

Table 1
Chemical composition of well waters from Bangladesh[a]

Location	Type	Screen (m) top	Screen (m) base	pH	E.C. (μS cm^{-1})	DO_2(%)	Temp (°C)	Na (mg l^{-1})	K (mg l^{-1})	Ca (mg l^{-1})	Mg (mg l^{-1})	Fe (mg l^{-1})	Mn (mg l^{-1})	HCO_3 (mg l^{-1})	Cl (mg l^{-1})	SO_4 (mg l^{-1})	NO_3 (mg l^{-1})	As (μg l^{-1})
Dhaka																		
Banam Rd. 18	PTW	75	157	5.95	180	66	28.9	17.1	1.7	15.5	4.90	< 0.03	0.03	81	5.7	0.8	1.0	< 10
Nayanagar	PTW	68	138	6.38	210	10	28.9	21.4	1.6	16.9	5.20	0.04	0.01	103	5.7	0.6	< 0.3	< 10
Mohakhali DOHS	PTW	63	132	5.99	210	22	26.2	16.8	1.7	27.3	6.69	< 0.03	0.02	81	13.0	0.7	3.8	< 10
Goran-1	PTW	70	173	6.14	210	51	26.1	20.9	1.5	16.5	7.21	0.11	0.08	122	3.7	< 0.1	0.9	< 10
Magdapara	PTW	67	165	6.06	210	82	26.4	20.4	1.5	16.7	6.21	< 0.03	0.07	116	4.6	1.9	0.3	< 10
Bijoynagar OHT	PTW			6.31	340	84	26.0	21.9	2.0	29.6	11.9	< 0.03	0.09	122	25.4	14.6	10.1	< 10
Armanitala	PTW			7.00	470	85	26.2	27.6	2.4	49.5	19.3	0.07	0.12	222	41.2	7.2	< 0.3	< 10
Nilkhet W.H.	PTW			5.97	500	21	25.5	29.1	2.1	48.8	1.39	0.07	0.05	117	56.0	31.8	21.8	< 10
Dhanmondi No.8	PTW			6.08	460	122	25.2	31.0	1.7	44.4	1.39	< 0.03	0.10	164	50.1	15.5	9.7	< 10
Pallabi	PTW	58	144	6.04	210	58	25.8	17.8	1.5	20.1	0.61	< 0.03	0.03	105	9.8	0.6	3.0	< 10
Sec. 10	PTW	60	133	5.82	250	116	26.4	16.4	1.3	21.9	0.70	< 0.03	0.04	101	13.4	0.1	3.8	< 10
BIBM Mirpur	PTW	57	114	5.51	180	105	25.3	15.1	2.0	14.6	0.41	0.06	0.08	66	12.9	0.3	8.9	< 10
Shamali	PTW	60	170	5.85	240	61	25.4	20.1	1.7	24.0	0.64	< 0.03	0.01	109	11.3	3.3	< 0.3	< 10
Ulan	PTW	59	158	6.25	330	148	24.8	53.9	1.1	14.5	0.43	< 0.03	0.03	152	22.2	2.8	1.4	< 10
Hazaribagh-4	PTW	69	141	6.76	540	27	26.3	43.5	1.9	45.0	16.9	< 0.03	0.09	179	46.8	31.6	< 0.3	< 10
WAPDA colony	PTW	68	149	6.64	360	26	25.8	26.2	1.6	31.8	11.5	0.29	0.14	150	25.3	9.9	< 0.3	< 10
Tejgaon	PTW	67	146	6.40	330	82	25.9	24.3	2.0	27.7	9.02	0.11	0.05	86	56.7	19.5	20.9	< 10
Narayanganj																		
Palpara 992/2	HTW		58	6.89	970	1	27.6	150	5.8	46.9	33.5	0.46	2.39	478	121	< 0.1	< 0.3	< 10
Palpara 307/3	HTW		58	6.74	960	12	25.8	151	2.5	48.7	31.7	1.71	2.66	477	105	< 0.1	< 0.3	17
Palpara 311	HTW		58	6.84	960	12	26.6	151	2.2	44.9	32.3	0.37	2.31	465	132	3.3	1.3	< 10
Majdair	PTW	128	170	6.25	860	12	26.8	72.8	3.1	76.6	35.2	0.21	0.59	199	211	8.8	< 0.3	< 10
Dewbogs OHT	PTW	28	68	6.58	840	0	27.8	110	3.3	49.9	27.1	2.46	1.81	353	147	5.8	< 0.3	< 10
Baruhall Pump	PTW	128	177	6.55	250	0	26.3	24.6	1.6	26.4	11.9	0.05	0.10	158	12.5	34.4	< 0.3	< 10
Manikganj																		
Prod. Well 1	PTW	91	116	6.72	510	0	25.2	27.1	4.0	66.9	21.9	8.09	0.25	375	11.4	< 0.1	< 0.3	77
Prod. Well 2	PTW	75	99	6.97	480	0	25.3	21.4	4.2	68.9	22.5	8.36	0.25	381	11.0	< 0.1	< 0.3	95
Well 6 (Beutha)	HTW	37		6.92	690	18	24.9	13.8	2.7	103	33.5	10.4	0.46	491	8.1	39.7	< 0.3	58
Well 11 (Dashora)	HTW	23		6.92	670	20	24.5	19.5	4.0	92.2	29.8	10.2	0.50	386	58.4	0.7	< 0.3	60
Well 10 (Dashora)	11M	42		6.92	550	4	24.2	10.4	3.9	69.5	25.6	10.1	0.94	322	12.5	34.4	< 0.3	47
Savar																		
Irrigation well No.6	IRR	31	68	5.88	300	30	26.0	7.5	2.0	6.4	1.54	0.03	0.02	27	2.8	< 0.1	9.0	< 10

(continued on next page)

Table 1 (*continued*)

Location	Type	Screen (m)		pH	E.C. (µS cm^{-1})	DO$_2$(%)	Temp (°C)	Na (mg l^{-1})	K (mg l^{-1})	Ca (mg l^{-1})	Mg (mg l^{-1})	Fe (mg l^{-1})	Mn (mg l^{-1})	HCO$_3$ (mg l^{-1})	Cl (mg l^{-1})	SO$_4$ (mg l^{-1})	NO$_3$ (mg l^{-1})	As (µg l^{-1})
		top	base															
Dhamrai																		
Well 74	IRR	26	56	6.77	420	7	25.1	16.5	2.4	50.3	18.9	10.4	0.98	302	7.8	0.2	< 0.3	14
Saturia																		
Well 58	RR	28	52	7.00	520	14	25.2	11.8	3.7	70.6	22.7	8.00	1.19	355	10.8	0.2	< 0.3	34
Keraniganj																		
Cornakhula	ITW		68	7.14	600	0	24.9	26.8	3.6	105	48.0	10.4	0.12	382	61.4	55.6	1.8	< 10
Sakta No. 3	IRR	38	87	6.84		0	24.9	25.2	3.0	78.1	24.3	15.9	0.86	399	40.9	16.9	< 0.3	49
Harirampur																		
H.C. PTW	PTW		> 90	7.04		0	25.5	20.9	4.9	105	36.9	21.8	0.20	597	9.0	0.2	< 0.3	159
H.C. HTW	HTW	18	30	7.26		0	25.3	11.3	4.1	111	24.3	3.07	0.82	399	4.3	35.7	< 0.3	107
T.H. PTW	PTW		90	7.17		6	25.4	12.7	4.1	115	31.6	5.87	0.51	508	4.9	1.0	< 0.3	164
T.H. HTW	HTW			7.17		0	24.9	13.7	4.5	137	28.3	7.63	1.27	577	3.3	< 0.1	< 0.3	152
Faridpur																		
PTW 12 (after)	PM	70	100	7.40	900	0	26.0	35.6	4.8	127	36.1	< 0.03	0.16	542	22.1	0.4	< 0.3	42
PTW 12 (before)	PTW	70	100	7.12	870	0	26.0	29.8	4.7	110	36.3	7.82	0.19	580	14.2	< 0.1	< 0.3	220
Jhiltuli HTW	HTW		9	7.25	950	0	25.2	29.8	4.5	142	28.1	0.12	0.69	504	47.7	< 0.1	< 0.3	26
PTW 10	PTW	64	98	7.11	1010	0	26.1	49.1	5.4	123	36.2	6.87	0.31	595	37.2	< 0.1	< 0.3	191
PTW 11 (new)	PTW	52	83	7.07	980	0	25.6	48.6	5.7	111	40.2	6.90	0.14	639	18.0	< 0.1	< 0.3	268
HTW near PTW 11	HTW	18	23	7.03	1330	0	25.5	76.2	5.6	158	37.6	2.46	2.03	702	101	40.4	< 0.3	49
Gopalganj																		
HTW2	HTW	8	18	6.69	2800	9	25.0	118	8.8	296	71.5	29.2	0.31	697	631	< 0.1	< 0.3	178
HTW3	HTW		46	6.83	7890	12	25.0	882	17.0	219	120	21.8	0.19	654	2380	< 0.1	7.7	332
HTW4 (new)	RTW	7	9	6.63	1900	21	25.4	79.6	7.0	214	52.7	24.7	0.32	642	493	1.8	1.8	118
Rainwater								2.50	1.4	6.40	1.18	0.39	0.05		6.2	1.6	0.8	< 10

[a] Wells may contain several screened sections; values are top of highest screen and bottom of lowest screen. Values for PTW 12 in Faridpur refer to before and after Fe treatment. HTW = hand pumped water supply tubewell; PTW = water supply tubewell with electric pump; IRR = irrigation well; ITW = pumped tubewell for industrial water supply; HC = health complex; TH = Thana headquarters.

Table 2
Chemical parameters of sediments from Bangladesh

Sample	Depth mbgl	Total		Diagenetically available				Pyrite (equiv. %)	Total C (%)	Org C (%)
		Fe (%)	As (ppm)	Fe (%)	As (ppm)	Al (%)	S (%)			
Dark grey clay	3.0	3.15	24	3.12	24	2.51	0.17	0.32		
Grey clay	4.6	3.26	28	3.19	26	2.92	0.16	0.29		
Grey clayey silt	6.1	3.07	26	2.72	22	1.49	0.21	0.39		
Grey silty sand	7.6	2.69	17	2.60	17	1.56	0.16	0.29		
Grey sand	9.1	1.47	9	1.46	7	0.58	0.09	0.18		
Brown clay	1.8	3.93	28	3.74	26	0.71	0.14	0.26	0.63	0.48
Grey clay	2.1	1.81	12	1.55	9	0.56	0.17	0.33	6.21	6.20
Grey silty clay	4.3	3.42	26	3.30	24	1.96	0.11	0.21	0.71	0.61
Grey silt	5.2	2.73	25	2.59	21	1.25	0.12	0.23	0.59	0.47
Grey silty sand	7.6	3.11	26	2.91	22	1.76	0.17	0.33	0.65	0.18

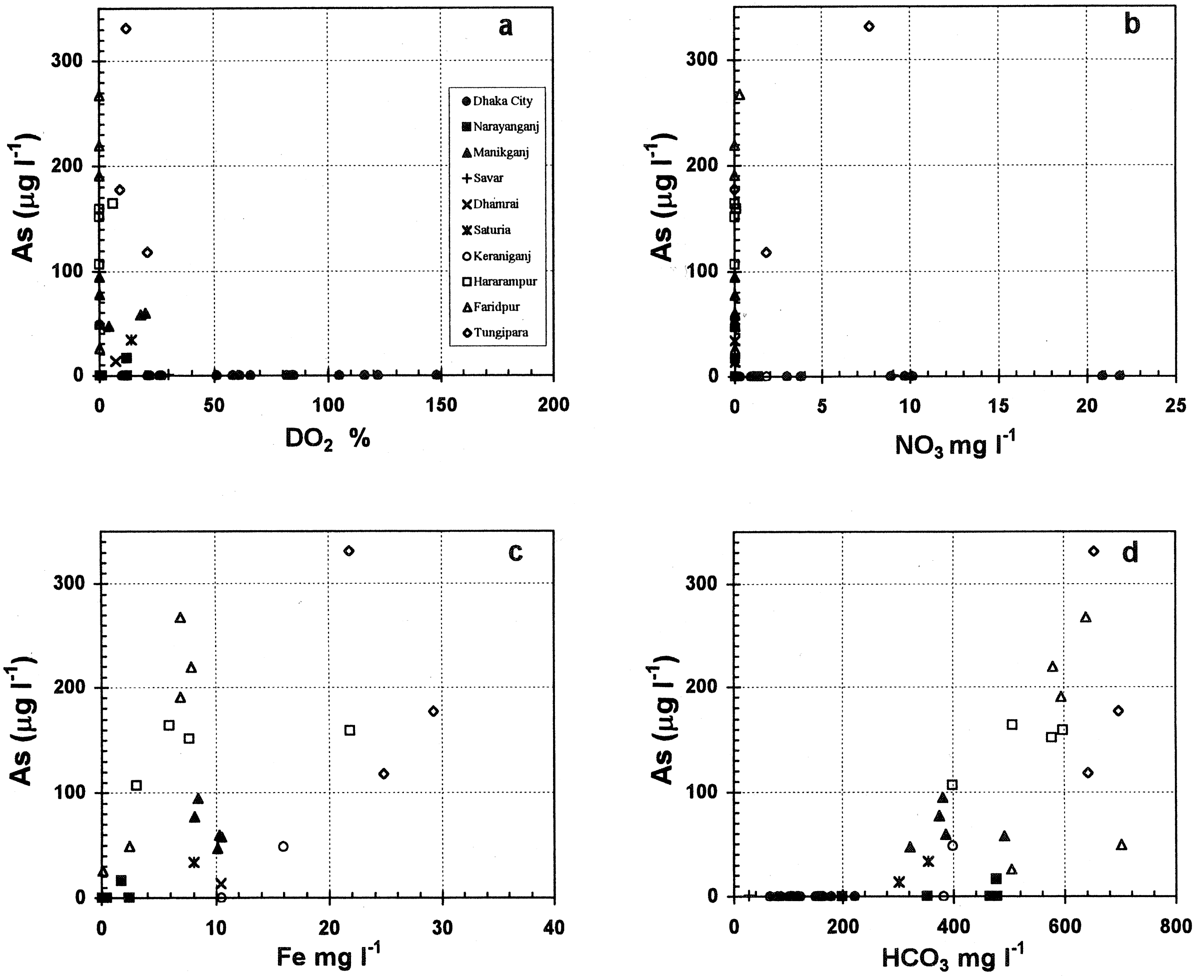

Fig. 2. Chemistry of Bangladesh well water. Relation of (a) As to dissolved O_2; (b) As to NO_3^-; (c) As to Fe; (d) As to HCO_3^-.

Tract and from 28 wells that tap the alluvial aquifers comprised of the late Pleistocene-Holocene sediments of the Brahmaputra and Ganges Rivers. These latter wells were sited within 50 km of Dhaka City at Dhamrai, Faridpur, Harirampur, Keraniganj, Manikganj, Narayanganj, Savar, Saturia and at Tungipara, district of Gopalganj, which is 100 km further to the southwest; locations are shown in Fig. 1 and well details are given in Table 1. Water samples were filtered on site using 0.45 μm membrane filters. Samples for cation analysis were acidified to pH 2, those used for anion analysis were not acidified. Measurements of dissolved O_2, conductivity and alkalinity were made at the well head. With some wells, measurement of dissolved O_2 was affected by contamination with atmosphere and values for such wells are therefore spuriously high. Alkalinity is reported as equivalent HCO_3^- and is corrected for acidity produced by oxidation of Fe(II) during the titration, as many samples precipitated Fe oxyhydroxides soon after exposure to atmosphere. Sediment samples were collected from two borehole cores taken in the late Pleistocene-Holocene sediments at Gopalganj, 100 km SW of Dhaka (Fig. 1).

For waters, cation analysis was done using ICP-AES and anion analysis was done using ion chromatography. Concentrations of As were measured on acidified samples using graphite-furnace AAS (detection limit 10 μg l^{-1}). The amount of diagenetically-available Fe, As, Al and S, in sediments was determined by extraction with hot concentrated HCl acid (Raiswell et al., 1994) followed by analysis of extracts with flame-AAS for Fe and Al, graphite-furnace AAS for As and ion chromatography for SO_4^{2-}. For the determination of total Fe, As, S and Al, samples were fused with lithium metaborate and the fusion dissolved in dilute acid for analysis by ICP-AES and graphite-furnace AAS (for As). Analyses for organic C and total C were done with a LECO C/S 125 Analyser; for organic C, samples were pretreated with 10% v/v HCl to remove inorganic carbonate. Chemical data are given in Table 2. Analytical precision was <5% for all determinations.

4. Results and discussion

4.1. Water Analysis

The data (Table 1) show that well waters contain dissolved O_2 concentrations that range from zero to 148% saturation. Values above 100% are due to pump aeration; many of the values that are between 0 and 22% (e.g. Palpara, Saturia) almost certainly result from contamination by atmosphere during measurement since such waters contain dissolved Fe^{2+} but no NO_3^-. Water from wells sited in Dhaka City and tapping the Plio-Pleistocene Madhupur Tract have As concentrations that are mostly below 50 μg l^{-1}; most contain appreciable concentrations of dissolved O_2. In waters from wells in the Ganges Plain where dissolved oxygen is absent (or arises from contamination), concentrations of As reach 330 μg l^{-1} and concentrations of dissolved Fe reach 29 mg l^{-1} (Table 1; Fig. 2a). Higher concentrations of As have been reported to occur in groundwaters from other sites in the Ganges Plain (PHED, 1991; Bhattacharaya et al., 1997; Safiullah, 1998).

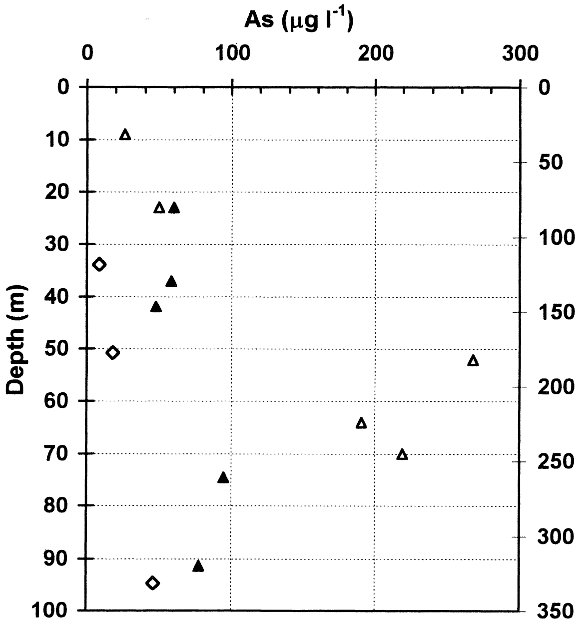

Fig. 3. Relation of dissolved As concentration to depth of wells at Manikganj, Faridpur and Tungipara, Gopalganj. Symbols as for Fig. 2.

As would be expected from thermodynamic considerations of redox reactions (Appelo and Postma 1993; Drever 1997), well waters containing dissolved Fe are free of NO_3^- (Fig. 2b) (with the exception of two at Gopalganj, which the authors believe results from local NO_3^- pollution accessing a poorly-constructed casing). Microbiological reduction of Fe oxyhydroxide occurs after reduction of free molecular O_2 and NO_3^- has exhausted these more thermodynamically favourable oxygen sources. Also (apart from the exceptions noted above) waters that contain NO_3^- do not contain detectable amounts of dissolved As.

In the study waters, concentrations of As correlate poorly with concentrations of dissolved Fe (Fig. 2c) but correlate better with concentrations of HCO_3^- (Fig. 2d). The latter relation with As shows an axial intercept ≈220 mg l^{-1} of HCO_3^- which must represent the

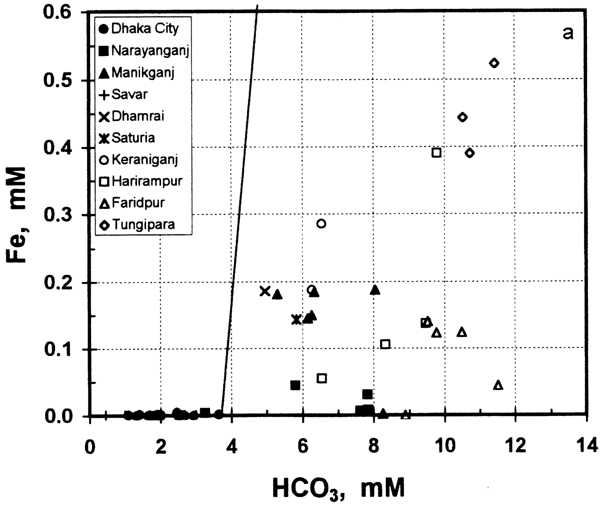

Fig. 4. Relation of HCO_3^- to (a) Fe^{2+}; the line shows the HCO_3/Fe^{2+} production ratio of 2; all data plot well to the right of the line showing that Fe^{2+} is not conservative in solution; (b) Ca + Mg + Fe; the good linear correlation with a slope of 2 suggests that simple mineral dissolution dominates the groundwater chemistry. Symbols as for Fig. 2.

local baseline alkalinity that results from mineral weathering, O_2 consumption and NO_3^- reduction. Arsenic concentrations increase with depth in wells at Manikganj, Faridpur and Gopalganj (Fig. 3), but other trends are reported to occur elsewhere, in particular, a maximum As concentration at 20 to 40 m depth has been reported (Karim et al., 1997; S.K. Acharyya, pers. comm., 1999; T. Roy Chowdhuri, pers. comm. 1999), below which As concentrations decline.

The present data suggest that As is released to groundwater through reduction of arseniferous iron-oxyhydroxides when anoxic conditions develop during sediment burial (Nickson, 1997; Nickson et al., 1998). This process is driven by the microbial oxidation of organic C, concentrations of which reach 6% C in aquifer sediment (Table 2). This mechanism is considered by Bhattacharaya et al. (1997) to be a more likely As source than is pyrite oxidation and the process has been documented to occur in groundwater elsewhere (e.g. Matisoff et al., 1982; Welch and Lico, 1998). The process dissolves Fe oxyhydroxide and releases to groundwater both Fe^{2+} and the sorbed load of the Fe oxyhydroxide, which includes As. The process generates HCO_3^- ions and so produces the relationship between HCO_3^- and As shown in Fig. 2d. The stoichiometry of the reaction yields HCO_3^- and Fe^{2+} in a mole ratio of 2 according to the reaction:

$$4FeOOH + CH_2O + 7H_2CO_3 \rightarrow 4Fe^{2+} + 8HCO_3^- + 6H_2O$$

(modified from de Lange 1986; Lovley, 1987; Drever 1997; where CH_2O represents organic matter). Yet $HCO_3^-/\ Fe^{2+}$ values (adjusted for a background concentration of HCO_3^- of 220 mg l^{-1}) greatly exceed 2 (Fig. 4). The present data also show that a poor correlation exists between Fe^{2+} and As, a finding that confirms similar observations by Safiullah (1998). Presumably, Fe^{2+} does not behave conservatively in these waters, probably because it precipitates as $FeCO_3$ (Sracek et al., 1998; Welch and Lico, 1998). Samples with high concentrations of Fe^{2+} and HCO_3^- (Tungipara, Gopalganj; Table 1) are oversaturated with siderite (S.I. of 1.2; Plummer et al., 1995) and slightly oversaturated with calcite (S.I. of 0.3) and

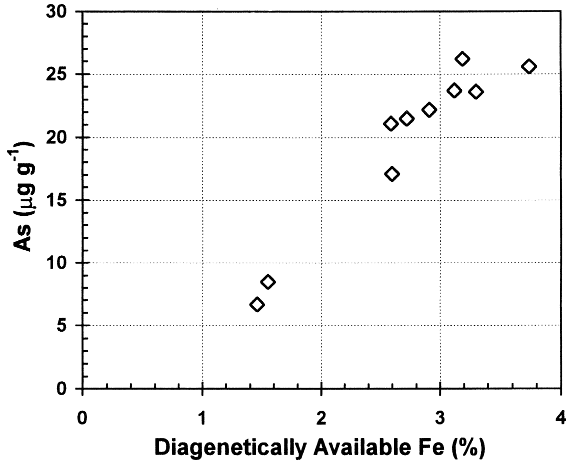

Fig. 5. Relation of diagenetically-available Fe and As in sediments from Tungipara, Gopalganj.

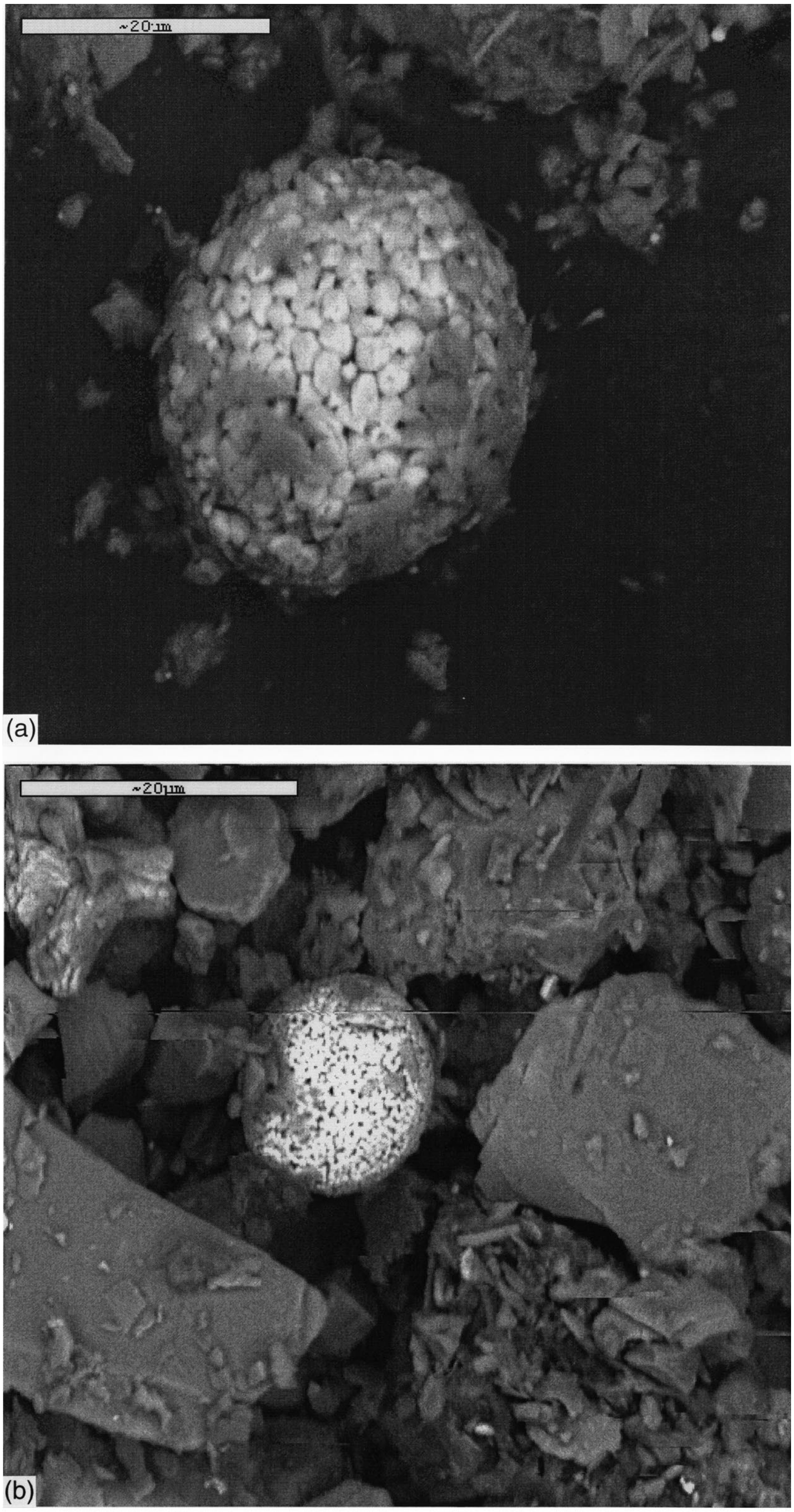

Fig. 6. Framboidal early-diagenetic pyrite in Ganges sediments from Tungipara, Gopalganj.

dolomite (S.I. of 0.3), owing to the high concentrations of HCO_3^-.

4.1.1. Sediment analysis

Sedimentary Fe oxyhydroxides are known to scavenge As (Mok and Wai, 1994; Thornton, 1996; Joshi and Chaudhuri, 1996; references therein). In the sediment samples, concentrations of As correlate with concentrations of diagenetically-available Fe (Fig. 5); an axial intercept of 0.5% Fe represents Fe in phases resistant to our chemical leaches. The concentrations of diagenetically-available S in the sediments is equivalent to between 0.18 and 0.39% pyrite (Table 2). There is no correlation between As and S in the sediments (Table 2). Recalculated to a pure FeOOH (63% Fe) basis from the amounts of diagenetically available Fe (1.4 to 3.6%, corrected for Fe potentially in pyrite; Table 2), the concentration of diagenetically-available As (7–26 ppm; Table 2) represents 289–517 ppm of As in FeOOH.

The current mechanism explaining As contamination of Ganges groundwater via pyrite oxidation owes something to the presence within the aquifer of sedimentary units that contain small amounts of pyrite (Das et al., 1995; Nickson 1997; Fig 6) and the well known association of As with sedimentary pyrite (Ferguson and Garvis, 1972; McArthur, 1978; Thornton, 1996). Under today's wet and oxidising (21% O_2) atmosphere, pyrite does not survive the natural weathering processes and so does not occur naturally as a detrital mineral. Pyrite in Ganges sediments must be diagenetic and must form during the SO_4-reduction stage of diagenesis, which occurs after sediment deposition. Study of our sediments with SEM revealed rare framboidal pyrite of the type typical of that formed during early diagenesis (Fig. 6) and similar studies by others (e.g. Das et al., 1995) have also identified sedimentary pyrite in Ganges sediments. Pyrite formation was limited by low concentrations of SO_4 in the fresh water recharge to the Ganges alluvial aquifers (<20 mg l^{-1}; Table 1).

4.1.2. Sources of arsenic to Ganges sediments

The source of As sorbed to Fe oxyhydroxides must lie upstream of Bangladesh. According to Ghosh and De (1995), the more arseniferous subsurface sediments in the district of N-24 Paraganas (West Bengal) are derived from the Rajmahal–Chotonagpur Plateau to the west, whilst less arseniferous sediment derives from other regions of the Bihar Plateau and from the Himalayas. Contrary to the statement in Nickson et al. (1998), the base-metal deposits upstream of the Ganges Plain are too small in scale to be a likely source for the As (pers. comm. S.K. Acharyya et al., 1999). Potential sources identified by S.K. Acharyya, B.C. Raymahashay and colleagues include the coal of the Rajmahal basin and its overlying basaltic rocks; isolated outcrops of sulfide containing up to 0.8% As in the Darjeeling Himalaya; and the Gondwana coal belt, which is drained by the Damodar River. Weathering of As-rich minerals releases finely divided Fe oxyhydroxides which would strongly sorb co-weathered As (Mok and Wai, 1994; Thornton, 1996; references therein). This process would have supplied As-containing Fe oxyhydroxide to Ganges sediments since the late Pleistocene i.e. since the last glacial maximum (about 18 ka), particularly during the period when rising sea level provided accommodation space for sediment accumulation (post 10 ka, C. Bristow, pers. comm. 1998). Furthermore, As concentrations are higher in fine overbank sediments than in the coarser channel fill. This might be anticipated on grain size considerations alone; Fe oxyhydroxide films coat detrital particles, so their abundance as a fraction of a sedimentary mass increases as grain-size decreases and the surface area of particles increases.

5. Water treatment

In the short term, the fact that dissolved As is often accompanied by dissolved Fe provides an emergency solution to As removal from arseniferous waters. Aeration of Fe-rich water will precipitate Fe oxyhydroxide which will, in turn, coprecipitate some of the As from solution (Pierce and Moore, 1980). Water treatment methods based upon this process have been described by Jekel (1994), Joshi and Chaudhuri (1996), Bhattacharaya et al. (1997) and Safiullah (1998) and show promise for local use. At a water-treatment plant in Faridpur, aeration, coagulation and sand-filtration removes a substantial amount of the As by co-precipitation with Fe: at the time of sampling, As concentrations fell from 220 $\mu g\ l^{-1}$ before treatment to 42 $\mu g\ l^{-1}$ after treatment (Table 1). In Bangladesh, a common treatment applied to clarify river water for domestic use has been to stir water in a vessel with an alum stick and leave the water to settle overnight before decantation or filtration through sand or finely-woven cloth. This procedure might aid the flocculation of Fe oxyhydroxides and has the advantage of being known to the population. Such a practice may alleviate As intake in the short term until more effective solutions to the problem can be found.

6. Conclusions

In the late Pleistocene-Recent alluvial aquifers of the Ganges Plain, concentrations of As correlate with concentrations of HCO_3^- and poorly with concentrations of iron. The relations strongly suggest that the As in groundwater beneath the Ganges Plain is derived by reductive dissolution of Fe oxyhydroxides in the sediment. Oxidised groundwaters, common in the Dupi Tila aquifer of the Madhupur Tract (Plio-Pleistocene), contain less As than do anoxic waters from late Pleistocene-Recent sedimentary aquifers. Where arsenical waters contain high concentrations of Fe^{2+}, As may be removed partially by aeration (oxidation), flocculation and filtration of Fe oxyhydroxide, which sorbs As strongly.

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