

Technical Problems with Home-Scale Coagulation for Removing Arsenic and Other Toxic Elements from Western Bangladesh's Drinking Water

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Abstract

Groundwater samples were collected from 4 neighborhoods in western Bangladesh. These samples were analyzed for every toxic element that has ever been found above World Health Organization (WHO) guidelines in Bangladesh's drinking water: As, B, Ba, Cr, Mn, Mo, Ni, Pb, Sb, and U. In this study, As, Cr, Mn, Ni, Pb, Sb, and U were found above WHO drinking water guidelines. These results were used to formulate a representative influent, called synthetic tubewell water in this study, for evaluating treatment options under laboratory conditions. The effect of safe, simple, and affordable materials (wood ashes, agricultural calcic limestone, agricultural dolomitic limestone, powdered brick, and iron(III) chloride) on As removal and pH were evaluated using this synthetic tubewell water. Effective treatment to remove As requires the oxidation of soluble As(III) to insoluble As(V), the coagulation of insoluble As(V), the

decolorization of Fe(III), the control of pH, and the proper disposal of sludge. Effective treatment to remove the other toxic elements will likely cause even more problems. Therefore, home-scale coagulation for removing As and other toxic elements from western Bangladesh's drinking water is most likely too problematic for effective use.

Experimental Section

A. Sample Collection, Preservation, Shipment, and Analyses. Groundwater samples were collected from 4 neighborhoods in western Bangladesh (Figure 1). A total of 71 random samples were collected from 67 tubewells in these 4 neighborhoods. A total of 18 random samples were collected from 17 tubewells in each of 3 neighborhoods (Bualda, Fulbaria, and Jamjami). Access was denied at 1 sampling location; therefore, a total of 17 random samples were collected from 16 tubewells in the fourth neighborhood (Komlapur). To the extent possible, the sampled tubewells in each neighborhood were distributed at 500-meter intervals along perpendicular axes that radiated in 4 equal lengths from the center (Figure 1). Two samples were collected from the centermost tubewell in each neighborhood. The results for each analyte from each of these 4 centermost tubewells were averaged. One sample was collected from each of the remaining tubewells. The northings and eastings of these tubewells were measured using a Garmin Global Positioning System 12 Channel Personal Navigator™.

Established collection, preservation, and storage methodologies were used to ensure that each sample was representative of groundwater quality (APHA et al. 2005; Stumm and Morgan 1981). Accordingly, all sampled tubewells were purged by pumping vigorously for 10 minutes (min) immediately before sample collection. All samples were collected directly into polyethylene bottles. These samples were not filtered. Samples were analyzed

immediately after collection with pH paper, preserved by acidification to pH <2 with 5.0 Molar (M) hydrochloric acid (HCl; BDH Laboratory Supplies, product number 101256J, Poole, England), and stored in ice-packed coolers. The temperature of all stored samples was maintained at 0° to 4° Celsius (C) until immediately before analysis at laboratories in Dubai, France, and Vermont.

These samples were shipped to Dubai and analyzed for arsenic (As) by the arsenomolybdate method (Frisbie et al. 2005). After which, these samples were shipped to France and analyzed for barium (Ba), chromium (Cr), manganese (Mn), molybdenum (Mo), nickel (Ni), lead (Pb), and uranium (U) by inductively coupled plasma mass spectrometry (ICPMS; APHA et al. 2005). Finally, these samples were shipped to Vermont and analyzed for boron (B) by the azomethine H method (LaMotte Company 2005), iron (Fe) by flame atomic absorption spectroscopy (FAAS; APHA et al. 2005), and antimony (Sb) by graphite furnace atomic absorption spectroscopy (GFAAS; APHA et al. 2005).

B. Optimizing Water Treatment in the Laboratory. Synthetic tubewell water was used to evaluate treatment options under laboratory conditions. This synthetic water contained 84, 9.5, 870, 31, 1.2, 2.3, and 0.93 micrograms per liter ($\mu\text{g/L}$) of As, Cr, Mn, Ni, Pb, Sb, and U, respectively. It was prepared immediately prior to use from the following stock solutions. Stock As(III) (product number A-222), Sb (product number AA-110), and U (product number AA-325) solutions each with 1.00 gram (g) of analyte per L were purchased from Spectrum Chemicals & Laboratory Products, Gardena, CA, USA. Stock Cr(III) solution was prepared by dissolving 0.487 g of chromium(III) chloride hexahydrate ($\text{CrCl}_3 \cdot 6\text{H}_2\text{O}$; Sigma-Aldrich, Inc., product number 230723, St. Louis, MO, USA) in 1 L of deionized water. Stock Mn(II) solution was prepared by dissolving 0.312 g of manganese(II) chloride

tetrahydrate ($\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$; Sigma-Aldrich, Inc., product number 221279) in 1 L of deionized water. Stock Ni(II) solution was prepared by dissolving 0.126 g of nickel(II) chloride hexahydrate ($\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$; Sigma-Aldrich, Inc., product number 223387) in 1 L of deionized water. Concentrated Pb(II) solution was prepared by dissolving 0.164 g of lead(II) chloride (PbCl_2 ; Sigma-Aldrich, Inc., product number 268690) in 1 L of deionized water. Stock Pb(II) solution was prepared by diluting 10.0 mL of concentrated Pb(II) solution to 1 L with deionized water. If required, the synthetic tubewell water was made to 7,300 $\mu\text{g/L}$ of Fe(II) by the dissolution of iron(II) chloride tetrahydrate ($\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$; Sigma-Aldrich, Inc., product number 220299). This was $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ added to the synthetic water immediately prior to use.

The effects of wood ashes, agricultural calcic limestone, agricultural dolomitic limestone, powdered brick, and iron(III) chloride (FeCl_3) on As removal and pH were evaluated using synthetic tubewell water. These ashes were a combustion byproduct from mixed hardwoods harvested in Vermont. They were sieved to >30 mesh prior to use. The calcic limestone (product name Cal-Carb) was purchased from The Old Mill, Inc., North Troy, VT, USA. The dolomitic limestone (product name Pulverized Limestone) was purchased from the Southern States Cooperative, Inc., Richmond, VA, USA. Three grades of bricks were purchased from 6 brickyards in western Bangladesh. These 3 grades of brick were *pora int* (often black or brown, misshapen by excessive heat in the kiln, used to build roads), *ek nombor it* (deep red, not misshapen, used to build houses), and *dui nombor it* (yellowish red, not misshapen, used to build walls). These 6 brickyards were owned by Abdul Hye of Chorhush, Naosher Ali of Boriya, Abdul Sobhan of Baburhut, Abdul Hussain of West Mozompur, Nawab Ali of Nawa Para, and Mamun and Shihab of Nalkupa, respectively.

Prior to use, these 18 bricks were crushed, sieved to >50 mesh, and combined to make 1 composite sample. The FeCl₃ (product number I89) was purchased from Fisher Scientific International, Inc., Hampton, NH, USA.

The effects of these ashes, limestones, powdered brick, and FeCl₃ on As removal and pH were evaluated during jar tests. A 6-position gang stirrer was used for all jar tests (Phipps & Bird, Inc., model number 7790-400, Richmond, VA, USA). Six 1-L beakers were used as jars. Each beaker received 1 L of synthetic tubewell water. The first beaker was a control. The second received 1 g of wood ashes or 1 g of agricultural limestone. The third received 1 g of >50 mesh brick. The fourth received 50 milligrams (mg) of FeCl₃. The fifth received 1 g of wood ashes or 1 g of agricultural limestone immediately followed by 1 g of >50 mesh brick. And the sixth received 1 g of wood ashes or 1 g of agricultural limestone immediately followed by 50 mg of FeCl₃. A second set of these 6 solutions was prepared as a duplicate. All solutions were stirred for 30 min at 120 revolutions per min. After which, all solutions were allowed to settle for 30 min. The pH of each final solution was measured by glass electrode (APHA et al. 2005). Finally, a sample of each solution was passed through a 0.45-micron filter, preserved by acidification to pH <2 with concentrated nitric acid (HNO₃; Spectrum Chemicals & Laboratory Products, product number N1079, Gardena, CA, USA), and measured for dissolved As by GFAAS (APHA et al. 2005; Frisbie et al. 2005).

C. Characterizing Limestone. At least 0.1000 g of each limestone was dissolved in separate 10 mL aliquots of concentrated HNO₃. Two extracts of each limestone were prepared. Each extract was made to 100 mL with deionized water. Each diluted extract was analyzed for Ca²⁺ and Mg²⁺ by FAAS (APHA et al. 2005). These results were expressed as percent (%) CaCO₃ (weight/weight) and % MgCO₃ (weight/weight), respectively.

Results and Discussion

A. The Distributions of Toxic Elements. All 71 random groundwater samples from Bualda, Fulbaria, Jamjami, and Komlapur were analyzed for every toxic element that has ever been found above World Health Organization (WHO) guidelines in Bangladesh's drinking water: As, B, Ba, Cr, Mn, Mo, Ni, Pb, Sb, and U (BGS/DPHE 2001; Frisbie et al. 2002). In this study, As, Cr, Mn, Ni, Pb, Sb, and U were found above WHO drinking water guidelines. Conversely, B, Ba, and Mo were not found above these guidelines. In addition, these samples were analyzed for Fe (Tables 1, 2).

B. Optimizing Water Treatment in the Laboratory.

1. The Composition of Synthetic Tubewell Water. A representative and uniform supply of untreated water is required to evaluate treatment options under laboratory conditions. The formulation of this untreated water, called synthetic tubewell water in this study, was based on the health risks and concentrations of toxic elements from Bualda, Fulbaria, Jamjami, and Komlapur's groundwater (Table 1). More specifically, since chronic As poisoning was by far the most significant health risk caused by drinking tubewell water from these 4 neighborhoods, the first step was to sort these results by As concentration. All samples with As concentrations less than or equal to the 10 µg/L WHO guideline were classified as relatively safe and were not used to formulate this synthetic water. In contrast, all samples with As concentrations greater than 10 µg/L were classified as unsafe and used to formulate this synthetic water. The second step was to identify any other elements that cause a significant risk to public health. In addition to As, the toxic elements Cr, Mn, Ni, Pb, Sb, and U were found above WHO drinking water guidelines in at least 1 sample from this subset of samples with As concentrations greater than 10

$\mu\text{g/L}$. Therefore, these elements cause a significant risk to public health. The third step was to use the average concentrations of As, Cr, Mn, Ni, Pb, Sb, and U from this subset of samples with As concentrations greater than $10 \mu\text{g/L}$ to make synthetic tubewell water (Table 3).

In addition, ferrous iron (Fe^{2+}) at natural concentrations in Bangladesh's tubewell water can significantly increase As removal by oxidation and coagulation. That is, the oxidation of soluble As(III) and soluble Fe(II) by dissolved oxygen (O_2), chlorinated lime (a locally available disinfectant; $a\text{Ca}(\text{OCl})_2 \cdot b\text{CaCl}_2 \cdot c\text{Ca}(\text{OH})_2 \cdot d\text{H}_2\text{O}$), or some other oxidizing agent yields insoluble As(V) and insoluble Fe(III) that coprecipitate and settle out of solution (Frisbie et al. 1999; USAID 1997). In this study the ambient concentration of Fe^{2+} is $7,300 \mu\text{g/L}$, the average Fe concentration from the subset of samples with As concentrations greater than $10 \mu\text{g/L}$ (Table 3). The effect of ambient Fe^{2+} on As removal was evaluated by using synthetic tubewell water with $7,300 \mu\text{g/L}$ of Fe^{2+} and $0 \mu\text{g/L}$ of Fe^{2+} during laboratory testing (Table 4).

2. Jar Testing. As predicted, ambient Fe^{2+} did significantly increase As removal at the 95% confidence level, according to a paired t-test of % As removal from jar tests that did not use FeCl_3 as a coagulant ($p\text{-value} = 0.0003$; Table 4). Moreover, 7 of 12 treatments that used water with ambient Fe^{2+} removed As to less than or equal to the $50 \mu\text{g/L}$ Bangladesh standard, but only 1 of 12 treatments that used water without ambient Fe^{2+} removed As to these concentrations (Table 4). However, the distribution of these elements in Bangladesh's groundwater is variable and individual tubewells might not have enough Fe to coagulate As. For example, a tubewell from Bualda had $100 \mu\text{g/L}$ of As and $4,700 \mu\text{g/L}$ of Fe compared to a tubewell from Fulbaria that had $200 \mu\text{g/L}$ of As

and only 42 µg/L of Fe. Therefore, the effect of a proposed treatment on As removal must be tested using water with and without ambient Fe²⁺ so that a system can be designed to provide safe water at a variety of influent Fe concentrations.

In contrast, ambient Fe²⁺ did not significantly increase As removal if 50 mg/L of FeCl₃ was added as a coagulant, according to a paired t-test of % As removal from jar tests that did use FeCl₃ (p-value = 0.18; Table 4). That is, adding 50 mg/L of FeCl₃ (17,000 µg/L of ferric iron, Fe³⁺) increased As removal from water without ambient Fe²⁺ (Table 4).

Wood ashes, calcic limestone (CaCO₃), and dolomitic limestone (CaMg(CO₃)₂) are common and affordable in Bangladesh. Wood ashes are a byproduct from cooking fires, pottery kilns, and brick kilns. These limestones are used in agriculture to increase soil pH and provide nutrients for crop growth. In this study, wood ashes, calcic limestone, and dolomitic limestone were used to increase solution pH and promote the alkaline oxidation of soluble As(III) and soluble Fe(II) to insoluble As(V) and insoluble Fe(III). Oxidation with 1,000 mg/L of wood ashes followed by coagulation with 50 mg/L of FeCl₃ was the only treatment that removed As to less than the 10 µg/L WHO drinking water guideline from water with and without ambient Fe²⁺ (Table 4).

It was originally thought that powdered brick might be an affordable oxidant and adsorbent for As removal. That is, Fe(III) from the brick might both oxidize As(III) and adsorb As(V). This mechanism, if present at all, provided negligible As removal (Table 4). However, 1,000 mg/L of powdered brick did on all occasions remove the yellow color caused by the oxidation of ambient Fe²⁺ and the use of FeCl₃ as a coagulant (Table 4).

The control of the final pH must be improved (Table 4). The final pH must range from 5.5 to 8.5 (WHO 1984). Limestone was primarily used in these jar tests to promote the alkaline oxidation of As(III) and Fe(II) (Table 4). However, calcic limestone yields a highly buffered solution with a maximum pH of 8.2 and might best be used to adjust the final pH (Stumm and Morgan 1981).

Conclusions

Ambient Fe^{2+} did significantly increase As removal (p-value = 0.0003; Table 4). However, the distribution of elements in Bangladesh's groundwater is highly variable and individual tubewells might not have enough Fe to coagulate As. In contrast, ambient Fe^{2+} did not significantly increase As removal if 50 mg/L of FeCl_3 was added as a coagulant (p-value = 0.18; Table 4). Therefore, the need to use FeCl_3 or a similar coagulant for affective As removal is a technical and economic problem.

In this study, wood ashes, calcic limestone, and dolomitic limestone were used to increase solution pH and promote the alkaline oxidation of soluble As(III) and soluble Fe(II) to insoluble As(V) and insoluble Fe(III). Oxidation with 1,000 mg/L of wood ashes followed by coagulation with 50 mg/L of FeCl_3 was the only treatment that removed As to less than the 10 $\mu\text{g/L}$ WHO drinking water guideline from water with and without ambient Fe^{2+} (Table 4). Therefore, the need to use wood ashes or a similar oxidizing agent for affective As removal is another technical and economic problem.

Unfortunately, the oxidation of ambient Fe^{2+} by wood ashes with the use of FeCl_3 as a coagulant made the water yellow and the pH unacceptably high ($\text{pH} = 9.8 \pm 0.0$; Table 4). Therefore, the need to use powdered brick for decolorization, and the need for calcic limestone or some other material for pH control are other technical and economic problems.

In summary, home-scale coagulation for removing As from western Bangladesh's drinking water is most likely too problematic for effective use. First, soluble As(III) must be oxidized to insoluble As(V). Second, insoluble As(V) must be coagulated. Third, the water likely needed to be decolorized. Fourth, the pH must be controlled. Fifth, the inputs must be safe, simple, and affordable. Sixth, the sludge must be properly disposed. Moreover, effective treatment to remove the other toxic elements will likely cause even more problems.

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Table 1. The average concentrations of toxic elements in Bualda, Fulbaria, Jamjami, and Komlapur's groundwater, the WHO drinking water guidelines for these toxins, and the percent of tubewells exceeding these guidelines (WHO 1996; WHO 1998).

Element	Average Concentration (µg/L)	WHO Guideline (µg/L)	% of Unsafe Tubewells
As	29	10	33
B	19	500	0
Ba	140	700	0
Cr	4.7	50	1
Fe	2,700	NA ^a	NA
Mn	800	500	75
Mo	1.4	70	0
Ni	11	20	3
Pb	0.52	10	1
Sb	1.6	5	3
U	2.5	2	48

^a The WHO has not established a drinking water guideline for Fe (WHO 1996; WHO 1998).

Table 2. Correlation coefficients (r) for the concentrations of toxic elements in tubewell water from Bualda, Fulbaria, Jamjami, and Komlapur. Significant linear relationships at the 99% confidence level are shown with a superscript “a”. Significant linear relationships at the 95% confidence level are shown with a superscript “b”. No significant linear relationships at either confidence level do not have a superscript.

	As	B	Ba	Cr	Fe	Mn	Mo	Ni	Pb	Sb	U
As	1.00 ^a										
B	0.81 ^a	1.00 ^a									
Ba	0.26 ^b	0.40 ^a	1.00 ^a								
Cr	0.82 ^a	0.92 ^a	0.30 ^b	1.00 ^a							
Fe	0.82 ^a	0.92 ^a	0.40 ^a	0.97 ^a	1.00 ^a						
Mn	0.46 ^a	0.31 ^b	0.19	0.26 ^b	0.21	1.00 ^a					
Mo	0.28 ^b	0.05	0.16	-0.03	-0.01	0.28 ^b	1.00 ^a				
Ni	0.13	0.07	0.07	0.10	0.09	-0.09	-0.05	1.00 ^a			
Pb	0.83 ^a	0.94 ^a	0.33 ^a	0.98 ^a	0.96 ^a	0.28 ^b	-0.02	0.09	1.00 ^a		
Sb	0.34 ^a	0.33 ^a	0.56 ^a	0.24	0.31 ^b	0.38 ^a	0.29 ^b	0.40 ^a	0.28 ^b	1.00 ^a	
U	-0.02	0.07	-0.27 ^b	0.04	-0.05	0.18	-0.21	-0.02	0.08	0.04	1.00 ^a

Table 3. The concentrations of toxic elements in synthetic tubewell water based on the health risks from drinking Bualda, Fulbaria, Jamjami, and Komlapur's groundwater (WHO 1996; WHO 1998).

Element	Concentration ($\mu\text{g/L}$)	WHO Guideline ($\mu\text{g/L}$)	% of Unsafe Tubewells
As	84	10	100
Cr	9.5	50	5
Fe ^a	7,300	NA	NA
Mn	870	500	59
Ni	31	20	9
Pb	1.2	10	5
Sb	2.3	5	9
U	0.93	2	14

^a Synthetic tubewell water was prepared with ambient iron Fe^{2+} (7,300 $\mu\text{g/L}$ of Fe^{2+}) and without ambient iron Fe^{2+} (0 $\mu\text{g/L}$ of Fe^{2+}). The WHO has not established a drinking water guideline for Fe (WHO 1996; WHO 1998).

Table 4. The percent As removals and final pH values for jar tests using synthetic tubewell water prepared with and without ambient Fe²⁺. Each test used 1 L of this synthetic water. Each result is an average plus or minus its standard deviation.

Treatment	Synthetic Water with Ambient Fe ²⁺		Synthetic Water without Ambient Fe ²⁺	
	% As Removal	Final pH	% As Removal	Final pH
Control	6.6 ± 6.2	5.6 ± 0.1 ^c	0.0 ± 0.1	5.6 ± 0.2 ^c
1,000 mg Brick	19.3 ± 3.7	5.9 ± 0.4 ^c	0.0 ± 0.0	6.5 ± 0.9 ^c
50 mg FeCl ₃	29.0 ± 3.5	3.3 ± 0.1	30.9 ± 6.8	2.9 ± 0.1
1,000 mg Wood Ashes	64.3 ± 0.4 ^a	10.2 ± 0.5	11.5 ± 0.6	10.2 ± 0.1
1,000 mg Wood Ashes + 1,000 mg Brick	65.9 ± 1.0 ^a	10.5 ± 0.2	11.9 ± 0.2	10.3 ± 0.1
1,000 mg Wood Ashes + 50 mg FeCl ₃	96.7 ± 0.6 ^{a, b}	9.8 ± 0.0	89.9 ± 0.7 ^{a, b}	9.6 ± 0.1
1,000 mg Calcic Limestone ^d	68.2 ± 2.8 ^a	6.5 ± 0.0 ^c	0.6 ± 0.7	6.8 ± 0.1 ^c
1,000 mg Calcic Limestone ^d + 1,000 mg Brick	70.3 ± 0.8 ^a	6.6 ± 0.0 ^c	1.6 ± 0.4	6.9 ± 1.0 ^c
1,000 mg Calcic Limestone ^d + 50 mg FeCl ₃	16.3 ± 0.2	4.9 ± 0.3	13.9 ± 1.5	4.6 ± 0.0
1,000 mg Dolomitic Limestone ^e	53.5 ± 8.5 ^a	5.6 ± 0.1 ^c	1.3 ± 1.0	5.6 ± 0.0 ^c
1,000 mg Dolomitic Limestone ^e + 1,000 mg Brick	50.8 ± 3.2 ^a	5.4 ± 0.2	1.2 ± 0.5	5.6 ± 0.1 ^c
1,000 mg Dolomitic Limestone ^e + 50 mg FeCl ₃	17.7 ± 1.7	4.7 ± 0.1	17.2 ± 0.0	4.2 ± 0.4

^a Meets the 50 µg/L Bangladesh drinking water standard for As on average.

^b Meets the 10 µg/L WHO drinking water guideline for As on average.

^c Meets the 5.5 to 8.5 WHO drinking water guideline for pH on average (WHO 1984).

^d Calcic limestone was 95.5 ± 0.7 % CaCO₃ and 5.0 ± 0.5 % MgCO₃.

^e Dolomitic limestone was 55.3 ± 0.1 % CaCO₃ and 45.0 ± 0.5 % MgCO₃.

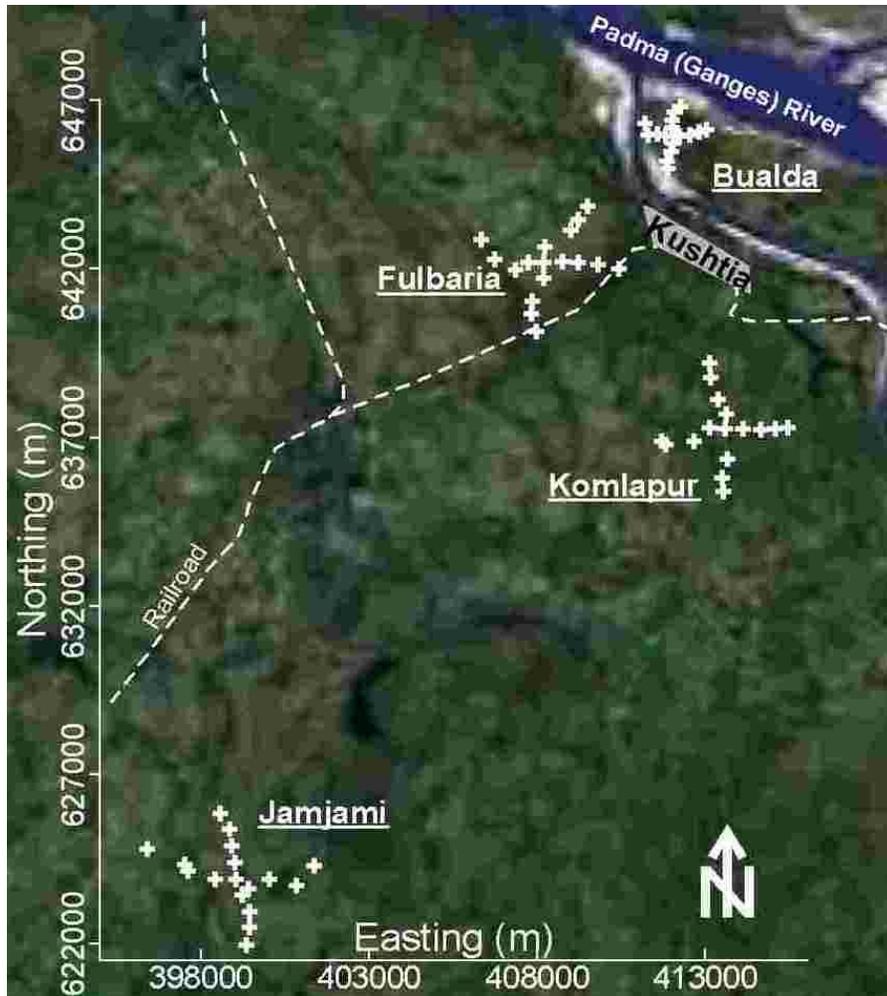


Figure 1. Satellite image of western Bangladesh showing the 4 neighborhoods where groundwater samples were collected from tubewells. These 4 neighborhoods are centered in the villages of Bualda, Fulbaria, Jamjami, and Komlapur. Each sampling location is labeled with a +. Kushtia is a major city (GlobeXplorer™ 2005).