Results from the First-Round of Bangladesh Environmental Technology Verification - Support to Arsenic Mitigation (BETV-SAM) Field Test

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Abstract

The Bangladesh Environmental Technology Verification - Support to Arsenic Mitigation (BETV-SAM) program objectively assessed thirteen arsenic removal technologies (ARTs) that had been submitted for performance verification, and selected seven technologies for the field testing and performance verification. Each technology was deployed to five hydrogeologically distinct regions of Bangladesh and was tested on five wells (with replicate units installed and tested on two wells). Raw and treated water samples were collected at regular intervals and were analysed for arsenic and other water quality parameters. The data were analysed using standard statistical software and the performance of individual ARTs have been evaluated and compared with the proponent's claim. Two technologies (STAR & CIWP) performed well while five other technologies performed poorly and their claims were rejected. The results of the first-round of the BETV-SAM field testing and performance verification program presented and discussed in this paper, may be important from a technological design and development point of view.

Key words: Arsenic removal technology, Field testing, Performance verification, Water quality parameters.

Introduction

Arsenic contamination of groundwater in Bangladesh is widespread and acute and an estimated 30 million Bangladeshi (Heikens 2006) obtain their drinking water from shallow tubewells that are contaminated with arsenic above the national drinking water standard of 50 mg/l (Government of Bangladesh [GoB], 1997). Arsenic is poisonous and a known cancer causing agent (Centeno *et al.* 2002, Loewenberg 2007) and approximately 10,000 - 30,000 Bangladeshis have already been diagnosed with arsenicosis (Heikens 2006). Thus, meeting the need for arsenic-safe[3] drinking water in Bangladesh is urgent.

Approximately 30% of shallow tubewells in Bangladesh are contaminated with concentrations of arsenic in the range of 100 - 1000 µg/L. The occurrence of high concentrations of arsenic is often associated with elevated levels of iron, phosphate, and manganese. A recent survey found that more than 80% of arsenic-affected tubewells (i.e. [As]>50 µg/l) also contain equal or greater than 2 mg/l of iron (Ahmed, 2001; Ahmed, 2003). In areas with an acute iron problem, the iron concentrations in wells can reach more than 10 mg/l.

A variety of Arsenic Removal Technologies (ARTs) could be employed to remove arsenic from arsenic contaminated groundwater; however, the technologies may not last long, perform as well as expected, and may not produce as much arsenic-safe water as expected. The technologies that were marketed in Bangladesh in the past had not been tested adequately and appropriately in the country and their performance claims are not guaranteed. The international community was also reluctant to deploy those ARTs to address the problem associated with the shortage of arsenic-safe drinking water, due to potential liabilities associated with the use of technologies that might not work "as advertised".

The Government of Bangladesh (GoB) sought means of validating the performance of any ART before permitting it to be marketed and sold in the country and designated the Bangladesh Council of Scientific and Industrial Research (BCSIR) as the national verification authority. The Bangladesh Environmental Technology Verification-Support to Arsenic Mitigation (BETV-SAM) program, a bilateral

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[3] The term arsenic-safe refers to potable water with arsenic concentrations below the permissible Bangladesh drinking water standard level of 50 µg/L.

project between the Governments of Bangladesh and Canada, was established in order to verify that ARTS: A) demonstrate the ability to consistently produce potable water with arsenic concentrations below 50 μg/l from arsenic-contaminated well water at a prescribed minimum daily volume, B) produce the quantity of arsenic-safe water the proponents claim that their technologies are able to generate, and C) don't add harmful substances to the treated water. In addition, the BETV-SAM also analysed treated water for other regulated species, such as manganese, boron, etc.

Thirteen candidate ARTs registered through BCSIR for performance verification in the first round. None of the proponent provided sufficient, independent, and verifiable data that could be used to verify their performance claim. Therefore, there was the need to field test each technology to collect independent, credible and verifiable data in order to be able to evaluate technology performance and to either verify or reject a technology proponent's claim. After screening all applications, the BETV-SAM program, in its first-round of field testing and performance verification, selected, deployed, and field tested seven (7) ARTs in five hydrogeologically different regions of Bangladesh.

In addition to arsenic, groundwater in Bangladesh is often contaminated with iron, phosphate, manganese, and other inorganic species. Removal of these latter species has important implications for the technology performance and can be achieved through proper design and O&M procedures. The influence of water quality parameters, removal of iron dissolved in groundwater, and other O&M procedures on ARTs performance are discussed in the paper.

Materials and Methods

A. Technology Screening and Selection

The program screened and ranked all ARTs on the basis of available experimental data for arsenic removal ability, ease of operation and maintenance, ability to treat well water with different concentrations of dissolved iron, phosphate, silicate, alkalinity, etc., and other technical requirements and selected seven ARTs for the field testing and performance verification. These include Apyron, Canadian International Water Purification Limited (CIWPL), MAGC/Alcan (referred to as Alcan afterwards), Nelima, Shapla, STAR, and Wholly Water. Nelima and Shapla were developed locally and the rest were either imported from overseas or the media and concepts were developed abroad and the units were designed and built locally. The CIWPL and STAR employ coagulant, ferric sulphate, and the rest employ arsenic adsorption media for the capture and removal of arsenic dissolved in groundwater. The types and quantity of media and/or coagulants employed by each technology and their modes of operations are presented in Table I.

B. Testing Procedures

When applied for performance verification, each technology proponent made a performance claim, which specified that: a) the ART can produce arsenic-safe, potable water from arsenic contaminated groundwater that contains known concentrations of As, iron, phosphate, silicate, sulphate (SO_4^{2-}), etc, and within a certain range of pH, and b) the technology can generate X and Y litres of arsenic-safe water in a day (or per batch) and over the useful lifespan of the media (for ARTs that employ sorption media), respectively.

Table I: The types of media and the production capacities of different ARTs

Technology	Media	Quantity	Polishing Media	Operation
Apyron	Alumina and Modified Alumina	48.0 - 63.0 kg	Granular Activated Carbon (GAC)	Flow-Through
CIWPL	Ferric Sulphate	200 - 600 mg/20 L	Special slow CIWP sand filter	Batch
Alcan	Modified Alumina	40 kg	None	Flow-Through
Nelima	Granular Ferric Oxide	1.0 kg	Sand filter	Batch
Shapla	Fe_2O_3 impregnated Brick chip	40.0 kg	None	Batch
STAR	Ferric Sulphate	1.5 g/20 L	Sand and GAC	Batch
Wholly Water	MnO_2, Brass Powder, Reducing agent and Alumina	~28.0 kg	GAC/Modified alumina	Flow-Through

The BETV-SAM program in consultation with proponents developed Technology Specific Test Plans (TSTP) for each technology and tested each technology on five wells (35 wells in total) in Chapai, Dohar, Kolaroa, Ishwardi, and in Nabinagar. The wells, selected for field testing and performance verification in each area, met proponents' water quality criteria such as concentrations of As, PO_4^{3-}, Fe, pH, etc, as well as the ease of access, free from the influence of point sources of pollution, and availability of space for testing, etc.

The technologies were installed either by proponents or by field staff that were trained by proponents. Each technology was operated and maintained following proponents Operation and Maintenance (O&M) procedures. The coagulation-flocculation technologies were tested for 10 - 20 days and the adsorption technologies were operated until: a) arsenic breakthrough (i.e. effluent [As])> 50 μg/l) occurred, or b) produced enough arsenic-safe water and met the proponent's performance claim. A brief description of each technology and the operating procedures is provided here.

ALCAN

The ALCAN ARTs consist of two trapezoidal tanks (approximate dimensions: 0.46 m × 0.46 m × 0.46 m) that are piped together and each tank houses about 20 kg of arsenic removal media. The media was covered with a perforated plate and a furnace filter and were suspended off the bottom of the tank by a perforated acrylic sheet. During the normal operation, the tubewell water was pumped, at a flow rate of ≤ 5l/min, directly into the first tank using an electrical pump. The water flows in a down-flow mode through the first tank and an up-flow mode through the second one. The media was backwashed at least once a week for approximately 15 minutes to remove silts, sands, and iron flocs.

Apyron

An Apyron ART consists of a chlorination chamber for the oxidation and removal of dissolved iron as well as arsenite, a 25 cm × 165 cm PVC column that houses media (three different types of media that were packed in three different layers in the column), flow distribution plates, and frits. The unit was connected to a pump that was in turn attached to the well head. The well water was pumped, at a flow rate of <10 l/min, directly into the unit in a downward direction, and treated effluent water was collected in a container or released into the field. The unit was backwashed periodically in order to remove accumulated silts, sands, and iron flocs and to restore normal flow rate.

Wholly Water

A Wholly Water ART unit consists of an electrical pump, two arsenic removal filters, an overhead tank, a backwash tank, and other accessories. The first arsenic removal filter was filled with four different layers of manganese dioxide, brass powders, ferric sulfate modified alumina, and granular activated carbons successively from top to the bottom. The second filter was only filled with modified alumina. The well water was pumped into the overhead tank and was then spiked with sodium hypochlorite to oxidize and remove dissolved iron. The pre-treated water was then flowed into the arsenic filtration units at a flow rate ≤4 l/min and the treated water was either collected in a clean container or released into the field. The filter was regularly backwashed for at least 10 minutes and at a rate of approximately 4 l/min for the removal of accumulated silts, sands, and iron flocs and restoration of normal flow rate.

Nelima

The Nelima ART consists of two 30 L buckets, each with a lid and a water tap that was attached on the sidewall and slightly above the bottom, a sand filter, and an arsenic removal filter. The two buckets are placed one above the other, on a folding tripod steel stand. The top bucket was perforated along the brim and its lid was fitted with a PVC rod that extended into the bucket. The sand filter was placed in the bottom of the top bucket, and the arsenic removal filter was placed in the bottom bucket. The filters and water taps were connected to each other with flexible rubber tubing. During normal operation, the water taps on both the top and bottom buckets were closed, the lid from the top bucket was removed and approximately 20 L of well water was poured into the top bucket. The lid was re-secured and the solution was stirred with the PVC rod about 3 or 4 times to increase the aeration rate and to accelerate the oxidation and flocculation. If the influent water contained more than 3 mg/l of dissolved iron, the water was allowed to stand in the top bucket for approximately 1.5 to 2 hours. Otherwise, the water tap on the top bucket was opened and the pretreated water flowed to the lower bucket, through first the sand filter and then the sand arsenic removal filter, and accumulated in the lower bucket, which acts as a reservoir for the treated water.

Shapla

The Shapla ART consists of two earthenware columns, approximately 56 cm × 87 cm each, with two lids of the

same materials and a single point of water delivery system. Each column housed approximately 20 kg of filter media. Each column was filled slowly with about 12 L of well water and the water was allowed to flow through the column by carefully opening the draining valve and allowing treated water to leave the column. The Shapla ART was operated intermittently, in a batch-flow mode, with an average flow rate of 5.5 l/h. The unit was able to treat about 24 litres of contaminated water in approximately 4.3 hours and was operated twice in 12 hours, with two hours resting period in between.

STAR

The STAR ART consists of two empty buckets, one of which contains the clean, coarse filter sand media and is fitted near the bottom with a spout, flexible tubing, and packets of the reagent chemical which is composed of ferric sulphate and calcium hypochlorite. In a normal operation, the empty bucket was filled with approximately 16 L of well water, a packet of chemical reagent was added, the solution was mixed well with a clean stick for one minute, and allowed to stand for approximately 10 - 15 minutes. It was then gently poured through an inverted plastic bottle whose bottom was cut-off onto the sand filter in the filtration bucket to remove the suspended solids, including the arsenic-containing iron flocs. Water was drained from the sand filter bucket.

CIWPL

The CIWP ART consist of the filter apparatus (NSF food grade PVC piping and CIWPL sand filter media), reagent chemical (ferric sulphate and 5% sodium hypochlorite solution), a rod to stir the raw water after chemical additions, a ladle to remove solids from suspension, two raw water buckets and a food grade bucket for collection of arsenic-safe water, and an additional bucket for sludge collection. In a normal operation, approximately 20 L of well water was added to a plastic bucket, spiked with sufficient volumes of sodium hypochlorite such that a distinct chlorine odour is detected after stirring the bucket. This was followed by the addition of 200, 400, or 600 mg of ferric sulfate to well water containing <1 mg/l, 1-2 mg/L, or >2 mg/l of phosphate, respectively. The solution was stirred for about two minutes, allowed to stand for an hour, and filtered through a filter assembly to remove the suspended solids, including the arsenic containing iron hydroxide particulates.

C. Sampling and Analysis

During the testing period, samples were taken at regular intervals and were analysed for concentrations of arsenic, iron, phosphate, silicate, alkalinity, bacterial contamination, total metal scan, etc. following the procedures outlined below.

* Total Arsenic Analysis: Five (5) to seven (7) raw groundwater samples and 10 - 20 treated groundwater samples were collected from each technology and analysed for total arsenic (using atomic absorption spectroscopy-hydrate generation (AAS-HG) procedures) in one of the designated analytical laboratories[4] following the Standard Procedures (Clesceri *et al.* 1998), thereby ensuring that both the influent and effluent water were sufficiently characterised.

* As(III), Total Iron, Phosphate and Microbial Analyses: At least three influent and three effluent samples are collected from each technology and analyzed in one of the designated analytical laboratories in Bangladesh for concentrations of As(III), Fe (total), PO_4^{3-}, and faecal and total coliforms[5] following Standard Procedures (Clesceri *et al.* 1998).

* Field Analyses: Five (5) influent and five (5) effluent samples were analysed in the field for alkalinity and chlorine (for ARTs that employed chlorine) analysis. In addition, the pH, conductivity, and turbidity of 5 influent samples and 10 treated effluent samples were measured using portable meters, following standard protocols. The pH and conductivity were measured employing a Hach Multimeter, model Sension 156, Chlorine was measured using a Hach Chlorine (total) test kit model CN-66T, Turbidity was measured with a Hach Turbidimeter model 2100, and alkalinity was determined with a Hach Alkalinity test kit model AL-AP MG-L.

* QA/QC Samples: Duplicate, field blank, and reference standard samples were also collected and sent to the Maxxam Analytical Laboratory, Mississauga, Ontario, Canada, an ISO 17025 certified analytical laboratory, for QA/QC purposes. Sample analysis at Maxxam was performed employing inductively coupled plasma - mass spectrometry (ICP-MS) (Clesceri *et al.* 1998).

* Other Water Quality Parameters: A number of influent and effluent water samples were also collected and analysed for anions, metal scan (Reference Lab using

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[4] The designated laboratories were selected amongst laboratories that are located in Dhaka, participated in a round-robin sample analysis prior to the field testing, and performed well.

[5] Bacteriological samples were analysed for faecal coliform; 10% were tested for total coliform.

ICP-MS techniques), and volatile organic compounds and chlorinated phenols (for ARTs employing chlorine). A summary of the analytical methods employed for the BETV-SAM field testing program are outlined in Table II.

Where appropriate, the effluent arsenic data has been analysed statistically using MINITAB14 statistical software to show that: a) the data are randomly distributed around a mean value prior to arsenic breakthrough, and b) the mean

Table II: Summary of Sampling Requirements and Analytical Methods

Parameter	Sample volume (mL)	Reference Method	Preservation	Recommended Storage time	Field QC
Total As - lab analysis	100	Standard Method 3114 B	Acidify with HNO_3 to pH < 2	2 months	B, D[7]
As(III)	100	Standard Method 3114 B	Acidify with HNO_3 to pH < 2	2 months	B,D
Total Fe	100	Standard Method; Spectrophotometery	Acidify with HNO_3 to pH < 2	6 months	B,D
Orthophosphate (PO_4^{3-})	100	Spectrophotometry	Acidify with HNO_3 to pH < 2	48 hours	B,D
Metal scan[8]	100	ICP-AES, ICP-MS or ICP-AAS	Acidify with HNO_3 to pH < 2	6 months	B,D
Anion scan	100	Ion Chromatography	Refrigerate at 4°C, keep in the dark	48 hours	B,D
Faecal and total Coliforms	200	Membrane Filter method	Refrigerate at 4°C	24 hours	B,D
Sulphate (SO_4^{2-})	100	Ion Chromatography	Refrigerate at 4°C, keep in the dark	48 hours	B,D
Total As - field analysis	50	Arsenator™ portable test kit	Not applicable (NA)	At the well site	B,D
pH, turbidity, conductivity	200	Use Hach field kits	NA	At the well site	NA
Alkalinity	200	Hach kit	NA	At the well site	NA

[6] Recommended storage times are maximum storage times. For best analytical results, samples should be analysed as soon as possible.
[7] Blank and Duplicate samples are collected and analysed.
[8] Metals scan includes all required parameters, including Al, Ca, Cr, K, Mn, Na, Si, and U.
[9] Field tests were carried out as soon as possible after sampling.

* Arsenic-Safe Water Production Capacity: The field testing program measured the rate of production of treated water (l/h and l/day) and cumulative volume (CV) of water that was generated by each technology over the duration of the technology testing period (for technologies that employ sorptive media), respectively. The effluent arsenic concentrations ($[As]_T$) on each well site were plotted against the cumulative volume of treated water (for sorption technologies) or against days of technology operation (for coagulation technologies). The data were then analyzed using MINITAB14 statistical software to find the correlation between $[As]_T$ and CV or days of technology operation.

effluent arsenic concentration prior to breakthrough is less than 50 µg/l with 95% confidence.

Results and Discussions

Each proponent made a performance claim when it applied for the technology performance verification. The performance claims for each of the seven technologies that were tested by the BETV-SAM program are presented in Table III and the range of concentrations of As, Fe, PO_4^{3-}, and the pH of well waters selected for different ARTs are presented in Tables IV - X. The data clearly demonstrate that: a) wells selected for each technology met water quality parameters specified by the vendors in almost all cases, and b) water quality parameters covered a range of values from low to

Table III: The performance claims of ARTs

ART\WQP	[As]/ µg/l	[Fe]/ mg/l	[PO_4^{3-}]/ mg/l	pH	Daily/Batch Capacity	Total Capacity
Apyron	$\leq$1,200	$\leq$10	$\leq$5	5.0- 8.0	4800 L/day	160,000 L
CIWPL	$\leq$1000	$\leq$20.0	$\leq$10.0	C[10]	20 L/batch	NA
Alcan	1,500	Any	5	C	2400 L/day	80,000 L
Nelima	$\leq$350	<4	<10	5.5 - 8.0	20 L/batch	8,000 L
Shapla	450 - 700	$\leq$40.0	$\leq$4.0	C	24 L/batch	7,000 L
STAR	$\leq$750	$\leq$21	$\leq$10	C	20 L/batch	NA
Wholly Water	$\leq$1000	$\leq$10	$\leq$6	5.5 - 8.5	2080 L/d.	295,000 L

Table IV: Summary of the well water quality parameters for Apyron; Replicate units were installed on wells in Chapai and Kolaroa

Location and Well Number	[As]/ µg/l Mean ± CI[11]	[As(III)]/ [As]$_T$	[Fe^{+2}]/ mg/l Mean ± CI	[PO_4^{3-}]/ mg/l Mean ± CI	pH Mean ± CI
Chapai/W21	866 ± 156	0.80	0.6 ± 0.3	0.4 ± 0.1	7.12 ± 0.27
Dohar/W52	265 ± 32	0.93	13.0 ± 0.1	3.8 ± 0.3	7.05 ± 0.11
Ishwardi/W65	900 ± 120	0.73	7.8 ± 2.2	1.2 ± 1.2	6.93 ± 0.04
Kolaroa/W07	228 ± 25	0.91	5.0 ± 1.0	3.8 ± 0.9	7.14 ± 0.13
Nabinagar/W75	344 ± 42	1.08	4. 5 ± 0.8	7.9 ± 0.5	7.31 ± 0.50

Table V: Summary of the well water quality parameters for CIWPL; Replicate units were installed on wells in Chapai and Nabinagar

Location and Well Number	[As]/ µg/l Mean ± CI	[As(III)]/ [As]$_T$	[Fe^{+2}]/ mg/l Mean ± CI	[PO_4^{3-}]/ mg/l Mean ± CI	pH Mean ± CI
Chapai/W21	622 ± 23	0.76	3.3 ± 0.5	1.1 ± 0.2	7.02 ± 0.14
Dohar/W31	348 ± 31	0.82	11.3 ± 3.9	4.6 ± 0.6	7.23 ± 0.17
Kolaroa/W07	295 ± 7	0.94	8.6 ± 0.4	2.4 ± 0.3	6.60 ± 0.24
Ishwardi/W65	482 ± 37	1.01[12]	4.7 ± 0.5	1.5 ± 0.3	6.97 ± 0.04
Nabinagar/W20	323 ± 14	1.04[12]	5.1 ± 0.3	7.5 ± 0.3	7.38 ± 0.32

[10] C - Circumneutral pH   [11] CI is Confidence Interval; Mean ± CI shows 95% confidence intervals
        [12] These are probably too high because of analytical error

Table VI: Summary of the well water quality parameters for MAGC/ALCAN; Replicate units were installed on wells in Chapai and Kolaroa

Location and Well Number	[As]/ µg/l Mean ± CI	[As(III)]/ [As]$_T$	[Fe^{+2}]/ mg/l Mean ± CI	[PO_4^{3-}]/ mg/l Mean ± CI	pH Mean ± CI
Chapai/W21	519 ± 13	1.0[13]	0.9 ± 0.3	0.2 ± 0.1	7.02 ± 0.11
Dohar/W52	358 ± 35	0.86	6.9 ± 0.8	2.9 ± 0.2	7.02 ± 0.18
Ishwardi/W65	1151 ± 310	0.80	10.2 ± 1.8	1.0 ± 0.5	6.91 ± 0.05
Kolaroa/W07	257 ± 50	0.95	7.9 ± 0.5	4.5 ± 1.1	7.10 ± 0.12
Nabinagar/W75	396 ± 50	0.85	3.0 ± 0.4	7.9 ± 1.0	7.64 ± 0.16

[13] This is high because of the analytical error involved in measuring As concentration

Table VII: Summary of the well water quality parameters for Nelima; Replicate units were installed on wells in Chapai and Dohar

Location and Well Number	$[As]/$ µg/l Mean $\pm$ CI	$[As(III)]/$ $[As]_T$	$[Fe^{+2}]/$ mg/l Mean $\pm$ CI	$[PO_4^{3-}]/$ mg/l Mean $\pm$ CI	pH Mean $\pm$ CI
Chapai/W24	274 $\pm$ 10	0.8	4.2 $\pm$ 0.8	1.4 $\pm$ 0.3	7.06 $\pm$ 0.07
Dohar/W57	283 $\pm$ 20	0.83	2.3 $\pm$ 0.3	1.1 $\pm$ 0.2	7.05 $\pm$ 0.25
Ishwardi/W93	269 $\pm$ 38	1.00[12]	2.3 $\pm$ 0.2	0.8 $\pm$ 0.3	6.85 $\pm$ 0.03
Kolaroa/W07	301 $\pm$ 16	0.90	8.6 $\pm$ 0.6	2.4 $\pm$ 0.6	6.69 $\pm$ 0.23
Nabinagar/W60	221 $\pm$ 12	1.00[12]	1.2 $\pm$ 0.1	2.4 $\pm$ 0.1	7.66 $\pm$ 0.20

Table VIII: Summary of the well water quality parameters for Shapla; Replicate units were installed on wells in Chapai and Dohar

Location and Well Number	$[As]/$ µg/l Mean $\pm$ CI	$[As(III)]/$ $[As]_T$	$[Fe^{+2}]/$ mg/l Mean $\pm$ CI	$[PO_4^{3-}]/$ mg/l Mean $\pm$ CI	pH Mean $\pm$ CI
Chapai/W24	278 $\pm$ 17	0.80	4.3 $\pm$ 0.8	1.5 $\pm$ 0.2	7.10 $\pm$ 0.05
Dohar/W57	283 $\pm$ 20	0.83	2.3 $\pm$ 0.3	1.1 $\pm$ 0.2	7.16 $\pm$ 0.39
Ishwardi/W93	269 $\pm$ 38	0.97[12]	2.1 $\pm$ 0.4	0.8 $\pm$ 0.1	6.90 $\pm$ 0.04
Kolaroa/W07	301 $\pm$ 16	0.89	8.7 $\pm$ 0.8	3.2 $\pm$ 0.2	6.90 $\pm$ 0.30
Nabinagar/W60	221 $\pm$ 12	0.61	1.2 $\pm$ 0.1	2.3 $\pm$ 0.2	7.52 $\pm$ 0.33

Table IX: Summary of the well water quality parameters for STAR; Replicate units were installed on wells in Chapai and Nabinagar

Location and Well Number	$[As]/$ µg/l Mean $\pm$ CI	$[As(III)]/$ $[As]_T$	$[Fe^{+2}]/$ mg/l Mean $\pm$ CI	$[PO_4^{3-}]/$ mg/l Mean $\pm$ CI	pH Mean $\pm$ CI
Chapai/W31	621 $\pm$ 3	0.76	3.1 $\pm$ 0.1	1.0 $\pm$ 0.1	7.02 $\pm$ 0.13
Dohar/W21	347 $\pm$ 40	0.82	12.6 $\pm$ 0.6	4.5 $\pm$ 0.8	7.02 $\pm$ 0.56
Kolaroa/W07	298 $\pm$ 11	0.94	8.5 $\pm$ 0.4	2.3 $\pm$ 0.5	6.95 $\pm$ 0.06
Ishwardi/W65	526 $\pm$ 53	1.01[12]	4.6 $\pm$ 0.5	1.4 $\pm$ 0.2	6.97 $\pm$ 0.05
Nabinagar/W75	355 $\pm$ 15	1.04[12]	3.1 $\pm$ 0.2	10.4 $\pm$ 0.8	7.36 $\pm$ 0.36
Nabinagar/W20	309 $\pm$ 23	1.04[12]	5.2 $\pm$ 0.3	7.9 $\pm$ 0.4	7.45 $\pm$ 0.34

Table X: Summary of the well water quality parameters for Wholly Water; Replicate units were installed on wells in Chapai and Kolaroa

Location and Well Number	$[As]/$ µg/l Mean $\pm$ CI	$[As(III)]/$ $[As]_T$	$[Fe^{+2}]/$ mg/l Mean $\pm$ CI	$[PO_4^{3-}]/$ mg/l Mean $\pm$ CI	pH Mean $\pm$ CI
Chapai/W21	675 $\pm$ 23	0.84	1.1 $\pm$ 0.4	1.6 $\pm$ 0.2	7.19 $\pm$ 0.24
Dohar/W52	420 $\pm$ 22	0.72	5.45 $\pm$ 0.4	2.8 $\pm$ 0.7	7.18 $\pm$ 0.35
Ishwardi/W65	641 $\pm$ 75	0.98	4.1 $\pm$ 0.6	0.5[14]	7.04 $\pm$ 0.14
Kolaroa/W07	239 $\pm$ 17	0.93	4.7 $\pm$ 0.9	4.0 $\pm$ 0.4	6.97 $\pm$ 0.56
Nabinagar/W75	341[14]	0.43	5.2[14]	7.4 $\pm$ 1.7	7.44 $\pm$ 0.60

[14] Average of two measurements and hence no precision is reported for these data

Table XI: Summary of statistical analysis (*t-statistic*) of effluent arsenic concentrations in Apyron treated water samples, prior to arsenic breakthrough, in different locations.

Location/Unit	Mean effluent [As]/ μg/L; Mean ± CI	Treated water volume/L	Claimed Volume/ L	Claim Status
Chapai/U3	2.0 ± 2.4	61,300	175,000	Denied
Chapai/U7	1.5 ± 0.7	63,500	175,000	Denied
Dohar/U1	31 ± 11	169,300	175,000	Verified
Ishwardi/U4	3.0[15]	27,119	175,000	Denied
Kolaroa/U2	4.5[15]	42,149	175,000	Denied
Kolaroa/U6	20.0[15]	44,743	175,000	Denied
Nabinagar/U7	5.0[15]	37,224	175,000	Denied

[15] Average of two to three measurements

Table XII: Summary of statistical analysis (*t-statistic*) of arsenic concentrations in CIWPL treated water samples in different locations.

Location/Unit	No. of Data Points	Mean effluent [As]/ μg/l; Mean ± CI	P [16]	Claim Status
Chapai/U3	14	37.1 ± 8.9	0.008	Verified
Chapai/U6	19	37.9 ± 10.7	0.030	Verified
Dohar/U1	13	7.7 ± 1.3	0.000	Verified
Ishwardi/U4	12	14.7 ± 4.2	0.000	Verified
Kolaroa/U2	13	7.2 ± 1.1	0.000	Verified
Nabinagar/U5	11	60.0 ± 18.2	0.249	Denied
Nabinagar/U7	10	50.7 ± 14	0.907	Denied

Table XIII: Summary of statistical analysis (*t-statistic*) of arsenic concentrations in Alcan treated water samples, prior to arsenic breakthrough, in different locations.

Location/Unit	Mean effluent [As]/ μg/L; Mean ± CI	Treated water volume/L	Claimed Volume/ L	Claim Status
Chapai/U3	25.4 ± 8.4	≥ 81,000	80,000	Verified
Chapai/U7	27.9 ± 16.0	~ 40,000	80,000	Denied
Dohar/U1	26.4 ± 16.9	~ 73,000	80,000	Verified
Ishwardi/U4	15.7 ± 11.8[17]	~ 8,000	80,000	Denied
Kolaroa/U2	39.9 ± 10	~ 27,000	80,000	Denied
Kolaroa/U6	38.9 ± 10.5	~ 23,000	80,000	Denied
Nabinagar/U7	39.2 ± 10.4	~ 32,000	80,000	Denied

[16] The probability of obtaining a sample mean if the true sample mean is really equal to 50 mg/L as was hypothesized. If the p-value is less than or equal to the corresponding a-level (0.05 in this case), the null hypothesis (mean = 50 mg/L) can be rejected.

[17] This is a tentative value since the unit at this location behaved badly from the beginning.
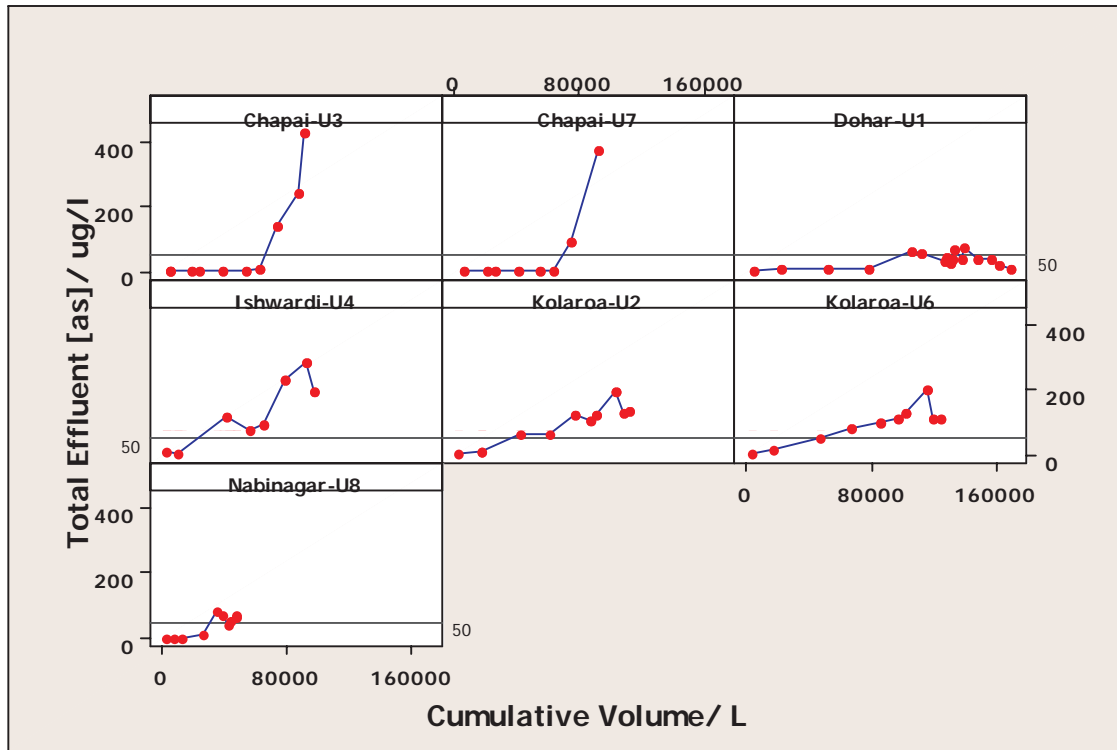
Figure 1: Plots showing effluent As concentrations vs cumulative volume of treated water for a sorption ART. The technology was operated in flow-through mode.
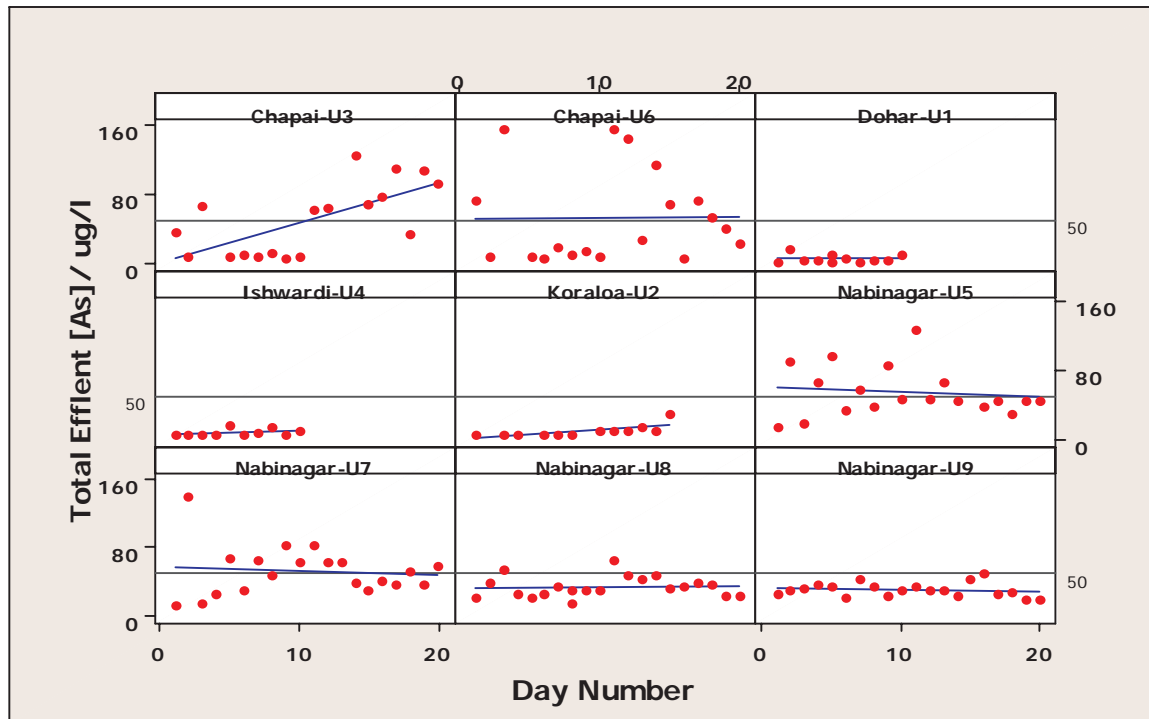

Figure 2: Plots showing effluent As concentrations vs testing days for a coagulation ART

high and the high values were close to those claimed by the proponents.

All sorptive technologies failed prematurely and did not meet proponents' performance claims while the coagulation-flocculation technologies performed well and were able to generate arsenic-safe water as was claimed by their proponents. Typical plots for both the coagulation and sorption technologies are presented in Figures 1 and 2. In general, the effluent arsenic concentrations for the sorption technologies were found to be randomly distributed around a mean value prior to arsenic breakthrough, except in the case of Shapla, where there was an approximate linear relation between $[As]_T$ and CV. In the case of coagulation technologies, the concentrations of arsenic in the effluent were found to fluctuate around a mean value at each site.

Apyron technology failed prematurely at Chapai, Ishwardi, Kolaroa, and Nabinagar but was able to generate 169,000 L of arsenic-safe potable water in Dohar. The water flow rate at Dohar dropped quickly from the prescribed rate of 10 l/min to around 2 l/min (perhaps due to filling of the empty bed space by silts, sands, and ferric hydroxide) and the unit was operated at this flow rate for most of the testing period; hence the performance was better at this site. Nelima, Shapla, and Wholly Water technologies performed similarly; the first two technologies failed prematurely on four (4) sites out of five and the last one failed on all five sites.

Two Alcan units, one of the replicates at Chapai and the one in Dohar, performed well and generated $\geq$ 81,000 L and about 73,000 L of arsenic-safe potable water, respectively, over the duration of the testing. These two units were backwashed frequently and the effluent after backwashing was discarded for some time because it was found that the concentrations of arsenic in the treated water was high immedi

ately after backwashing and decreased gradually to below 50 mg/l.

The STAR technology performed well on three wells initially and consistently produced arsenic-safe water, failed on the fourth well (replicate units, Nabinagar), and its performance on the fifth well (replicate units, Chapai) could not be confirmed. The failure at Nabinagar was attributed to high concentrations of phosphate (10.5 mg/l) in this well water. The technology performed well when transferred to and tested on a different well in the same area but with lower phosphate concentration (7.5 mg/l). The inconsistent performance at Chapai is attributed to operating procedures, a low quantity of sand and sand-boiling as water is poured over the sand by operators. The CIWPL technology also performed well and consistently produced arsenic-safe water on four wells but failed on a well at Nabinagar. This is also attributed to the high concentration of phosphate in this well.

The performances of the two coagulation technologies, CIWP and STAR, were verified while those of the five sorption technologies were denied. Summaries of all field data are presented in Tables XI-XVII.

The failure of the sorption technologies to meet their performance claims and the premature arsenic breakthrough is attributed to the following:

1) These technologies were either not tested with Bangladesh groundwater or tested inadequately by the proponents and the performance claims were not based on realistic field observations.

2) The influence of WQPs such as Fe, PO_4^{3-}, alkalinity, salinity, etc. on technology performance was not considered (Cheng *et al.* 2004, Meng & Korfiatis 2001, Meng *et al.* 2000, Meng *et al.* 2001, Roberts *et al.* 2004).

Table XIV: Summary of statistical analysis (*t-statistic*) of arsenic concentrations in Nelima technology treated water samples, prior to breakthrough, in different locations

Location/Unit	Mean effluent [As]/ µg/L; Mean ± CI	Treated water volume/L	Claimed Volume/ L	Claim Status
Chapai/U3	10.0 ± 19.4	1500	8100	denied
Chapai/U7	25.3 ± 10.4	3710	8100	denied
Dohar/U1	14.0 ± 23.3	< 1000	8100	denied
Dohar/U6	8.7 ± 26.8	790	8100	denied
Ishwardi/U4	22.9 ± 10.4	3000	8100	denied
Kolaroa/U2	21.4 ± 6.5	> 8050	8100	validated
Nabinagar/U5	19 ± 18.5	2890	8100	denied

[18]   Field test was terminated prior to breakthrough, when the technology had produced 8050 L of potable water

3) Dissolved iron was not removed properly (Ali *et al.* 2001).

4) The flow rates were often set too high.

5) The O&M were based on limited laboratory and/or field test data.

The WQPs in general and the concentrations of arsenic, iron, and phosphate in groundwater and groundwater pH in particular play crucial roles in the performance of the arsenic removal technologies (Cheng *et al.* 2004, Meng & Korfiatis 2001, Meng *et al.* 2000, Meng *et al.* 2001, Roberts *et al.* 2004). Ferric oxide/hydroxide and aluminum oxide have limited but reasonable capacities for arsenic adsorption. For example, theoretically, one kilogram of hydroxy ferric oxide (HFO) can adsorb about 148 g of arsenic (Dzomback & Mofrel, 1990), which is much higher than the experimentally determined values of (16g As- 40g As)/(kg of HFO) (Hussam *et al.* 2003, Pal 2001, Pierce & Moore 1982).

Aluminum oxide, on the other hands, adsorbs a lower quantity of arsenic: a kilogram of Aluminum oxide adsorbs about 4 g of arsenic (ALCAN 2006). These values indicated that the Alcan, Apyron, Wholly Water, and Nelima ARTs should be able to generate $\geq 4 \times 10^5$ L, 3.2×10^5 L, 2.2×10^5 L and 11000 L of arsenic-safe water, respectively, from groundwater contaminated with 500 mg/l of As. The above estimations assume that iron and phosphate dissolved in groundwater do not affect media capacity, which is an incorrect assumption. Dissolved iron should assist (Meng & Korfiatis 2001) and phosphate should hinder (Roberts *et al*. 2004) arsenic removal ability of adsorption media. For example, our field observations show that the oxidation and removal of iron naturally present in water can remove up to 75% of arsenic dissolved in groundwater.

Iron dissolved in groundwater can have both positive and negative effects on media capacity. The half-life of iron dissolved in groundwater, Fe(II), at circumneutral pH and in the

Table XV: **Summary of statistical analysis (*t-statistic*) of arsenic concentrations in Nelima technology treated water samples, prior to breakthrough, in different locations**

Location/Unit	Treated water volume/L	Claimed Volume/ L	Claim Status
Chapai/U3	6370 ± 920	7600	Verified
Chapai/U7	4940 ± 640	7600	Verified
Dohar/U1	1020 ± 370	7600	Denied
Dohar/U6	970 ± 490	7600	Denied
Ishwardi/U4	3200 ± 1200	7600	Denied
Kolaroa/U2	1140 ± 985	7600	Denied
Nabinagar/U5	530 ± 520	7600	Denied

Table XVI: **Summary of statistical analysis (*t-statistic*) of arsenic concentrations in STAR treated water samples in different locations**

Location/Unit	No. of Data Points	Mean effluent [As]/ µg/l; Mean ± CI	P[19]	Claim Status
Chapai/U3	19	57 ± 26	0.531	Denied
Chapai/U6	19	53 ± 26	0.819	Denied
Dohar/U1	17	5 ± 2	0.000	Verified
Ishwardi/U4	18	8 ± 3	0.000	Verified
Kolaroa/U2	15	9 ± 4	0.000	Verified
Nabinagar/U5	20	55 ± 14	0.506	Denied
Nabinagar/U7	20	53 ± 13	0.620	Denied
Nabinagar/U8	20	35 ± 5	0.000	Verified
Nabinagar/U9	19	32 ± 4	0.000	Verified

[19] The probability of obtaining a sample mean if the true sample mean is really equal to 50 mg/L as was hypothesized. If the p-value is less than or equal to the corresponding a-level (0.05 in this case), the null hypothesis (mean = 50 mg/L) can be rejected.

presence of air is approximately 4 minutes (Hussam *et al.* 2003). When exposed to air, the ferrous ion, Fe(II), dissolved in groundwater oxidises rapidly to ferric iron, [Fe(III)]. The latter is insoluble in water under circumneutral pH conditions and hydrolyses rapidly; forming ferric hydroxide (Stumm & Morgan 1996), which then forms hydroxy ferric oxide (HFO) through coagulation and flocculation processes.

$$Fe^{3+} + 3\,H_2O \rightarrow Fe(OH)_3 + 3\,H^+ \qquad (1)$$
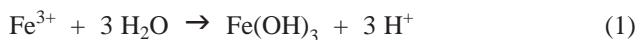
The dissolved arsenic is either trapped within the HFO flocs during floc formation, or adsorbs onto the floc surfaces and subsequently reacts with the surface active -OH group. Both of these processes sequester dissolved arsenic, generate particulate arsenic, and reduce arsenic concentrations in water (Cheng *et al.* 2004, Meng & Korfiatis 2001, Meng *et al.* 2000, Meng *et al.* 2001, Roberts *et al.* 2004). In addition to arsenic, HFO can also remove phosphate, fluoride, silicate, etc. (Cheng *et al.* 2004, Meng & Korfiatis 2001, Meng *et al.* 2000, Meng *et al.* 2001, Roberts *et al.* 2004).

and removal of HFO. The data also show that the concentration of iron was also reduced to below the detection limit of 0.01 mg/l following filtration and removal of HFO. Although reduction of arsenic concentrations was not significant in all cases, the data never-the-less shows that HFO can end-up in the effluent and bring with it arsenic if not removed.

Phosphate, at concentrations greater than 1 mg/l, is also found in approximately 46% of the shallow tubewells in Bangladesh that are contaminated with arsenic [Ahmed, 2001 & 2003]. Phosphate is chemically similar to arsenate [As(V)], as indicated by the sorption equilibrium constants (see Eqs. 1 - 4), and competes effectively with the latter for the active sites on the surface of iron hydroxide (Roberts *et al.* 2004). It is important to note that the sorption constants presented here were calculated using experimental data obtained from the removal of arsenic, phosphate and silicate

A Virgin Media Particle   A Particle Exposed to Groundwater

Figure 3: Schematic showing iron oxide filling media cracks and crevasses

The HFO flocs do not adsorb and attach to media. If not removed properly before coming in contact with media, HFO will accumulate in the media cracks and crevasses as illustrated schematically in Figure 3, fill the empty bed spaces (Ali *et al* 2001), and will slowly wash out into the effluent stream. The net effect of accumulation of iron flocs is blockage of media active sites, blocking of water flow paths and creation of short-cuts that lead to reduced residence times, and reduction of system capacity and premature arsenic breakthrough. This is believed to be partially responsible for the failure of sorption ARTs.

Table XVIII presents concentrations of arsenic and iron in the treated water samples taken from an Alcan and an Apyron unit before and after filtration and removal of HFO, demonstrating that iron flocs can breakthrough the system. The data clearly show that arsenic concentrations in the treated water samples were invariably reduced after filtration

by Fe(III), and competitive sorption modeling (Roberts et al. 2004). Thus, the sorption constants provide qualitative comparison of the intrinsic adsorption capacity of HFO for As(V), As(III), phosphate and silicate.

Sorption Equation	Log K	
=Fe-OH + As(v) $\leftrightarrow$ =Fe-As(v)	5.93	(2)
=Fe-OH + As(III) $\leftrightarrow$ =Fe-As(III)	4.46	(3)
=Fe-OH + P $\leftrightarrow$ =Fe-P; P = Phosphate	6.07	(4)
=Fe-OH + Si $\leftrightarrow$ =Fe-Si; Si = Silicate	3.36	(5)

It has been reported that in addition to removing arsenic, HFO also removes 85% - 99% of the dissolved phosphate from an aqueous environment (Cheng *et al.* 2004, Meng & Korfiatis 2001, Meng *et al.* 2000, Meng *et al.* 2001, Mukherjee *et al.* 2007, Roberts et al. 2004). It is perhaps for this reason that the amount of iron required to remove arsenic from phosphate-contaminated water is much higher than in the absence of phosphate.

Table XVII: Summary of the performance of Wholly Water ART in Various Locations

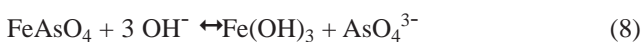| Location/Unit	Treated water Volume/L	Claimed Volume/L	Claim Status
Chapai/U3	< 20,000	300,000	Denied
Chapai/U7	< 20,000	300,000	Denied
Dohar/U1	< 20,000	300,000	Denied
Ishwardi/U4	< 20,000	300,000	Denied
Kolaroa/U2	< 10,000	300,000	Denied
Kolaroa/U6	~ 23,000	300,000	Denied
Nabinagar/U5	<10,000	300,000	Denied

Table XVIII: Arsenic and iron concentrations in the Alcan and Apyron effluent before filtration and removal of HFO

Technology	Effluent Water	[As]/ µg/l	[Fe]/ mg/l
Alcan	Unfiltered	65	--
	Filtered	40	--
Alcan	Unfiltered	51	0.5
	Filtered	47	<0.02
Apyron	Unfiltered	235	--
	Filtered	190	--
Apyron	Unfiltered	180	0.2
	Filtered	170	<0.01

Other species dissolved in groundwater that could adversely influence the performance of arsenic removal technologies are silicate, fluoride, alkalinity, etc (Cheng *et al.* 2004, Meeussen *et al.* 1996, Meng & Korfiatis 2001, Meng *et al.* 2000, Meng *et al.* 2001, Roberts *et al.* 2004). Silicate and fluoride compete with arsenate and arsenite for the surface active sites, and alkalinity regulates the solution pH, the ionization of As (V), As(III), phosphate ions, and adsorption of these species as well as regulating solubility of -Fe-As(V)/As(III) species as illustrated (Dzomback & Mofrel 1990) by the following equations.

$$FeAsO_4 + H^+ \leftrightarrow Fe^{3+} + HAsO_4^{2-} \qquad (6)$$
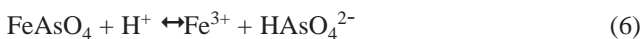
$$K = K_{SP}(FA)/K_3(AA) = 1.1 \times 10^{-18}, [HAsO_4^{2-}]$$

$$= \{1.8 \times 10^{-9}[H^+]\}^{1/2} \qquad (7)$$

$$FeAsO_4 + 3\ OH^- \leftrightarrow Fe(OH)_3 + AsO_4^{3-} \qquad (8)$$

$$K = K_{SP}(FA)/K_{SP}(FH) = 1.4 \times 10^{17}, [AsO_4^{3-}]$$

$$= 1.4 \times 10^{+17}[OH^-]^3 \qquad (9)$$

All sorption ARTs performed well, both in terms of volume of water generated in a day and removal of arsenic from well water, and according to specifications for the first couple of weeks. Surface active sites, easily accessible or buried, are free and available initially. The sites that are easily accessible will be occupied by arsenic leaving inaccessible and hard

to reach active sites free.   The longer a technology operates the longer the residence time required to reach these latter sites, assuming that they are free and are not blocked by either HFO or the sands and silts pumped from the ground. If not removed, sands and silts will fill the empty bed volume and the cracks and crevasses on media and block access to active sites, create short paths, and reduce arsenic removal efficiency.

Conclusion

The coagulation technologies performed reasonably well and were able to consistently generate arsenic-safe water in 4 out of the five locations where they were tested. The sorption ARTs, on the other hands, have performed poorly and failed prematurely in most field locations after only a few weeks of operation. The reasons for the failure of these technologies are in part due to the design flows, inadequate field testing in Bangladesh, and inappropriate O&M procedures.

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