A performance assessment of arsenic-iron removal plants in the Manikganj District of Bangladesh

R. Brennan and E. McBean

ABSTRACT

In Bangladesh, arsenic contamination of groundwater, microbial contamination of surface water and seasonally variable rainfall make reliable access to acceptable quality drinking water a challenge. Arsenic-iron removal plants (AIRPs) are a relatively inexpensive way of removing arsenic from groundwater for access to safer drinking water. This study evaluated the performance of 21 (of 105) AIRPs installed by a local non-governmental organization (Society for People's Action in Change and Equity) with financial assistance from the Australian High Commission, Dhaka, under the Direct Aid Program of the Australian Government. All AIRPs achieved the Bangladesh standard for arsenic in drinking water of 50 μg L^{-1} and 17 achieved the World Health Organization guideline of 10 μg L^{-1}. The AIRPs removed 87% of influent arsenic, on average. After cleaning, poor arsenic and iron removal was observed for about 2 days due to inadequate residence time. Chemical processes that may influence AIRP performance are also discussed herein, including iron and arsenic oxidation, arsenic co-precipitation with iron, multiple iron additions, interference by organics, and iron crystallization. Effluent faecal coliform counts were generally low, though were slightly higher than influent counts. Overall, AIRPs were shown to possess considerable promise for use in areas with high natural iron where users are concerned about arsenic and/or iron in their drinking water.

Key words | AIRP, arsenic, Bangladesh, coliforms, drinking water, low-cost technology

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INTRODUCTION

Reliable access to drinking water of an acceptable quality is one of many challenges facing Bangladesh, which ranked 146th of 182 countries on the Human Development Index (UNDP 2009). Historically, sources such as surface water, ponds, and dug wells were used for drinking water, but a high prevalence of diarrhoeal diseases led to a transition to groundwater use. The installation of shallow tubewells, beginning in the 1970s, to obtain microbially safe drinking water, was initially deemed a major success but has since been tarnished by the discovery of arsenic in groundwater at high concentrations across much of the country. Regular flooding and the annual cycle of monsoons and drought further complicate reliable access to good quality drinking water.

The shift of drinking water sources, initiated in the 1970s by international organizations, from surface water to arsenic-contaminated groundwater, has been referred to as 'the largest mass poisoning of a population in history' (Smith, quoted in WHO 2000). Ten million tubewells were installed across the country before arsenic testing (Sambu & Wilson 2008). It is reported that 97% of rural Bangladeshis now obtain drinking water from groundwater (NAISU 2003). Approximately 25–40% of shallow tubewells have been found to contain arsenic levels above the Bangladesh drinking water standard of 50 μg L^{-1} (estimates from Johnston *et al.* 2001; Smedley & Kinniburgh 2001; Rahman & Rahman 2004; Ravenscroft *et al.* 2005), and in the south-eastern part of Bangladesh, 90% of wells are affected (Smedley & Kinniburgh 2001); 61 of 64 districts are affected by arsenic contamination (Rahman & Rahman 2004). Concentrations of arsenic in groundwater have been reported to range from 0.5 to 2,500 μg L^{-1} (Department of Public

Health Engineering (DPHE)/British Geological Survey (BGS)/Mott MacDonald Limited 1999 and BGS & DPHE 2001 in Smedley & Kinniburgh 2001).

Approximately 21–40 million people have been exposed to arsenic in drinking water exceeding the Bangladesh standard of 50 µg L^{-1} (estimates from Regional Committee for South-East Asia 2001; BGS & DPHE 2001 in Safiuddin & Karim 2001; Smedley & Kinniburgh 2001; Caldwell *et al.* 2005; Ravenscroft *et al.* 2005), although this may be as high as 77 million (WHO 2000). It is thought that 42–60 million people are exposed to drinking water above 10 µg L^{-1} (estimates from Caldwell *et al.* 2005; Sambu & Wilson 2008), which is the World Health Organization (WHO) guideline for arsenic (WHO 2008). Although numbers vary, it is evident that vast numbers of Bangladeshis are consuming arsenic in their drinking water above the Bangladesh standard and WHO guideline. The total number suffering from arsenic poisoning is unclear, but the United Nations Children's Fund (UNICEF 2008) reports that 40,000 people have been identified as having arsenicosis, as arsenic poisoning is termed.

Arsenic-iron removal plants (AIRPs) show promise as a method of removing arsenic from groundwater in areas with high iron, for access to safer drinking water. AIRPs have low capital costs compared with other arsenic-removal technologies, essentially no maintenance costs, require no added chemicals, are easily maintained, can be constructed with locally available materials by local contractors, and are based on a technology used for three decades in Bangladesh, in the form of iron-removal plants (IRPs). The first IRPs in Bangladesh were designed by the Bangladesh University of Engineering and Technology (BUET) in the 1980s and involved aeration, sedimentation and filtration (Ahmed 2003), to address concerns of high iron in drinking water. Though high iron is not a health concern, many users desire iron removal because iron imparts taste and colour to water and food.

The Society for People's Action in Change and Equity (SPACE) has installed 105 AIRPs (Figure 1) in the Manikganj District of Bangladesh (as of January 2011; Azahar Pramanik, SPACE Director, personal communication). These AIRPs are located in the Mohadebpur and Baliakhora Unions, just west of the City of Manikganj in the Manikganj District of Bangladesh, and are contained within an area
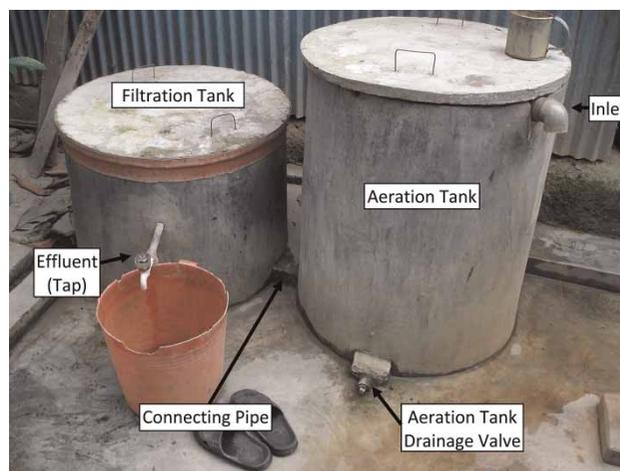

Figure 1 | Typical AIRP.

approximately 5 km by 5 km. The AIRPs are designed for multi-household use; each unit is designed to serve the drinking water needs of approximately 20 people. These AIRPs can hold about 350 L of water. Typical use is 150–200 L day^{-1}, giving a residence time of about 2 days.

SPACE AIRPs consist of two tanks approximately 65 cm in diameter constructed from pre-made sanitary latrine rings. The aeration tank is about 90 cm in height, and the filtration tank is about 60 cm in height. Source water from the tubewell is pumped into the aeration tray (Figure 2), at the top of the aeration tank. Bangladesh's groundwater is strongly reducing and at neutral pH (Smedley & Kinniburgh 2001); iron is therefore expected to

Figure 2 | Inside aeration tank.

exist mostly as Fe(II) and arsenic as As(III). Iron naturally in the water is oxidized through aeration and subsequently precipitates in the aeration tank. Arsenic co-precipitates with the iron. Co-precipitation refers to the process of adsorption that occurs as solid precipitates are forming, rather than adsorption to precipitates that have already formed. In the field experiments described herein, this oxidation and precipitation of iron was observed to occur in minutes upon entering the aeration tank. Following precipitation, ageing for 2–4 h is reportedly necessary to stabilize the suspensions (Jambor & Dutrizac 1998). Settling takes much longer, and is on the order of several hours.

Water leaves the aeration tank through the connecting pipe at the bottom of the tank, and passes into the upflow filtration tank. Iron, and co-precipitated arsenic, is removed both by settling in the aeration tank and filtration in the filtration tank. The filter (Figure 3) comprises brick chips, charcoal, and sand. The brick chip layer is about 12 cm thick, the charcoal about 4 cm and the sand about 8 cm. Brick chips provide coarse filtration, and lengthen the flow path which improves flocculation (Ahmed 2005). Charcoal is used to remove organics, and sand is used for fine filtration.

AIRPs are particularly suited for use in areas with high natural iron because natural iron would clog many other types of arsenic-removal filter quickly, many users desire iron removal because iron imparts taste and colour to water and food, and the existence of natural iron in solution

Figure 3 │ Filtration tank.

as Fe(II) has advantages in arsenic removal. Fe(II) was found to be more effective than Fe(III) in arsenic removal in laboratory batch experiments because of higher sorption capacities of iron (hydr)oxides formed from Fe(II) (Roberts et al. 2004). Additionally, co-precipitation with iron removes more As(V) than adsorption to pre-formed iron (Waychunas et al. 1993), in part because adsorption happens before aggregates form: there are more adsorption sites, and the process is not diffusion controlled (Paige et al. 1996; Fuller et al. 1993). Additionally, oxidation of Fe(II) has been reported to cause partial oxidation of As(III), which can improve arsenic removal efficiency.

It is widely reported that most arsenate (As(V)) removal efficiencies by adsorption to iron oxides are higher than arsenite (As(III)) (e.g. Leupin & Hug 2005). However, other authors (Raven et al. 1998; Dixit & Hering 2003) report that this is not always the case; removal of arsenite and arsenate from solution by adsorption to iron oxides depends on pH, the iron to arsenic ratio, the type and specific surface area of the iron minerals, and the concentration of phosphate (Dixit & Hering 2003), for example. At concentrations of arsenic, iron and orthophosphate as seen in this study (and typical of Bangladesh), and at near-neutral pH, it appears that arsenate removal by adsorption to hydrous ferric oxide (HFO) (Dixit & Hering 2003) or ferrihydrite (Raven et al. 1998) is in fact greater than arsenite. It is believed that the iron precipitates, as seen in this study, commonly referred to as iron hydr(oxides), iron oxyhydroxides and HFO are, more specifically, ferrihydrite as described by Jambor & Dutrizac (1998).

Oxidation of arsenite to arsenate could therefore improve arsenic removal efficiency for AIRPs in Bangladesh. Though aeration is not effective in oxidizing As(III) (Lowry & Lowry 2002), Roberts et al. (2004), in their laboratory studies, report that oxidation of Fe(II) causes partial oxidation of As(III), which 'proves to be a clear advantage because As(V) competes much better with P [phosphorus] for the available sorption sites than As(III)'. For a further discussion of competition with phosphorus in AIRPs, see Brennan & McBean (2011). Leupin & Hug (2005) report that 'it appears that the oxidation of As(III) is linked to the presence of dissolved, adsorbed, or structural Fe(II) and the presence of dissolved oxygen', and is likely oxidized by 'reactive intermediates such as Fe(IV), reactive oxygen

species and/or carbonate radicals'. Therefore, with adequate aeration, AIRPs can make use of natural Fe(II) in the water to co-precipitate and adsorb arsenic, and to oxidize As(III) to As(V), its more removable form.

Despite their promise, and current use in many parts of Bangladesh, little work has been done on characterizing the performance of AIRPs. Laboratory experiments have been performed on the removal of arsenic with iron, but few field experiments have been identified. This paper discusses the arsenic removal performance, factors that may impact performance, and the microbial effectiveness of the SPACE AIRPs in the Manikganj District. Another paper (Brennan & McBean 2011) discusses the impact of orthophosphate and role of dissolved oxygen on arsenic removal in these AIRPs.

METHODS

Field sampling was conducted between October and December 2009. To assess arsenic removal, 21 AIRPs were tested. Initially, eight AIRPs were selected based on location; the study area was divided into eight geographic regions and one site from each was randomly selected. Influent and effluent samples for arsenic and iron were taken at all eight sites, at least once. Upon completing testing at these eight sites, influent and effluent samples were collected from another 13 AIRPs, selected based on suspected high arsenic in the raw water.

At four of the eight sites tested initially, samples were collected a second time (sites 16, 7, 53, and 42). The second sampling event at these four sites involved sample collection at the influent, the aeration tank (at approximately 30 cm depth), the bottom of the filtration tank (at the valve), and the effluent. Sampling locations are shown in Figure 4. The aeration tank and filtration tank samples (locations 2 and 3) were filtered through filter paper to remove iron precipitates and any co-precipitated ions. Both unfiltered and filtered aeration tank samples and filtration tank samples were analysed. These extra sampling locations were included to provide insight into the kinetics and mechanisms of arsenic removal.

Cleaning cycle experiment

At one of these four sites sampled a second time (site 16), additional testing was performed throughout a cleaning cycle to observe the effect of cleaning on AIRP performance. Samples were collected before cleaning, immediately after cleaning, and then 1 day, 3 days, and 8 days after cleaning. Again, samples were collected at the influent, aeration tank (filtered and unfiltered samples), filtration tank (filtered and unfiltered samples), and effluent.

PVC elbow experiment

In the standard design of AIRPs the connecting pipe is at the very bottom of the aeration tank. At another of these four sites sampled a second time (site 7), a polyvinyl chloride (PVC) elbow (diameter about 4.5 cm; Figure 5) was inserted into the connecting pipe, such that the bottom of the elbow inlet was about 8 cm above the bottom of the aeration tank, to reduce the movement of settled iron particles into the
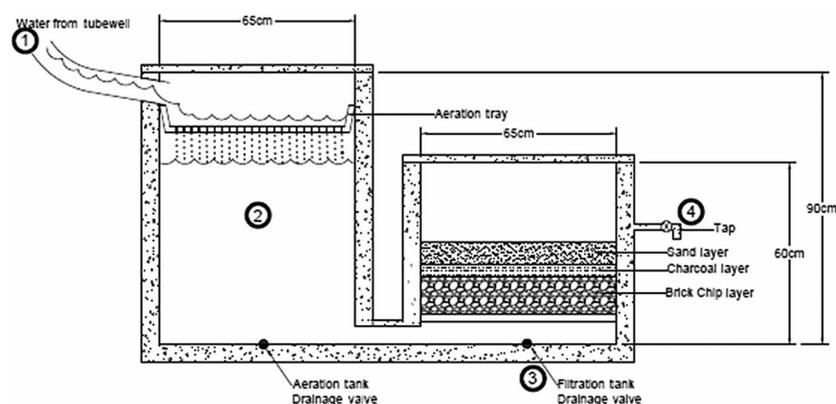

Figure 4 │ AIRP sampling locations.

Figure 5 | Aeration tank with elbow installed.

filtration tank. Samples were collected before and after installation of this elbow to observe the impact on AIRP arsenic removal efficiency.

Batch experiment

A batch experiment was also conducted at site 7 to gauge the rate of iron precipitation and arsenic co-precipitation. First, a number of 500 mL plastic sample bottles were filled to the top with aerated influent water. At given time intervals, the water was filtered and then tested for arsenic and iron. Initially, the containers were left open to the atmosphere. After 4.5 h the lids were closed and the containers were mixed approximately once each 2 h to simulate iron precipitates moving through the water, as would occur in an actual AIRP. There were a few notable differences between this batch test and field conditions. Firstly, settling time was much shorter in the batch test, owing to the smaller settling depth. Secondly, in the field, water in the aeration tank was not well mixed, but would have been influenced by the addition of new water. As new water was added, more precipitates would have formed and settled, potentially removing dissolved arsenic at lower depths.

Analysis

Temperature, pH and oxidation-reduction potential (ORP) were measured *in situ* for each sampling location with field-level probes. For each sample location (influent, aeration tank unfiltered, aeration tank filtered, filtration tank filtered, filtration tank unfiltered, and effluent) two samples were acid-preserved in 100 mL plastic sample bottles for later analysis in Dhaka. One of these was used to test arsenic, the other iron. A third sample at each location was collected in a 100 mL glass sample bottle, was not acid-preserved, and was used to analyse orthophosphate on a Hach DR 2400 Spectrophotometer, and was analysed within 8 h of collection. Coliform sampling was done separately from regular chemical sampling, as coliform analysis required use of the BUET laboratory in Dhaka and results were time-sensitive.

Atomic absorption spectrometry (AAS) at BUET was used to analyse arsenic and iron for all samples. About 15% of the arsenic results were then cross-checked with the Bangladesh Council of Scientific and Industrial Research (BCSIR) Dhaka laboratory, which recently received International Organization for Standardization (ISO) 17025 certification. Good correlation was observed between BUET and BCSIR results: 24 tests had a line of best fit of $y = 1.03x$ and an R^2 of 0.965, where x represents BCSIR results and y represents BUET results. The sample set of relative error between BUET and BCSIR results had a standard deviation of 13%.

Iron results were cross-checked with the DR 2400 Spectrophotometer. Hach Method 8008 (the FerroVer Method) was used, which has a range of 0.02–3.00 mg L^{-1}. Dilutions were therefore needed for samples with higher iron concentrations. Good correlation was seen between DR 2400 and BUET results: 52 tests had a line of best fit of $y = 0.96x$ and an R^2 of 0.974, where x represents BUET results and y represents DR 2400 results.

Orthophosphate, was measured on the DR 2400 Spectrophotometer using Hach Method 8114 (the Molybdovanadate Method). The range of detection is 0.3–45.0 mg L^{-1} PO$_4^{3-}$. For six tests done on standards ranging from 1 to 6 mg L^{-1} the line of best fit between measured values (y-values) and expected values (x-values) was $y = 0.99x$. The R^2 value was 0.975. The sample set of relative error had a standard deviation of 8%.

Sample blanks, reagent blanks, duplicate readings, and calibration against standard solutions were used for quality control. All orthophosphate tests were done in duplicate. Some iron and arsenic duplicate tests were done; however, because of cost, most samples were just tested once. Two samples were taken at the same sampling location on five different occasions to

observe the differences sampling techniques have on results. Table 1 provides a summary of these results.

Arsenic results for duplicate samples were within 10% of each other in all cases. The same was true for iron with one exception; the effluent iron at site 7 was so low that even the slight discrepancy of 0.03 mg L^{-1} between results works out to be a 27% difference. The influent and effluent orthophosphate samples were within 10% of each other, however aeration tank orthophosphate results exhibited differences above 10%; the potential for variability at this location was high because of variations in the depth of sampling and the time since water was added to the tank.

Coliform sampling was completed according to *Standard Method 9222* (Membrane Filter Technique, in Standard Methods APHA/AWWA/WEF 2005) with a few slight modifications. Method D was used for faecal coliforms and Method B was used for total coliforms. Rather than incubating samples for 24 ± 2 h, samples were left for approximately 18 ± 2 h, following recommendations from BUET lab technicians (and subsequent observations) that coliform groups lost their colour after this time and were difficult to count. The filter-holding assemblies were not autoclaved, but rather cleaned with alcohol between samples. Plates were also sometimes re-used, after cleaning with acid, followed by cleaning with alcohol. Sample bottles were plastic, and were sterilized by soaking in boiling water. Pipette tips were also sterilized in boiling water. Ice, frozen after being boiled, was used to cool samples between the time of collection and analysis, which was within 9 h. Distilled water was used as rinse water and was coliform-free.

RESULTS AND DISCUSSION

Influent pH ranged from 6.6 to 7.3, ORP ranged from -146 to -76 mV, and temperatures ranged from 20 to 27 °C. At the pH and ORP values observed, arsenic is expected to have been present mostly as As(III) and iron as Fe(II). Influent arsenic at the 21 sites ranged from 1 to 232 µg L^{-1}. Iron ranged from 5.4 to 24.4 mg L^{-1}, and orthophosphate ranged from 1.5 to 5.7 mg L^{-1}. Eleven of 21 sites tested exceeded the Bangladesh standard of 50 µg L^{-1} arsenic in the influent, and 20 exceeded the WHO guideline of 10 µg L^{-1}. All 21 AIRPs were able to achieve the Bangladesh standard in the effluent, and 17 of the 21 achieved the WHO guideline in the effluent.

All AIRPs are identical in construction (except one in which the two tanks are the same height). Therefore, performance variations are due to influent water chemistry or usage patterns and methods. As discussed later, the time since the last cleaning impacts performance as well. At the initial eight sites tested, cleaning was reported to have been as recent as 3 days before testing occurred, and as distant as one month before testing occurred. Cleaning dates and times at the other 13 sites were unknown.

AIRP removal efficiencies

AIRP removal efficiencies are listed in Table 2. Iron removal was quite high, averaging 98%. Arsenic and orthophosphate removal were similar, averaging 87 and 89%, respectively. The 99% confidence interval for the mean arsenic removal efficiency is 87 ± 5%. One data point was removed when

Table 1 | Summary of duplicate samples taken

Parameter		Sample Site 7					Site 16 (Day 1 after cleaning)				
		I	I-2	AU-D	AU-S	E	E-2	AU-1	AU-2	E-1	E-2
Iron	C (mg L^{-1})	5.58	5.18	1.25	1.23	0.11	0.08	3.24	3.59	2.48	
	dif. (%)	7		2		27		10			
Arsenic	C (µg L^{-1})	92	85	40	43	29	30	57	60	53	
	dif. (%)	8		7		3		5			
Orthophosphate	C (mg L^{-1})	3.9	4.1	1.7	1.5	0.9	0.85	1.9	2.6	2.7	2.5
	dif. (%)	5		12		6		27		7	

Influent (I), sampling location 1; aeration unfiltered samples (AU), sampling location 2; and effluent (E), sampling location 4 as shown in Figure 4. Designations –1 and –2 differentiate between samples taken at the same location. AU-D, sample taken at approximately 30 cm depth (deep) in the aeration tank; AU-S, 5 cm depth (shallow) to assess variability as a result of aeration tank sampling depth. Percent difference refers to the difference between values as a percentage of the larger value.

Table 2 | Arsenic, iron, and orthophosphate removal efficiency

| Parameter	No. of samples	Removal efficiency (%)			
		Range	Average	Median	SD
Iron	21	94–100	98	99	2
Orthophosphate	20	77–97	89	90	6
Arsenic	20	60–96	87	90	8

calculating arsenic removal efficiency statistics; at this site the influent arsenic was 1 μg/L, the detection limit of AAS. For orthophosphate, all values below the detection limit of $0.3 \, \text{mg L}^{-1}$ were set to $0.15 \, \text{mg L}^{-1}$ when calculating removal efficiency.

Other authors report similar, or lower, arsenic removal efficiencies. Removal efficiencies of 80–95% were observed in municipal systems in the United States when Fe(II) was greater than $1.5 \, \text{mg L}^{-1}$ (McNeill & Edwards 1995, as reported in Fields et al. 2000). The United States Environmental Protection Agency (USEPA 2010) recommends a 20:1 mass ratio of iron to arsenic in order to achieve arsenic removal efficiencies of 80–90%. Municipal systems in the United States would typically add chemicals to oxidize As(III), however. Ahmed (2005), in a study of five community level AIRPs in Bangladesh, concluded that over 75% of arsenic can be removed without using chemicals, if arsenic concentrations in the raw water are below $200 \, \mu\text{g L}^{-1}$, and the iron to arsenic ratio is above 30. Higher arsenic concentrations and lower ratios of iron to arsenic require intermittent dosing of an oxidizing agent to oxidize As(III), according to Ahmed (2005). Rahman & Rahman (2004) performed a field study on 60 AIRPs across Bangladesh. Arsenic removal efficiencies varied from 50 to 90%, with most operating in the 60–70% range.

In laboratory experiments, iron requirements are reportedly higher than the 20:1 iron to arsenic ratio recommended by the USEPA. Meng et al. (2001) found that removal of arsenic in water containing $1.9 \, \text{mg L}^{-1}$ P and $18 \, \text{mg L}^{-1}$ Si required an iron to arsenic ratio of about 40, after oxidation of arsenic. In their batch experiments, Roberts et al. (2004) added iron to a sample of synthetic groundwater (with P $3 \, \text{mg L}^{-1}$ and Si $30 \, \text{mg L}^{-1}$). These samples were well mixed and then left to settle for 4 h. Achieving 90% removal of influent water containing $500 \, \mu\text{g L}^{-1}$ As(III) required $50–55 \, \text{mg L}^{-1}$ of Fe(II) (Roberts et al. 2004), an iron to arsenic ratio of 100.

At the 21 sites tested in this study, iron to arsenic ratios ranged from 36 to 1,920. As a result, influent iron is not considered to be have limited arsenic removal for these raw water sources. Only low correlations were seen between arsenic removal efficiencies and iron to arsenic ratios, iron concentrations, and arsenic concentrations ($R^2 < 0.07$). This is likely because there were adequate adsorption sites for arsenic. If iron concentrations had been limiting removal efficiencies, as could be the case in other regions, iron could be added. Fe(II) is continuously released during the corrosion of metallic iron (Roberts et al. 2004), which is widely available (Leupin & Hug 2005).

Figure 6 shows arsenic removal at each of the 21 sites. The single result for which 60% removal was observed (at site 7) is considered an anomaly. Variations in performance were seen throughout a cleaning cycle at site 16, as discussed below. However, at site 16, poor performance was only seen up to 3 days after cleaning. At site 7, removal was found to be 60% after reported cleaning 3–4 days earlier, and on another occasion, after reported cleaning 7–10 days earlier. Dissolved oxygen in the effluent at site 7 was noticeably lower than at other sites, perhaps caused by the presence of organics. Organics can interfere with the formation of flocs (Roberts et al. 2004; Pallier et al. 2010), and can play a role in the settling of iron precipitates (Roberts et al. 2004). Munter et al. (2005) report the potential for both organic substances and silica to form complexes with iron. As organics may influence the complexes formed during iron oxidation, and arsenite oxidation is linked to iron oxidation (Roberts et al. 2004; Leupin & Hug 2005),
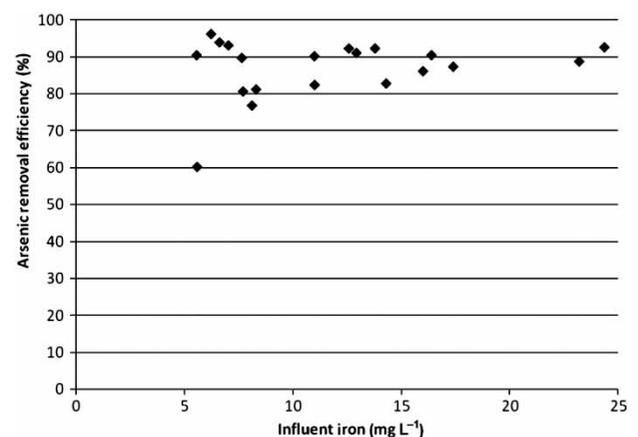
Figure 6 | Arsenic removal efficiency of AIRPs.

the presence of organics may interfere with the oxidation of As(III) to As(V). Organics can also be oxidized by Fe(III), reducing Fe(III) back to Fe(II) (Munter *et al.* 2005). This process may cause iron, arsenic and orthophosphate to re-enter solution, leading to poor removal efficiencies.

Cleaning cycle experiment results

Some of the variability in arsenic removal performance shown in Figure 6 could be a result of the natural changes in performance over a cleaning cycle. Site 16 was sampled immediately before cleaning, immediately after cleaning, and on Days 1, 3, and 8 after cleaning. The sample collected before cleaning was assumed to be 21 days after the previous cleaning; owners reported that they typically clean their AIRP every 3–4 weeks.

Figure 7 shows iron concentrations at various sampling locations over time, after cleaning, and Figure 8 shows arsenic concentrations at various sampling locations over time, after cleaning. Influent iron was 11.0 mg L^{-1} at this site and influent arsenic was 139 μg L^{-1}. Initially, iron concentrations were quite high at all sampling locations. The same trend was seen for arsenic as it adsorbs to, and co-precipitates with, iron. Typically, the residence time in the AIRP is about 2 days. However, during cleaning, both the aeration and filtration tanks were drained completely.
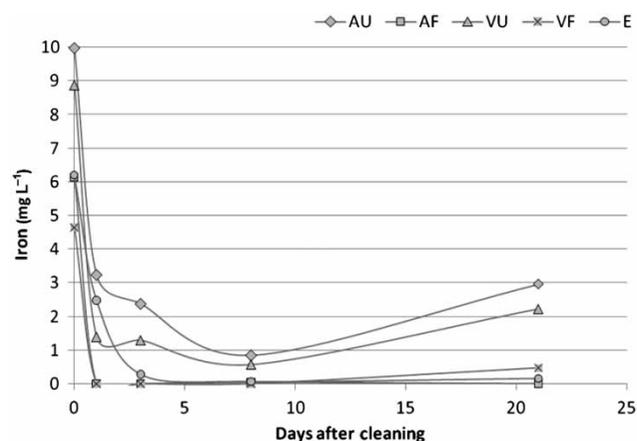
Figure 8 | Arsenic concentrations at various locations throughout the cleaning cycle. Filtered samples were passed through filter paper to remove precipitated iron, and hence any co-precipitated arsenic and orthophosphate (see Figure 7 for abbreviations).

After cleaning, they were filled in about five minutes. This low residence time was insufficient to allow iron to precipitate and arsenic to be oxidized and co-precipitate with iron. To complete oxidation and precipitation of iron, and sorption of anions, 4 h are needed after aeration of groundwater; then, 10–20 h are required for precipitated iron to flocculate and settle (Roberts *et al.* 2004). Owing to insufficient residence time after cleaning, iron probably precipitated and settled in the stored water after the filtration stage; naturally, it would have taken time to dissipate this build-up of iron, as seen in the steady drop in the effluent iron concentration over 3 days.

Placing a valve between the aeration and filtration tanks would allow for increased residence time after cleaning. The valve could be closed during the draining and cleaning of the filtration tank, and opened again to fill the filtration tank after cleaning. It could then be closed to drain and clean the aeration tank. This would ensure a residence time of at least ½ day.

In the aeration and filtration tank unfiltered samples, a minimum concentration of iron and arsenic was reached around Day 8. Concentrations rose just before cleaning. This trend was also seen in the effluent, although the concentration increase just before cleaning was less pronounced; the effect of an increase in iron in the aeration tank and filtration tank was diminished across the filter. This suggests breakthrough had not occurred and there was some other process at work. Although only one cleaning cycle was

Figure 7 | Iron concentrations at various locations throughout the cleaning cycle. AU, aeration tank unfiltered sample (sampling location 2); AF, aeration tank filtered sample (sampling location 2); VU, valve unfiltered sample (sampling location 3, at the bottom of the filtration tank); VF, valve filtered sample (sampling location 3); and E, effluent sample (sampling location 4). Filtered samples were passed through filter paper to remove precipitated iron, and hence any co-precipitated arsenic and orthophosphate.
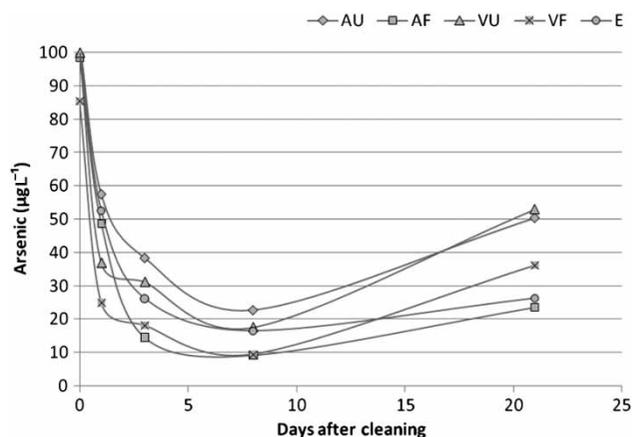
observed, similar results of increasing iron concentrations (though after 3–4 weeks) were seen in community level AIRPs studied by Ahmed (2005).

Desorption of arsenic can occur over time (Fuller et al. 1993), but the corresponding increase in iron concentrations is still not explained. Ahmed's (2005) explanation of increased sloughing of particulates and decreased contact time due to increased flow rates is also insufficient to explain this phenomenon; flow rates did not increase over time. Velocities were low even with the tap open, and particulates sloughed in the bottom of the aeration tank or in the connecting pipe would have had ample time to re-settle. Over time, ferrihydrite will convert to more stable iron oxides, goethite or haematite (Jambor & Dutrizac 1998). Conversion to haematite was thought to be direct, but is now thought to undergo solution-reprecipitation in a manner similar to the conversion to geothite (Jambor & Dutrizac 1998). This process may have occurred in the AIRP and caused the dissolution of both iron and adsorbed anions. The timescale of this transformation is difficult to pinpoint, though is expected to be on the order of months, rather than 8 days.

Alternatively, reducing conditions may have developed in at various locations in the system, causing iron and arsenic to re-dissolve. Reducing conditions could have been created as a result of biological activity, particularly in the sand filter. At the bottom of the tanks burying of precipitates could lead to reducing conditions. An ORP profile through the system would therefore be valuable.

PVC elbow experiment results

Site 7, where the elbow was installed in the connecting pipe between the aeration tank and filtration tank, was the site at which 60% arsenic removal was observed in the initial round of testing. Cleaning was performed 7–10 days prior to testing the first time, and cleaning was performed 6 days prior to testing after elbow installation. Influent arsenic at this site was 97 $\mu g\,L^{-1}$, iron was 5.6 mg L^{-1} and orthophosphate was 3.7 mg L^{-1}. After insertion of the PVC elbow (Figure 5), 67% removal was observed. This improved arsenic removal efficiency was coupled with decreased orthophosphate removal efficiency (96% to 77%). Differences in arsenic and orthophosphate results were also apparent in the aeration tank. In the aeration tank, 70% of arsenic had co-precipitated in the tests done with the elbow, compared to 44% without the elbow, and only 80% of orthophosphate had co-precipitated in tests done with the elbow, compared to 96% without the elbow. However, this was only one test at one site, so general conclusions are not possible.

If the presence of organics did in fact lead to the initial poor performance of this AIRP (60% removal efficiency), oxidation of these organics at the bottom of the aeration tank could reduce Fe(III) to Fe(II) and re-dissolve iron, along with co-precipitated arsenic and orthophosphate. As these processes would likely occur at the bottom of the aeration tank, raising the connecting pipe would have led to a higher chance that iron flocs settling in the aeration tank adsorbed some of these ions, thereby improving removal. However, reasons for decreased orthophosphate removal efficiency with the elbow are still unexplained by such a process, as are differences in the aeration tank results. These differences could be a result of events out of our control: perhaps the source water organics levels had changed, or users had changed their habits of AIRP use.

Batch experiment results

Observed AIRP removal efficiencies were much higher than the removal efficiency measured in the batch experiment conducted at site 7 on aerated influent. In the batch study, only 30–35% arsenic was removed, compared with 60% in the AIRP. Arsenic removal in the batch experiment occurred in the first 2 h. This arsenic removal efficiency is similar to results obtained by Roberts et al. (2004) in their passive sedimentation field trials on Bangladesh groundwater. AIRPs therefore have some advantage not seen in batch experiments.

The improved performance of AIRPs over batch experiments may be a result of multiple iron additions (addition of water each time the tank is filled). With each new addition, more iron flocs move downward through the tank, increasing the contact between dissolved arsenic and iron flocs. Owing to the importance of contact time with iron flocs, for sufficient oxidation and co-precipitation of arsenic, a mixing stage could be considered to achieve better removal in AIRPs.

Multiple additions of Fe(II) have been shown to be advantageous in arsenic removal in batch studies (Roberts *et al.* 2004). The experiments by Leupin & Hug (2005) investigated arsenic removal with Fe(0) in aerated columns. The authors explain that 'addition of Fe(II) in multiple doses with sufficient time for Fe(II) oxidation leads to higher fractions of oxidized As(III) than one single addition of the same total amount of Fe(II)', and 'slow and continuous release of Fe(II) in a column of corroding iron thus provides ideal conditions for the oxidation of As(III), as long as the water contains dissolved O_2 and there is sufficient time for the oxidation of Fe(II)'. Additionally, multiple additions give phosphorus 'less of a headstart on As(V) in the competition for sorption sites' (Roberts *et al.* 2004). Multiple additions of water to the aeration tank in AIRPs are therefore thought to be ideal for arsenic removal.

Microbiological results

The performance of AIRPs with respect to arsenic has been discussed above. It is important to consider the microbial performance of AIRPs as well. Results of microbiological tests are presented in Table 3. Samples collected aseptically were taken after applying alcohol to the tap, and burning it with a flame. Influent samples were collected directly from the tubewell (or the hose attached to an electric pump, if used) while effluent samples were collected from the AIRP tap. The faecal coliform counts from a pond are shown for comparison, as this pond was an alternative drinking water source used by some households in one village.

Influent samples were generally low in faecal coliforms (<4 CFU/100 mL), although still above the WHO guideline and Bangladesh standard of <1 CFU/100 mL. Results for faecal coliforms in influent samples were found to be less than the detection limit of 1 CFU/100 mL five times out of eleven. At three of those five sites, however, faecal coliforms were present when tested on another occasion. Site 77 on one occasion had a high influent count of 61 CFU/100 mL.

Faecal coliform counts generally increased from influent to effluent (normal sample collection), by anywhere from 0 to 22 CFU/100 mL, except at site 77. At this site, faecal coliform counts were high in the influent during the first visit (61 CFU/100 mL), and decreased throughout the AIRP (to 51 CFU/100 mL). During the second visit, influent counts

Table 3 | Coliform counts

| | | Coliform count (CFU/100 mL) | | | |
| | | Influent | | Effluent | |
Site	Sampling date	Aseptic sample collection	Normal sample collection	Aseptic sample collection	Normal sample collection
53	04-Nov-09		<1		4
7	03-Nov-09		<1		1
16	03-Nov-09		4		9
62	04-Nov-09		1[a]		1
32	03-Nov-09		1		2
77	04-Nov-09		61		51
	05-Dec-09	<1	<1	122	145
67	04-Nov-09		2		13
	05-Dec-09		<1[a]	<1	1
	05-Dec-09		7[a]	32	46
42	03-Nov-09		4		26
	05-Dec-09	<1	<1	3	3
	05-Dec-09		11	73	
Pond	05-Dec-09		1,120		

Shaded rows show total coliforms; all other rows show faecal coliforms.
[a]Sample taken from an electric pump attached to the tubewell.

were low (0 CFU/100 mL), and effluent counts were high (145 CFU/100 mL). The owners of the AIRP at site 77 did not use their AIRP regularly, which was uncommon for AIRP owners. Due to high coliform counts and high variability between sampling events, it is believed that contamination was caused by users at this site. Faecal coliform results depend on hygiene, and whether users touch the tap outlet when taking water.

Table 3 shows that some microbial contamination was introduced in the AIRP itself, as effects of the user at the tap were eliminated through aseptically collecting effluent samples. The increase in total coliforms between influent samples collected without alcohol and flame sterilization and effluent samples collected aseptically further support this conclusion. Microbial contamination could have been introduced through handling of the sand when cleaning. After cleaning AIRPs, owners sometimes poured hot water on the sand, but this was likely insufficient to kill all coliforms, if present.

Despite this increase in faecal coliforms through the AIRP, the average effluent faecal coliform counts remained low; the average effluent count was 6 CFU/100 mL, and the

average influent count was 1 CFU/100 mL (excluding site 77). This can be compared with the pond sample (1,120 CFU/100 mL faecal coliforms), where some households got drinking water in one of the villages visited.

Overall, the AIRPs studied were very well maintained, and users were capable of the prescribed maintenance procedures. However, educating users on the importance of avoiding touching the tap, and hand washing prior to cleaning the AIRP, may decrease effluent faecal coliform counts.

CONCLUSIONS

Influent concentrations of arsenic, iron and orthophosphate were variable (arsenic 1–230 $\mu g\,L^{-1}$, iron 5.6–24.4 $mg\,L^{-1}$, and orthophosphate 1.5–5.7 $mg\,L^{-1}$), among the 21 sites tested even in a small geographic area. The influent at 11 of the 21 AIRPs tested exceeded the Bangladesh standard for arsenic of 50 $\mu g\,L^{-1}$, and 20 exceeded the WHO guideline of 10 $\mu g\,L^{-1}$. All 21 AIRPs achieved the Bangladesh standard in the effluent and 17 were able to achieve the WHO guideline.

AIRPs were able to remove, on average, 87% of influent arsenic. The standard deviation of arsenic removal efficiency among 20 AIRPs (one AIRP with 1 $\mu g\,L^{-1}$ influent was ignored for statistics purposes) was 8%. Only low correlations were found between arsenic removal efficiencies and iron to arsenic ratios, iron concentrations, and arsenic concentrations. This is believed to be a result of abundant natural iron in the influent water, resulting in adequate adsorption sites.

All 21 sites exhibited greater than 80% removal of arsenic except two; one had 77% removal and the other 60%. At the site with 60% removal (site 7), organics may have played a role in influencing iron flocculation and settling, arsenic oxidation, and/or re-dissolution of iron and co-precipitated ions. Low dissolved oxygen in the effluent provides support for this theory. Elevating the connecting pipe at this site (site 7) showed a slight improvement in arsenic removal efficiency (67% removal) on one sampling occasion.

After cleaning at site 16, arsenic and iron concentrations were high because of the inadequate residence time. This could be rectified through the installation of a valve between the aeration and filtration tanks, and cleaning each tank

separately. Effluent arsenic and iron concentrations decreased steadily until 8 days after cleaning, but were higher just before cleaning. Iron crystallization, or the generation of reducing conditions, may have played a role in this rise in concentrations.

A batch experiment at site 7 resulted in only 30–35% arsenic removal, whereas in the AIRP 60% removal was observed. The superior performance of AIRPs in the field is thought to be a result of multiple iron additions; adding water to the tank a number of times throughout the day is thought to leads to a higher fraction of oxidized As(III), gives orthophosphate less of a head start in competing for adsorption sites, and increases contact time of dissolved arsenic with iron flocs.

The tested AIRPs decreased arsenic risk, but faecal coliform counts generally increased from 1 to 6 CFU/100 mL through AIRPs, on average. This highlights the importance of proper AIRP cleaning and user hygiene. Faecal coliform counts remained low, however, and compared with an alternative drinking water source, AIRP water quality was quite good.

Overall, reasonable arsenic removal from drinking water was obtained (arsenic removal met the Bangladesh standard, and in most cases the WHO guideline) in the study area, as a result of both AIRP performance and influent concentrations of arsenic and iron in the area. Additionally, most users were pleased with their AIRP performance, the AIRPs studied were well-maintained, users were capable of the prescribed maintenance procedures, and AIRPs are inexpensive compared with other arsenic removal devices. For these reasons, this technology shows promise for use in areas of Bangladesh with high natural iron, where users are concerned with arsenic, iron, or both, in their drinking water.

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