Application of a simple arsenic removal filter in a rural area of Bangladesh
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ABSTRACT
A simple and low-cost household-based arsenic (As) removal filter (ARF) was tested under actual field conditions in a rural area of Bangladesh. The ARF consisted of a ceramic filter made of clay soil and rice bran collected on-site, iron netting and iron bacterial sludge liquor. Fifteen ARFs (14 shallow and one deep tubewells) were installed in three villages (five in each area) in the Khulna region (southwestern region of Bangladesh), and their performance was evaluated. More than 60% of ARFs produced effluent with As <50 μg/L (Bangladesh standard level). The effects of Fe and P on As removal were the same as in laboratory experiments. X-ray Absorption Fine Structure (XAFS) analysis showed the adsorption of primarily As(V), with lesser amounts of As(III). Continuous As removal performance was observed over 1 year of ARF use. By introducing a double ARF system, the As removal was significantly enhanced for the region with high As contamination levels. The ARF manufacturing cost was estimated to be US$4–5, which is low and affordable to the rural households of Bangladesh. The ARF, made of locally available materials, had a low cost and minimal maintenance and showed high user acceptance, satisfaction and sustained use.

Key words | arsenic removal filter, groundwater contamination, iron oxidation, rural area of Bangladesh, simple treatment

INTRODUCTION
In the 1970s, millions of tubewells were installed to improve the health of people in Bangladesh and India by replacing their dependence on polluted surface waters (Nicksen et al. 1998). Unfortunately, after two decades of use, in the early 1990s, the tubewell water in Bangladesh was found to be highly contaminated with arsenic (As) (Hossain 2006). Today, in Bangladesh and India, over 100 million people consume drinking water with As concentrations exceeding the guideline values of the WHO and EPA (10 μg/L) and the Bangladesh standard (50 μg/L) (Bangladesh Bureau of Statistics/UNICEF 2009; Sarkar et al. 2010). This crisis is expanding to Southeast Asia and affects several countries including China, Cambodia and Vietnam (Berg et al. 2007). Several technologies have been developed and tested to mitigate the As, and some of them have gained wide-scale application in the field (Hussam & Munir 2007). Although the As removal efficiency of these technologies is high, they have had several drawbacks, including being expensive and difficult to maintain because of their use of a sand bed for filtration (Shafiquzzaman et al. 2009). Moreover, community-based technologies are difficult to maintain after installation because of the lack of ownership and social mobilization. Household water treatment plays an important role in improving drinking water in developing countries (Rosa & Clasen 2010). Ceramic water filters were applied to two household-scale drinking water systems to obtain successful reduction of diarrheal disease (Brown et al. 2008). Therefore, when using existing, shallow tubewells, a simple, cost-effective, household-based technology would be a good option for providing safe drinking water in rural areas.

In our previous study, an arsenic removal filter (ARF) was developed and its ability to remove As from synthetic...
groundwater at the laboratory scale was assessed (Shafiquzzaman et al. 2011). This ARF was operated based on the principle of biological Fe(II) oxidation, coprecipitation and subsequent removal of As, Fe and others contaminants by filtering through a simple ceramic filter. In this system, As was removed via the formation of biological Fe(III) oxides from Fe(II) in groundwater and those released from iron netting corrosion. Use of the iron-oxidizing bacteria in the system helped to form biological iron oxides, and As was removed by adsorption and coprecipitation (Shafiquzzaman et al. 2008). Iron netting was used as a source of zero-valent iron to supply soluble iron to the reactor, which is the most prominent technology for removing As (Manning et al. 2002; Leupin & Hug 2005). The precipitate formed was subsequently trapped by the ceramic filter, thus removing As from the water. Although enough data have not been collected yet, ARF is also expected to remove rather harmful microorganisms as well as the other ceramic filters do (Brown et al. 2008).

This study evaluated the long-term field-level performance of the ARF for As removal. Accordingly, 15 ARFs were installed at household level in the southwestern region of Bangladesh, and the removal performance of As and other contaminants was monitored for 1 year. Social acceptability was also investigated by a questionnaire survey to improve the design and assist in decision-making regarding the widespread implementation of ARFs.

MATERIALS AND METHODS

The ARF

A detailed description of the ARF design is given in our previous study and in our patent (Shafiquzzaman et al. 2011). The ARF was composed of a 14–15 L clay pot, which was used as a reactor and placed on a wooden stand, and an effluent plastic bucket (Figure 1). The reactor consisted of a ceramic filter (10 cm diameter, 10 cm height and 2 cm thickness (Figure 1(a)), iron netting (600 g) and iron bacterial sludge liquor (dry weight of 5 g). The ceramic filter was made by mixing clay soil and rice bran in a proportion of 80:20 (by weight), sun-drying for 48–72 h and then firing in a local small-scale pottery kiln at 900 °C. The opening face of the ceramic filter was attached to the bottom of the reactor using cement paste and was covered by a cube-shaped box made of iron netting.

Site selection, ARF installation and operation

The field study was conducted in three villages located in the Khulna region (in the southwestern region) of Bangladesh: Fakirhat, Mollahat and Rupsa. Recent reports showed that 66% of groundwater from the shallow aquifer of this region exceeded the Bangladesh standard for As level of 50 μg/L (Kinniburgh & Smedley 2001). The socio-economic

Figure 1 | (a) Schematic representation of the ceramic filter, (b) ceramic filter inside the reactor with iron netting, (c) schematic representation of the ARF, (d) the ARF in operation and (e) a double ARF system.
profile of the area was shown in Shafiquzzaman et al. (2009). In August 2009, 15 ARFs (14 in shallow and one in deep tubewells, five in each area: F1-F5 in Fakirhat, M1-M5 in Mollahat and R1-R5 in Rupsa) were freely provided and installed. The groundwater was poured into the ARF (at the maximum capacity of approximately 14 L) once a day and all of it allowed to filtrate by gravitational pressure to the effluent bucket. The amount of dissolved oxygen (DO) in the freshly withdrawn groundwater from the tubewell was increased by aeration; the groundwater was poured from one bucket to another a minimum of five times before it was poured into the ARF. The initial flow rate at the beginning of the filtration was measured while the rate decreased according to the decrease of water level. The water quality of the influent groundwater and the effluent of the ARFs were monitored for 33 days.

**Long-term monitoring of the ARFs**

Five ARFs (F2, F5, M2, M4 and M5) were selected from the 15 ARFs. They were kept in operation by pouring water once a day for 1 year and their performance was monitored in March (dry season) and in September (wet season) 2010. The ARFs were maintained by the inhabitants of the households, including simple physical cleaning of the filter. Questionnaires on user acceptance were conducted in both periods.

**Double ARF systems**

Two tubewells in Rupsa (R2 and R3) whose raw As concentration were more than 500 μg/L were selected for installation of a double ARF system in September 2010. In the system, two ARFs were connected in series (Figure 1e), and a two-step filtration was performed using the two reactors. The raw water was poured in the first reactor and its filtrate then passed through the second reactor to the effluent bucket.

**Analytical methods**

The following values were measured on-site: pH, oxidation reduction potential (ORP) and electric conductivity (EC) (HORIBA-D-545); DO and temperature (HACH-HQ30d); alkalinity and total hardness (Aqua Check ECO, Siemens Medical); and Fe(II), phosphate and silicate (Pack Test, Kyoritsu). The filtration rate was estimated by measuring the filtrate water amount immediately after pouring the raw water. The following values were measured in the laboratory: As (ICP-MS, YOKOGAWA, HP 4500); T-Fe, Mn, Ca, Mg and Si (ICP spectrometry, Seiko SPS 4000); DOC (total organic carbon (TOC) after filtration, Shimadzu TOC-VCPH/CPN); and phosphorus (JIS K 0102).

**XAFS analysis for removed arsenic form**

To clarify the As form (trivalent or pentavalent), the As L3-edge spectra of the XAFS (X-ray Absorption Fine Structure) of the sludge produced in the ARFs were measured and compared with the spectra of standard compounds using the BL-10 in the SR center of Ritsumeikan University.

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**RESULTS AND DISCUSSION**

**Influent characteristics**

The average and standard deviation of the pH, ORP, EC, temperature and DO of the 15 tubewells at 33 different monitoring times are shown in Table 1. The values of the influent DO before and after aeration are shown in the table, and the enhancement of the DO by the simple aeration technique is clear. The influent pH and the conductivity of F1 were higher because it was a deep tubewell. The pH (neutral), ORP (reductive), DO (2.5–3.5 mg/L) and temperature (approximately 27 °C) of all three areas were almost the same, except for F1. The EC varied with the area and was lower in the Rupsa area. The alkalinity and the hardness were 180 mg/L and approximately 425 mg/L, respectively, in all of the areas. The influent As in Fakirhat, Mollahat and Rupsa ranged from 207 to 219, 178 to 395 and 340 to 585 μg/L, respectively, except for F1 (26 μg/L), as shown in Figure 2. The As in Rupsa was high, and its average exceeded 500 μg/L for R2 and R3. The influent Fe, P, Mn and DOC ranged from 6.7 to 14.5, 1.2 to 2.4 and 2.0 to 12.3 mg/L, respectively, except for F1, as shown in Figure 3. In Rupsa, the average Fe, P and DOC were low, whereas Mn was high. Fe and DOC were high in Fakirhat. These differences
Removal performance of the AR

Higher than the others, respectively, except for Ca and Mg, in F1, which were within the range of 95–135, 23.6–81.0, and 141–237 mg/L, could be caused by the differing soil contents in the three areas (Azam et al. 2009). Influent Ca, Mg, and Si exhibited small variation in the three areas, and their values were (81.0 and 14.1 mg/L, respectively). The DOC concentrations decreased from 10 to 6 mg/L within 15 days followed by gradual decrease and then stabilised at approximately 4.1 mg/L after 1 month. The pH fluctuated from 7.0 to 7.2 in F1, 7.2 to 7.4 in F2, and 7.3 to 7.5 in F3. ORP in F1, F2, and F3 was 95.4, 99.8, and 101.5 mV, respectively. TheDO in F1, F2, F3, M1, M2, M3, M4, M5, R1, R2, R3, R4, R5 was 4.1, 4.2, 4.3, 4.4, 4.5, 4.6, 4.7, 4.8, 4.9, 5.0, 5.1, 5.2, 5.3, 5.4, 5.5 mg/L respectively.
pH and EC showed no change between the influent and the effluent, as shown in Table 1. The effluent temperature increased by 1 °C, while DO decreased a little during filtration. The effluent ORP changed to oxidative conditions after filtration. Alkalinity and hardness were the same for the influent and the effluent. The effluent As concentrations in Fakirhat, Mollahat and Rupsa ranged from 35 to 51 (removal efficiency 77–85%), 32 to 163 (60–93%) and 36 to 190 μg/L (64–85%), respectively, except for F1 (10 μg/L and 60%), as shown in Figure 2.

The results showed that all of the ARFs in Fakirhat (F1-F5) and three in Mollahat (M2, M4 and M5) of which influent As concentration was around 200 μg/L reduced the As level to the Bangladesh standard level (<50 μg/L). Effluent Fe, P and Mn were low (<0.6, <0.2 and <0.3 mg/L, respectively) because of the filtration, but the removal of DOC was not significant, as shown in Figure 3. Although Ca was reduced after the filtration to less than 50% of its initial value, the effluent Mg and Si were almost the same as the influent.

The reduction of As, Fe and P suggested that oxidation of iron and adsorption of As and P by iron floc effectively occurred in the ARFs. A model equation was developed from our laboratory study with synthetic groundwater which described the reduction of As concentration through the ARF using influent Fe(II) and P concentrations (Shafiquzzaman et al. 2011).

As removal (μg/L) = 33 (μg/mg) × Fe (mg/L) − 45 (μg/mg) × P (mg/L) + 430 (μg/L)

The equation showed that P inhibited the As removal. The inhibition of As removal by P was caused by competition of As and P for sorption sites on the precipitated iron hydroxides surfaces (Shafiquzzaman et al. 2011). The calculated As reduction using the influent Fe(II) and P obtained in this study were compared with the actual reduction of As (Figure 4). Although the actual reduction was slightly lower than the calculated value in the high As region, the calculated and actual As reductions were in good agreement, as shown in Figure 4. The agreement suggested that Fe(II) accelerated while P inhibited the As reduction in actual groundwater in Bangladesh in the same manner as in the laboratory study.

The L3-edge spectra of the XAFS of the As standards and the sludge taken from the ARFs of three areas are shown in Figure 5. The difference in the As(III) and As(V) standard spectra were clear for the peaks of the absorption between 1,325 and 1,335 eV. All of the sludge seemed to primarily consist of As(V) when compared with the standard spectra, which have a right peak. However, the spectra also suggested the existence of some As(III) in the sludge because of the left peak, especially in F3 and F4. By using As K-edge XAFS analysis, Fujikawa et al. (2007) reported that As(III) and As(V) adsorbed to iron floc, which agreed with our results.

The long-term performance of the ARFs

All five ARFs continued to be used for 7 months and were found to be in use during the March survey (dry season). The As concentrations in the influent and the effluent are shown in Figure 6 (left and right, respectively), along with those from August 2009. Three of the five ARFs were found not to be in use in the August 2010 (wet season) survey because the households used rainwater. The five ARFs including the three not in use were filled with influent, and their As concentrations and the effluent concentrations are shown in Figure 6.

The influent As was found to be higher in the wet season than the dry season, similar to the seasonal change reported by Tanabe et al. (2001), although Tareq et al. (2003) reported
no clear seasonal change. One possible reason for the increase of influent As in the wet season is acceleration of As release by rainwater (Azam et al. 2009b). Figure 6 (right) shows that the ARFs could remove As to the level of the Bangladesh standard consistently through 1 year of use, including intermittent use.

The ARFs were occasionally physically cleaned by hand during the period of use. The filtration rate had been almost stable at approximately 4 L/h during a year. The simple physical cleaning seemed to maintain the long-term As removal performance of the ARFs, as well as a stable filtration rate.

**Performance of the double ARF system**

Figure 7 (left) showed the As concentrations of R2 (influent: $443 \pm 41 \mu g/L$, the first reactor effluent: $141 \pm 31 \mu g/L$ and the second reactor effluent: $59 \pm 16 \mu g/L$) and R3 (influent: $475 \pm 25$, the first reactor effluent: $169 \pm 56$ and the second reactor effluent: $85 \pm 20 \mu g/L$). The overall As removal efficiency of R2 was 87% (first reactor: 68±7%, second reactor: 58±11%) and that of R3 was 82% (first reactor: 64±12%, second reactor: 49±12%). These results are...
shown in Figure 7 (right). The results indicated that the As removal performance could be enhanced by introducing a double ARF system for the region contaminated with high levels of As. Almost all phosphorus was removed by the first filtration, and the supplied Fe remained in the first reactor. The removal of As in the second reactor suggested that the iron netting provided enough Fe to remove the remaining As without the interference of the phosphorus.

Acceptance of ARF and its sustainability

The questionnaire survey showed that the users had high acceptance of the ARFs and were satisfied with them. Eighty percent of users reported that the filter was easy to maintain and that the filtered water appeared clear, tasted better and was odorless. Some of the iron netting was found to be broken into several parts from dissolution. This observation indicated that the iron netting should be replaced after 1 year of use. The ARF cost was estimated to be US$4–5 by the total expense for manufacturing. It is less expensive than the other ceramic water filter (Brown et al. 2008) and is easily affordable by the rural people of Bangladesh. The operation was simple, and no chemicals needed to be added. The ARF did not require any special maintenance other than cleaning the ceramic filter by scrubbing with a soft cloth or brush. Careful handling of the iron sludge after cleaning and appropriate use according to the manual are recommended. The only operation and maintenance cost to the users would be the cost of replacing the iron netting once a year.

CONCLUSIONS

This study evaluated the performance and sustainability of a simple household-based ARF in a rural area of Bangladesh over the course of 1 year. The main conclusions of this study were the following:

- The ARF could remove As from actual contaminated groundwater with a concentration of around 200 μg/L to levels below the Bangladesh standard level.
- The developed model equation could be applied to the field data, suggesting that Fe(II) accelerated and P inhibited the As reduction.
- The As L3-edge spectra of the XAFS confirmed that As(V) mainly adsorbed to iron floc; however, some adsorption of As(III) also occurred.
- The ARFs could be used continuously to remove As over a 1 year period.
- The As removal performance could be enhanced by introducing a double ARF system in the region of Bangladesh with high As contamination levels.
- The ARF, made of locally available materials, had a low cost and minimal maintenance and showed high user acceptance, satisfaction and sustained use.

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REFERENCES

environmental engineering research forum, Yamaguchi, Japan 110, 227–229.


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