

Hand-pump subsurface arsenic removal: the effect of groundwater conditions and intermittent operation

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ABSTRACT

Hand-pump subsurface arsenic removal (SAR) has been investigated in rural Bangladesh with different groundwater conditions and intermittent operation modes. Multiple injection-abstraction cycles were performed after injection of 1 m³ of aerated water. From these experiments it can be concluded that hand-pump SAR, in the traditional injection-abstraction design, does not provide drinking water below the WHO arsenic guideline of 10 µg/L. Results show that arsenic removal was not enhanced by: (i) injection of O₂-rich water, (ii) higher Fe:As ratios in the groundwater, or by (iii) multiple injection-abstraction cycles, i.e. at location 1, the breakthrough occurred at abstraction-injection ratios of $V_a/V_i = 2$, for cycle 23. It is proposed that dissolved organic carbon (DOC), bicarbonate and phosphate have a significant effect on the arsenic adsorption process. However, iron removal was very efficient and abstraction-injection ratios increased within successive cycles, with $V_a/V_i > 8$ for cycle 23. Furthermore, intermittent operation reduced arsenic concentrations after stop and restart, suggesting insufficient contact time between soluble arsenic and oxidized iron surfaces around the tube well.

Key words | Bangladesh, groundwater conditions, hand-pump, intermittent operation, iron, subsurface arsenic removal

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INTRODUCTION

Arsenic contamination of groundwater occurs naturally in Bangladesh and has become a serious problem due to the extensive use of tube wells for drinking water abstraction during the last decade. Analyses of groundwater have revealed that at many locations arsenic exceeds the WHO guideline and the Bangladesh drinking water standard, 10 and 50 µg/L, respectively (BGS/DPHE 2001; WHO 2006). This metalloid threatens the long-term sustainability of the local ecosystem and, when it enters the food chain, is a serious hazard to human health (Smith *et al.* 2002; WHO 2006; Dittmar *et al.* 2007).

For many decades, subsurface iron removal (SIR) or *in situ* iron removal, has been used in Europe (von Oesten 1900; Van Beek 1985; Mettler 2002). The same principle

of injection-abstraction has been proposed as a low-cost technology to remove arsenic from groundwater, namely subsurface arsenic removal (SAR) (van Halem *et al.* 2010). During SAR, aerated water is periodically injected into an aquifer through a tube well, forming an oxidation zone in the subsurface. Thus adsorbed Fe²⁺ is oxidized, creating new adsorptive iron hydroxide surface areas. When abstraction starts, groundwater with soluble Fe²⁺ and trace elements passes through this oxidation zone. Consequently, the formed iron hydroxides adsorb soluble constituents, such as iron, arsenic and other dissolved anions and cations (Figure 1). This process is explained by adsorptive-catalytic oxidation theory (Van Beek 1985). When SAR treatment is applied, higher volumes of

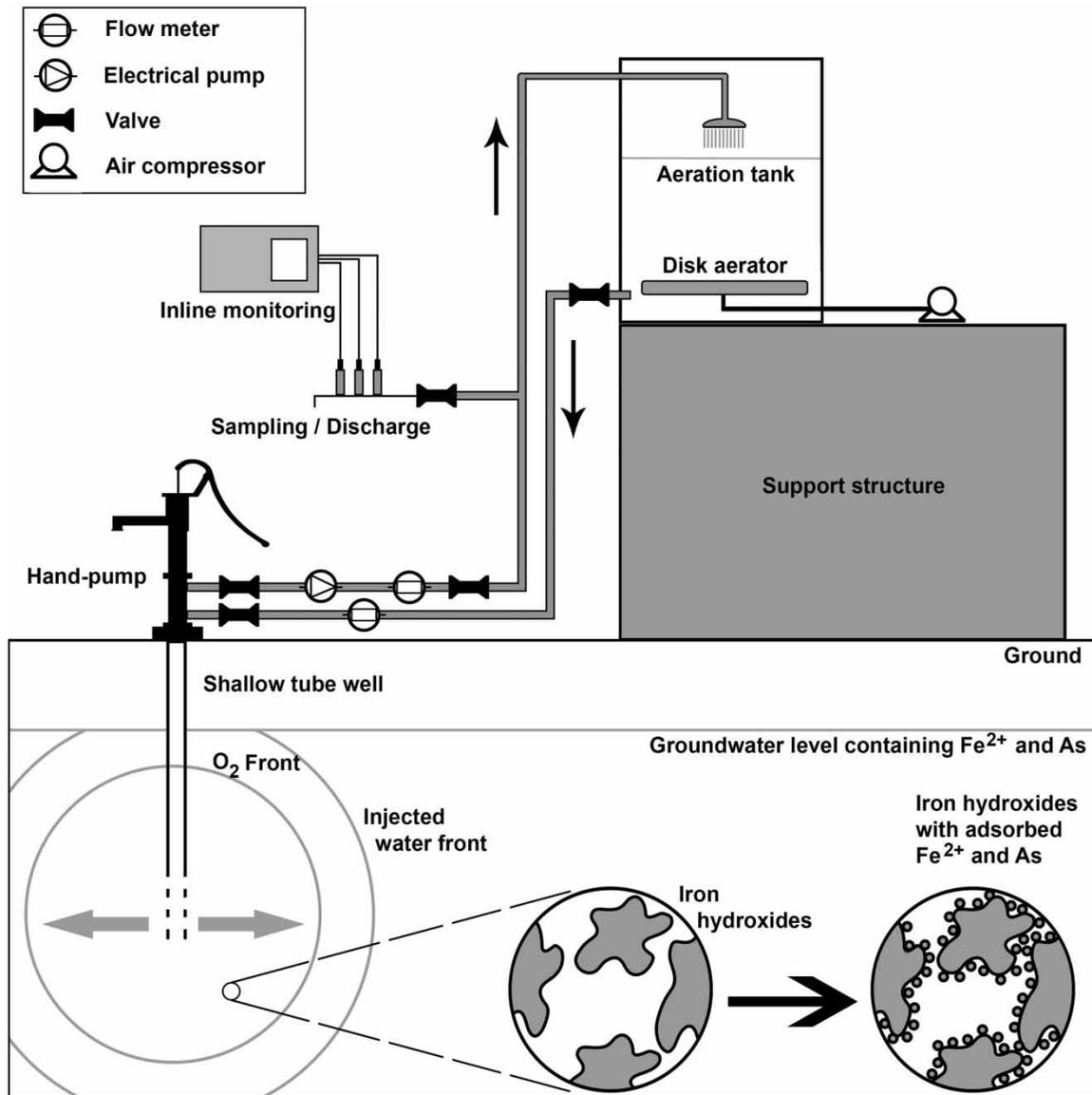


Figure 1 | Schematic of SAR principle and hand-pump SAR experimental setup.

groundwater with lower iron and arsenic concentrations can be abstracted (V_a) than the volume of oxygen-rich water that was injected (V_i). The SAR technology makes use of consecutive sequences of injection (V_i) – abstraction (V_a), referred to as a cycle. The normalized concentration, C/C_0 , as a function of the volumetric ratio V_a/V_i defines the effectiveness of the method.

Previous research has shown that the groundwater composition has a large influence on the efficiency of the SAR process (Kent & Fox 2004; Moed *et al.* 2012). Arsenic

removal depends on the iron removal processes, as precipitated iron hydroxides form the adsorptive surfaces for the arsenic adsorption. In theory, the more oxygen is injected, the more iron is oxidized and the more sorption sites are available (Sharma *et al.* 2001; Dixit & Hering 2003). Therefore, for arsenic removal during SAR, the Fe:As molar ratio and contact time are proposed to be important factors (van Halem *et al.* 2010). The presence of anions such as sulphate, bicarbonate and phosphate play an important role in inhibiting arsenic removal by iron hydroxides (Appelo *et al.*

2002). The competition for adsorptive sites between arsenic species and other ions in groundwater (PO_4^{3-} , SO_4^{2-} , HCO_3^- and dissolved organic carbon (DOC), amongst others) has been investigated in laboratory settings (Meng *et al.* 2000; Stachowicz *et al.* 2008; van Halem *et al.* 2012). Due to the specific binding at the ferric hydroxide surface, phosphate competes directly with arsenic for sorption sites on soil iron hydroxides (Davenport & Peryea 1991). DOC has also been found to affect arsenic adsorption (Xu *et al.* 1991; Grafe *et al.* 2002). Organic matter forms complexes with surfaces of the iron hydroxides, hindering the arsenic adsorption. Thus, the presence of high DOC (>5 mg/L) enhances arsenic leaching from soil grains increasing its mobility (Lin *et al.* 2002).

The effectiveness of SAR is expected to depend largely on local groundwater conditions. Previous SAR studies have been performed at high phosphate and low DOC levels (van Halem *et al.* 2010) or with limited field measurements (Sarkar & Rahman 2001; Sen Gupta *et al.* 2009). The objective of this study was to gain insight into the application and performance of hand-pump SAR in rural Bangladesh: (a) in diverse groundwater conditions; and (b) studying the influence of intermittent operation on the subsurface arsenic adsorption process, i.e., the role of kinetics during SAR.

MATERIAL AND METHODS

The Muradnagar *upazila*, located about 100 km southeast of Dhaka, was selected for the field application of hand-pump SAR. The local groundwater is known to have high iron and arsenic concentrations, low phosphate and pH 6.9–7.0. New wells were drilled using the ‘*sludger*’ method, a manual drilling technique (BGS/DPHE 2001). Each 38 mm (1.5 inch) tube well has a depth of 20.5 to 21.5 meters with a well screen of 3 meters length placed at the bottom of the tube well. The SAR installations were connected to the hand pumps fixed in the new wells. Pipes, valves and flow meters were installed to monitor injection and abstraction processes. At each SAR unit a storage tank combined with showerheads and disk aerators was placed in an elevated area to be used for injection. The experimental setup of both SAR units, referred to as location 1 and 2, is illustrated in Figure 1.

Table 1 | Water quality conditions of the investigated wells

	units	Location 1	Location 2	Location A	Location B
Depth	m	20.5	21.4	*n.m.	*n.m.
pH	–	6.98	6.95	6.85	6.85
EC	$\mu\text{S}/\text{cm}$	630	580	*n.m.	*n.m.
Eh	mV	–130	–137	*n.m.	–170
T	$^{\circ}\text{C}$	26.5	26.5	*n.m.	*n.m.
As	$\mu\text{mol}/\text{L}$	3.59	2.71	1.94	1.94
As(III)		2.94	2.20	1.6	1.66
Fe		0.14	0.23	0.018	0.27
Fe:As		38	83	9	139
Mn		0.00	0.00	0.045	0.006
Na^+		1.29	1.09	0.39	1.04
Ca^{2+}		1.46	0.61	2.1	2.8
Mg^{2+}		1.65	1.65	*n.m.	*n.m.
K^+	mmol/L	0.18	0.19	*n.m.	*n.m.
HCO_3^-		4.87	3.20	*n.m.	*n.m.
PO_4^{3-}		0.001	0.001	0.035	0.053
SO_4^{2-}		0.01	<DL	*n.m.	*n.m.
NO_3^-		0.12	0.04	*n.m.	*n.m.
Cl^-		1.13	1.13	*n.m.	*n.m.
DOC	mg/L	5.05	4.82	1.47	*n.m.

*n.m. – not measured.

The present study – locations 1 and 2 – is compared with a previous SAR study in Manikganj district (locations A and B), with different groundwater conditions, specifically high phosphate and low levels of DOC (van Halem *et al.* 2010). The groundwater composition of the locations 1, 2, A and B is given in Table 1.

Injection-abstraction cycles

The combination of showerheads and disk aerators provided high oxygen concentrations (0.28 mmol/L) in the storage tank. The injection volumes were limited to 1 m³ of aerated groundwater, performed every evening 15 to 18 h before abstraction started. The subsurface treated water was abstracted using two different electrical suction pumps with different pumping rates: 19 L/min at location 1, 22 L/min at locations A and B, and 28 L/min at location 2.

To monitor the injection-abstraction processes, samples were collected and analysed with field test kits,

EconoArsenic Quick™. Arsenic speciation was done using the field Clifford method (Clifford *et al.* 2004). All analyses were confirmed in the laboratory (Perkin-Elmer Flame AAS 3110, Perkin-Elmer Flame GF-AAS 5100PC and Perkin-Elmer 4300 ICP-OES). Sampling points were installed and multimeters were fixed inline to monitor pH (WTW SenTix41), oxidation-redox potential (WTW Sentix ORP), dissolved oxygen (WTW Cellox 325) and electrical conductivity (EC) (TetraCon 325). The EC of injected water is lower than that of the groundwater, indicating when groundwater arrives at the well, and is thus used as a tracer.

Intermittent operation

In rural Bangladesh, shallow tube wells with suction hand pumps are a very popular technology at the household level because they are easy to operate and cost-effective (BGS/DPHE 2001). In order to simulate the 'typical' hand-pump operation, the SAR unit at location 1 was operated intermittently, characterized by starts, stops and re-starts. This simulation is referred to as *intermittent* operation.

Different intermittent experiments were performed: (A) cycle 18, run abstraction until $V_a/V_i \sim 1$ and stop; and (B) cycle 20, run abstraction until $V_a/V_i \sim 2.5$ and stop. In both experiments (A) and (B), the abstraction process re-started again 4 hours after the stop without any injection process. Another final regular cycle (23) was performed as a reference. During the intermittent operation experiments the abstraction rate was decreased to 13 L/min.

RESULTS AND DISCUSSION

Iron and arsenic removal

The purpose of this study was to gain insight into the application and performance of hand-pump SAR in diverse groundwater conditions in rural Bangladesh. The breakthrough curves of iron and arsenic during injection-abstraction cycles 1 and 6, at locations 1, 2, A and B are illustrated in Figure 2 ((a) cycle 1-Fe, (b) cycle 6-Fe, (c) cycle 1-As, (d) cycle 6-As).

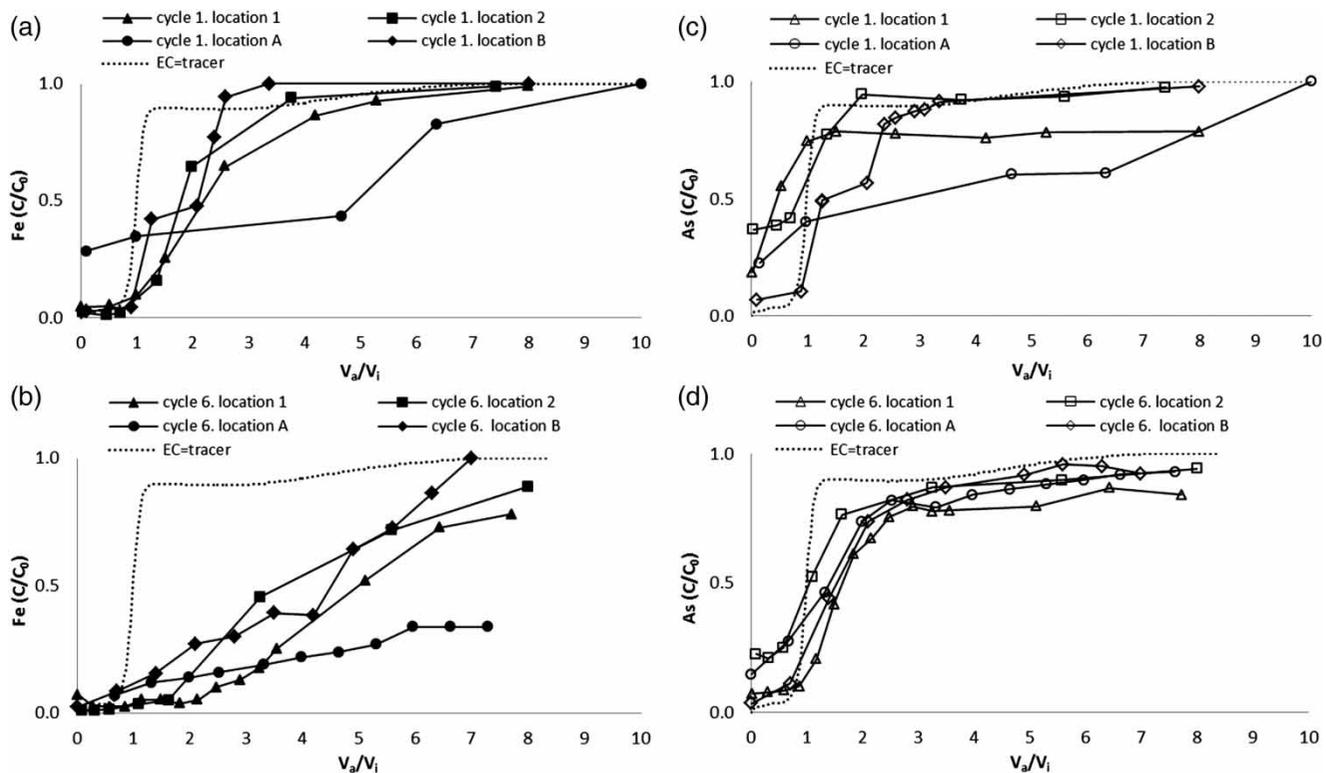


Figure 2 | Iron and arsenic breakthrough curves at locations 1, 2, A and B during: (a) cycle 1-Fe, (b) cycle 6-Fe, (c) cycle 1-As, and (d) cycle 6-As.

Figure 2(a) shows that during cycle 1 an iron breakthrough of $C/C_0 = 0.5$ occurred around $V_a/V_i \sim 2$ for location 1, 2 and B. At location A the breakthrough curve was somewhat peculiar, due to the presence of low concentrations of iron (<0.018 mmol/L). Figures 2(a) and 2(b) clearly show that the removal efficiency of iron improved after successive cycles, Figure 2(b) shows that iron breakthrough ($C/C_0 = 0.5$) occurred around $V_a/V_i \sim 5$, $V_a/V_i \sim 3.5$ and $V_a/V_i \sim 4.5$ for location 1, 2 and B. With breakthrough curves like this, mass balances were calculated. For cycle 6, an estimated iron removal was found to be 0.58, 0.90 and 0.94 mmol in total at locations 1, 2 and B, respectively. At location A, small amounts of iron were removed (~ 0.18 mmol), given the small levels of iron present in the groundwater. It is noteworthy that, during both cycles 1 and 6, the iron breakthrough was delayed compared to the arrival of the groundwater front (=tracer) at the well, when $V_a/V_i = 1$.

Figure 2(c) shows that arsenic levels increased rapidly, simultaneously with the groundwater front, $V_a/V_i = 1$, for cycle 1. At location A and cycle 1, a peculiar arsenic breakthrough curve, similar to that of iron, is illustrated. For cycle 6, Figure 2(d), arsenic breakthrough occurred after $V_a/V_i = 1.5$. At location B, arsenic breakthrough ($C/C_0 = 0.5$) occurred after $V_a/V_i = 1$ and $V_a/V_i = 1.5$, respectively, for cycles 1 and 6. It can be observed that at location 1 arsenic breakthrough occurred after $V_a/V_i = 0.5$ and $V_a/V_i = 1.8$, in cycles 1 and 6, respectively. Two locations, 1 and B, show a slight enhancement in arsenic removal after successive cycles. Therefore, results show that arsenic breakthrough has a noteworthy delay compared to the tracer, i.e., the arrival of the groundwater front at the well. At location 2, arsenic breakthrough occurred around $V_a/V_i = 1$ for both cycles 1 and 6, not showing any improvement of removal efficiency. Given the different settings, the tailings displayed by the arsenic breakthrough curves were very similar at all locations. Compared to iron removal efficiencies, arsenic was removed only in small amounts.

Based on the operational conditions mentioned in the 'Material and methods' section, calculations show that the contact time in the oxidation zone between fresh groundwater and formed iron hydroxides was approximately 25 and 17 min, at locations 1 and 2, respectively, and 20 min

at both locations A and B. Figure 2(d) shows that arsenic adsorption slightly improves with higher contact time. However, an incomplete breakthrough curve for arsenic was observed at all locations. The contact time in the oxidation zone was still insufficient, while adsorption sites for arsenic were still available.

An additional finding was that at locations 1 and 2 the dissolved oxygen levels in the injection water were higher than at locations A and B; thus highly aerated injection water does not increase arsenic removal. Furthermore, higher Fe:As molar ratios, of 38, 83, 9 and 139, at locations 1, 2, A and B, respectively, do not increase arsenic removal.

Complex groundwater matrices have a large influence on the arsenic adsorption process. At locations 1 and 2, the DOC and bicarbonate concentrations were high (Table 1). Therefore, interference with the arsenic adsorption process could have occurred. Organic matter and bicarbonate ions do not directly occupy the available sorption sites, but can cover and 'mask' their surfaces, as a shield, not allowing other ions to pass through and be adsorbed (Harvey *et al.* 2002; Appelo & de Vet 2003; Peyton *et al.* 2006). Contrary to locations 1 and 2, the levels of phosphate were high at locations A and B, in concentrations high enough to interfere with arsenic adsorption (Appelo & de Vet 2003; van Halem *et al.* 2010). At locations A and B, phosphate is identified as the major competing anion, while at locations 1 and 2, DOC and bicarbonate are proposed to interfere, inhibiting higher arsenic adsorption.

Intermittent operation

Figure 2(d) shows incomplete arsenic breakthrough curves, suggesting an insufficient contact time. In order to understand the kinetics of arsenic adsorption in the subsurface, the hand-pump behaviour was simulated. Figures 3(a1) and 3(b1) illustrate the curves for arsenic and Figures 3(a2) and 3(b2) show the curves for iron during intermittent operation and a regular cycle.

The intermittent operation experiments (A) and (B) show that at the start of the process the arsenic curve follows the same pattern as a regular cycle during SAR. The process was stopped after $V_a/V_i \sim 1$ and $C/C_0 = 0.35$ in experiment A, and stopped after $V_a/V_i \sim 2.5$ and $C/C_0 = 0.70$ in

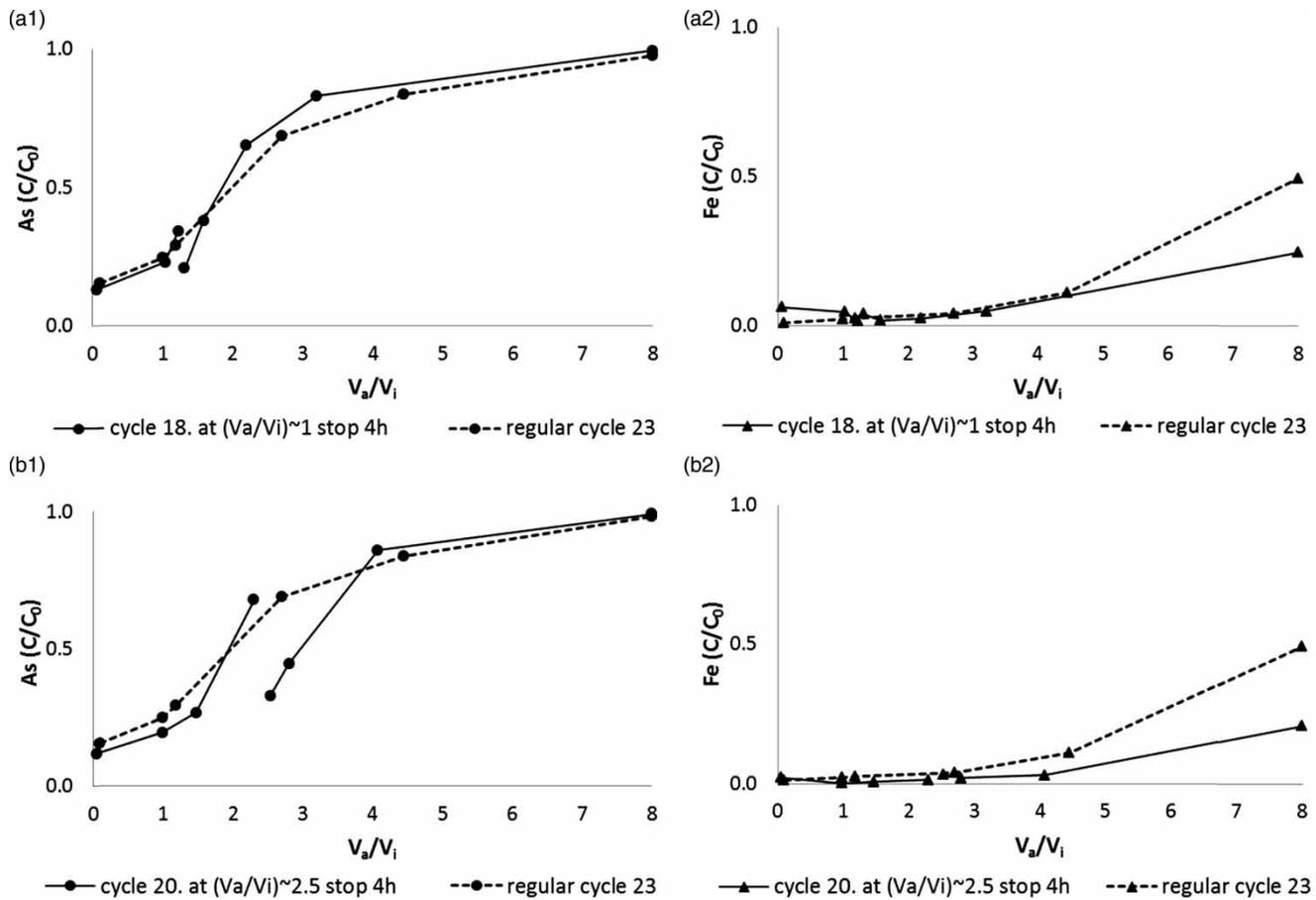


Figure 3 | Arsenic and iron breakthrough curves during different intermittent operation experiments and a regular cycle.

experiment B. After the shutdown of the process, the arsenic concentration was reduced, with a decrease in arsenic levels of 0.49 and 1.25 $\mu\text{mol/L}$, respectively. After restarting the process, the breakthrough of arsenic was resumed, reaching $C/C_0 \sim 1$ at $V_a/V_i = 8$, for all experiments. The results show that if a repeated intermittent operation were used, partial arsenic would be continuously adsorbed after every stop mode and the arsenic breakthrough would be delayed.

The regular cycle 23, shows $C/C_0 = 0.5$ was reached for iron and arsenic at $V_a/V_i = 7$ and $V_a/V_i = 2$, respectively. From these results, it is concluded that arsenic removal slightly improved when compared to cycle 6 in Figure 2(d), but clearer is the improved iron removal after successive cycles. The total arsenic removal was higher during intermittent operation, compared to reference cycle 23. In the subsurface the contact time between the arsenic and

formed iron hydroxides was increased to approximately 37 min. SAR was slightly more effective at a longer contact time, but still insufficient. Based on these results, calculations show that if the contact time is longer than 90 min, with a pumping rate lower than 5 L/min, the more efficient the arsenic adsorption will be. Longer contact time with the iron hydroxides enhances the equilibrium and kinetics of arsenic adsorption.

The intermittent operation also had an effect on iron removal. When abstraction was resumed following a stop, the concentration of iron increased compared to the decreased concentration of arsenic. A peak in iron levels of 0.004 and 0.002 $\mu\text{mol/L}$ was observed in Figures 3(a2) and 3(b2), followed by a small drop, before iron levels increased again. During the stop, mobilization of adsorbed Fe^{2+} occurs. Although, as expected, SIR improved considerably after successive cycles.

Under different groundwater compositions and intermittent operation, the results show that drinking water can be provided below the Bangladesh arsenic guideline of 50 µg/L. Although the WHO guideline of 10 µg/L was not achieved. Therefore, under these conditions, the conventional operation as a 'single' injection-abstraction mode with injection volumes limited to 1 m³ does not seem to be a feasible option for arsenic removal. Iron removal, however, was very efficient and abstraction-injection ratios (V_a/V_i) increased every successive cycle. SIR showed great potential for decentralized application at community-scale in Bangladesh.

CONCLUSIONS

Hand-pump SAR has been investigated at different locations in Bangladesh with the traditional single injection-abstraction mode. From these experiments is concluded that, for common groundwater conditions in Bangladesh, SAR at the hand-pump level does not provide drinking water that meets the WHO arsenic guideline of 10 µg/L. The removal of arsenic was not enhanced by: (i) higher oxygen concentration in the injection water; (ii) higher Fe:As ratios in the groundwater; or (iii) multiple injection-abstraction cycles. It is proposed that DOC, bicarbonate and phosphate have a significant effect on the arsenic adsorption process. The removal of iron, however, was very efficient and injection-abstraction ratios (V_a/V_i) increased with successive cycles. Furthermore, intermittent operation reduced arsenic concentrations, indicating the importance of adsorption kinetics during hand-pump operation.

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