

Well-head arsenic removal units in remote villages of Indian subcontinent: Field results and performance evaluation

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Abstract

Since 1997, over 135 well-head arsenic removal units have been installed in remote villages in the Indian state of West Bengal bordering Bangladesh. Every component of the arsenic removal treatment system including activated alumina sorbent is procured indigenously. Each unit serves approximately 200–300 households and contains about 100 L of activated alumina. No chemical addition, pH adjustment or electricity is required for operating these units. The arsenic concentration in the influent varies from around 100 µg/L to greater than 500 µg/L. In the treated water, arsenic concentration is consistently below 50 µg/L. The units are capable of removing both arsenites and arsenates from the contaminated groundwater for several months, often exceeding 10,000 bed volumes. In the top portion of the column, the dissolved iron present in ground water is oxidized by atmospheric oxygen into hydrated Fe(III) oxides or HFO particles which in turn selectively bind both As(III) and As(V). Upon exhaustion, these units are regenerated by caustic soda solution followed by acid wash. The arsenic-laden spent regenerant is converted into a small volume sludge (less than 500 g) and contained over a coarse sand filter in the same premise requiring no disposal. Many units have been operating for several years without any significant operational difficulty. The treated water is used for drinking and cooking. Most importantly, the villagers are responsible for the day to day operation and the upkeep of the units.

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1. Introduction

In Bangladesh and India, drinking water drawn from underground sources has been responsible for wide-

spread arsenic poisoning affecting nearly 100 million people (Bagla and Kaiser, 1996; Bearak, 1998; Lepkowski, 1998). According to current estimates, the adverse health effects caused by arsenic poisoning in this geographic area are far more catastrophic than any other natural calamity throughout the world in recent times. Although the genesis of arsenic contamination in groundwater in this region is yet to be fully understood, natural geochemical weathering of subsurface soil and

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not industrial pollution is the sole contributor of dissolved arsenic in groundwater (Acharyya et al., 1999; Dhar et al., 1997; Nickson et al., 1998). Concentrations of dissolved arsenic in many existing wells providing drinking water exceed over 200 µg/L, while the maximum permissible arsenic concentration in India and Bangladesh is 50 µg/L. The presence of unacceptable levels of arsenic does not alter the taste, color or odor of water. Also, the health-related impairments caused by arsenic poisoning from drinking water is slow and takes several years before becoming fatal and irreversible from a life-threatening viewpoint.

Rainfall in this region is quite significant and often exceeds 1500 mm/year. Ironically, however, arsenic-free surface water is practically unusable due to poor sanitation practices in the region and consequent contamination with the potential for an outbreak of water borne diseases. In many remote villages, arsenic-contaminated groundwater is the only viable source of drinking water and cost-effective arsenic removal technology is a bare necessity to provide safe drinking water. Besides the presence of an unacceptable level of arsenic, the groundwater is otherwise quite fit for drinking.

Since 1997, Bengal Engineering College in Howrah, India, in association with Lehigh University in Pennsylvania, USA, has installed over 135 well-head arsenic removal units in remote villages in the state of West Bengal, India, which borders Bangladesh. These are essentially fixed-bed columns mounted at the top of the existing hand-pump units. The following guidelines were used in the design, installation and operation of these well-head units:

- chemical addition, pH adjustment and electricity are to be avoided;
- the entire operation should be simple and manual;
- the materials to be used including the sorbent are to be procured from indigenous source only;
- each unit should serve approximately 200 households living within a walking distance of the unit;
- each unit in every village is to be run, maintained and monitored through a committee appointed by the villagers;
- arsenic removed from the contaminated groundwater must be retained in the same premise without any indiscriminate disposal.

Several treatment strategies and equipment configurations are available for removal of trace concentrations of arsenic (Chwirka et al., 2000; DeMarco et al., 2003; SenGupta and Greenleaf, 2002). After taking into consideration the above-mentioned guidelines, the fixed-bed activated alumina column process was adopted for well-head arsenic removal process. The specific

objectives of this communication are to present: pertinent design details of the well-head units; results pertaining to arsenic removal for a prolonged period of time; and scientific analysis of the data for elucidation of arsenic removal mechanism and arsenic containment.

2. Materials and methods

2.1. The well-head treatment unit

The adsorption column mounted on top of the existing well-head hand pump is essentially a cylindrical stainless-steel (SS-304) tank containing two distinct compartments. The upper chamber or head space of the column contains a splash distributor and atmospheric vent connections. This chamber ensures oxidation of dissolved iron into insoluble hydrated Fe(III) oxides or HFO particles. Underneath the head space is the fixed-bed activated alumina followed by graded gravels and the treated water collection chamber. The general configuration of the sorption column and its operational features are schematically presented in Fig. 1A while the specific details are provided in Fig. 1B. The design flow rate of the column operating under gravity is 8–10 L/min. The column is routinely backwashed for 10–15 min everyday and the backwash water is passed through a coarse sand filter to retain the HFO particulates. Fig. 1C provides the pertinent details of the coarse sand filter.

Activated alumina was procured from an indigenous manufacturer (Oxide India Limited, Durgapur, West Bengal) after performing a series of laboratory studies validating its suitability for long-term column operation. The average size of the spherical adsorbent particle varied between 600 and 900 µm; active alumina content was 93.6% by mass. Each column is provided with a flow totalizer to record the total volume of water treated.

2.2. Sample preparation and analysis

For analysis of As(V) and As(III), samples collected at site are first adjusted to a pH near 4.0 and then they are immediately separated using a strong-base anion exchange resin mini-column in accordance with the technique developed by Ficklin (1983) and Clifford et al. (1983). Total arsenic is determined from the original sample, As(III) from the sample collected at the exit of the anion exchange column and As(V) by difference. The technique is validated using samples of known As(III) and As(V) concentrations.

In Bengal Engineering College, India, arsenic is analyzed using an automatic flow injection atomic absorption spectrophotometer (Chemito, India) with hydride vapor generation accessory. At Lehigh University,

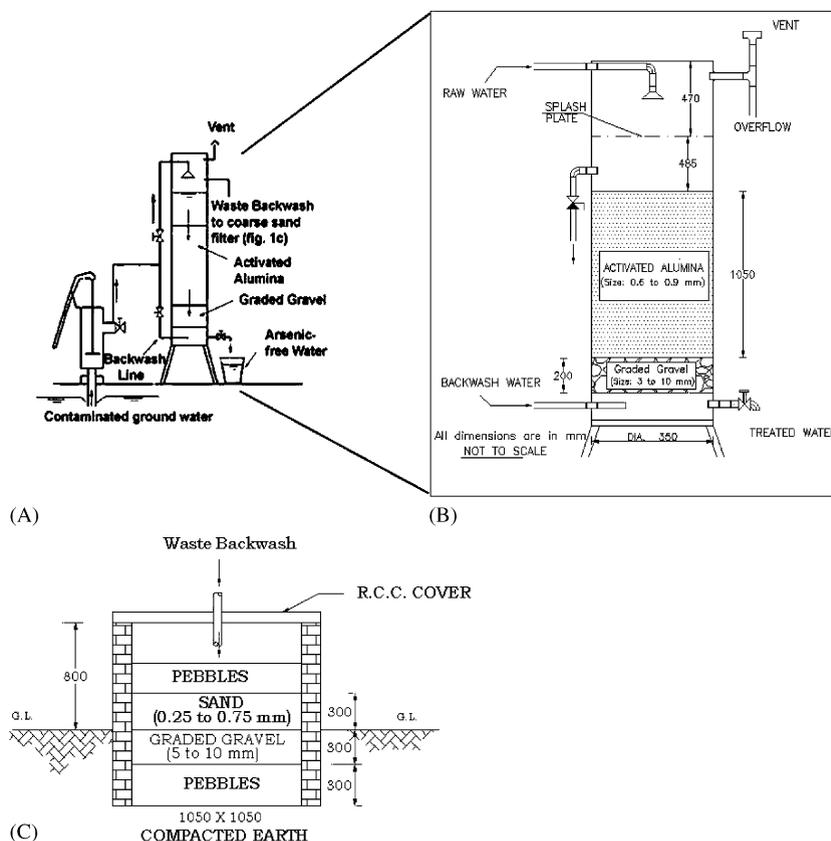


Fig. 1. (A) Schematic of the well-head arsenic removal unit; (B) details of the activated alumina adsorption column; and (C) the coarse sand filter.

USA, arsenic is analyzed using an atomic absorption spectrophotometer with graphite furnace accessory (Perkin Elmer model SIMAA 6000). Samples for analysis of iron are preserved at pH < 2 through addition of a few drops of 8 M HNO₃.

Analysis of dissolved oxygen (DO), hardness, alkalinity, silica and phosphate are carried out in Lehigh University and Bengal Engineering College in accordance with the procedure of Standard Methods (APHA and AWWA and WEF, 1998).

2.3. In situ regeneration

Upon exhaustion of the adsorption column, i.e., after total arsenic concentration in the treated water approaches 50 µg/L, each unit is regenerated. Nearly two bed volumes (175 L) of 4% NaOH are recirculated through the bed for about 90 min. During the entire period, pH of the regenerant solution remains in the vicinity 12.0. The spent alkali is subsequently drained off in the down-flow mode. The bed is subsequently

neutralized with 150 L of 1% HCl. Subsequently, the spent acid is drained and collected in a separate plastic drum. The entire bed is then rinsed with tube-well water for half an hour. At the end of the regeneration, spent acid, alkali and rinse water are mixed and pH is adjusted to around 6.5 by adding 10% hydrochloric acid. Thick brown slurry is immediately formed and disposed of at the top of the coarse sand filter. The arsenic-laden solids with HFO particles are intercepted and retained at the top of the filter. The entire regeneration including the spent regenerant treatment is completed in about 10 h.

3. Results

3.1. Column run performance

Fig. 2 shows the concentrations of dissolved arsenic in both contaminated groundwater (i.e., influent) and treated water from a representative well-head unit for a period of nearly 16 months in Sangrampur village in

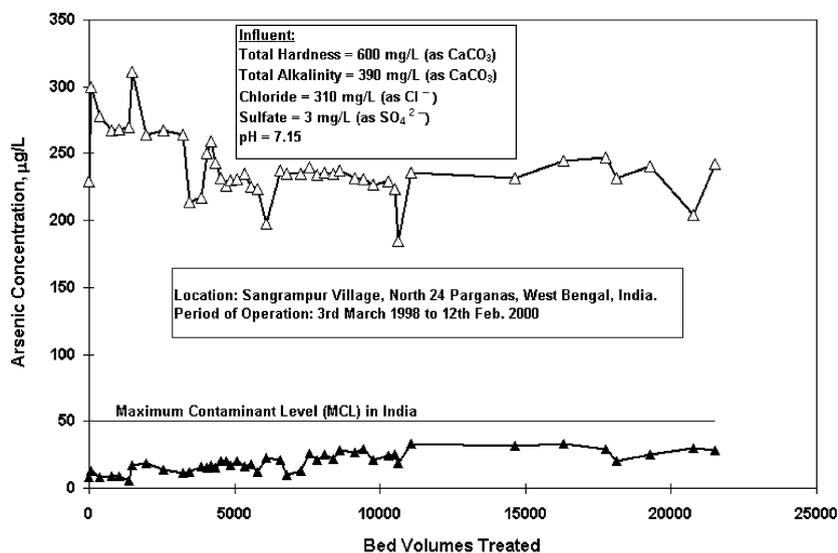


Fig. 2. Arsenic concentration in the influent (open triangle) and treated effluent (closed triangle) versus bed volumes of water fed to the well-head unit.

West Bengal bordering Bangladesh. During this period, the unit served nearly 200 households, i.e., approximately 1000 villagers living within 1.5 km of the treatment unit. Note that while the arsenic in the influent varied between 200 and 300 µg/L, the arsenic concentration was consistently below the maximum contaminant level (MCL) of 50 µg/L. Also, the increase in arsenic concentration in the treated water was very gradual and the samples could, therefore, be collected and analyzed every 2 weeks without the risk of suddenly exceeding the MCL. Phosphate and dissolved silica concentrations in the groundwater did not change significantly with time. While the phosphate concentration was 80–90 µg/L, the silica concentration varied between 25 and 30 mg/L.

3.2. Dissolved iron content and arsenic removal

Fig. 3 shows the influent and effluent arsenic histories for a well-head unit at Debnagar village in Murshidabad district. Note that the arsenic concentration of contaminated groundwater hovers around 500 µg/L and is significantly greater than the arsenic concentrations for the groundwater shown in Fig. 2. Nevertheless, the well-head unit was able to treat nearly 12,000 bed volumes before 50 µg/L of arsenic breakthrough was observed. Total dissolved iron content in this particular location was quite high, i.e., in the vicinity of 10 mg/L. Fig. 4 shows the effluent history of total iron at the same site; note that over 90% iron removal was obtained within the column primarily due to the oxidation of Fe(II) into

hydrated Fe(III) oxide (HFO) precipitates. HFO particles can selectively bind both ligands (i.e., Lewis bases) and toxic metals (i.e., Lewis acids) at different pHs (Clifford, 1999; Gao et al., 1995).

It is well recognized that the presence of dissolved iron, although aesthetically displeasing, contributes toward arsenic removal. As demonstrated later, the well-head unit accomplishes near-complete oxidation of dissolved iron into insoluble Fe(III) hydroxide precipitates that have an ability to sorb both As(III) and As(V) species. Oxidation of dissolved iron alone in the absence of activated alumina is, however, unable to bring the total arsenic concentration below 50 µg/L, as evidenced from independent laboratory batch studies.

3.3. Distribution and removal of arsenic(III) and arsenic(V)

Every water well studied to date has been found to contain both As(III) and As(V) but their relative distribution varies; the presence of As(III) along with As(V) has also been reported in Bangladesh and elsewhere (Hering and Chiu, 2000; Safiullah et al., 1998). The effectiveness of the well-head unit is assessed by its overall arsenic removal ability, i.e., for removal of both arsenates and arsenites. Selected sets of influent and treated water analyses were carried out for a number of well-head units in the region after significant arsenic breakthroughs took place. Table 1 provides the names of the villages, As(V) and As(III) analyses of

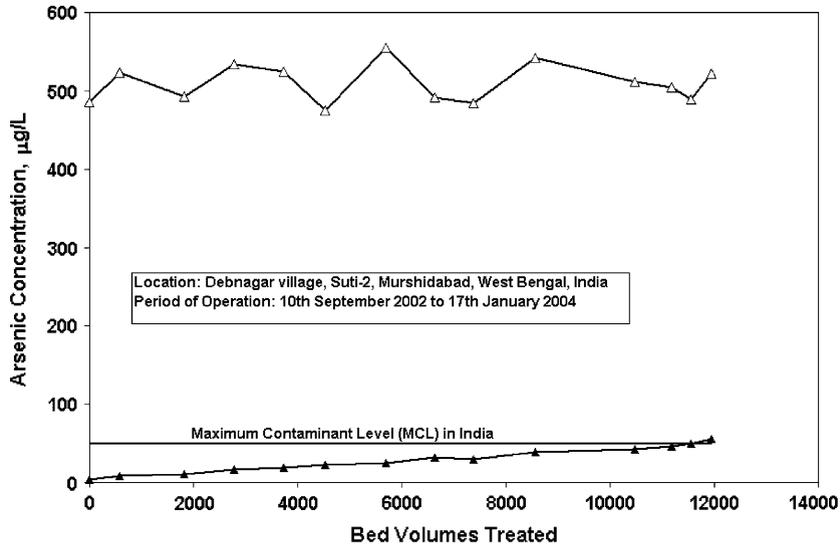


Fig. 3. Arsenic concentration in the influent (open triangle) and treated effluent (closed triangle) versus bed volumes of water fed to the well-head unit.

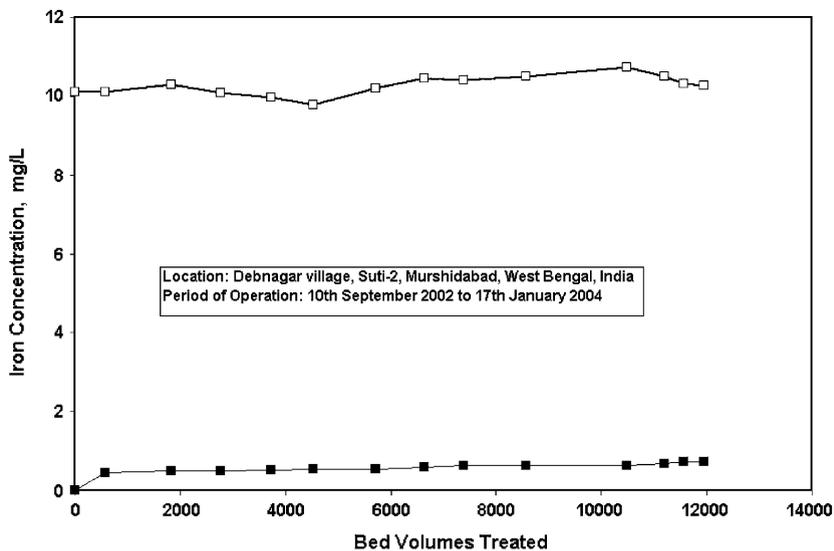


Fig. 4. Iron concentration in the influent (open rectangle) and treated effluent (closed rectangle) versus bed volumes of water fed to the well-head unit.

influent and effluent, and their percentage removal. Note that As(III) removal is equally good or even better in several locations than As(V) removal. Since activated alumina is not effective in removing As(III), the role of dissolved iron or, more specifically, HFO precipitates in arsenic removal process can be readily recognized (Clifford, 1999; Frank and Clifford, 1986; Hering and Elimelech, 1996).

3.4. Regeneration

Barring a few minor exceptions, in situ regeneration follows an identical protocol for each well-head unit as described earlier. Since its installation in December 1999, the unit at Maslandpur village in West Bengal was regenerated several times. The pertinent details of the first regeneration carried out on 22nd July, 2000, are

Table 1
Distribution of As(III) and As(V) in well water and their percentage removal

Location of well-head units	Arsenic ($\mu\text{g/L}$)						% Removal	
	Influent			Effluent			As(III)	As(V)
	As(III)	As(V)	Total	As(III)	As(V)	Total		
Debnagar, Murshidabad	219	144	363	10	23	33	95.43	84.03
Rampur, N 24 Parganas	140	80	220	13	25	38	90.71	68.75
Prithiba, Guma, N 24 Parganas	90	57	147	3	25	28	96.67	56.14
Banipur, Habra, N 24 Parganas	154	63	217	13	47	60	91.56	25.40
Dakshin Chatra, N 24 Parganas	53	53	106	8	13	21	84.91	75.47
South Betpool, N 24 Parganas	108	144	252	22	2	24	79.63	98.61
South Chatra, N 24 Parganas	50	30	80	14	4	18	72.00	86.67
Sendanga, N 24 Parganas	70	30	100	9	12	21	87.14	60.00
Potapara, N 24 Parganas	62	58	120	20	15	35	67.74	74.14
Ashrafabad, N 24 Parganas	115	78	193	20	20	40	82.61	74.36

Table 2
Volumes and compositions of individual spent regenerant streams at Maslandpur Village in West Bengal, India

Description	Volume (L)	pH	Total arsenic (mg/L)	Total iron (mg/L)
Spent caustic	175	12.2	16	74
Spent acid	150	3.0	0.65	130
Spent rinse water	150	3.5	0.1	1

provided in Table 2 which includes the volumes and pHs of spent alkali, spent acid and rinse water and the concentrations of total arsenic and total iron in them. As expected, arsenic concentration in the spent alkali regenerant was by far the greatest. Also, laboratory analyses confirmed that arsenic in spent regenerant was present solely as As(V) or arsenates.

3.5. Dissolved oxygen, dissolved iron and Arsenic(III) removal

Arsenic(III) was significantly present along with arsenic(V) in every water well and it contributed 30–90% of total arsenic for over 100 wells investigated to date. It was well recognized from the very beginning that activated alumina in conjunction with dissolved iron develop a synergy leading to enhanced arsenic removal. In order to develop an insight into the roles of oxygen and dissolved iron, one well-head removal unit in Potapara village, N. 24 Parganas district, West Bengal, where the dissolved arsenic is present almost solely as arsenite or As(III), was identified for an independent field study. Four separate sample ports were provided in the unit as shown in Fig. 5.

At three different time intervals (i.e., at different bed volumes), samples were collected from individual ports and appropriate steps were taken at site to analyze arsenic(III) and arsenic(V) in accordance with the procedure described earlier. Figs. 6A–C show the concentrations of DO, total Fe and As(III) at four different ports for samples collected at different times or different bed volumes of water fed. The following observations are noteworthy: first, regardless of the time of sample collection, the DO is high in the influent and drops to near-zero value at the end of the column; second, iron precipitates or HFO particles are well retained in the column; and third, As(III) removal is quite good in the beginning but it gradually starts breaking through the column as more water is fed.

3.6. Spent regenerant treatment and leachate characteristics

Spent alkali, spent acid and spent rinse water were mixed after each regeneration and the pH adjusted to around 6.5 by adding hydrochloric acid. Laboratory tests with mixed regenerant wastes clearly demonstrated

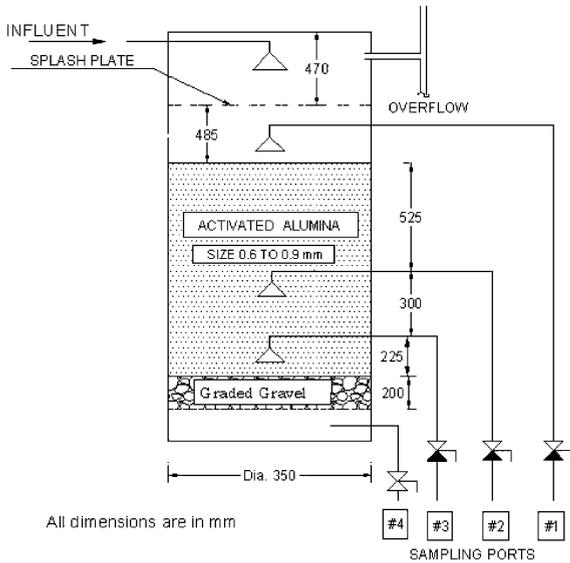


Fig. 5. Schematic of the well-head arsenic removal unit in Potapara village, North 24 Parganas with four sampling ports at different bed depths of the adsorption column.

that the concentrations of dissolved arsenic and iron were minimum in the vicinity of $\text{pH} = 6.5$. A thick brown precipitate formed rapidly following the pH adjustment and the volume of the settled solids in the mixed spent regenerant was less than 5.0 L. Subsequently, the entire amount of slurry was slowly disposed of at the top of the coarse sand filter. Arsenic in the spent regenerant was converted to a solid waste along with iron at near-neutral pH and collected at the top of the coarse sand filter located in the same premise. For every regeneration, the mass of the sludge (dry basis) added to the top of the coarse filter is less than 400 g. According to our estimate, every on-site sand filter may collect and contain the spent sludge for over 30 years. If upon exhaustion, activated alumina is to be replaced in every village with a fresh batch without being regenerated, it would require a disposal of 100 kg of arsenic-loaded medium in a region where such practices currently do not exist.

These filters are open to the atmosphere and hence, besides backwash water, they may also come in contact with rainwater. Laboratory tests were carried out with the mixed regenerant sludge from Maslandpur site including pH adjustment and leachate analyses. Fig. 7 provides a plot of equilibrium pH and dissolved arsenic and iron concentration in the leachates. The results demonstrate that the concentrations of dissolved arsenic and iron tend to be minimum in the vicinity of $\text{pH} = 6.0$. Also, arsenic in the dissolved phase was present solely as arsenate or As(V) .

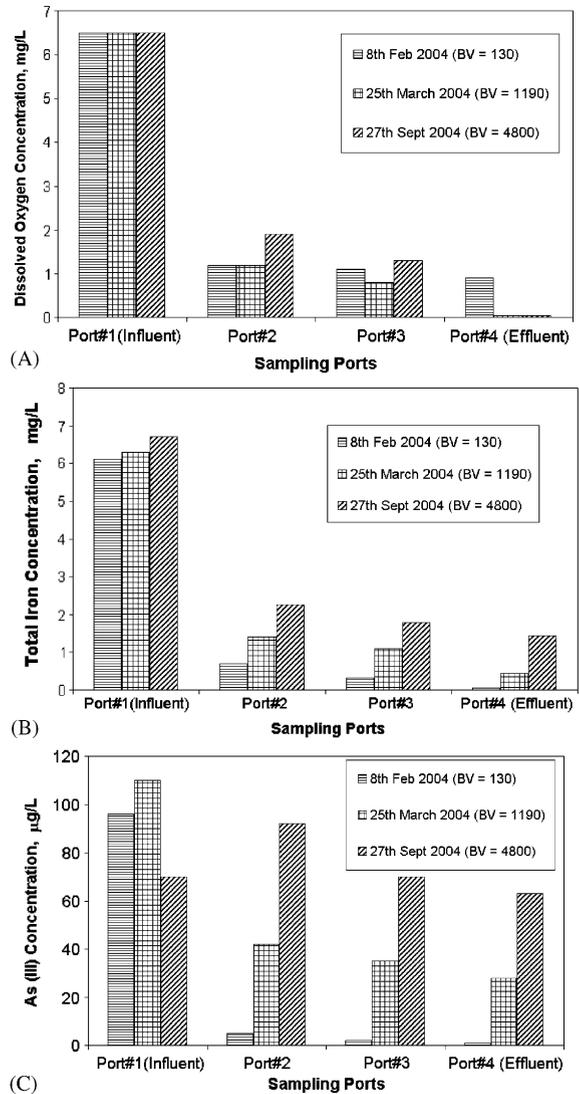


Fig. 6. Concentrations of (A) dissolved oxygen, (B) total iron and (C) As(III) at four different bed depths of the adsorption column at three different times.

4. Discussion

4.1. Arsenic removal: interplay of different variables

Arsenic removal data presented previously clearly suggest that the following three variables greatly influence the overall arsenic removal: first, dissolution of oxygen, second, the presence of iron in well water and its rapid oxidation to HFO and third, the relative percentage of As(III) or arsenite in the groundwater. The design of the top part of the column with stainless-steel splash distributor ensures near-complete oxidation

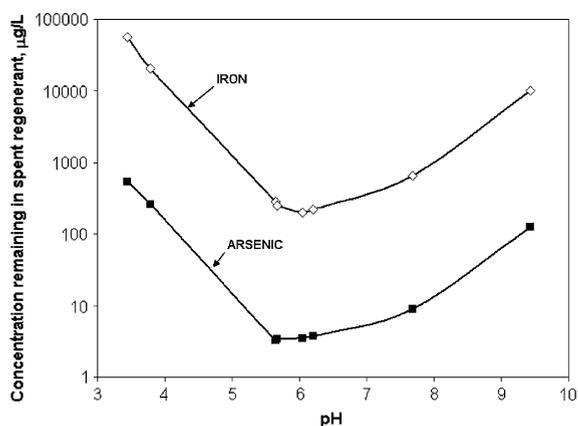


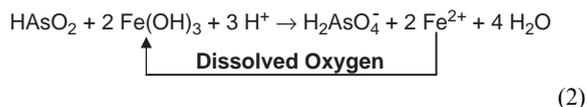
Fig. 7. Concentrations of dissolved arsenic and iron in the leachate from the mixed spent regenerant at different pHs.

of all dissolved iron to hydrated ferric hydroxide or HFO by oxygen as shown below (Morel and Hering, 1993):



Note that the standard state free energy change for the above reaction is a high negative value implying that the forward reaction is very favorable thermodynamically. Hydrogen ions generated by the precipitation reaction are neutralized instantaneously by the alkalinity (HCO_3^-) present in every groundwater. No significant pH change in the water has been observed at any site regardless of its dissolved iron content. The HFO particles are intercepted primarily in the top part of the column. X-ray diffraction (XRD) analysis confirmed that HFO particles are present in the amorphous state and no crystalline iron oxide (e.g., goethite, hematite, magnetite) was formed even after several weeks.

As(III) oxidation to As(V) by DO or precipitated HFO particles is thermodynamically feasible. In regard to the removal of As(III) or arsenites, it may be hypothesized that a portion of precipitated HFO particles first oxidize As(III) to As(V), which is subsequently sorbed strongly onto HFO particles. Dissolved Fe(II), thus formed, is oxidized back to ferric hydroxide by oxygen in accordance with the following scheme:



However, analyses of samples collected at different bed depths from the arsenic removal unit in Potapara village (Fig. 6C) suggested minimal oxidation of As(III)

to As(V). Even after complete exhaustion of the unit, less than 25% of the total arsenic was present as As(V). Several previous studies also confirmed that As(III) oxidation by DO and/or hydrated Fe(III) oxide are kinetically slow and significant only in plug-flow-type fixed beds containing HFO particles (Scott and Morgan, 1995; Greenleaf et al., 2003). Significant As(III) or arsenite removal in every well-head unit is attributed to selective arsenite sorption onto freshly precipitated hydrated Fe(III) oxide or HFO particles (Pierce and Moore, 1982). Previous spectroscopic studies validated arsenite binding onto HFO particles through the formation of inner sphere complexes (Manning and Goldberg, 1997; Manning et al., 1998). In fact at pH around 7.5, As(III) sorption capacity of HFO is significantly greater than that of As(V) (Manning et al., 1998). Activated alumina obviously plays a significant role in removing bulk of the arsenates or arsenic(V), originally present in well water or formed within the column due to partial oxidation of As(III). Laboratory batch studies independently confirmed that dissolved iron alone is not capable of reducing total arsenic to less than 50 µg/L for several contaminated well waters. The observation that arsenic breakthrough gradually increases to 50 µg/L for every well-head unit over a prolonged period of time is a confirmation that activate alumina acts as an arsenic-selective sorbent. Otherwise, the arsenic breakthrough curve would not be gradually increasing in a consistent manner as observed for every well-head unit.

A scientific hypothesis delineating the important steps leading to high arsenic removal may be presented as follows: first, oxygen dissolution and oxidation of Fe(II) to HFO particles take place in the top portion of the column; HFO particles are amorphous and intercepted in the top one-third of the column; arsenites or As(III) species are selectively sorbed onto HFO particles; activated alumina removes mostly arsenates and a minor amount of arsenites. Fig. 8 attempts to illustrate roles of individual process variables for arsenic removal in the well-head units.

4.2. Arsenic containment/disposal

Currently, there are no specific regulations in regard to the disposal of arsenic-laden spent regenerant on the Indian subcontinent. It will also be extremely difficult to enforce any such regulation, if promulgated later, in hundreds of remote villages. Arsenic in the spent regenerant is converted to solid waste along with iron at near-neutral pH and collected at the top of the coarse sand filter located in the same premise. For every regeneration, the mass of sludge (dry basis) added to the top of the coarse filter is less than 400 g and contains mostly ferric and aluminum hydroxide. According to our estimate, every on-site sand filter may collect and

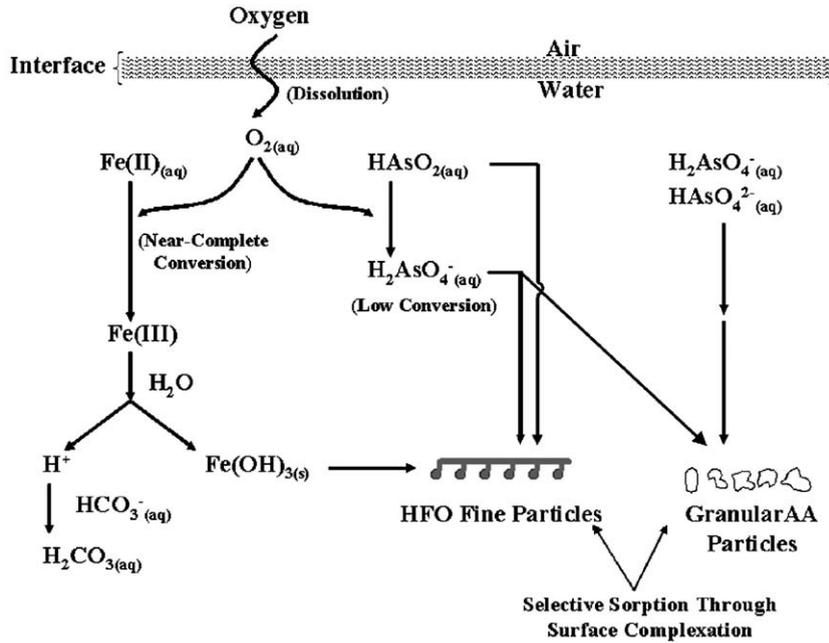
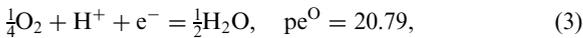


Fig. 8. An illustration depicting interplay of different variables for simultaneous removal of As(III) and As(V) in the adsorption column, AA—activated alumina, HFO—hydrated Fe(III) oxides.

contain the spent sludge for over 30 years. Arsenic in contaminated groundwater contains both As(III) and As(V). However, As(III) or arsenite was absent altogether in the spent alkali regenerant and in the leachates collected from the solids in the coarse sand filters, i.e., the total dissolved arsenic was present solely as As(V) or arsenate.

Fig. 9 shows the predominance or pe–pH diagram for various As(III) and As(V) species developed from thermodynamic data. The small open square near the middle of the diagram represents prevailing redox and pH conditions in groundwaters in the region favoring the presence of As(III) or arsenite along with As(V) or arsenate. During the regeneration procedure, all the solutions are nearly saturated with atmospheric oxygen. The following half reaction and the resulting pe value tend to determine the redox environment for them (Morel and Hering, 1993):



$$pe = pe^0 + \frac{1}{4} \log PO_2 - pH, \quad (4)$$

where PO_2 is the partial pressure of oxygen, = 0.21 atm for atmospheric oxygen, (i) for spent alkali regenerant, at pH = 11.5–12.5 and $PO_2 = 0.21$ atm, $pe = 9.12$ –8.12, (ii) for precipitated sludge, at pH = 6–7, $PO_2 = 0.21$ atm, $pe = 14.62$ –13.62.

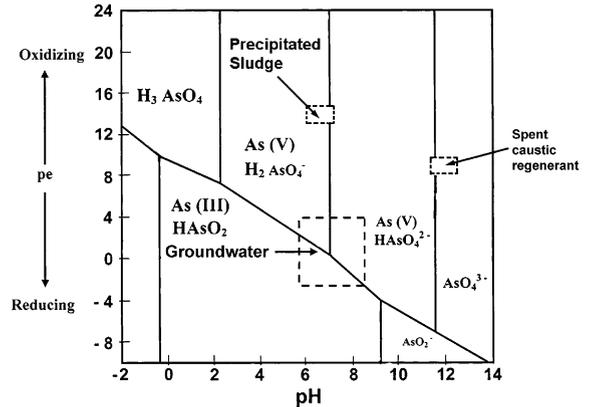


Fig. 9. Predominance or pe–pH diagram showing thermodynamically favorable arsenic species in the spent caustic solution, precipitated sludge and groundwater.

Superimposed on Fig. 9 are marked the estimated pe–pH boundaries of the alkali regenerant and the spent regenerant sludge. Note that As(V) is the predominant species for both and that is consistent with the experimental observations at individual sites. Since any reduction of insoluble HFO particles in the sludge to dissolved Fe(II) may result in an enhanced leaching of arsenic, the coarse sand filter is deliberately kept

accessible to atmospheric oxygen and Fe(III) is thermodynamically stable under this condition.

5. Concluding remarks

Approximately 135 well-head arsenic removal units are currently operating in the Indian state of West Bengal bordering Bangladesh. These units are essentially operated and maintained by the local villagers and the well depths vary from 100 to 200 ft. No pH adjustment of groundwater or chemical addition is needed for day to day operation. These locations were chosen because the villagers around these units are unlikely to have access to arsenic-free drinking water from any other source in the near future. Each unit serves approximately 200 households and the treated water collection rate using hand pump is 8–10 L/min. Significant findings from this long-term study can be summarized as follows:

- Dissolved arsenic is present in groundwater as both arsenite or As(III) and arsenate or As(V). The well-head units remove both of them to attain a total arsenic concentration significantly less than 50 µg/L in the treated water.
- Independent experiments carried out in the field demonstrated that the dissolved iron is oxidized by DO to hydrated Fe oxides or HFO precipitates at the top portion of the column. HFO particles play a significant role in selectively removing arsenites or As(III). However, HFO particles alone in the absence of activated alumina cannot consistently reduce arsenic level to less than 50 µg/L.
- Chemical regeneration for each unit is done every year or so after the total arsenic concentration in the effluent has reached near 50 µg/L. The entire regeneration operation including the treatment of spent regenerant takes nearly 10 h. In the spent regenerant, arsenic is present only as As(V) or arsenates.
- Arsenic present in the spent regenerant is transformed into a low-volume sludge containing primarily ferric and aluminum hydroxide. The sludge is retained on the top of the coarse sand filter located in the same premise and no external disposal of sludge is necessary. Every regeneration produces a sludge weighing less than 400 g on dry basis. Concentration of arsenic in the leachate is consistently below 50 µg/L.

Last but not least, natural arsenic contamination of groundwater has also been reported in other developing countries, namely, Vietnam, Nepal and Mongolia (Christen, 2001; Berg et al., 2001). Similar community-based easy-to-operate treatment units requiring no

electricity and external addition of chemicals may be found viable in these countries.

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