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Mitigating arsenic crisis in the developing world: Role of robust, reusable and selective hybrid anion exchanger (HAIX)

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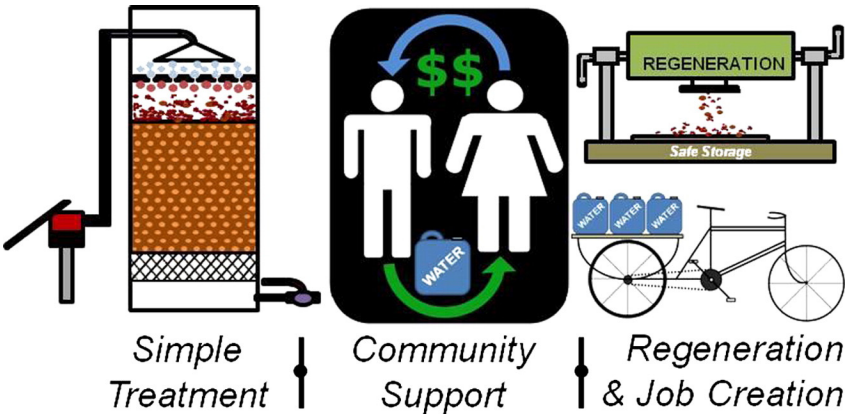
HIGHLIGHTS

- Durable adsorbent-based systems provide arsenic-safe water for many years.
- Nanoparticle infused polymers are durable and selective trace contaminant adsorbents.
- Hybrid anion exchange resins create a synergy between metal oxide nanoparticles and their polymer support.
- Appropriate design and operation can make water systems profitable in remote and rural locations.

GRAPHICAL ABSTRACT



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ABSTRACT

In trying to address the public health crisis from the lack of potable water, millions of tube wells have been installed across the world. From these tube wells, natural groundwater contamination from arsenic regularly puts at risk the health of over 100 million people in South and Southeast Asia. Although there have been many research projects, awards and publications, appropriate treatment technology has not been matched to ground level realities and water solutions have not scaled to reach millions of people. For thousands of people from Nepal to India to Cambodia, hybrid anion exchange (HAIX) resins have provided arsenic-safe water for up to nine years. Synthesis of HAIX resins has been commercialized and they are now available globally. Robust, reusable and arsenic-selective, HAIX has been in operation in rural communities over numerous cycles of exhaustion–regeneration. All necessary testing and system maintenance is organized by community-level water staff. Removed arsenic is safely stored in a scientifically and environmentally appropriate manner to prevent future hazards to animals or people. Recent installations have shown the profitability of HAIX-based arsenic treatment, with capital payback periods of only two years in ideal locations. With an appropriate implementation model, HAIX-based treatment can rapidly scale and provide arsenic-safe water to at-risk populations.
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Abbreviations: AA, Activated alumina; BESU, Bengal Engineering Science University; BVs, Bed volumes; EBCT, Empty bed contact time; GFH, Granular ferric hydroxide; GFO, Granular ferric oxide; HAIX, Hybrid anion exchange; HFOs, Hydrated ferric oxides; HMOs, Hydrated metal oxides; ITC, Institute of Technology in Cambodia; LAB, Lewis acid–base; NF, Nanofiltration; RO, Reverse osmosis; Rs, Indian rupees (or INR); SARSACs, Sustainable arsenic removal systems for affected communities; TCLP, Toxicity characteristic leaching protocol; TDS, Total dissolved solids; THF, Technology with a Human Face; TSF, Tagore-SenGupta Foundation; USEPA, United States Environmental Protection Agency; WHO, World Health Organization.

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1. Introduction

1.1. Problem

When selecting a water source in rural and urban communities of developing countries, groundwater is often favored over surface water. Local availability, biological safety and low capital costs create low barriers to access versus typical surface water schemes. Proper physical and human infrastructure for treatment and distribution of surface water is an ideal long-term development goal for dense urban populations, but it is less appropriate for rural communities with lower population densities and higher per capita operating costs. In the 1970s, large social engineering efforts pushed mass installations of tube wells as "safe" potable water sources, especially for rural communities. In Bangladesh alone, there are over 10 million hand pump wells (Kinniburgh and Smedley, 2001; Cheng et al., 2005). In the short-term, the quality of life improved greatly from the decrease in acute waterborne illnesses. However, not all groundwater is safe. Arsenic groundwater contamination across the Gangetic delta extends over a large area of Bangladesh and India and is one of the worst calamities of the world in recent times. Over 100 million people are at-risk by drinking water well above the WHO recommended limit of 10 ppb As (Chatterjee et al., 1995; Bearak, 1998). The crisis also affects several countries in South and East Asia, including Nepal, Burma, Vietnam, Cambodia, Laos, China, etc (Berg et al., 2007; Stanger et al., 2005; Christen, 2001; Sun, 2004). Arsenic in groundwater has been a focus for public health scientists and engineers over the last twenty years (Bagla and Kaiser, 1996; Ng et al., 2003; Ravenscroft et al., 2005, 2009; Bhattacharya et al., 2007; Bundschuh et al., 2009). But, long-term health studies in Bangladesh during the previous decade, noted significant associations between arsenic exposure through drinking water and mortality rates (Argos et al., 2010).

1.2. Past technologies

It is common for groundwater to be potable and palatable, with only minimal treatment beyond trace contaminant removal. When people are accustomed to groundwater, it is often described as "sweet" versus reverse osmosis (RO)-treated packaged water being "bitter" because of the difference in total dissolved solids (TDS). Furthermore, membrane processes have high energy use, water rejection (up to 60%) and operational costs for trace contaminant removal. Thus, a highly specific adsorbent is more ideal, especially for regions with concerns of water and electricity availability. A significant proportion of arsenic adsorption technologies, if not all, use innocuous hydrated metal oxides (HMOs) with high arsenic affinity, the metals being iron, aluminum, titanium and zirconium (Bang et al., 2005; Driehaus et al., 1998; DeMarco et al., 2003; Dutta et al., 2004; Suzuki et al., 2000). Polymeric ligand exchange has also been demonstrated to be a viable sorbent medium for selective arsenic removal (Ramana and SenGupta, 1992). In developed countries, many different adsorbents based on HMOs have been successfully commercialized for specific uses, including Siemens granular ferric hydroxide (GFH), Severn Trent GFO/AdEdge Sorb33 and DOW Adsorbsia (Westerhoff et al., 2008). But, these materials lack mechanical stability and regenerability, making them inappropriate options for rural communities in developing countries, unless local production facilities were created.

2. Hybrid anion exchanger: Underlying scientific approach

2.1. Adsorption

Aluminum oxides, such as activated alumina or AA (Al_2O_3), have high capacity at certain conditions for arsenate or As(V). However, AA has poor arsenite or As(III) capacity (Cumbal, 2004). Hydrated ferric oxides (HFOs) have high affinity for both As(III) and As(V) because of the functional surface groups, $FeOH_2^+$ and FeOH, at relevant groundwater pH conditions. HFOs form monodentate or bidentate inner-sphere complexes where Fe(III), a transition metal, serves as the electron-pair acceptor or Lewis acid, Fig. S1 (Dzombak and Morel, 1990). The inner-sphere complexes between arsenic and Fe(III) are stable across the necessary operational pH and pE conditions for groundwater arsenic removal. After adsorbent exhaustion, efficient regeneration of HFOs can be achieved by raising the pH, making iron oxide functional groups negatively charged and repelling the arsenic.

2.2. Nanoparticle support and immobilization

Electrocoagulation-based processes generate fresh nanoparticles of HFOs during operation. However, the nanoparticle structure is not maintained after release into the bulk solution, where particles are free to cluster and aggregate. Maintaining the morphology of the desired metal oxide nanoparticles in a continuous process with low head losses, e.g., a packed bed, is optimal for efficient long-term adsorption of trace contaminants. Numerous types of support structures for nanoparticles have been proposed and tested in the past, e.g., alginate, zeolite, activated carbon, chitosan and sulfonic acid-functionalized polymer resin. All of these supports have undesirable intraparticle diffusion of arsenic because of the presence of numerous cation exchange groups and the Donnan co-ion exclusion effect. When metal oxide nanoparticles are impregnated in quaternary ammonium-functionalized polymer beads, the non-diffusible, positive functional groups enhance the local concentration of anions, e.g., As(V), near the hydrated metal oxides. In this hybrid anion exchange (HAIX) resin, there is a synergy of intraparticle diffusion, contaminant selectivity and mechanical strength, Fig. 1 (Donnan, 1911, 1995; Cumbal and SenGupta, 2005). As(V) and As(III) capacity comparisons of HAIX and AA are in Figs. S2A–B, respectively. The synergy in HAIX allows for highly scalable system designs similar to any other ion exchange process with the major design parameter being a minimum empty bed contact time (EBCT) of 2 min.

The specific objective of this paper is to provide evidence of community-scale HAIX installations as effective long-term solutions for arsenic contaminated groundwater with the potential to operate as a local for-profit business.

3. Materials and methods

3.1. HAIX synthesis

Impregnation or dispersion of hydrated Fe(III) oxide or HFO nanoparticles within an anion exchanger posed major challenges because

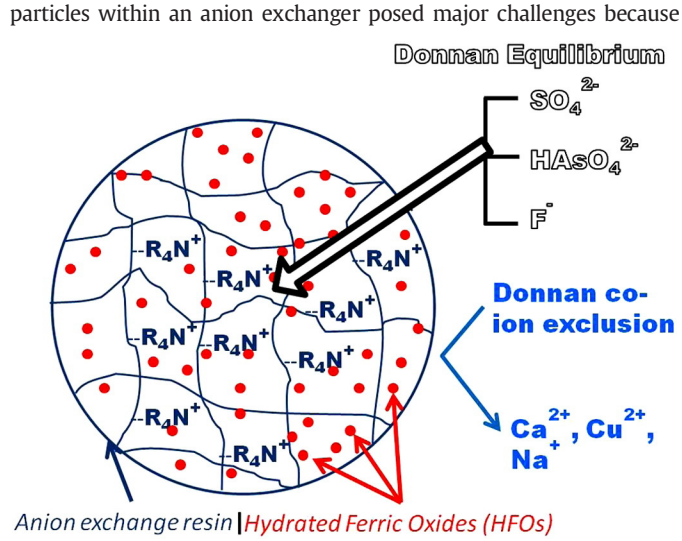

Fig. 1. Diagram of an HAIX resin bead with the anion exchange resin support impregnated with nanoparticles of HFOs. Note the transport of counterions (i.e., anions) through the resin per the Donnan membrane equilibrium and rejection of co-ions (i.e., cations) per Donnan co-ion exclusion.

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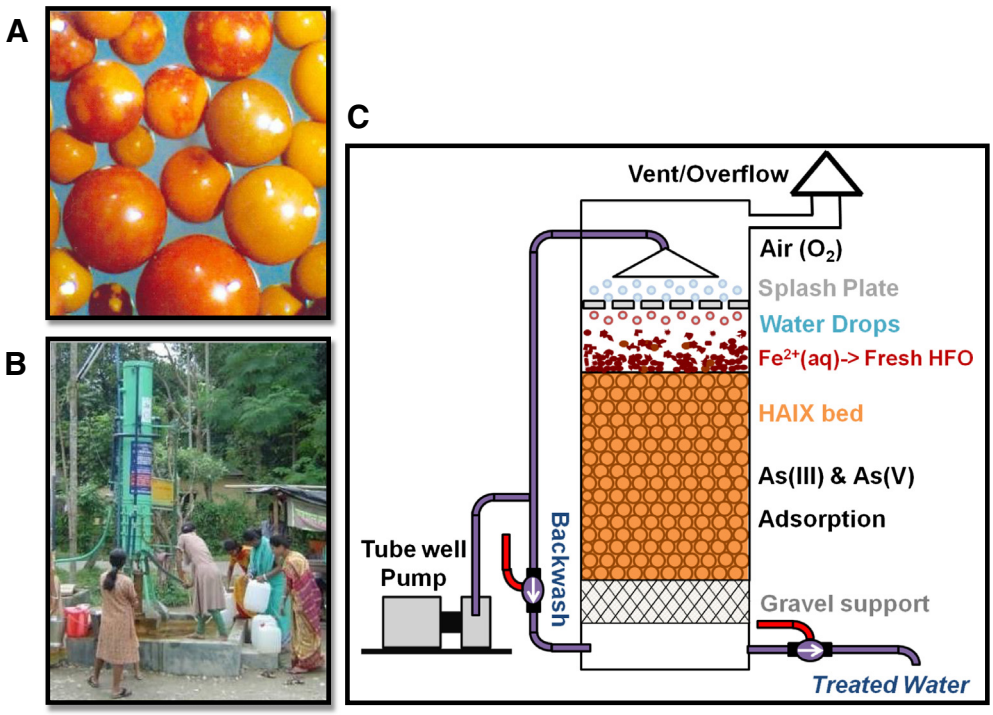

Fig. 2. A) HAIX resin bead impregnated with HFO nanoparticles, B) women using a hand pump-attached arsenic treatment column at the first HAIX installation, and C) diagram of arsenic filtration column mechanics.

of the associated difficulty in introducing positively charged ferric ions (Fe^{3+}) within an anion exchange resin with positively charged quaternary ammonium functional groups. Through several iterations, a reproducible, operationally simple protocol was developed in 2004 and described in detail in United States Patent US 7291578 (SenGupta and Cumbal, 2007), a simple overview of synthesis is available in literature (Sarkar et al., 2007). Under this patent, HAIX is now commercially available globally as LayneRT™ from Layne Christensen Co. (LayneRT).

3.2. Field work

Over the past 15 years, 200 sustainable arsenic removal systems for affected communities (SARSACs) have been successfully installed and operated in rural communities of West Bengal and Bihar (India), Cambodia, Laos and Nepal. Today over 200,000 people have benefited from the past arsenic treatment systems in South and Southeast Asia. The progression in system design has been detailed in literature (Sarkar et al., 2005, 2007, 2010, 2012). Note that the national standard for arsenic in drinking water was 50 ppb in India until 2012. Over time, these efforts have been assisted and coordinated by several organizations, including Bengal Engineering Science University (BESU), Institute of Technology in Cambodia (ITC), Tagore-SenGupta Foundation (TSF), Technology with a Human Face (THF), and Shibpur Association for Technological Humanitarian and Environmental Endeavours (SATHEE). At this time, communities are fully responsible for operation, testing and maintenance of SARSACs. Local testing labs are chosen by communities for arsenic analysis to be confident their water is safe, conform to NGO operating rules and identify when regeneration is

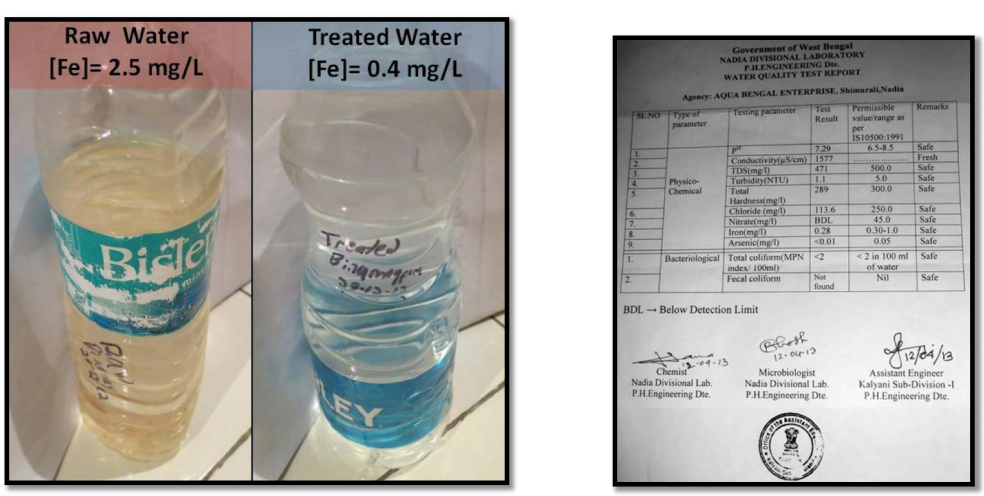

Fig. 3. A) Iron removal during HAIX arsenic treatment at Binimaypara, N. 24 Parganas, West Bengal, India (left: raw water at 2.5 mg/L, right: treated water at 0.4 mg/L). B) Treated water analysis at a recent SARSAC installation; water analysis completed by West Bengal Public Health Engineering Department.

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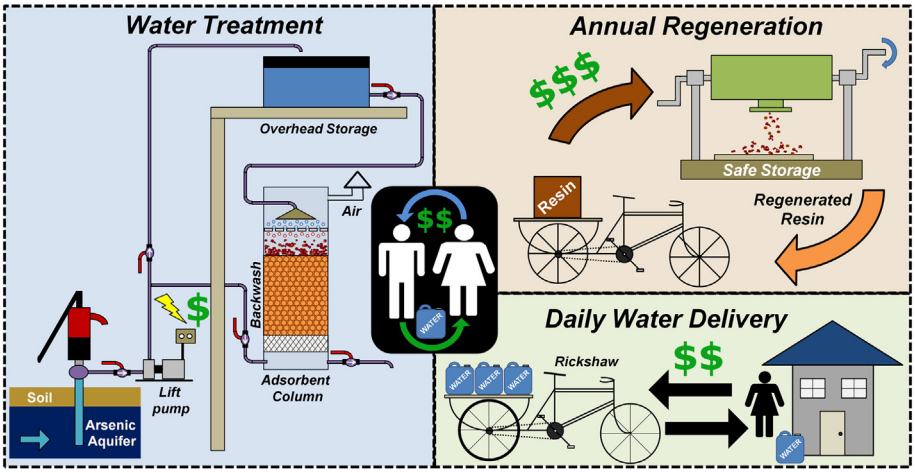

Fig. 4. Overview of holistic arsenic treatment in operation: water treatment, daily water delivery, and annual resin regeneration.

necessary. Local regeneration of materials is ordered by the managing community club and performed by trained local organizations when effluent arsenic is above the local arsenic standard; a gradual breakthrough curve prevents unsafe water reaching consumers before regeneration.

4. Results and discussion

4.1. Long-term performance in West Bengal, India

Amorphous HFOs of sizes 20–100 nm were precipitated on the gel-phase of macroporous anion exchange resins (DeMarco et al., 2003; Cumbal et al., 2003), Fig. 2A. HAIX loaded with high surface area HFOs has similar capacity to other synthetic iron hydroxide adsorbents (e.g., Bayoxide E33) for both As(III) and As(V), even though the ferric oxide content of HAIX is much less on a mass basis: 40% vs. 70%, respectively (Sarkar et al., 2007; Lanxess Bayoxide E33®, 2008). The mechanical integrity of polymer resins allow for several cycles of use and regeneration with minimal changes in metal oxide morphology and content; Bayoxide E33 is a single-use adsorbent (Sarkar et al., 2008).

In early 2004 the first public use of HAIX was in a community-scale installation for a rural community in Ashok Nagar, West Bengal, India, Fig. 2B. Since 2004, the fixed-bed adsorption process using HAIX has been scaled for personal use in the USA from household-scale to municipality-scale (>20,000 households), based on a minimum empty bed contact time of 2 min. In the Ashok Nagar design, groundwater is first pumped by hand to the top of the column. Upon entering the column, water is aerated, Fe(II) is oxidized and Fe(III) solids are filtered by the adsorbent bed, Fig. 2C. Iron removal, caused primarily by oxidation of Fe(II) into Fe(III) by oxygen, is a simple, but a key step for aesthetic appeal and consumer confidence in water purity. Incipient precipitation in the top portion of the column leading to the formation of HFO nanoparticles also renders removal of both dissolved As(III) and As(V) (Sarkar et al., 2007). Remaining arsenic is then removed by HAIX. Fig. 3A shows clear evidence of iron removal and the transparency of water is greatly improved following iron removal. Fig. 3B shows the scanned copy of treated water analysis from a recently installed arsenic removal system, note the low iron and arsenic concentrations; treated water arsenic concentration is made available to consumers.

Over the past 10 years, the arsenic treatment process has changed to maximize convenience for end-users, Fig. 4. Many rural communities in West Bengal are electrified with limited "load shedding" or power cuts. While electricity is available, arsenic contaminated groundwater can be pumped to and aerated within an elevated water storage tank. At low elevation head (5 m), water can be fed by gravity through aeration and adsorption treatment. Treated water is available to people for on-site pick-up or rickshaw delivery. All labors in the process are appropriately compensated: construction, operation, maintenance and delivery. Adequate economic incentives are necessary for long-term sustainability.

Today, communities in Ashok Nagar continue to purchase arsenic-safe water from HAIX treatment systems up to nine years after installation. In Nabarun Sangha the most recent water tests indicate both columns are producing water with arsenic <50 ppb. Eventually, the annual regeneration requirement will be necessary and performed by a local company of trained staff. Column No. 1 has produced 6.5 million liters of safe water (2004–2013) and Column No. 2 has produced 5.5 million liters of safe water (2010–2013); high water demand on Column No. 1 necessitated a parallel column to boost output. Consistent operation in Ashok Nagar shows the long-term durability and high arsenic capacity of HAIX treatment has been proven in both lab-scale (Sarkar et al., 2007) and community-scale systems, Fig. 5.

4.2. On-going efforts in Cambodia

Cambodia has very high levels of arsenic (up to 1600 ppb) with the majority of impacted people near the Phnom Penh province (Buschmann et al., 2007; Berg et al., 2007). Currently, there are tens of HAIX arsenic treatment systems in operation from Nepal to Cambodia that produce safe water. In the Sambour District of Kratie Province,

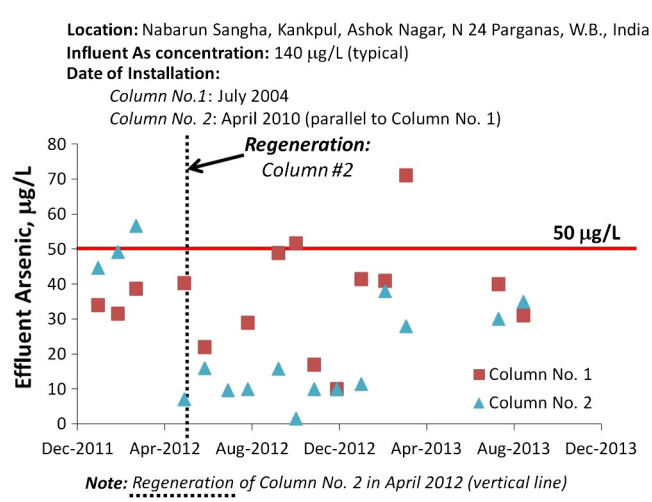

Fig. 5. Recent performance of two HAIX columns at Nabarun Sangha, Ashok Nagar, West Bengal India.

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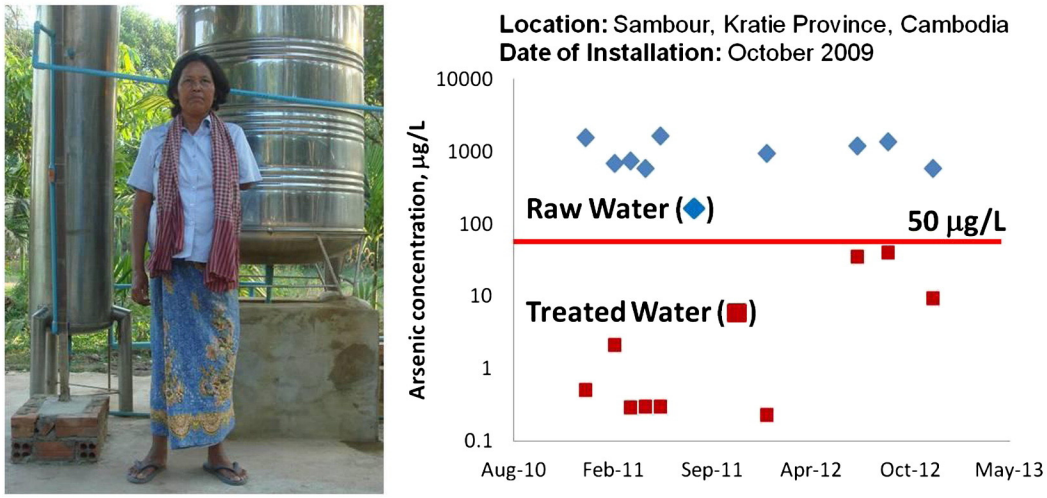

Fig. 6. Arsenic-safe water was produced in Sambour District, Kratie Province, Cambodia from 2010–2013 by a SARSAC using HAIX; the system is operated by a woman affected by arsenic (left).

Cambodia, local women have operated SARSACs using HAIX since October 2009. Over the years, arsenic-safe water has been provided throughout the community at an affordable rate of 500 riel ($0.12) per 20 L or at 1000 riel ($0.25) per 20 L for door-to-door delivery of water, Fig. 6. In Cambodia, rain water is preferred when available during the wet season and a SARSAC may be underutilized for several months, but it still operates as designed upon restart; note the consistent performance over several years. Such reliability is an important, non-quantitative operational consideration when systems are remote; technical expertise is distant and maintenance can be costly, e.g., adsorbent or RO membrane replacement.

4.3. Economic sustainability: From water crisis to water business

A business opportunity exists when no other reliable and desirable water supply has been made available to a population: typical packaged water containers are too expensive, piped water is nonexistent and groundwater quality is undesirable. When there is a high density of customers willing to pay for safe and convenient water, a centralized system is more affordable and better able to be monitored than domestic units. Regular water quality testing and monitoring of waste disposal at one location protect the customer from unsafe water or improper waste handling that could happen at 500 households. To ensure safety for the community, local employees are paid a fair wage to take care of the necessary maintenance, upkeep and management.

In Ashok Nagar, India, there are over 1300 families who purchase safe water from three HAIX based arsenic treatment systems. The families pay Rs. 20–30 ($0.40–$0.60) per month for daily pick-up of 20 L potable water; people pay extra for cycle rickshaw delivery. Both communities in Nabarun Sangha and Binimaypara have had HAIX installations selling safe water since 2005, Fig. 7. In 2010, a new treatment column was installed in Nabarun Sangha to boost production and meet user's demand.

At the new Sakthi Sadhana Community Club installation in Ashok Nagar (2010–present) there are over 600 families currently purchasing water at Rs. 30 per month, Table 1. Revenue was great enough for the club to use a small diesel truck for the delivery of water. Now if a family desires daily water delivery they pay the local truck drivers an additional Rs. 60–150 per month, depending on the distance.

The most recent installation, as of the writing of this manuscript, was commissioned on April 15, 2013; the capital budget is in Table 2. Note the cost of resin is only 15% of the overall system costs, a minor portion of the overall expense. Many past systems have used AA instead of HAIX because of the lower initial costs. Using AA would have saved $450 in initial costs, or 10%, but this cost is insignificant when looking at the overall revenue during operation and the improvement in treatment performance (Sarkar et al., 2010).

Franchising decentralized water treatment plants is seen as one option for rapidly scaling access to safe water (Mukherjee, 2010). A recent installation in coordination with a private, female entrepreneur was the first privately-owned HAIX system in a rural community. Her aim is to

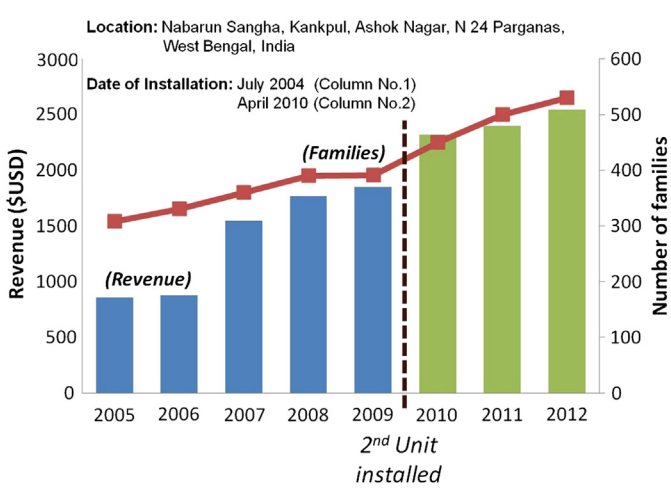

Fig. 7. Annual revenue generated at Nabarun Sangha from sales of HAIX-treated water (2005–2012). Note: total annual revenue is denoted by the bars and number of families is denoted by the line plot.

Table 1
The financials of annual water operation and maintenance using HAIX at Sakthi Sadhana Community Club in Ashok Nagar, West Bengal.

	Unit amount (Rs.)	Annual frequency	Total (Rs.)	($US)
Revenue				
Water purchases	30	7200	216,000	4000
Costs				
Caretaker	−3000	12	−36,000	−667
Electricity	−5000	12	−60,000	−1111
Water testing	−200	12	−2400	−44
HAIX regeneration	−8000	1	−8000	−148
Total			109,600	2030

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Table 2
Capital expenses for a recent installation.

Item	Number	Total (Rs.)	($US)
Materials			
Stainless steel HAIX columns	2	56,000	1037
Plumbing and accessories	1	13,800	256
Water tanks	3	38,250	708
Well pump	1	6000	111
Air compressor	1	1500	28
HAIX resin	100 L	36,160	670
Secure building	1	60,000	1111
Subtotal + VAT tax (4%)		220,200	4100
Labor and services			
Transportation		3000	58
Installation and commissioning		3500	67
Plumbing		4500	85
Civil work		7500	140
Subtotal		18,500	350
Total		238,700	4450

operate as an entrepreneur selling packaged water in an area at-risk of arsenic consumption.

5. Closing remarks

The objective of this paper was to present data indicating the long-term success of HAIX for treating arsenic contaminated water and generating revenue in rural communities of South and Southeast Asia. Community-scale arsenic removal with HAIX, referred to as SARSAC, is a robust process that has been effectively used for several years in rural communities of India and Cambodia. Regular operation, maintenance and testing of the system are managed locally by representative villagers' committee. The arsenic data points presented were tested by laboratories paid with funds collected from the local consumers. Upon exhaustion, HAIX is able to be repeatedly regenerated by removing the adsorbed arsenic because HFOs are chemically resilient and the parent anion exchange resin is physically robust (Cumbal, 2004; Sarkar et al., 2010).

During regeneration, arsenic is removed from the adsorbent by creating Donnan co-ion exclusion of arsenic by the ferric nanoparticles with a high pH solution. Afterwards, the pH is readjusted to neutral and the adsorbent is reactivated (Sarkar et al., 2007). The removed arsenic is concentrated in the waste regenerant. Preventing concentrated arsenic from re-entering the ecosystem is important for the well-being of all life in the community; improper disposal could lead to wildlife consumption, acute poisoning and death. Landfill disposal of exhausted iron-based arsenic adsorbents is inadvisable because in a landfill Fe(III) can reduce to Fe(II), become aqueous and leach arsenic, although the EPA TCLP indicates that landfill disposal is safe (Ghosh et al., 2004). Concentrated arsenic is safely stored at the community-level in a scientifically sound manner that is environmentally benign: 1) excess iron is added to arsenic-rich regenerant, 2) fresh ferric oxide is formed and arsenic is adsorbed, 3) ferric oxide is precipitated and arsenic-safe water is passed through a soak pit filter, Figs. S3, 4) iron–arsenic complexes are retained on top of the soak pit in an oxidizing environment, and 5) iron remains oxidized as Fe(III) and arsenic is safely retained. Central regeneration facilities in rural West Bengal have safely stored the arsenic waste from over one hundred regeneration cycles; a similar central regeneration facility has been constructed in Cambodia under the auspices of ITC. The underlying chemistry has been presented and substantiated through field data in the literature (Blaney and SenGupta, 2006; Delemos et al., 2006; Ghosh et al., 2004; Sarkar et al., 2005, 2007, 2010).

Past rural efforts pushed by governmental and non-governmental organizations in South Asia have not had appropriate feedback cycles for ensuring long-term use. Countless arsenic systems have had technical or social failures within one year of installation with no consequences for the responsible party (Hossain et al., 2005). Local, privately-owned water businesses are more sustainable than contracted installations from non-local organizations because there is a large incentive for high quality product delivery to generate revenue and maintain local employment.

Robustness, reusability and excellent arsenic removal capacity of HAIX have catalyzed community participation in tens of rural communities in arsenic affected countries in South and Southeast Asia. Not only is arsenic-safe water provision possible in remote communities, it can also be profitable, as demonstrated in many communities. Simple system operation, low operational costs and high water quality have been possible with HAIX.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2013.10.092.

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