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Locally-affordable arsenic remediation for rural South Asia using electrocoagulation

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Bangladesh and neighbouring areas face health threats from drinking arsenic-contaminated groundwater. The challenge is to develop arsenic remediation that is (1) affordable to most of the local population, (2) robust and easy to maintain long term, (3) technically effective for removing arsenic down to 10 µg/L in the presence of other competing ions in the water, and (4) does not require hazardous chemicals or produce excess levels of arsenic-laden waste. Electrochemical Arsenic Removal (ECAR) uses a small DC current and ordinary steel electrodes to produce iron rust in the arsenic-contaminated groundwater that binds arsenic and can be removed by filtration. We describe performance results using synthetic and real groundwater and describe the design of a 100L reactor. We demonstrate low production of waste sludge that is non-hazardous according to US EPA standards, and show preliminary results of successful sludge stabilization in concrete. Finally we estimate the operating costs.

Introduction

Naturally occurring arsenic contamination in groundwater has been discovered in Argentina, Chile, Mexico, China, Hungary, Vietnam, Cambodia, United States, West Bengal (India), and Bangladesh. In Bangladesh alone, 57 million people are exposed to arsenic levels well in excess of the maximum contaminant level (MCL) recommended by the World Health Organization of 10 µg/L (WHO 1993; Kinniburgh and Smedley 2001). Exposure to arsenic causes dermatologic, neurologic, vascular, and fatal carcinogenic effects (Chowdhury et al. 2000). A recent decade-long cohort study published in *The Lancet* found that 1 in 5 (21.3%) of all deaths in Bangladesh are now attributable to arsenic in drinking water (Argos et al. 2010). Exposed children also suffer from a demonstrable decrease in intellectual function indicated by IQ (Wasserman et al. 2004; Wang et al. 2007). Increased health care and loss of income cost the average household in rural West Bengal as much as $84 per year (Roy 2008) - a crippling burden on yearly income for those making US $1-2 per day.

Over twenty years has passed since arsenic was first discovered in groundwater, and yet no effective and affordable arsenic treatment technology has been implemented on a large scale (Hossain et al. 2005; Ahmed et al. 2006; Kabir and Howard 2007). Arsenic treatment units (ATUs) have failed to reach even 1% of the population at risk in Bangladesh (Ahmed et al. 2006). The same is true for many safe water alternative methods such as pond sand filters, rainwater harvesting, shrouded shallow dug wells, and piped water systems (Ahmed et al. 2006). While surface water treatment and alternatives to groundwater should be explored, locally affordable arsenic removal technologies are essential to account for areas with no alternative drinking water source and to prepare for the expected increase in such areas due to global climate change.

Evaluations of ATU failure frequently cite the lack of proper maintenance as a major factor (Ahmad et al. 2003; Hoque et al. 2004; Hossain et al. 2005; Ahmed et al. 2006; Kabir and Howard 2007). When ATUs are successfully maintained, high upfront or ongoing costs prevent poor users from gaining access. The challenge is to develop a method for arsenic remediation that is (1) affordable to most of the local population, (2) robust

and easy to operate and maintain long term, (3) technically effective for removing arsenic down to 10 μg/L in the presence of other competing ions, and (4) does not require use of hazardous chemicals or produce excess levels of arsenic-laden waste at any point in the supply chain.

ElectroChemical Arsenic Remediation (ECAR) is a form of electrocoagulation (EC) that has been developed to meet the needs of an appropriate community scale implementation scheme that is financially viable, locally affordable, and offers long-term sustainable safe water access in rural areas (Addy 2008; Addy et al. 2008). In ECAR, electrolytic oxidation of a sacrificial iron anode produces Hydrous Ferric Oxide (HFO; also called Fe(III) precipitates) in arsenic-contaminated water. Arsenic forms complexes with HFO, which then aggregate to form a floc that can be separated from water. As(III) oxidation to As(V) occurs during the ECAR process either through electrolytic action at the electrode or via highly reactive radical species produced by the oxidation of Fe(II) by dissolved oxygen (Hug and Leupin 2003; Roberts et al. 2004). ECAR is promising due to many advantages over chemical coagulation - including pH buffering ability, ease of operation, amenability to automation, low maintenance, low sludge production, small system size, and the benefit of side reactions like electro-oxidation and electro-flotation (Kumar et al. 2004; Mollah et al. 2004; Addy 2008). In ECAR, adsorbent media with a high capacity is generated during treatment, with no need for media regeneration or hazardous chemicals.

In this paper, we describe ECAR treatment results using arsenic-contaminated synthetic groundwater prepared in the laboratory, and also preliminary results from real groundwater in Bangladesh and Cambodia. We describe the design of a small ECAR reactor to treat 100 L of water at time, for a planned technical trial in West Bengal (India). We demonstrate the low production of waste sludge and characterize it as non-hazardous according to US EPA regulations. We show preliminary results of successful sludge stabilization in concrete, potentially offering an alternative to landfill disposal. Finally we estimate the operating costs and benefits of ECAR for potential use in community scale system capable of providing clean water at a locally affordable price.

ECAR Performance

Performance in synthetic groundwater

Arsenic removal is known to be highly sensitive to groundwater composition, specifically the presence of phosphate, silicate, natural iron and, to a lesser extent, carbonate, and calcium (Roberts et al. 2004; Hug et al. 2008; Guan et al. 2009). Each of these ions is present in contaminated Bangladesh groundwater (Kinniburgh and Smedley 2001). Co-occurring ions affect arsenic removal by competing with arsenic for sorption sites on HFO (Meng et al. 2002; Roberts et al. 2004), or by affecting the structure of HFO formed from Fe(II) oxidation (Ahmad et al. 2006; Voegelin et al. 2010). To understand ECAR arsenic removal performance in a relevant groundwater composition, batch tests were performed in Synthetic Bangladesh Groundwater (SBGW; Table 1) designed to mimic tubewell-water parameters in Bangladesh measured by BGS (Kinniburgh and Smedley 2001).

Table 1. Composition of Synthetic Bangladesh Groundwater (SBGW) for a representative initial total arsenic concentration of 600 μg/L.

Ion	Na^+ (mM)	Ca^{2+} (mM)	Mg^{2+} (mM)	Cl^- (mM)	HCO_3^- (mM)	SiO_3^{2-} (mM)	SO_4^{2-} (μM)	PO_4^{3-} (μM)	AsIII (μM)	AsV (μM)	AsTOT (μM)	pH
SBGW	6.0	1.5	0.33	3.5	4.5	0.70	84	42	4.0	4.0	8.0	7.0

Batch experiments were performed in an electrochemical cell using an iron anode and copper electrode in arsenic-spiked SBGW, containing both As(III) and As(V). Contaminated water was treated in 3-liter batches using a galvanostatic current of 110 mA (current density of 1.1 mA/cm^2). Treated solution was stirred for an additional 60 minutes with no electrodes. All tests were duplicated and the results averaged.
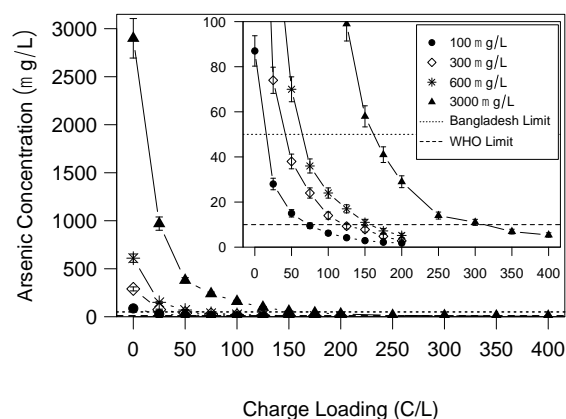
**Figure 1. Aqueous arsenic concentration as a function of charge loading
in Synthetic Bangladesh Groundwater (SBGW) for initial arsenic concentrations
of 100 – 3000 μg/L (1:1 As(V):As(III))**

Dotted and dashed lines indicate the legal Bangladesh limit (50 μg/L)
and the WHO MCL (10 μg/L) respectively
Detail near the WHO MCL is shown in the inset

Aqueous arsenic concentrations as a function of charge loading (i.e. the total charged passed through the electrochemical cell during electrolysis, measured in coulombs per liter, C/L) for initial arsenic concentrations of 100 – 3000 μg/L are shown in Figure 1. In every case, ECAR reduces total arsenic in SBGW to well below the WHO MCL of 10 μg/L. The effect of initial concentration is to increase the charge loading required to reach the WHO MCL (see inset, Figure 1). Charge loading is directly related to the concentration of dissolved iron in solution by Faraday's law (Gu et al. 2009; Lakshmanan et al. 2009), and thus can be thought of as a proxy for the HFO adsorbent dosage. An increase in the initial concentration is expected to require an increase in HFO adsorbent dosage to reach the WHO MCL.

It should be noted that numerous complicated processes are occurring simultaneously during ECAR, including the electrochemical dissolution of the electrode, Fe(II) oxidation and HFO hydrolysis, surface adsorption, and coagulation (Mollah et al. 2004; Holt 2005). ECAR operating parameters, such as the current density (current per active electrode area), charge loading, operating current, and post-electrolysis mixing time all have the potential to affect arsenic removal in subtle and sometimes complicated ways. It is beyond the scope of this paper to discuss or test the effect of each, though many have been explored to some extent in our lab (Addy 2008).

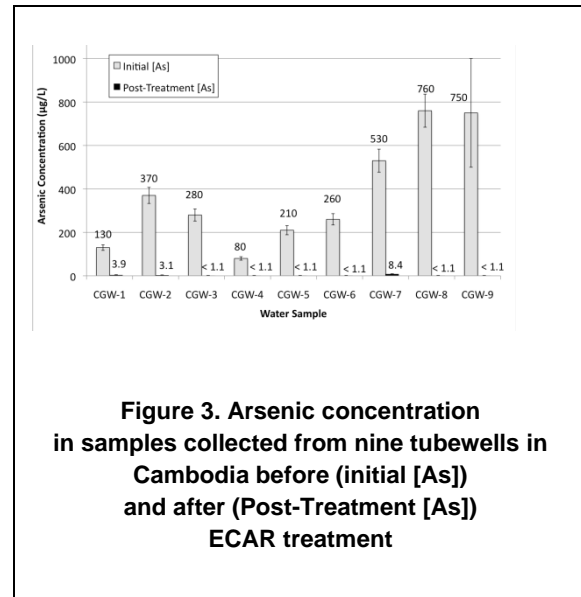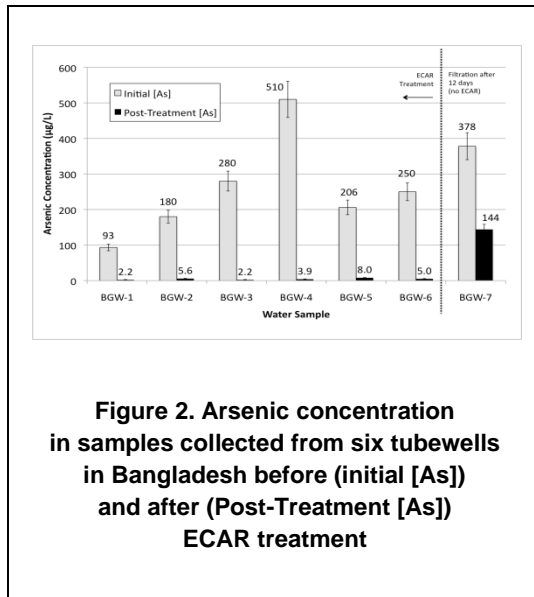## Performance in real groundwater
Bangladesh
Six groundwater samples were obtained from arsenic contaminated tubewells in rural villages of Jhikargachha, Abhaynagar, and Sonargaon Upazilas in Bangladesh. One liter of water from each well was collected after approximately 5 minutes of pumping (to avoid bacterial contamination and oxygenated water in the well-head) and stored in tightly capped polyethylene bottles filled to the brim (for full sample collection and treatment procedures, see Addy (2008)).

Figure 2 shows the initial (immediately before treatment) and final (post-treatment) aqueous arsenic concentrations for Bangladesh groundwater samples treated with ECAR. In every case, ECAR is able to reduce initial arsenic concentrations of up to 510 μg/L to less than the WHO MCL of 10 μg/L in real groundwater matrices. Samples BGS-1 to -4 were treated in the beaker scale setup described above (current density 1.1 mA/cm^2). Samples BGW-5 and BGW-6, were treated in a bench-scale continuous flow prototype

(described in Addy (2008)). One sample (BGW-7) was stored for 12 days and then filtered (no ECAR treatment) to determine the removal effect of naturally occurring iron precipitates in the tubewell water. Consistent with other passive sedimentation tests (WaterAid 2001; Roberts et al. 2004), only a fraction of the arsenic was removed, and the final arsenic concentration (144 μg/L) was well above both the WHO MCL and the legal Bangladesh limit (50 μg/L).
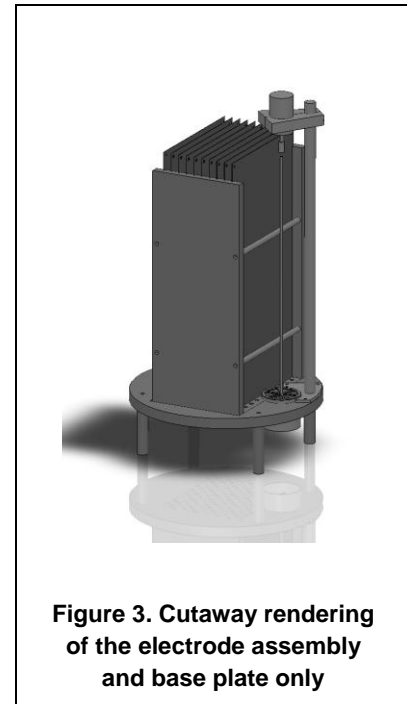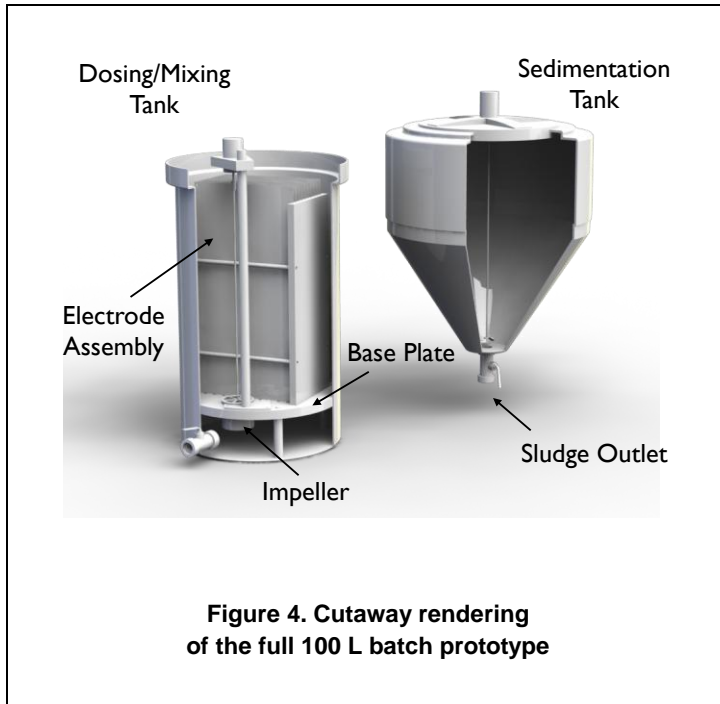
Cambodia

Up to 1 million are at risk due to arsenic in drinking water in the Mekong Delta region of Cambodia and South Vietnam (Berg et al. 2007), primarily from aquifers with a different overall water composition than Bangladesh (Hug et al. 2008). Nine arsenic-contaminated tube wells were chosen from the three communes Preak Russei, Dei Edth, and Preak Aeng in the Mekong Delta region of Kandal Province, Cambodia (initial arsenic concentrations 80 – 750 μg/L). Arsenic concentrations before and after treatment with ECAR (using the bench-scale continuous flow prototype) are shown in Figure 3. In every case, ECAR-treated water contains less than 10 μg/L arsenic. In 6 of the 9 cases, final arsenic concentrations were below the detection limit of ICP-MS (< 1.1 μg/L).

Figure 2. Arsenic concentration in samples collected from six tubewells in Bangladesh before (initial [As]) and after (Post-Treatment [As]) ECAR treatment

Figure 3. Arsenic concentration in samples collected from nine tubewells in Cambodia before (initial [As]) and after (Post-Treatment [As]) ECAR treatment

Performance and development of 100L batch prototype

A small 100L batch scale ECAR prototype has been designed, built, and tested in the lab for removing arsenic from SBGW. The prototype (Figure 4 - 5) comprises a cylindrical tank for dosing and mixing connected to a sedimentation tank for coagulant addition and solid/solution separation. The electrode assembly (Figure 5) consists of 10 parallel mild steel plates (5 anode and 5 cathode) with alternate plates connected in series. The configuration allows for easy reversal of current, allowing each plate to alternate between anode and cathode to minimize extensive rust build up and passivation. A DC motor attached to a small impeller pushes water under the base plate and up between the electrode plates, allowing for efficient and uniform mixing between the plates. The voltage across the electrodes was < 3V.

The prototype has successfully and repeatedly remediated SBGW with up to 3000 μg/L initial arsenic (with equal parts of As(III) and As(V)) to less than the WHO MCL in the lab, showing equal or better performance (in terms of required charge loading) than the bench-scale electrochemical cell. Electrolysis time (not optimized) was around 2 hours. With the addition of a small amount of alum (5 - 25 mg/L as Al), settling occurs within 2 - 4 hours, leaving a clear supernatant solution with < 10 μg/L arsenic. Sludge is easily accessed and collected through a valve at the cusp of the sedimentation tank. The prototype will be field tested in the highly arsenic-contaminated Murshidabad district of West Bengal, India, beginning in November of 2010.

**Figure 4. Cutaway rendering
of the full 100 L batch prototype**

**Figure 3. Cutaway rendering
of the electrode assembly
and base plate only**

Arsenic-laden waste

All arsenic removal technologies produce arsenic-laden sludge or waste that must be disposed of. The amount of sludge collected per 100 L batch (including alum) is 10 – 20g for initial arsenic concentrations of 600 – 3000 µg/L. Leachates from waste sludge generated during batch reactor tests were prepared via a Toxicity Characteristic Leaching Procedure (TCLP) using US EPA Method 1311. The resulting leachate was analyzed for Ag, As, Ba, Cd, Cr, Hg, Pb, and Se metals and all were found to be well below regulatory limits (most were not detected). Arsenic in the leachate was 160 µg/L, well below the regulatory limits of 5000 µg/L. Thus ECAR waste sludge is not considered hazardous waste in the U.S.

Previous studies suggest that leaching of arsenic may be enhanced in a landfill or hazardous waste site (Delemos et al, 2006; Ghosh et al 2004). Therefore alternative methods of waste stabilization should be explored. Banerjee and Chakraborty (2005) have demonstrated that preparation of briquettes, cement sand-mortar, and concrete with a similar arsenic-laden sludge mixed in at up to 10, 18, and 40% of volume respectively produced a TCLP leachate below the Government of India inland water discharge standards. Concrete used to stabilize arsenic-laden sludge could potentially be packed into roadways with minimal risk of arsenic leaching. To test this option, arsenic-laden sludge was generated using ECAR in SBGW (estimated arsenic loading was 1.5% of sludge weight), and subsequently mixed into the aggregate of a Portland-cement concrete mixture, replacing 40% concrete by volume and 6% by weight. Two resulting 100g concrete cylinders were cast, cured for 7 days and then crushed to particles of < 9.5 mm diameter. A TCLP leachate preparation of the crushed concrete showed no detectable arsenic via ICP-MS (detection limit 1 µg/L). Other tested metals (Ag, Ba, Cd, Cr, Hg, Pb, and Se) were either not detected or well below US EPA regulatory limits. This suggests that concrete stabilization may be a viable alternative to landfill disposal for ECAR.

Cost estimate

Consumable costs for ECAR include the iron consumed in the sacrificial anode, electricity required by the electrodes and required to mix the water during electrolysis, and a coagulant to aid settling. The exact electricity consumption of ECAR will depend on the final device design, electrode plate area, resistivity of the source water, and operating current among other factors. However, we can estimate this cost using batch prototype tests (not optimized for energy consumption) operating with parameters found to reduce up to 600 µg/L arsenic to less than 10 µg/L in SBGW.

The input cost for electricity is assumed to be \$0.50/kWh. This is purposely higher than average grid electricity to account for the possibility of using a small solar cell, including amortized capital over several years. Some wells may have higher initial arsenic values, or harsher water compositions than the ones tested – therefore we have increased the measured dose required to reduce 600 µg/L to less than 10 µg/L arsenic. For consumable iron dissolved from the anode, we assume \$0.71/tonne, the March 2010 global average transaction cost for hot rolled plate carbon steel (compiled by Worldsteelprices.com, http://www.worldsteelprices.com/). For a coagulant, we assume alum (which has successfully been used with the 100L prototype) at a cost of \$0.16/kg (following Jiang et al. (2005)), plus the electricity cost of flash mixing for 5 minutes followed by slow mixing for 25 minutes using a small impeller. The measured power required for mixing during electrolysis in the 100L batch prototype is $4 \cdot 10^{-6}$ kWh/L/min (not optimized).

The resulting estimated consumable cost for ECAR treatment is \$0.00031/L. Electricity costs account for about 82% of the total, with material iron and alum accounting for 18%. Electricity and material costs associated with alum addition account for 8% of the total. Assuming per capita consumption of 10L/day, this amounts to \$1.13 per capita/year, or \$7.93 per family/year, assuming 7 people/family. For comparison, Roy (2008) has estimated the health care and loss of income cost to an average family in West Bengal drinking water with arsenic greater than 50 µg/L as approximately \$84 per year.

Implementation

ECAR is well suited to a community scale clean water center. It requires few consumable inputs, uses a simple and robust electrical system that can be maintained locally, requires no hazardous chemicals for operation or maintenance, produces very little waste, and is expected to be easy to maintain (electrochemical cleaning of the electrodes can be achieved by switching the current direction once per day). A community scale center is advantageous for arsenic removal because it (1) removes the burden of maintenance from the households, (2) allows for centralized monitoring of water quality, both ensuring that the treated water continues to be safe and allowing treatment to be tailored to local water conditions, lowering costs and waste, and (3) allows for rapid upgrades as new safer or lower cost technology is developed. Significantly, the operating cost of ECAR, including provisions for a small solar system, is low enough to envision clean water sold at a locally affordable price (perhaps \$0.002 per liter, or about 6 times the operating cost) with full cost recovery including capital costs, consumables, maintenance, and operation. Private sector involvement via a public-private partnership can further incentivize continued maintenance, quality control, and promote rapid scale-up.

Conclusions

Electrochemical arsenic remediation (ECAR) has been successfully tested in synthetic Bangladesh water and found capable of reducing up to 3000 µg/L initial arsenic (including As(III)) to below the WHO MCL of 10 µg/L. Solid/solution separation by low cost sedimentation can occur in < 4 hours with the addition of alum. ECAR treatment has been used to treat real Bangladesh groundwater (initial arsenic 93 – 510 µg/L) and real Cambodia groundwater (initial arsenic 80 – 760 µg/L). In all cases, ECAR was able to reduce arsenic levels to below the WHO MCL of 10 µg/L. A small-scale batch 100 L prototype has been fabricated and successfully tested in the lab. It will be field tested in West Bengal, India beginning in November 2010. The prototype produces little arsenic-laden waste, which has been characterized as non-hazardous using EPA methods. Initial tests show that up to 34% of concrete aggregate (by volume) can be replaced with ECAR generated sludge and show no detectable (< 1 µg/L) arsenic in leachate tests, making waste stabilization in concrete used for roadways a possible alternative to landfill disposal.

The low operating cost of ECAR (\$0.00031/L), along with its low waste production, lack of any hazardous materials, simplicity, ease of maintenance, and effectiveness at reducing arsenic below the WHO MCL of 10 µg/L in synthetic and real groundwater make it a promising candidate technology to operate in community scale clean water centers under public-private partnerships.

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