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Electro-chemical arsenic remediation: Field trials in West Bengal



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HIGHLIGHTS

- Product water consistently $< 5 \mu g/L$ arsenic during 3.5 mo field trial of ECAR reactor.
- Consumable and amortized capital at 5% (10 years) is \$0.83/m³ before optimization.
- Iron and aluminum concentrations in product water are below WHO recommended levels.
- ECAR supports a sustainable, scalable model addressing causes of previous failures.

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ABSTRACT

Millions of people in rural South Asia are exposed to high levels of arsenic through groundwater used for drinking. Many deployed arsenic remediation technologies quickly fail because they are not maintained, repaired, accepted, or affordable. It is therefore imperative that arsenic remediation technologies be evaluated for their ability to perform within a sustainable and scalable business model that addresses these challenges. We present field trial results of a 600 L Electro-Chemical Arsenic Remediation (ECAR) reactor operating over 3.5 months in West Bengal. These results are evaluated through the lens of a community scale micro-utility business model as a potential sustainable and scalable safe water solution for rural communities in South Asia. We demonstrate ECAR's ability to consistently reduce arsenic concentrations of ~266 μ g/L to <5 μ g/L in real groundwater, simultaneously meeting the international standards for iron and aluminum in drinking water. ECAR operating costs (amortized capital plus consumables) are estimated as \$0.83-\$1.04/m³ under realistic conditions. We discuss the implications of these results against the constraints of a sustainable and scalable business model to argue that ECAR is a promising technology to help provide a clean water solution in arsenic-affected areas of South Asia.

1. Introduction

Tens of millions of people in Bangladesh and India (particularly the Ganga river corridor in Uttar Pradesh, Bihar, and West Bengal) are exposed to high levels of naturally occurring arsenic in groundwater used for drinking (Rahman et al., 2009). Chronic ingestion of arsenic causes cancer, painful skin lesions, cardiovascular and reproductive problems, and may have a detrimental effect on the IQ of children (Kapaj et al., 2006). In the last decade the WHO reduced their recommended maximum contaminant level (WHO-MCL) from 50 $\mu g/L$ to 10 $\mu g/L$ based on emerging knowledge of cancer risks (WHO, 1993).

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Arsenic-related healthcare and loss of income costs a poor family in West Bengal ~\$84 per year (Roy, 2008).

Household and community scale arsenic treatment units have been quickly abandoned because they are not maintained, repaired, acceptable, or affordable (Das, 2011; Kabir and Howard, 2007). Technology evaluations often focus on the ability of a technology to solve these problems, ignoring or minimizing the role of the business model for going to financially viable large-scale implementation. These evaluations fail to account for key aspects of sustainability that a business model addresses, such as strategic investment in marketing to increase acceptability, or developing efficient supply chains to drastically reduce costs. These aspects play a crucial role in determining whether a technology is maintained, repaired, accepted, or affordable.

A technology can be designed to meet the needs of a business model rather than just the physical operating environment. Through this lens, it is possible to target sustainability at scale from the beginning of

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technology development. The full system comprising both the technology and the business model can then be iterated and evaluated together as a possible solution to address a complex problem, such as the arsenic crisis in South Asia.

Community scale micro-utilities based on public-private partnerships are attractive business models for arsenic remediation because they directly address the causes of previous failures. These models are amenable to rapid scale-up, provide incentives for ongoing maintenance and education, allow for centralized quality control (enabling education and increasing acceptability), remove undesired maintenance burden from the end-user, and eliminate the first cost barrier to direct ownership by allowing the user to "pay-as-you-go." Public-private partnerships based on a build-operate-transfer model have successfully sold treated water at locally affordable prices without subsidies in India, Bangladesh, and Ghana (Novogratz, 2007).

To successfully support this type of model, an arsenic removal technology must be (a) consistently effective to international and local arsenic standards in diverse and relevant groundwater compositions, (b) reliable and robust in the field with minimal and low-skilled maintenance, (c) low cost enough for clean water to be locally affordable with necessary business margins, (d) operable with minimal risk to safety and the environment, and (e) culturally acceptable to the local population. Each criterion supports the ability of a community scale micro-utility to reliably provide locally affordable water without subsidies.

Electro-chemical arsenic remediation (ECAR) is a form of electro-coagulation (EC) that has been developed to support a community scale micro-utility business model (Amrose et al., 2013). In EC, electrolytic oxidation of a sacrificial iron anode produces Fe(III) (oxyhydr)oxides (also called Fe(III) precipitates) in arsenic-contaminated water. Arsenic forms binuclear, inner-sphere complexes with Fe(III) precipitates (van Genuchten et al., 2012), which then aggregate to form a floc (Fig. 1). In ECAR, arsenic-laden flocs are separated from clean water through gravitational settling aided by a small amount of alum as a coagulant.

Many properties inherent in the ECAR process have advantages with respect to criteria *a–e* above. In ECAR, high capacity adsorbent media is generated in-situ, removing the need for a central adsorbent manufacturing plant or importing media from abroad. There is no difficult-to-predict breakthrough point, reducing possible failure modes. There is no media regeneration or hazardous chemical handling.

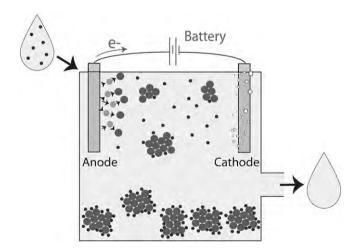


Fig. 1. The ECAR process is based on electrocoagulation (EC) with iron electrodes, depicted schematically above. Fe(II) dissolves from the iron anode (large light colored circles) where it is oxidized to Fe(III) and precipitated as Fe(III) (oxyhydr)oxide (large dark colored circles). Arsenic in solution (small black circles) binds to the surface of the Fe(III) (oxyhydr)oxide particles as they grow, and can be separated from product water via filtration or gravitational settling. The ECAR process uses gravitational settling with alum addition to reduce settling time (not depicted above).

The system is small and requires a minimal supply chain (e.g. steel plates, low-voltage electricity, and readily available alum coagulant). The process is highly amenable to automation, which can improve system reliability, reduce operator error and system downtime, and enable nighttime operation to take advantage of intermittent power when it tends to be most available. These properties help increase reliability, decrease operating costs, and decrease time to scale.

Prior research suggests further advantages of ECAR with respect to criteria a–e. ECAR effectiveness has been demonstrated using synthetic groundwater in lab studies, real contaminated groundwater from Bangladesh and Cambodia, and in short-duration field trials of two 100L batch reactors in West Bengal (Amrose et al., 2013). ECAR was found to consistently lower initial arsenic concentrations as high as 3000 $\mu g/L$ to below the WHO-MCL of 10 $\mu g/L$, and easily reached <5 μg/L. Strong oxidants produced during Fenton-type reactions were found to oxidize trivalent arsenite (As(III)) to pentavalent arsenate (As(V)) (Li et al., 2012). This is a key reaction for high effectiveness in the Bengal region because As(III) does not adsorb as strongly as As(V) to Fe(III) (oxyhydr)oxide surfaces in natural waters at neutral pH (Dixit and Hering, 2003), and both As(III) and As(V) are present in the groundwater (Kinniburgh and Smedley, 2001). Initial assessments of reliability, robustness, consumables cost, and sludge production from 100L reactor field trials were promising (Amrose et al., 2013), but very limited in scope due to the small system size and short duration. Longer term and larger scale field trials are needed to understand these properties over time and to understand whether effectiveness is consistent under realistic operating conditions. Field trials eventually need to be repeated across diverse aquifers to further understand and confirm broader ECAR applicability across South Asia.

In this paper we present field trial results of a 600 L ECAR reactor operating over 3.5 months in West Bengal. These results demonstrate ECAR's ability to consistently reduce arsenic concentrations to $<5~\mu g/L$ in a real arsenic contaminated groundwater source under realistic operating conditions. Operating cost estimates based on field trial results are in the range of \$0.83–\$1.04/m³. We discuss implications of the field trial on ECAR's ability to meet criteria a-e above and argue that ECAR remains a promising technology to help provide a sustainable and scalable clean water solution in arsenic-affected areas of South Asia.

2. Materials and methods

2.1. Chemical analysis

As(III) and total arsenic (As(tot)) were determined using an inductively coupled plasma optical emission spectrometer with hydride generation (HG-ICP-OES, Perkin Elmer5300 DV). P, Si, Al, Ca, Mg, and total Fe (Fe(tot)) were also determined with the same instrument (without hydride generation). The relative standard deviation (RSD) for the detection of all elements was <10% (typically <8%). Reported errors represent the larger of the standard deviation from repeated tests, \pm 10% measurement errors, and for arsenic concentrations a minimum measurement error of \pm 1 µg/L. Final arsenic concentrations reported for the 600 L reactor at coulombic dosages of 350 C/L and 250 C/L were measured using the arsenator field kit (PalinTest, UK) with a detection limit of 4 µg/L. Each sample was measured twice and the results averaged. This method produced concentrations within \pm 4 µg/L (27%) of HG-ICP-OES measurements based on 19 samples containing <20 µg/L arsenic (measured by HG-ICP-OES).

2.2. 600 L batch reactor

The 600 L ECAR batch reactor was fabricated in Mumbai using local materials and labor and shipped to West Bengal for testing. The reactor comprised three 750 L sintex tanks, one for electrolysis and two for settling. This allowed for simultaneous settling of two batches, though only

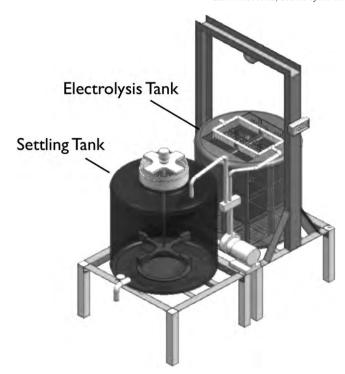


Fig. 2. Figure shows a schematic of the complete 600 L ECAR reactor assembly. The pump recirculates the water in reaction tank by withdrawing it from the bottom of the tank and pouring it into the tank through nozzles into each of the compartments formed by the iron plates. When the necessary residence-time has been reached, the same pump switches to delivering the water into the settling tank, by sending the water collected from the bottom of the reaction tank to the settling tank. A second settling tank is not shown.

one settling tank was used for field trials (Fig. 2). The electrolysis tank contained 4 cores, each with parallel interdigited mild-steel plates (5 anodes and 5 cathodes, each 625 mm \times 250 mm \times 4 mm). Alternate plates were electrically connected and spaced 2 cm apart. This design allows for easy scale-up by adding more cores to a larger tank. The design also allows for easy reversal of current, allowing each plate to be alternately deployed as anode and cathode to minimize extensive rust build-up and passivation. The prototype was equipped with an overhead beam and pulley system (included in the capital cost estimate) allowing the operator to easily add or remove individual cores of electrodes. A potentiostatic power supply provided current to the plates. A coulomb counter was set to the desired total coulombic dose (Coulombs per liter) and automatically ended electrolysis when that dosage was reached (taking into account current drift).

During electrolysis, water was recirculated through the tank at a turnover rate of ~3.4 reactor volumes per hour. After electrolysis, water was pumped into a settling tank and dissolved industrial grade non-ferric alum (Al₂(SO₄)₃·18 H₂O) was added to accelerate settling (Amrose et al., 2013). Alum that was purchased in the local market was added during rapid mixing with an impellor (130 rpm for ~2 min). Alum dosages were 6-15 mg/L (as Al). The batch was slowly agitated (15 rpm for 20 min) before being allowed to settle for at least 2 h. Settled sludge was removed with a pump after every 1–2 batches and stored onsite after most runs (electricity for pumping included in the cost estimate). This resulted in 520 L of product water per batch. There was no effort to maximize the amount of product water relative to source water at this stage. Some pre-field trial experiments were performed to benchmark reactor performance in the Civil Engineering Department of Jadavpur University (Kolkata, India). These tests used arsenic-free Kolkata groundwater (Table 1) from a shallow tubewell at Jadavpur University (32 m depth) spiked with 350 or 530 μg/L As(III) as NaAsO₂.

Table 1Groundwater composition for Kolkata groundwater, Dhopdhopi High School groundwater and published/derived values for Bangladesh.

		Kolkata groundwater ^a	Dhopdhopi Groundwater	Bangladesh ^d	
pН		7 ± 1 ^b	7 ± 1 ^b	7.05 ± 0.22	
As	$(\mu g/L)$	<4 ^c	266 ± 42	129 ± 155	
PO ₄ as P	(mg/L)	0.09 ± 0.01	1.4 ± 0.2	1.3 ± 1.5	
SiO₃ as Si	(mg/L)	17 ± 1.7	17.4 ± 1.7	19.7 ± 5.1	
SO ₄ as S	(mg/L)	-	3.3 ± 1.3	1.5 ± 5.8	
Ca	(mg/L)	120 ± 19	130 ± 13	66 ± 53	
Mg	(mg/L)	41 ± 4.7	35 ± 4.0	27 ± 21	
Fe	(mg/L)	0.24 ± 0.12	14.5 ± 2.1	5.6 ± 5.9	

- ^a A dash (-) indicates unmeasured ions.
- b During field trials, pH was measured using pH test strips (Merck).
- ^c This is the raw water arsenic concentration (before being spiked with As(III)) as measured by the arsenator field kit (Section 2.1). No arsenic was detected, so the detection limit is given as an upper bound.
- $^{
 m d}$ Groundwater parameters in Bangladesh were derived from the BGS (Kinniburgh and Smedley, 2001). pH was taken from the Special Study areas using 155 wells; all other values taken from the National Survey data using only wells with As > 10 μ g/L.

2.3. Field operation

The reactor was dismantled, shipped from its original setup at ladavpur University, and reassembled in an empty classroom at Dhopdhopi High School in South 24 Parganas, West Bengal, India (enrollment ~2500 coed students). The reactor was operated for 1–2 batches on most weekdays for 3.5 months from December to early March using raw water with composition shown in Table 1 pumped from a shallow tubewell (106 m deep, 266 \pm 42 µg/L initial arsenic). The reactor voltage was initially set to produce a current of 45 A except where noted, and generally drifted <5 A throughout electrolysis. Electrode polarity was reversed with each batch. Product water samples were collected from a tap ~10 cm above the bottom of the reactor tank for arsenic and iron measurements. Samples were stored onsite in 1 L bottles, which were later shaken before decanting into smaller 50 mL sample vials. Samples were acidified before chemical analysis. Raw water samples were collected from the well-head after approximately 5 min of continuous initial pumping to avoid bacterial contamination and oxygenated water in the well head. These samples were acidified immediately. As monitored by pH test strips (Merck, ± 1 pH unit), pH of the samples of raw water and those collected after electrolysis and settling, remained near pH 7. This was consistent with beaker batch tests in synthetic Bangladesh groundwater and field tests of a 100 L ECAR reactor in West Bengal (Amrose et al., 2013).

Over 3.5 months beginning in October of 2012, a total of 64,480 L of arsenic contaminated groundwater (composition in Table 1) was remediated in the 600 L reactor. Of these, approximately 34,000 L groundwater was remediated under variable operating configurations to understand the effect of different parameters on arsenic removal performance. The remaining 31,000 L was remediated under a standard common set of operating parameters listed in Table 2.

2.4. Routine reactor maintenance

The prototype was operated by a trained local operator and sometimes by research staff. Maintenance tasks included occasional light brushing of the plates, in situ, at intervals ranging from alternate days to alternate weeks. Maintenance was completed by a local operator using locally available materials and recorded into a maintenance log.

2.5. Quantifying dry waste sludge

To quantify the amount of dry sludge produced per batch, all of the wet waste sludge produced from four batches of the 600 L reactor was collected and dried in an oven at 100 °C over several days.

Table 2Arsenic and iron removal performance and operating parameters used for 600 L batch reactor field trials.

Exp series ^a	Water source	Liters treated	Charge dosage rate	Coulombic dose	Initial As	Final As	Initial Fe	Final Fe
		(kL)	(C/L/min)	(C/L)	(μg/L)	(µg/L)	(mg/L)	(mg/L)
KG-A	Kolkata groundwater	4.2	5.0	400	350 ± 61	9.3 ± 1.3	0.24 ± 0.12	
KG-B	Kolkata groundwater	1.6	5.0	400	532 ± 52	16 ± 3.3	0.24 ± 0.12	_
DG-A	Dhopdhopi groundwater	31	4.3	450	266 ± 42	5.3 ± 4.8	14.1 ± 2.1	0.04 ± 0.06
DG-A-Subset ^b	Dhopdhopi groundwater	15	4.3	450	266 ± 42	2.1 ± 1.0	14.1 ± 2.1	0.05 ± 0.07

- ^a The alum dosage for all experiments was 6–15 mg/L (as Al), Product water aluminum concentrations for 28 batches in series DG-A were all < 1 mg/L.
- ^b This is the subset of the experimental series DG-A during which electrode plates were brushed at least once per week.

3. Results and discussion

3.1. 600 L reactor performance in spiked Kolkata groundwater

The 600 L batch reactor was initially tested on Kolkata groundwater (sourced at Jadavpur University) spiked with As(III) to confirm replicable arsenic remediation under lab operation. Initial As(III) concentrations of 350 \pm 61 µg/L (8 batches, 4160 L total) and 532 \pm 52 µg/L (3 batches, 1560 L total) were remediated using a coulombic dose of 400 C/L. Final arsenic concentrations in the product water were 9.3 \pm 1.3 µg/L (\pm 14%) and 16 \pm 3.3 µg/L (\pm 21%) respectively, demonstrating replicable arsenic removal performance for final concentrations near the WHO-MCL. At the higher initial As(III) concentration, product water arsenic was slightly above the WHO-MCL. This suggests that dosages > 400 C/L may be necessary to consistently reach the WHO-MCL for initial As(III) concentrations in the range of 500 µg/L.

Arsenic removal efficiency (i.e. arsenic removed per unit coulombic dose) in spiked Kolkata groundwater was generally poorer than the efficiency observed in beaker batch tests with synthetic Bangladesh groundwater. In synthetic groundwater, 300 C/L was sufficient to lower initial arsenic concentrations of 2900 \pm 160 µg/L (half as As(III), half as As(V)) to below the WHO-MCL (Amrose et al., 2013). The significantly lower arsenic removal efficiency in Kolkata groundwater is surprising — even more so considering the much lower phosphate concentration of Kolkata groundwater relative to synthetic groundwater ([P] = 0.09 mg/L versus [P] = 1.3 mg/L). This is because phosphate ions are known to compete with arsenic for sorption sites (Roberts et al., 2004).

One possible explanation is that the process used to separate arsenic-laden sludge from product water in the 600 L reactor isn't as effective as the process used in beaker batch tests. The 600 L reactor relied on gravitational settling aided by a small amount of alum, whereas the beaker batch tests used 0.1 μm vacuum filtration. To compare filtration to gravitational settling with alum in the 600 L reactor, water samples from the electrolysis tank were filtered through 0.45 μm filters immediately after electrolysis and compared to the product water. Filtered and settled samples had arsenic concentrations of 7.1 \pm 1.1 $\mu g/L$ and 9.5 \pm 1.9 $\mu g/L$ respectively, indicating comparable performance between the two separation methods. This comparison does not rule out the possibility that some arsenic-laden particles can pass through the 0.45 μm filter but not the 0.1 μm filter, but it does constrain the hypothesis to this narrow size range.

The difference in arsenic removal efficiency between the 600 L reactor and beaker batch tests could also be due to the difference in charge dosage rate (Q = 5 C/L/min for reactor tests, versus Q = 2.2 C/L/min for beaker tests). In a previous study, we showed that an increase in charge dosage rate from 2 to 5 C/L/min in a comparable EC system resulted in lower efficiency – it led to an ~40% increase in the coulombic dose required to lower 500 μ g/L As(III) to 50 μ g/L total arsenic (Li et al., 2012). Computer modeling reproduced this effect and attributed it to increased competition between As(III) and Fe(II) for strong oxidants at higher charge dosage rates (Li et al., 2012). Higher competition for oxidants decreases the likelihood that As(III) will be oxidized to As(V), which in turn can decrease arsenic removal efficiency due to the lower uptake of As(III)

at neutral pH (Dixit and Hering, 2003). A direct comparison between the model and the 600 L reactor performance is not possible because the model assumes that equilibrium is reached after electrolysis, which is not true of the reactor. We do not currently have a non-equilibrium model of the reactor. However, a similar effect due to charge dosage rate might explain the difference in arsenic removal efficiency seen here.

3.2. Arsenic removal in the field

During 3.5 months of operation at Dhopdhopi High School, the 600 L ECAR reactor remediated 31,000 L (50 batches) of arsenic contaminated groundwater (Table 1, initial arsenic 266 \pm 42 µg/L) using a consistent set of operating parameters (Table 2). This produced 26,000 L of product water with arsenic concentrations of 5.3 \pm 4.8 µg/L. Final concentrations were less than the WHO-MCL and Indian regulatory limit (10 µg/L) in 86% of cases and less than the Bangladesh regulatory limit (50 µg/L) in 100% of cases respectively. The maximum final arsenic concentration was 22 \pm 5 µg/L.

The reactor voltage tended to be higher for batches with higher final arsenic concentrations. Lightly brushing the electrode plates with a steel brush decreased the voltage by ~33% and improved arsenic removal performance. This practice was adopted and continued at least once a week for a subset of 24 consecutive batches (14,880 L), producing 12,480 L of product water with arsenic concentrations of 2.1 \pm 1.0 $\mu g/L$ (Fig. 3). All final arsenic concentrations in this set were <5 $\mu g/L$ (less than 50% of the WHO-MCL) with a maximum final concentration of 3.7 \pm 1.0 $\mu g/L$. These results demonstrate that with a simple maintenance configuration, the 600 L ECAR reactor can consistently produce high quality arsenic-safe product water under real operating conditions.

The slow increase in voltage with time suggests that there may be a resistive layer forming over time on the electrode plates. However, the decrease in final arsenic concentration after lightly brushing the electrodes suggests that this simple solution could mitigate the adverse effects of a resistive layer. Longer-term field trials are needed to confirm the effect of electrode cleaning over longer time scales.

All of the above results were obtained using a conservative coulombic dose of 450 C/L. Lower coulombic dosages of 350 C/L and 250 C/L were used to remediate three additional batches each, resulting in average final arsenic concentrations of 2.7 \pm 2.4 µg/L and 6.7 \pm 6.3 µg/L respectively. This suggests that lower dosages may be sufficient to reach the WHO-MCL for the Dhopdhopi High School groundwater source. Longer duration field trials at lower coulombic dosages are needed to confirm this tentative finding.

Groundwater for this field trial contained relatively high natural iron ([Fe] = 14.1 ± 2.1 mg/L, Table 1). Naturally occurring iron in groundwater can oxidize when exposed to dissolved oxygen, potentially aiding arsenic removal. This process depends heavily on aquifer composition and generally does not produce final arsenic concentrations below $50 \, \mu \text{g/L}$ (BAMWSP, DFID, WaterAid, 2001). However, naturally occurring iron is expected to enhance arsenic removal during ECAR when it is present. This can potentially lower operating costs in areas with high natural iron, but may also enhance the arsenic removal effectiveness reported here compared to similar aquifers with less natural iron. Results from spiked Kolkata groundwater demonstrate that up to

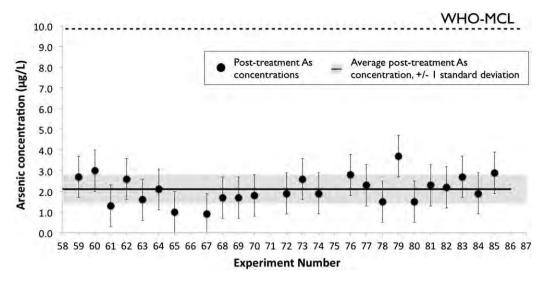


Fig. 3. Post-treatment arsenic concentrations for a series of experiments during the 600 L ECAR reactor trial at Dhopdhopi High School in West Bengal, India. For these experiments, electrode plates were lightly brushed at least once per week. Three post-treatment samples were lost (experiments 66, 71, and 75). The dashed line indicates the WHO maximum contaminant level (WHO-MCL) recommended for arsenic in drinking water.

 $350~\mu g/L~As(III)$ accompanied by much lower natural iron concentrations (0.24 $\pm~0.12~mg/L$) can be lowered to the WHO-MCL consistently (8 batches). This is consistent with beaker batch test results in synthetic groundwater containing no initial iron with up to 2900 $\mu g/L$ initial As(III) and As(V) (Amrose et al., 2013). However, results from the 600 L reactor field trial still need to be confirmed across diverse aquifers, including those with low natural iron concentration.

3.3. Post-treatment iron concentration and iron removal

In the ECAR process, iron is added to source water and then removed. This could result in high and aesthetically displeasing iron concentrations in treated water. Product water iron concentrations in the 26,000 L product water produced by the 600 L reactor at a coulombic dose of 450 C/L were 0.04 \pm 0.06 mg/L (Fig. 4). Thus iron concentrations in the product water were consistently below the human taste threshold and the maximum iron concentration recommended by the WHO of 0.3 mg/L (Mirlohi et al., 2011). This was true despite a relatively high iron concentration in the source water (14.1 \pm 2.1 mg/L) and the periodic brushing of electrodes, which introduced additional rust flakes into the source water during some batches. These results demonstrate

that the 600 L ECAR reactor is capable of consistently producing product water that meets international health and aesthetic standards for both arsenic and iron. In addition, researchers and local staff reported no objectionable tastes in the ECAR treated water, which was aesthetically preferred to source water due its much lower iron content.

3.4. Post-treatment aluminum concentrations

During this field trial, post-treatment aluminum concentrations were measured for 28 consecutive batches (16,120 L treated) from the 600 L reactor operating with 6–15 mg/L alum (as Al) added during the settling phase. Aluminum concentrations were below 0.1 mg/L for all samples. The WHO-MCL and Indian regulatory limit for aluminum is 0.2 mg/L (BIS, 2012; WHO, 2008). Thus, ECAR was able to consistently meet the WHO-MCL for aluminum using gravitational settling with no additional filtration.

3.5. Post-treatment turbidity

Product water turbidity across 31,000 L treated at a coulombic dose of 450 C/L was 7.9 \pm 4.2 NTU. This is higher than WHO-MCL and Indian

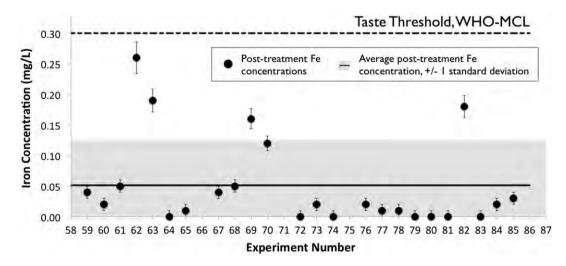


Fig. 4. Post-treatment total iron concentrations for a series of experiments during the 600 L ECAR reactor trial at Dhopdhopi High School in West Bengal, India. For these experiments, electrode plates were lightly brushed at least once per week. Three post-treatment samples were lost (experiments 66, 71, and 75). The dashed line indicates the human taste threshold for iron and the WHO aesthetic-based recommendation for iron in drinking water.

regulatory standards for drinking water of <1 NTU (BIS, 2012; WHO, 2008). Using only the subset of runs in which electrodes were brushed at least once per week (14,880 L), product water turbidity was 11 \pm 4.3 NTU. Alum dosages across batches varied from 6 to 15 mg/L (as Al). Turbidity did not differ significantly with alum dosage, but only few experiments were conducted with alum dosage as the only variable, making it difficult to draw conclusions. Consistent low iron concentrations in product water suggest that the excess turbidity is not due to residual Fe(II) oxidizing during the settling period.

Notably, there was no correlation between turbidity >1 NTU and arsenic, iron, or aluminum exceeding their respective WHO-MCLs during this trial. Consequently, these results suggest that turbidity was not a suitable proxy for arsenic, iron, or aluminum removal in our experiments.

3.6. Energy consumption in the field

Voltage across the electrode plates depends on a number of factors, including ionic strength of the source water, distance between plates, and operating current. For the 600 L reactor at Dhopdhopi High School the operating parameters listed in Table 2 plus electrode brushing at least once per week resulted in a stable voltage of 4.2 \pm 0.3 V by the end of electrolysis. Voltage drifted <0.5 V during treatment. Without routine maintenance the voltage rose to ~6 V on some occasions, but was quickly reduced back to ~4 V after maintenance was performed. The maximum measured voltage during the trial was 6.4 V \pm 0.1 V. To achieve the same current in Kolkata groundwater, the required voltage was only 2.1 \pm 0.3 V, demonstrating that the voltage can change significantly across different water sources. The low voltage in Kolkata groundwater was likely due to the salinity of the source, which had a noticeable salty taste. The higher voltage from the field trials was used in cost estimates.

There are tradeoffs between energy consumption, treatment time, and removal efficiency. To produce the same total coulombic dose in less time requires an increase in charge dosage rate (assuming the water volume being treated remains unchanged). This requires an increase in operating current and power, and may result in a lower As(III) removal efficiency due to competition between As(III) and Fe(II) for strong oxidants produced during Fe(II) oxidation with dissolved oxygen (Amrose et al., 2013; Li et al., 2012). Decreasing the treatment time may also decrease the average contact time between arsenic and the adsorbent decreasing both As(III) and As(V) removal efficiency (Amrose et al., 2013). In general, decreasing the treatment time will result in higher energy consumption, resulting in larger consumables cost, but also higher throughput for a given reactor, leading to smaller amortized capital costs.

For the operating parameters listed in Table 2, energy consumption of the 600 L reactor at Dhopdhopi school was 0.62 kWh/m³ (electrolysis only) when operating at 185 W. Under these conditions, the total coulombic dose of 450 C/L was applied in approximately 105 min. Time varies slightly between batches because electrolysis duration is set by an integrating coulomb counter to account for current drifts during treatment. Total throughput for this test reactor was 260 ± 30 L/h including time for alum addition, settling, and water transfer between tanks. The total energy consumption of the 600 L reactor, including recirculation during electrolysis and alum addition plus pumps to transfer water (e.g. from source to dosing tank, from dosing tank to settling tank, from settling tank to storage, and for sludge removal) was 2.31 kWh/m³ without any optimization. Several design changes to reduce energy consumption were noted for the next generation prototype.

3.7. Sludge output

All arsenic removal technologies produce arsenic-laden sludge or waste that must be disposed of properly. During operation at Dhopdhopi High School, the 600 L reactor produced ~245 mg/L dried

sludge (245 g/m³) at the highest coulombic dose (450 C/L). This dose was sufficient to consistently produce product water with $<5~\mu g/L$ arsenic. A previous 100 L ECAR test reactor produced $\sim0.4\%$ wet waste sludge by volume during field trials in West Bengal (Amrose et al., 2013). Neither the 100 L nor 600 L reactor design made any attempt to minimize sludge production at this stage in development. Opportunities to further reduce sludge volume were noted for future reactor designs.

3.8. Cost estimate

Consumables for ECAR include (1) iron consumed in the sacrificial anode, (2) electrical energy supplied for electrolysis, mixing, and pumping water (e.g. from source to dosing tank, from dosing tank to settling tank, from settling tank to storage, and for sludge removal), and (3) non-ferric commercial alum added to aid settling. Cost estimates are based on average measured power consumption during the 600 L prototype field trial with no optimization to reduce energy consumption. Input costs are actual (small-quantity) procurement costs for the field trial (~\$1/kg for steel plate, \$0.09/kWh for grid electricity, \$0.26/kg for commercial non-ferric alum). Consumables costs are \$0.44/m³ using a conservative charge loading shown to reliably achieve <5 µg/L. Electricity accounts for 49% and materials for 51% of the total cost. During the field trial, a smaller coulombic dose was delivered for several runs with no performance loss at a consumables cost of \$0.30/m³. The same level of performance was achieved in the 100 L ECAR reactor prototype for \$0.20/m³ (Amrose et al., 2013), suggesting the costs could be pushed lower with some optimization.

The 600 L prototype reactor cost \$5181 to custom build in India, including materials, labor, manufacturer overhead (including 20% margin), and retail purchase of all pumps and pipes. Table 3 contains a full breakdown of costs associated with the fabrication. Notably, labor alone accounts for 24% of the total, reflecting the custom fabrication. The reactor cost includes two settling tanks to increase throughput, as described in Section 2.2. To estimate reactor cost at a 10,000 L/day capacity, we assume 2 identical 600 L reactors (5500 L/day capacity each) with no economies of scale. Assuming 6 days per week of operation and amortizing over 10 years at 5% (assuming social rate for infrastructure investment) or 15% (assuming commercial rate for business investment) results in a total materials cost (amortized capital plus consumables) of \$0.83/m³ (\$0.0008/L) and \$1.04/m³ (\$0.001/L) respectively. This estimate ignores possible design optimization to minimize capital and consumables cost.

Sludge collection and stabilization is an important component of the operating cost. ECAR sludge is considered non-hazardous according to the US-EPA TCLP test (Amrose et al., 2013). However, we conducted preliminary cost calculations based on landfilling fees for the Haldia

Table 3Breakdown of costs for the 600 LECAR Batch Reactor used in this field trial. The reactor was custom built in Mumbai, India.

Item	Cost ^a		
	(USD)		
Electrodes	247		
Electrode assembly	149		
Power supply (including wiring)	864		
Voltage stabilizer	81		
Motors and impeller	647		
Piping	217		
Tanks	264		
Pumps	173		
Support structure	430		
Labor	1259		
Manufacturing overhead	850		
Total	5181		

^a Exchange rate from Indian Rupees is INR/USD 55.3649, taken from the time of fabrication

Hazardous Waste facility (West Bengal, India) and assuming transportation to the Haldia site from the farthest region with documented arsenic contamination within West Bengal. Our calculations suggest that these costs would add less than 5% to the cost per liter of arsenic-safe water production.

A viable business model must include additional costs for inventory, marketing, management, aesthetic enhancements (e.g. reducing turbidity to 1 NTU), a civil structure to house the equipment, quality control, and normal business margins, which have not been included in this cost estimate. Each will be incorporated as more data becomes available.

3.9. Implications for proposed business model

Implications of the new field trial results on criteria *a*–*e* outlined in the introduction are discussed below.

3.9.1. Effectiveness

ECAR's ability to consistently remediate arsenic to $<5~\mu g/L$ in the 600 L reactor has been demonstrated in an arsenic contaminated aquifer in West Bengal, addressing longer-duration performance consistency for the first time. This adds to previous work demonstrating highly effective arsenic removal across beaker batch tests, preliminary field studies in Bangladesh and Cambodia, and field trials of two 100 L reactors in West Bengal. Future field trials should target diverse aquifers, particularly with respect to natural iron content, and continue to verify effectiveness on larger scales.

3.9.2. Reliability and robustness

These trials produced a simple maintenance scheme (electrode brushing) that improved reliability and effectiveness of ECAR performance in the field. A trained local operator reported easy operation, maintenance, and undertook small repairs of the 600 L reactor without supervision. No unforeseen failure modes were discovered during this trial, and no downtime occurred because the reactor was in disrepair (downtime occurred due to holidays, demonstrations, and short term unavailability of trained operators). Additional longer-term field trials are needed to further validate ECAR robustness and reliability across seasons and to understand longer-term maintenance requirements.

3.9.3. Low cost

Several key components have been added to the cost estimate as a result of this 600 L field trial, making the cost estimate more robust. These include an estimate of capital costs for local manufacture, which is purposely conservative to act as an upper bound (i.e. assuming no optimization to reduce manufacturing costs and no economies of scale). Alum costs are more robust because they are based on the actual retail cost in the local market. Arsenic removal performance was also confirmed using the lower cost locally available industrial grade alum, which is of lower quality than the lab grade aluminum sulfate used in previous trials (Amrose et al., 2013). Electricity for the entire end-to-end treatment process, including the pumping of water from a tubewell source and the pumping of water to an overhead storage tank, has been taken into account.

Several costs still need to be assessed. Turbidity levels did not meet their respective WHO-MCL under the conditions tested here. More research is needed to understand the source of the turbidity and possibly minimize it by optimizing the alum dosage. The cost of a filter or other method to reduce turbidity from ~10 NTU to <1 NTU should be explored and added to cost estimates if necessary. In addition it is possible that the high naturally occurring iron concentration in the source water artificially lowered the cost estimate relative to aquifers with lower naturally occurring iron. The coulombic dosage used in the estimate was conservative (450 C/L), and lower dosages (350 and 250 C/L) were also able to reach the WHO-MCL for arsenic. This conservative choice can mitigate some of the uncertainty with respect to natural iron, but

additional field trials in aquifers with lower natural iron content are needed to confirm and refine these costs. Finally, more research is needed to understand the cost to collect and stabilize arsenic-laden waste sludge.

3.9.4. Minimal risk to safety and environment

The majority of the safety and environmental risk comes from the handling of arsenic-laden waste sludge. We previously demonstrated that ECAR waste sludge passes the Toxicity Characteristic Leaching Procedure (TCLP) using US EPA Method 1311 (and is therefore not considered hazardous waste in the U.S.) (Amrose et al., 2013). However, 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 stabilization methods have been explored. Banerjee and Chakraborty (2005) have demonstrated arsenic stabilization meeting Government of India standards in briquettes, cement sand-mortar, and concrete containing a similar arsenic-laden sludge mixed in at up to 10, 18, and 40% of volume respectively. This technique has been highlighted as the best option for arsenic wastes when compared to incineration and dilution/dispersion (Leist et al., 2000). ECAR sludge produced during beaker batch tests (arsenic loading ~1.5% by weight) was mixed into the aggregate of a Portland-cement concrete mixture, replacing 40% concrete by volume and 6% by weight. TCLP leachate of the crushed concrete showed no detectable arsenic (<1 µg/L). Other metals (Ag, Ba, Cd, Cr, Hg, Pb, and Se) were either not detected or well below US EPA regulatory limits (Gadgil et al., in press). This suggests that concrete stabilization may be a viable alternative to landfill disposal for ECAR.

Concrete used to stabilize arsenic-laden sludge could potentially be packed into roadways with minimal risk to the environment. This is the subject of ongoing investigation. All sludge from the 600 L reactor trials reported here was collected and is currently being used to study the long-term fate of arsenic when ECAR sludge is embedded in concrete.

The electrical systems of ECAR operate at low voltage (<6 V) and present minimal safety risk to operators.

3.9.5. Cultural acceptability

During this field trial, we met with local stakeholders for our host school (including a workshop with teachers, staff, and students to elicit feedback), host community, and with various Government officials (local and district). The technology was demonstrated and the process explained. No cultural objections to the technology arose in this limited setting. Longer-duration field trials are needed to better assess public acceptance.

The issue of safe sludge management has come up in various forums as a concern of all arsenic remediation technologies. Developing a plan to facilitate safe sludge stabilization into concrete will be an important aspect of the full arsenic solution.

4. Conclusions

Using the criteria a–e outlined in the Introduction, ECAR continues to be a promising candidate technology to operate in community scale micro-utilities offering clean water at a locally affordable price. The 600 L reactor field trial has confirmed consistent highly effective arsenic removal performance of ECAR under realistic conditions. The reactor consistently achieved <5 μ g/L arsenic in product water while meeting the WHO-MCL for iron and aluminum. The reactor operated reliably, robustly, and at a low cost while producing minimal waste. Additional work is needed to refine study stabilization of arsenic-laden sludge and to reduce turbidity in product water. Larger scale and longer-term field trials, particularly across diverse aquifers, with concurrent development and iteration of the proposed business model will continue to refine the assessment.

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References

- Amrose S, Gadgil A, Srinivasan V, Kowolik K, Muller M, Huang J, et al. Arsenic removal from groundwater using iron electrocoagulation: effect of charge dosage rate. J Environ Sci Health A 2013:48:1019–30
- BAMWSP, DFID, WaterAid. Rapid assessment of household level arsenic removal technologies. Phase I Final Draft Report. UK: BAMWSP/DFID/WaterAid; 2001.
- Banerjee G, Chakraborty R. Management of arsenic-laden water plant sludge by stabilization. Clean Technol Environ Policy 2005;7:270–8.
- BIS. 10500 Indian Standard drinking water-specification. Second revision. Delhi, India: Bureau of Indian Standards; 2012.
- Das A. PhD Thesis, The economic analysis of arsenic in water: a case study of West Bengal. Kolkata: Jadavpur University; 2011.
- Delemos JL, Bostick BC, Renshaw CE, Sturup S, Feng X. Landfill-stimulated iron reduction and arsenic release at the Coakly superfund site (NH). Environ Sci Technol 2006;40: 67–73
- Dixit S, Hering JG. Comparison of arsenic(V) and arsenic(III) sorption onto iron oxide minerals: implications for arsenic mobility. Environ Sci Technol 2003;37:4182–9.

- Gadgil A, Amrose S, Bandaru SRS, van Genuchten CM, Delaire C, Torkelson A, et al. Addressing arsenic mass poisoning in South Asia with electro-chemical arsenic remediation (ECAR). In: Ahuja S, editor. Water reclamation and sustainability. Elsevier; 2013s. [in press].
- Ghosh A, Mukiibi M, Ela W. TCLP underestimates leaching of arsenic from solid residuals under landfill conditions. Environ Sci Technol 2004;38:4677–82.
- Kabir A, Howard G. Sustainability of arsenic mitigation in Bangladesh: results of a functionality survey. Int J Environ Health Res 2007;17:207–18.
- Kapaj S, Peterson H, Liber K, Bhattacharya P. Human health effects from chronic arsenic poisoning—a review. J Environ Sci Health A 2006;41:2399–428.
- Kinniburgh DG, Smedley PL. Arsenic contamination of groundwater in Bangladesh, vol. 1–4. Keyworth, U.K.: British Geological Survey; 2001
- Leist M, Casey RJ, Caridi D. The management of arsenic wastes: problems and prospects. | Hazard Mater 2000;76:125–38.
- Li L, van Genuchten CM, Addy SEA, Yao J, Gao N, Gadgil A. Modeling As(III) oxidation and removal with iron electrocoagulation in groundwater. Environ Sci Technol 2012;46: 12038–45
- Mirlohi S, Dietrich AM, Duncan SE. Age-associated variation in sensory perception of iron in drinking water and the potential for overexposure in the human population. Environ Sci Technol 2011:45:6575–83.
- Novogratz J. Meeting urgent needs with patient capital. Innov: Technol, Governance, Globalization 2007;2:19–30.
- Rahman MM, Naidu R, Bhattacharya P. Arsenic contamination in groundwater in the Southeast Asia region. Environ Geochem Health 2009;31:9–21.
- Roberts LC, Hug SJ, Ruettimann T, Billah MM, Khan AW, Rahman MT. Arsenic removal with iron(II) and iron(III) in waters with high silicate and phosphate concentrations. Environ Sci Technol 2004;38:307–15.
- Roy J. Economic benefits of arsenic removal from ground water a case study from West Bengal, India. Sci Total Environ 2008;397:1–12.
- van Genuchten CM, Addy SEA, Peña J, Gadgil AJ. Removing arsenic from synthetic groundwater with iron electrocoagulation: an Fe and As K-edge EXAFS study. Environ Sci Tech 2012;46:986–94.
- WHO. Guidelines for drinking-water quality: recommendations, vol. 1. Geneva, Switzerland: The World Health Organization; 1993.
- WHO. Guidelines for drinking water quality: recommendations, incorporating the first and second addenda, vol. 1. Geneva, Switzerland: World Health Organization; 2008.