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Publication Date
2009-10-31

Peer reviewed

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September 17, 2009

This work was conducted at the University of California, Berkeley and Lawrence Berkeley National Laboratory under U.S. Department of Energy Contract No. DE-AC02-05CH11231. Funders include the National Collegiate Inventors and Innovators Alliance (NCIIA) and the University of California, Berkeley's Richard C. Blum Center for Developing Economies, Bears Breaking Boundaries Contest, Sustainable Products and Solutions (SPS) Program, and Big Ideas Marketplace. Past funders include the Lawrence Berkeley National Laboratory's Technology Transfer Office, the Blue Planet Run Foundation, the California Energy Commission (CEC), and the American Water Works Association Research Foundation (AwwaRF).

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This report is an improved version of a paper published in the proceedings of:
The 2008 University of North Carolina Environmental Symposium: Safe and Sustainable Drinking Water in Developing Countries: Where Science Meets Policy, Chapel Hill, NC, November 5-6, 2008.

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ACKNOWLEDGEMENTS
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A special thanks to our peer reviewers: Kayje Booker and Maithili Iyer.

DISCLAIMER

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Abstract
ARUBA (Arsenic Removal Using Bottom Ash) has proven effective at removing high concentrations of arsenic from drinking water in Bangladesh. During fieldwork in four sub-districts of the country, ARUBA reduced arsenic levels ranging from 200 to 900 ppb to below the Bangladesh standard of 50 ppb. The technology is cost-effective because the substrate—bottom ash from coal fired power plants—is a waste material readily available in South Asia. In comparison to similar technologies, ARUBA uses less media for arsenic removal due to its high surface area to volume ratio. Hence, less waste is produced. A number of experiments were conducted in Bangladesh to determine the effectiveness of various water treatment protocols. It was found that (1) ARUBA removes more than half of the arsenic from water within five minutes of treatment, (2) ARUBA, that has settled at the bottom of a treatment vessel, continues to remove arsenic for 2-3 days, (3) ARUBA's arsenic removal efficiency can be improved through sequential partial dosing (adding a given amount of ARUBA in fractions versus all at once), and (4) allowing water to first stand for two to three days followed by treatment with ARUBA produced final arsenic levels *ten times lower* than treating water directly out of the well. Our findings imply a number of tradeoffs between ARUBA's effective arsenic removal capacity, treatment system costs, and waste output. These tradeoffs, some a function of arsenic-related policies in Bangladesh (e.g., waste disposal regulations), must be considered when designing an arsenic removal system. We propose that the most attractive option is to use ARUBA in community-scale water treatment centers, installed as public-private partnerships, in Bangladeshi villages.

Introduction
Naturally-occurring arsenic in drinking water is a major public health problem threatening the well-being (and in many cases, lives) of more than a hundred million people worldwide. According to World Health Organization estimates, in Bangladesh alone, 28-77 million people drink arsenic-laden water from shallow tubewells (Ahmad et al. 2003). The vast majority of these wells were installed by the Bangladeshi government organizations, international agencies, various NGOs, and private citizens within the past 40 years as an alternative to drinking biologically contaminated surface water. High levels of arsenic were first noted in the shallow tubewells in the early 1990s. The country is currently experiencing the largest case of mass poisoning in human history. Arsenic-laden water is also known to exist in Argentina, Australia, Chile, China, Hungary, India, Mexico, Nepal, Peru, Thailand, and the United States.

It often takes many years before a person starts showing symptoms of arsenic poisoning (arsenicosis) so they may not immediately realize that they are being exposed to the toxin. The initial affects of arsenic poisoning are painful—lesions form on the hands and feet making daily chores difficult or impossible. Long-term chronic exposure leads to a variety of very serious health problems (e.g., liver and spleen enlargement and cirrhosis of the liver; myocardial degeneration and cardiac failure;

peripheral neuropathy affecting primary sensory functions; diabetes mellitus and goiter; and cancer of the bladder, kidney, lung, and skin (Chen and Ahsan 2004; Chowdhury 2004; Navas-Acien et al. 2008)). Vascular problems caused by arsenic can also lead to gangrene and amputations. People with poor nutrition are more likely to show signs of arsenicosis when exposed to a given dose of arsenic (WHO 2000, Biswas et al. 1998). Since current medical treatment cannot adequately address the long-term effects of arsenic poisoning, a preventative solution is necessary to address this issue.

For neighboring West Bengal, India, the welfare benefit of eliminating exposure to arsenic in drinking water (at a concentration of 400 ppb) is estimated to be approximately $48 per household per year (calculation based on that of Roy 2008, which takes into account costs associated with avoiding arsenic exposure, medical expenditures, and wealth loss due to sickness). We expect this figure to be similar in Bangladesh and, if so, this would imply that arsenicosis contributes to the continued poverty of the country. In 1997, Bangladesh was ranked 12th worldwide on the UNDP Human Poverty Index (UNDP 1997).

Scientists at the Lawrence Berkeley National Laboratory have developed a simple material named "ARUBA"—Arsenic Removal Using Bottom Ash that inexpensively and effectively removes arsenic from drinking water (Gadgil et al. 2008; Patel et al. 2006). ARUBA (also referred to as "media" in this document) uses bottom ash as a substrate. Bottom ash is obtained as a finely powdered, waste material from coal-fired power plants, which are common in India. The ash is sterile because the coal has been fired at extremely high temperatures (close to 800 ^{o}C). To make ARUBA, particles of bottom ash are coated with a complex consisting of oxides, hydroxides, and/or oxyhydroxides of iron, using relatively inexpensive chemicals (ferrous sulfate and sodium hydroxide). The process is conducted at room-temperature and atmospheric pressure. Thus, ARUBA can be produced with relatively simple equipment at low cost. Removing arsenic from contaminated drinking water is simple. ARUBA is mixed into the water, where it reacts with and immobilizes arsenic by adsorption and/or co-precipitation. The resulting complex can be settled out of the water, and is safe enough for disposal in most U.S. municipal landfills. Importantly, given ARUBA's large surface area to volume ratio, little material is needed to remove a given amount of arsenic, meaning that ARUBA treatment produces less waste than most comparable technologies.

The cost of raw materials needed for ARUBA production is expected to be low– less than 0.5 cents ($0.005) per kg ARUBA. ARUBA handling, transport, storage, delivery, and margins for distribution and retailing are expected to add on the order of 10 cents ($0.10) per kg. This estimate is based on the assumption that these costs would be comparable to those associated with ground, iodized table salt in India. Costs associated with the centralized ARUBA manufacturing are still a subject of research.

ARUBA has proven effective at removing high levels of arsenic from contaminated drinking water in Bangladesh. Over the past two years the authors traveled to Bangladesh three times to conduct a number of experiments to test ARUBA's arsenic removal capacity with freshly-obtained, Bangladesh groundwater samples. Our findings have allowed us to develop possible protocols for full-scale rural arsenic treatment. We begin with a discussion of our research methods, and continue with a presentation of our key field results. We also present a discussion of tradeoffs between various treatment protocol modifications, and propose a plan for technology implementation and scalability. We end with a discussion of the policy implications of our work.

Methods

Making ARUBA

To make 100 g of ARUBA, 156.75 g of hydrated $FeSO_4$ is added to 600 ml of de-ionized water and stirred for 5 minutes. One hundred grams of bottom ash (obtained from a coal fired power plant in Eklahare, Nasik, Maharashtra, India) is added to the $FeSO_4$ solution and stirred for one hour. After 15 minutes of settling, the solution is decanted. Next, 100 ml of 0.5 M of NaOH solution is added and stirred for five minutes. Again the solution is left to settle for 15 minutes and then decanted. The remaining mixture is spread evenly onto a large Pyrex dish and set in a fume hood to air-dry overnight, allowing for oxidation of the ferric coating. The following day, the media is scraped into a beaker using a metal spatula and rinsed three consecutive times with ~500 ml of deionized water, decanting between each rinse. This process lowers its pH. The media is again spread onto Pyrex dish and dried overnight in a fume hood. On the third day the media is scraped and stored. In the past we made ARUBA in 100 g batches, though we are now able to make 1 kg per day through a direct scale-up of the 100 g protocol.

Testing ARUBA in the Laboratory

Arsenic removal capacity of a given batch of ARUBA is tested in the laboratory by adding 0.50 g of ARUBA to 250 ml of 2ppm As(V) spiked de-ionized water and stirring, using a magnetic stir plate, for 1 hour. The solution is then filtered through Whatman Grade Number 40 Quantitative filter paper (particle retention of 8 μm) with a vacuum pump. The filtered water is sampled and tested for total arsenic by Inductively Coupled Plasma- Mass Spectrometry (ICP-MS) at Curtis & Tompkins Ltd. (Berkeley, CA) a commercial laboratory certified with the US EPA.

Testing ARUBA in the Field

In Bangladesh, arsenic removal capacity is measured using a different protocol because logistical difficulties prevent the use of the laboratory equipment in the field. In addition, 1.0 g of ARUBA (instead of 0.50 g) is used to treat 250 ml of water because ARUBA's arsenic removal capacity has been shown to be lower when treating Bangladesh groundwater than arsenic-spiked de-ionized water. Reasons for this result are presented in the next section.

In order to test arsenic removal capacity in the field, we added 1.0 g of ARUBA to 250ml of arsenic contaminated groundwater, collected in a 250ml bottle at the tubewell. The bottle was shaken vigorously for 30 seconds and then set down. Every 30 seconds for a total of half an hour the bottle was flipped to prevent the ARUBA from settling. After 30 minutes the solution was filtered through Whatman Grade Number 1 filter paper (particle retention of 11 μm) using a plastic funnel positioned over a clean 250 ml bottle. The filtered water was sampled and tested for total arsenic. Two high precision arsenic concentration measurement methods were employed to test field samples (pre- and post- treatment) during the course of our research—ICP-MS in Berkeley (standard error +/- 10%) and Atomic Adsorption Spectroscopy (AAS) in Dhaka (standard error +/- 10%)[1]. We have indicated in the results section which data was obtained from which measuring technique. All samples were also tested onsite for total arsenic concentration with an arsenic testing field kit (Arsenic Quick test by Industrial Test Systems, SC) but results were generally inaccurate and are not presented here.

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[1] The error bars used in each figure in this document represent measurement error only. The error bars do not attempt to capture experimental errors. Quantifying experimental error is still a subject of our research.

Note that further modifications were made to these protocols to achieve various experimental results presented below. These modifications have been described in detail along with the associated results.

Results

Arsenic Removal

Laboratory-based arsenic removal capacity tests on eleven batches of ARUBA have shown that the media is able to lower As(V) concentrations in arsenic-spiked deionized water from 2000 ppb arsenic to below the Bangladesh arsenic standard of 50 ppb.[2] Field results have proven ARUBA to be effective at reducing high levels of arsenic (200 to 900 ppb) in drinking water to below the Bangladesh standard in 12 tubewells located in four *upazila*s (sub-districts) of Bangladesh (Jhikargachha in Jessore District, Abhaynagar in Jessore District, Sonargaon in Narayanganj District, and Sreenagar in Munshigonj District). Illustrative results from our first field visit in March 2007 are presented in Figure 1 and in Gadgil et al. 2008.

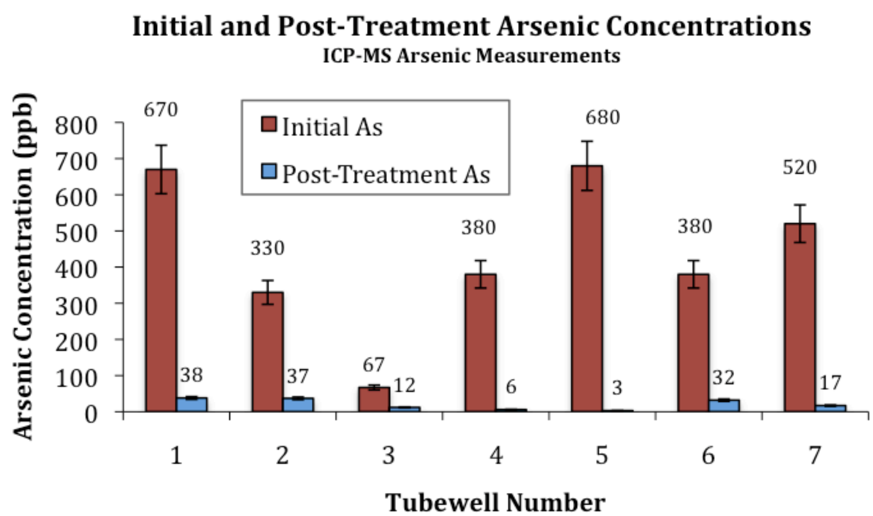

Figure 1. *Initial and Post-Treatment Arsenic Concentrations. ARUBA is effective at reducing high levels of arsenic in Bangladesh groundwater to below the Bangladesh standard of 50 ppb. In our initial field tests, one to four grams of ARUBA was used to treat 250 ml of water. (Jhikargaccha and Abhaynagar, March 2007)*

A comparison of field tests to lab test shows that more ARUBA is required to remove arsenic in Bangladesh groundwater than arsenic-spiked de-ionized water. In fact, using the arsenic treatment field protocol described above, we estimate that the ARUBA's arsenic removal capacity (measured in mg arsenic removed per gram of media used) decreases approximately ten-fold, from 0.96 mg As / g ARUBA (treating 2000 ppb arsenic-spiked de-ionized water in the laboratory) to 0.03-0.16 mg As / g ARUBA (treating 200 - 900 ppb Bangladesh groundwater in the field). The most likely reasons for diminished arsenic removal capacity in the field include the following:

(1) Arsenic removal capacity is dependant on the initial arsenic concentration. A high initial arsenic concentration leads to a high arsenic removal capacity, as defined above.

[2] The World Health Organization arsenic standard is 10 ppb. In both the laboratory and the field we have shown that it is possible to achieve post-treatment arsenic concentrations below 10 ppb by increasing the concentration of ARUBA used in treatment.

Concentrations of arsenic in Bangladesh groundwater are generally lower than the arsenic concentration tested in the laboratory (2000 ppb).

(2) Bangladesh groundwater has high concentrations of As(III), which ARUBA is less effective at removing (Patel et al. 2006). As(III) was not added to the arsenic-spiked de-ionized water made in the laboratory.
(3) Groundwater contains competing ions such as phosphate and silicate that bind to ARUBA's adsorption sites. Other researchers have quantified the effects of competing ions on iron hydroxide adsorbents (Manning and Goldberg 1996; Meng et al. 2002; Roberts et al. 2004; Su and Puls 2003; Tyrovola et al. 2006).

Using ARUBA's average arsenic removal capacity at an initial arsenic concentration of 400 ppb (0.08 mg As / g ARUBA), projected raw material costs remain minimal: 8 U.S. cents per person per year, assuming a person uses 10 liters of drinking-quality water per day. However, the coal ash required to treat that person's water would be approximately 50 grams per day. Reducing the amount of media required is essential for reducing costs (e.g., transport) and waste. Importantly, the amount of waste produced is directly tied to the cost of media since Bangladesh has very strict laws regarding the disposal of arsenic removal waste (discussed below).

Improving Arsenic Removal
A number of experiments were carried out to characterize ARUBA's interaction with Bangladesh groundwater and determine if modifications to the treatment protocol could increase ARUBA's arsenic removal capacity, thereby decreasing costs and wastes. These are discussed below.

Initially, ARUBA's kinetics were characterized by modifying the field treatment protocol. Water was treated for 60 minutes instead of 30 minutes and samples were filtered and stored at 5 minute intervals. Results for one tubewell in Neel Kanda Village, Sonargaon Upazila, Narayanganj District (July 2007) are shown in Figure 2. More than half of the arsenic is removed in the first five minutes of treatment. After 1 hour, the arsenic concentration is still decreasing, indicating that treatment is not yet complete. Arsenic removal kinetics results for water samples from other tubewells were similar.

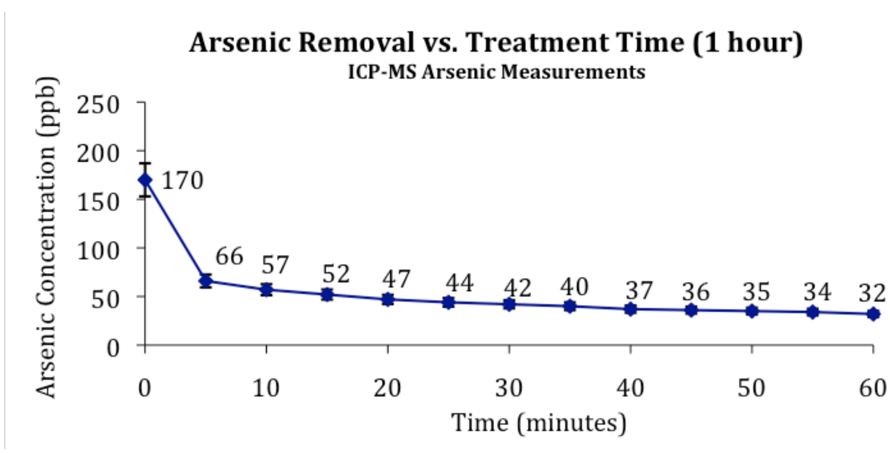

Figure 2. *Arsenic Removal vs. Treatment Time (1 hour). ARUBA removes more than half of the arsenic from Bangladesh groundwater after five minutes of treatment. Treatment does not seem to be completed after one hour. (Sonargaon, July 2007)*

Since ARUBA kinetic curves measured in July 2007 did not seem to reach a steady state, the experiment was repeated in June 2008, with several modifications to the protocol. Samples were taken every 15 minutes for the first hour of treatment (flipping the bottle every 30 seconds, as in the original protocol). After that hour, flipping was stopped and ARUBA was allowed to settle to the bottom of the container. At certain intervals, over the course of 1 week, samples were filtered and stored to determine if the settled ARUBA would continue to remove arsenic. Results from one tubewell in Besgao Village, Sreenagar Upazila, Munshigonj District (June 2008) are plotted in Figure 3. After one hour of treatment the arsenic concentration levels out; however, after 6 hours it begins to decrease again at a different rate. After 36 hours the minimum is reached. Similar results are seen for other tubewells tested. This can be explained by the hypothesis that ARUBA removes most of the existing As(V) in the sample very quickly, and only later begins to remove the As(III) as it slowly oxidizes to As(V). Therefore, leaving ARUBA in contact with the water in an uncapped treatment container improves its effective arsenic removal capacity.

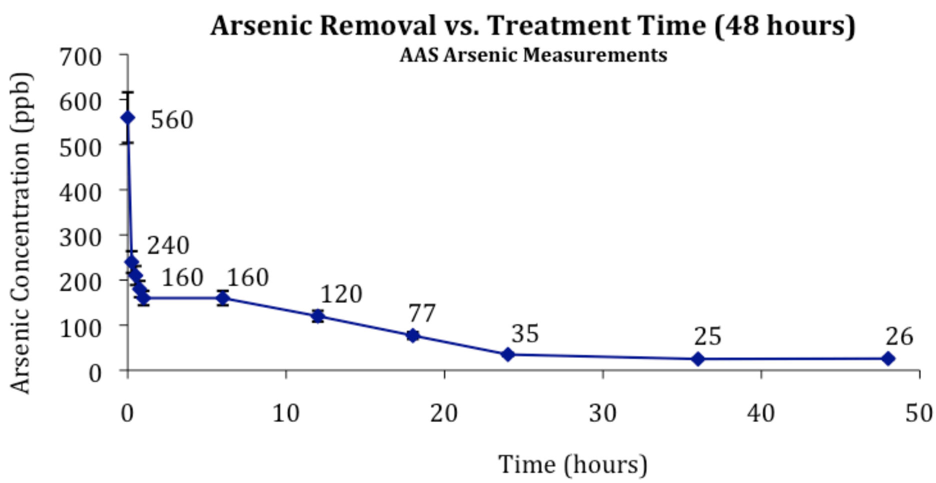

Figure 3. *Arsenic Removal vs. Treatment Time (48 hours). Continuing ARUBA treatment beyond 1 hour shows that the arsenic concentration temporarily plateaus between one and six hours. After six hours, the concentration diminishes further. We suspect this corresponds to the natural conversion of As(III) to As(V), which ARUBA is better able to remove. (Sreenagar, June 2008)*

Another method to increase the arsenic removal capacity of the media is to add ARUBA in sequential fractionated doses. This method is known to increase the arsenic removal capacity of iron-based arsenic adsorbents formed through the chemical addition of ferric or ferrous salts (Roberts et al. 2004). Water from two tubewells in Neel Kanda Village, Sonargaon Upazila, Narayanganj District (July 2007) was treated with ARUBA using the standard field protocol, and compared to water treated with various fractionated dosing schemes (Figure 4).

For each treatment with fractionated doses (except the last), ARUBA was filtered out of the water after the 30 minute flipping period, before the next fractionated dose was added and another 30 minute flipping period began. In the last case (marked with *), no filtration was performed between addition of sequential doses of ARUBA. Instead, the second fractionated dose was added directly to the treatment bottle containing the first fractionated dose. Despite this fact, the final arsenic concentration is still lower than when 1 g is added all at once.

These results may be due in part to the increased total contact time with the media. However, we also suspect that the initial dose of ARUBA removes ions that compete with arsenic for adsorption sites (e.g., phosphate and silicate) allowing subsequent ARUBA doses to remove more arsenic. Interestingly, an increase in arsenic removal capacity resulting from adding ARUBA in sequential fractionated doses was also seen in the laboratory, using de-ionized water spiked with As(V).

Water treated with three fractionated doses of ARUBA performed best indicating that increasing the number of doses increases ARUBA's arsenic removal efficiency. The choice of fractionation (e.g., ½ g + ½ g versus ¾ g + ¼ g of ARUBA) does not seem to have a significant affect on the final arsenic level.

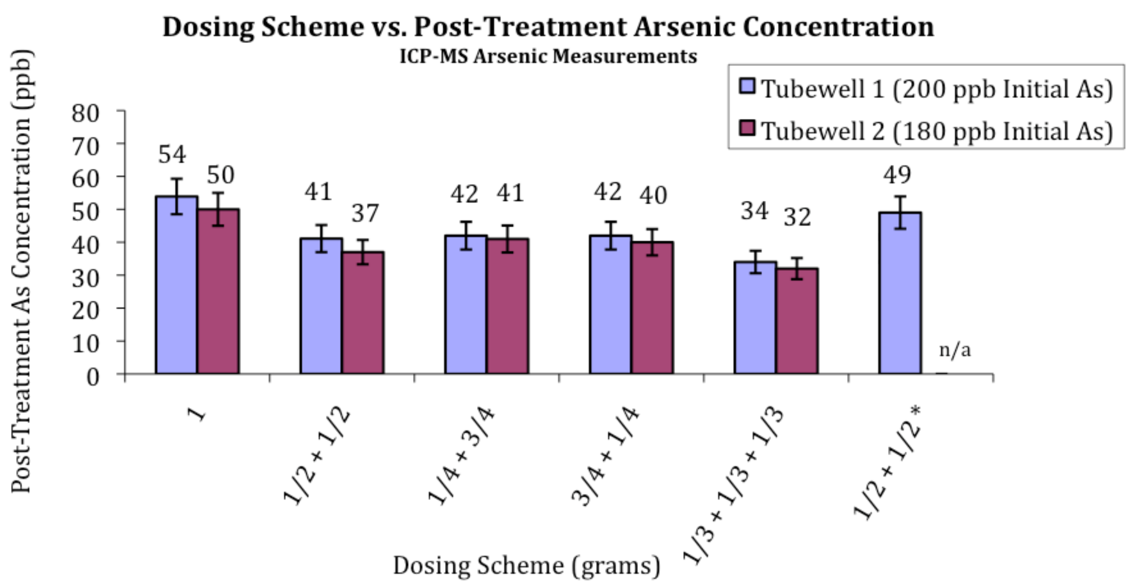

Figure 4. *Dosing Scheme vs. Post-Treatment Arsenic Concentration. One gram of ARUBA added in fractionated doses removes more arsenic than when added all as one dose. While the fractionation scheme (for a given number of doses) does not seem to matter, the number of fractionated doses does. Three doses are better than two doses, which are better than adding all of the ARUBA in a single dose. Not filtering (*) between two doses is better than a single dose, but worse than filtering the two doses. (Sonargaon, July 2007)*

We also tested the effect of water storage on arsenic removal capacity. Several liters of water were pumped from tubewells in Besgao Village, Sreenagar Upazila, Munshigonj District (June 2008) and stored uncapped for one week. At predetermined intervals, samples of the water were (1) tested for As(III) concentration, (2) filtered and tested for total arsenic concentration, and (3) treated with ARUBA using the standard field protocol and tested for total arsenic concentration.

As(III) concentrations were measured by passing 10 ml water samples through an arsenic speciation cartridge (manufactured by MetalSoft Center, NJ) and testing the effluent for total arsenic concentration. The speciation cartridge traps As(V) and also acts as a filter, trapping arsenic, including As(III), bound to larger particles. Therefore, we obtained only a lower bound for As(III).

The purpose of filtering the samples was to determine how much arsenic is settled through co-precipitation with naturally-occurring iron in the water sample. Filtration removed precipitates, including those to which arsenic binds, leaving us to measure total dissolved arsenic.

The data presented in Figure 5 shows results of the arsenic concentration measurements described above. Similar results are seen for three other tubewells. The concentration of As(III) decreases overtime as it oxidizes to As(V), increasing the performance of ARUBA. In fact, after storing water uncapped for 3 days, post-treatment arsenic concentrations are ten times lower than those of freshly pumped water. Note that natural co-precipitation followed by filtering removes some arsenic; however, this constitutes a small percentage of the total arsenic removed by ARUBA treatment.

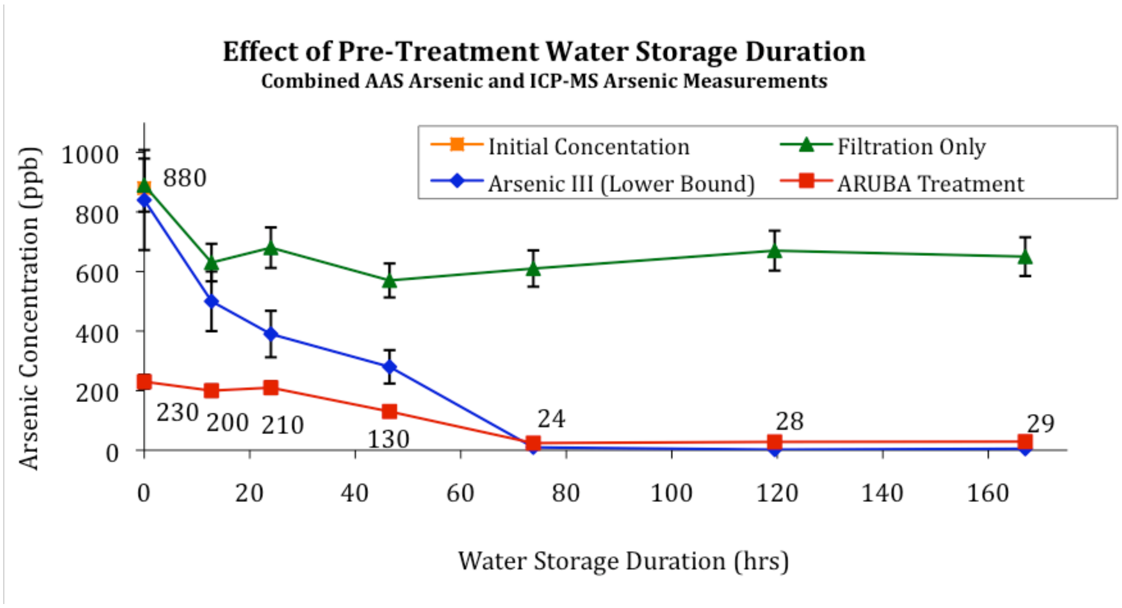

Figure 5. *Effect of Pre-Treatment Water Storage Duration. Arsenic removal capacity increases as pre-treatment water storage duration increases, up to 72 hours. While natural co-precipitation followed by filtering does remove some arsenic, concentrations remain well above the acceptable limit (50 ppb). ARUBA's ability to remove arsenic is approximately proportional to the water's remaining As(III) concentration. Note that we have used a combination of AAS (Atomic Adsorption Spectroscopy) data and ICP-MS (Inductively Coupled Plasma – Mass Spectrometry) data to produce this graph[3] (Sreenagar, June 2008).*

Tradeoffs: Arsenic Removal Capacity, Waste, and Costs

Our results imply a number of tradeoffs that must be made when designing an arsenic removal system using ARUBA. Storing water before treatment, increasing water treatment time, and/or using fractionated doses drives up treatment costs, while reducing the amount of media needed (though, we expect media costs will not dominate the total system costs). In turn, this decreases the amount of waste produced by the treatment system. It is likely that other adsorption-based arsenic removal media would exhibit properties and tradeoffs similar to those of ARUBA.

Despite the fact that spent ARUBA meets the U.S. EPA requirements for disposal in US municipal landfills, the disposal costs in Bangladesh could be substantial. We learned that Bangladesh policies require that spent arsenic removal media must be buried in concrete pits lined with thick plastic-membranes located far away from human habitation. Therefore, reducing waste is not only desirable from an environmental standpoint, but it is essential for lowering the cost of waste transport and containment. We do not currently have cost estimates for waste transport and disposal.

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[3] AAS data for Water Storage Duration = 0 hours did not match ICP-MS data. We expect this discrepancy arose because of the presence of a volatile chemical in the groundwater samples that affected initial AAS measurements. Storage containers were left uncapped and so the volatile chemical was able to escape. We noticed bubbles forming in the water samples and capped containers storing water from the same tubewell built up pressure over time. AAS and ICP-MS data corresponded well for Water Storage Duration ≥ 12 hours. Therefore, ICP-MS data has been used for Water Storage Duration = 0 hrs and 12 hrs, and AAS data has been used for the remaining Water Storage Durations.

Implementing just one treatment protocol modification—storing water for three days before treatment—decreases the amount of spent media per person per day from 50 grams to 41 grams (assuming treatment of 10 liters of water initially containing approximately 400 ppb arsenic). Over the course of the year this constitutes significant material and cost savings.

Scale, Technology Implementation, and Policy Implications

Our technical findings, in conjunction with field observations and informal discussions in Bangladeshi villages indicate that community-scale arsenic removal systems would be more acceptable to villagers than household-based technologies. Other studies have noted significant preference for community-based systems over household-based systems (Ahmad et al. 2003).

Community-scale water treatment provides a number of advantages over arsenic filters and other household treatment systems. Several studies have found that for villagers the most important attributes of a water treatment system are that it is low-cost and convenient to use (Ahmad et al. 2003; Caldwell et al. 2003). Many household filters have failed to gain acceptance because of maintenance and attention required (Ahmad et al. 2003). Importantly, community-scale treatment increases the feasibility of local water management, in addition to the effectiveness of local and national water policies. Moreover, water quality can be monitored and guaranteed, something that has been lacking in the ad-hoc development of rural clean water provision systems in Bangladesh (Ahmad et al. 2003).

Furthermore, at a large scale it is much easier to implement a number of the treatment protocol modifications described above. Water storage and increased water treatment time would only add minimal costs to a large-scale water treatment system; however, these modifications may not be feasible at the household scale. (While fractionated dosing would be possible in community-scale water treatment facilities as well, it would likely not provide enough material or cost savings to be worth the added infrastructure cost.) In addition, treating water in central locations simplifies waste management. The same vehicle that delivers ARUBA to a community treatment center could also take the spent media away for burial or reprocessing, if that proves feasible and affordable in the future.

Based on our observations and discussions with many leaders in Bangladesh's water sector, it seems as though the country's strict waste management policies have hindered the development and deployment of arsenic remediation systems, as all such systems produce some waste. Instead, many organizations promote switching to arsenic-free water sources (such as surface water or arsenic-free deep tubewell water) instead of removing arsenic from contaminated groundwater, in order to avoid the waste issue completely. While this may be appropriate in some parts of Bangladesh where arsenic-free, clean drinking water is easily available, many rural areas have no access to alternative sources and, without arsenic remediation, villagers are left to drink contaminated groundwater.

One possible implementation model is a public-private partnership based on that of WaterHealth International (www.waterhealth.com), which provides clean drinking water to more than a million people in rural villages in India through publicly-owned, privately-managed village-scale water treatment centers. Though a three-way partnership between a local financial institution, a local NGO, and a company responsible for constructing and maintaining the water treatment centers (all working together with the local village governments), community-scale arsenic-removal plants could be constructed in rural Bangladeshi villages. Users would pay a small fee for the water that they collect from the treatment center, but due to the low-cost of ARUBA the fee would remain affordable to those living on less than $2 a day *and* would be enough to cover both the capital and operating costs of the treatment center. For the example of WaterHealth, the water treatment centers are completely paid off after 8 years. Ahmad et al. (2003) has found that Bangladeshis are willing to pay around $9 per household per year for clean drinking water.

Conclusions

The key findings of our fieldwork experiments in Bangladesh to test and improve the arsenic removal capacity of ARUBA are highly encouraging. Modest changes to the treatment protocol such as water storage before treatment, increasing the treatment duration, and adding the media in fractionated doses, increase ARUBA's efficacy. Reducing the amount of media required for treatment decreases costs, especially the cost of waste transport and storage. Given Bangladesh arsenic removal waste management policies, reducing the cost of a water treatment system using ARUBA, which in turn reduces the cost of clean water, requires analysis of various tradeoffs. We propose that an affordable, technically effective, and financially viable solution would be to use ARUBA in public-private community-scale water treatment systems in rural Bangladesh.

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Acknowledgements

This work was conducted at the University of California, Berkeley and Lawrence Berkeley National Laboratory under U.S. Department of Energy Contract No. DE-AC02-05CH11231. Funders include the National Collegiate Inventors and Innovators Alliance (NCIIA) and the University of California, Berkeley’s Richard C. Blum Center for Developing Economies, Bears Breaking Boundaries Contest, Sustainable Products and Solutions (SPS) Program, and Big Ideas Marketplace. Past funders include the Lawrence Berkeley National Laboratory’s Technology Transfer Office, the Blue Planet Run Foundation, the California Energy Commission (CEC), and the American Water Works Association Research Foundation (AwwaRF). Thanks to the many researchers and students who built the foundation of this project: Yola Bayram, Ebere Chukwueke, Raymond Dod, Meghana Gadgil, Christie Galitsky, Lara Gundel, Mathew Jeung, Melissa Quemada, Heena Patel, Clete Reader, Mark Sippola, and Duo Wang. Also thank you to the rest of the Berkeley Arsenic Alleviation Group (BAAG): William Babbitt, Kosar Jahani, Tasnuva Khan, Suzanne Lippert, Nadia Madden, Shefah Qazi, and Mehmet Seflek. Iqbal and Kamal Quadir provided us funds and support for our first trip to Bangladesh and the Khan family greatly supported our second trip. Thanks to BRAC and Bangladesh University of Engineering and Technology (BUET), Dhaka. Finally, we would like to express extreme thanks to our Bangladeshi collaborators Dr. A.B.M. Badruzzaman and Mahbuba Iasmin Ahmed at BUET.