Development Engineering 3 (2018) 175–187

Contents lists available at ScienceDirect

Development Engineering

journal homepage: www.elsevier.com/locate/deveng

Addressing technical barriers for reliable, safe removal of fluoride from drinking water using minimally processed bauxite ores

Heather L. Buckley[a,b,*], Nusrat J. Molla[c], Katya Cherukumilli[c,d], Kathryn S. Boden[c], Ashok J. Gadgil[b,c]

[a] *Department of Civil Engineering, University of Victoria, Victoria, V8P 5C2 BC, Canada*
[b] *Energy Technology Area, Lawrence Berkeley National Laboratory, Berkeley, 94720 CA, USA*
[c] *Civil and Environmental Engineering, University of California Berkeley, Berkeley, 94720 CA, USA*
[d] *Department of Civil and Environmental Engineering, University of Washington, Seattle, 98195 WA, USA*

A B S T R A C T

Throughout the developing world, over 200 million people drink groundwater containing fluoride concentrations surpassing the World Health Organization's maximum recommended contaminant level (WHO-MCL) of 1.5 mg F^{-}/L, resulting in adverse health effects ranging from mottled tooth enamel to debilitating skeletal fluorosis.

Existing technologies to remove fluoride from water, such as reverse osmosis and filtration with activated alumina, are expensive and are not accessible for low-income communities. Our group and others have demonstrated that minimally-processed bauxite ores can remove fluoride to safe levels at a fraction of the cost of activated alumina. We report results from testing for some technical challenges that may arise in field deployment of this technology at large scale, particularly in a sufficiently robust manner for application in development contexts. Anticipating possible modes of failure and addressing these challenges in advance in the laboratory is particularly important for technologies for vulnerable communities where the opportunity to relaunch pilot projects is limited and small failures can keep solutions from the people that need them most.

This work addresses three potential technical barriers to reliable removal of fluoride from drinking water with bauxite ore from Visakhapatnam, Andhra Pradesh, India. We evaluate competition from co-occurring ions, adsorption reversibility, and potability of the product water with regards to leaching of undesirable ions during treatment with various adsorbent materials including raw and thermally activated bauxite, and synthetic gibbsite (a simple model system). Under the conditions tested, the presence of phosphate significantly impacts fluoride adsorption capacity on all adsorbents. Sulfate impacts fluoride adsorption on gibbsite, but not on either bauxite adsorbent. Nitrate and silicate (as silicic acid), tested only with gibbsite, do not affect fluoride adsorption capacity. Both thermally activated bauxite and gibbsite show non-reversible adsorption of fluoride at a pH of 6. Raw bauxite leached arsenic and manganese in a TCLP leaching test at levels indicating the need for ongoing monitoring of treated water, but not precluding safe deployment of bauxite as a fluoride remediation technology. Understanding these phenomena is crucial to ensure field deployment over large diverse geographical areas with aquifers varying in groundwater composition, and for ensuring that the appropriate engineering processes are designed for field implementation of this innovation.

1. Introduction

Throughout the developing world, over 200 million people drink groundwater containing fluoride concentrations (Edmunds and Smedley, 2013) that exceed the World Health Organization's maximum recommended contaminant level (WHO-MCL) of 1.5 mg F^{-}/L. (World Health Organization, 2006; World Health Organization, 2004b) In India alone, over 66 million people risk developing fluorosis due to natural contamination of their drinking water (UNICEF, 1999). In China, where other aspects of quality of life are rapidly improving, as much as 10% of the groundwater-based drinking water supply may contain dangerous levels of naturally occurring fluoride (Wu et al., 2011). The problem is widespread: dissolution of fluoride-rich granitic rocks in groundwater aquifers causes toxic levels of fluoride in arid regions of India, China,

* Corresponding author. Department of Civil Engineering, University of Victoria, Victoria, V8P 5C2 BC, Canada.
E-mail address: hbuckley@uvic.ca (H.L. Buckley).

https://doi.org/10.1016/j.deveng.2018.06.002
Received 30 November 2017; Received in revised form 2 June 2018; Accepted 4 June 2018
Available online 07 June 2018

the Middle East, the East African Rift Valley, central Argentina, and northern Mexico (Ozsvath, 2008; Jagtap et al., 2012), often in regions without reliable alternative water sources throughout much of the year. This exposes entire communities to devastating health effects, including anemia, reduced cognitive function and dental fluorosis (Brindha and Elango, 2011). At higher concentrations of fluoride in drinking water, skeletal fluorosis leads to irreversible spinal fusion and limb deformation in children, leaving victims severely disabled and often with chronic pain (Khairnar et al., 2015).

As with many public health challenges, poor, rural communities with limited access to healthcare are generally disproportionately affected by the lack of scalable solutions. Existing technologies to remove fluoride from water are both expensive and energy intensive from the perspective of low-income communities, and also have a significant greenhouse gas footprint (Chen and Graedel, 2012a). Many proposed alternative technologies have proven effective in the laboratory (Habuda-Stanic et al., 2014), but scaling these encounters challenges of reliability of water source (e.g., rainwater harvesting) (Mwenge Kahinda et al., 2007), availability of skilled labor for upkeep (e.g. Nalgonda technique) (Jagtap et al., 2012), cultural appropriateness (e.g. bone char in communities with dietary restrictions) (Osterwalder et al., 2014), and myriad challenges with cost, reliability of material sourcing, and wastefulness in water-stressed regions (reverse osmosis is widely used but recovers only about two thirds of the input water) (Mohapatra et al., 2009).

Despite some limitations, aluminum-based adsorbents offer an attractive approach to effective, selective fluoride removal due to the thermodynamic stability of the aluminum-fluoride bond (Haynes, 2009). Activated alumina is ubiquitous in utility-scale and household-scale water treatment in high income countries and middle class communities throughout the world, and effectively removes fluoride provided pH and co-occurring ions are appropriately managed (Dahi and Chiang MaiThailand, 2000; Choi and Chen, 1979; Farrah et al., 1987). However, the cost of activated alumina, which stems largely from the energy-intensive process of purifying (Chen and Graedel, 2012a; Patterson, 1967) and modifying raw bauxite ores at temperatures exceeding 1000 °C (Chen and Graedel, 2012b), makes the materials economically unattainable to low-income populations.

Previous work in our group (Cherukumilli et al., 2017a) and by others has demonstrated that raw bauxite ore from a variety of locations (including Iran (Malakootian et al., 2014), Ghana (Buamah et al., 2013), India (Das et al., 2005), Malawi (Kayira et al., 2014; Sajidu et al., 2008), and Turkey (Dilek et al., 2013)) can be used to remove fluoride from drinking water at significantly lower costs than activated alumina. Bauxite ore is composed largely of aluminum hydroxides, along with significant or trace quantities of iron-, silicon-, and sometimes calcium- and titanium-oxides. Our previous research shows that when pH is controlled between 5.5 and 6.5, pulverized bauxite ore used as a dispersive batch adsorbent can reliably bring fluoride levels to below the WHO-MCL (1.5 mg F^-/L) (Cherukumilli et al., 2017a). Bauxite that has been thermally activated at low temperatures (200–400 °C) has been further demonstrated to be a more effective fluoride adsorbent by Das and coworkers (Das et al., 2005), Peter (2009), and more recently by our group (Cherukumilli et al., 2018). Our work estimated that on a per-water-treated basis, raw bauxite costs roughly 23X less as a dispersive batch adsorbent for fluoride removal than activated alumina (Cherukumilli et al., 2017a), and that this cost is further reduced using thermally activated bauxite, accounting for increased bauxite treatment costs but reduced material transportation costs due to a lower required bauxite dose (Cherukumilli et al., 2018). Such a significant cost reduction clearly points to an opportunity to scale this research for the benefit of low-income communities.

One third of the globally reported cases of fluorosis occur in India (UNICEF, 1999), where the majority of states report regions with groundwater fluoride concentration in excess of the WHO MCL (Central Ground Water Board Ministry of Water Resources Government of India, 2010). The Nalgonda district in Telengana, India, where skeletal fluorosis is endemic, is relatively close (< 500 km distant) to Visakhapatnam in Andhra Pradesh, India, from where the bauxite used in this study is mined. Due to the geographically proximate abundant supply of bauxite, it is one example of an appropriate location to pilot a safe drinking water project in this region using locally sourced bauxite. The research presented in this work helps to fulfill the technical needs of such a pilot project. However, because technical readiness is only one component of successfully launching a technology, it is important to present this research in the broader context of technology implementation. To achieve successful technology integration through community partnerships and business practices, this project aims to follow models and lessons learned from the transition from laboratory to field pilot of electrochemical arsenic remediation (ECAR) for arsenic removal in Dhapdhapi, India (Amrose et al., 2014, 2015).

The model that our team followed when scaling ECAR from lab to field included four key steps. First, the team conducted social surveys to get an understanding of the community's risk-perception of arsenic and evaluate their interest in having a treatment facility installed (Das et al., 2016). In parallel, through cost-analyses, the team confirmed that the technology could produce healthy water at a locally affordable price, ensuring that it would be financially viable (Roy, 2008). Third, input from the community was gathered from open meetings and interviews and consultation with key community opinion leaders to ensure that ECAR design and operation would be culturally appropriate (Amrose et al., 2014; Delaire et al., 2017). Finally, after construction and commissioning, the plant was thoroughly tested for over a year before water was distributed to the community. This gave the team adequate time to confirm that the treated water fully met the standards for drinking water even under varying operating conditions, which included seasonal changes and occasional operator neglect. In conclusion in order for a technology to be useful, sustainable, and be considered as potentially scalable it must be (1) desired by the community, (2) affordable, (3) culturally appropriate, (4) technically effective, and (5) robust in the relevant operating environment.

The success-to-date of the ECAR model will guide the design of a pilot fluoride remediation plant, and this paper focuses on point 4–technical effectiveness. As we proceed, it is crucial to understand when bauxite can be safely and effectively used to treat drinking water, and if there are any major technical limitations. Indeed, this is the point at which many promising water treatment discoveries die at the laboratory bench; while initial scientific results are promising, the resources are often lacking to uncover latent problems and elucidate their resolution to create a technology. The “valley of death” faced in introducing a technology to market is often discussed as a business challenge; however, the barrier imposed by the details necessary to make the leap from test tube to pilot plant are equally daunting. In the development engineering context in particular, resource constraints often mean that once a technology enters an initial pilot stage, the technology implementers may get only “one shot” in terms of community perception and trust; the technology must succeed the first time it is unveiled, or lack of additional funding and loss-of-trust will not allow for a second attempt as it often possible for stable, large companies targeting high-income markets. Thus, exploring in a scientifically rigorous manner the technical constraints that may threaten the success of a new technology allows the operation of the first pilot to remain well inside the margins of failure, which is one crucial aspect of technology adoption.

Within the context of technical effectiveness, major parameters that can impact all water treatment technologies include pH, co-occurring ions (i.e. ions that are not themselves a health concern, but may interfere with removal of hazardous contaminants), and the inadvertent release of hazardous chemicals from the materials used in the water treatment process. An additional concern that is specific to adsorbent media for water treatment is the reversibility of binding of a target contaminant to the adsorbent. This is important both for the possible