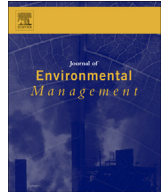




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Research article

Onsite defluoridation system for drinking water treatment using calcium carbonate

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ABSTRACT

Fluoride in drinking water has several effects on teeth and bones. At concentrations of 1–1.5 mg/L, fluoride can strengthen enamel, improving dental health, but at concentrations above 1.5 to 4 mg/L can cause dental fluorosis. At concentrations of 4–10 mg/L, skeletal fluorosis can occur. There are many areas of the world that have excessive fluoride in drinking water, such as China, India, Sri Lanka, and the Rift Valley countries in Africa. Treatment solutions are needed, especially in poor areas where drinking water treatment plants are not available. On-site or individual treatment alternatives can be attractive if constructed from common materials and if simple enough to be constructed and maintained by users. Advanced on-site methods, such as under sink reverse osmosis units, can remove fluoride but are too expensive for developing areas. This paper investigates calcium carbonate as a cost effective sorbent for an onsite defluoridation drinking water system. Batch and column experiments were performed to characterize F^- removal properties. Fluoride sorption was described by a Freundlich isotherm model, and it was found that the equilibrium time was approximately 3 h. Calcium carbonate was found to have comparable F^- removal abilities as the commercial ion exchange resins and possessed higher removal effectiveness compared to calcium containing eggshells and seashells. It was also found that the anion Cl^- did not compete with F^- at typical drinking water concentrations, having little impact on the effectiveness of the treatment system. A fluoride removal system is proposed that can be used at home and can be maintained by users. Through this work, we can be a step closer to bringing safe drinking water to those that do not have access to it.

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1. Introduction

In limited quantities, fluoride is beneficial and essential to the mineralization of bones and strengthening of dental enamel, which is why it is added into US drinking water supplies (Adler et al., 1970). The safe limit of fluoride in drinking water is 1.0 mg/L in the U.S. and the recommended dose varies by location and climate. The WHO guideline is 1.5 mg/L. However, at concentrations from 1.5 to 4 mg/L, fluoride in drinking water can cause dental fluorosis. At concentrations of 4–10 mg/L, skeletal fluorosis can occur (WHO, 2004). Excessive fluoride in drinking water is a detrimental problem to society, causing detrimental effects in 35 nations across the world and putting 200 million people in the world at risk of fluorosis, both skeletal and dental (Jha et al., 2013; Gupta and Ayoob, 2016). Observable symptoms of excessive fluoride include stained

teeth, bone diseases, tooth decay, stooped backs, and crooked hands and legs. Fluoride can also lead to non-skeletal fluorosis, such as harmful effects to erythrocytes, ligaments, spermatozoa, thyroid glands and destruction of filaments in the muscle tissues leading to muscle weakness. The gastrointestinal system is also adversely affected by high fluoride ingestion causing gastric irritation such as nausea, vomiting and gastric pain (Spak et al., 1989). These detrimental side effects cause excessive fluoride to be a pollutant of concern.

Fluoride occurs naturally in all waters. Excessive fluoride can also be found in large geographical belts in the mountains that have sediments of marine origins and in geographical belts that have volcanic activity, such as the mountainous regions from Iraq and Iran to Syria and Turkey to Algeria and Morocco and along the East African Rift from Eritrea to Malawi. Other examples include southern parts of the USA, Europe, and USSR (Fawell et al., 2006; Gupta and Ayoob, 2016; Manji and Kapila, 1986; Nair et al., 1984).

Groundwater can also be contaminated with fluoride when it

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comes into contact with rocks and soils that naturally contain fluoride. Locations that have contaminated groundwater include Southern and Western Africa, China, Thailand, Japan, Argentina, Persian Gulf states, Saudi Arabia, Europe, USA, Canada the Middle East, especially Pakistan, and southern Asia, but especially India and Sri Lanka (WHO, 2005) (Susheela, 1995). There have been fluoride concentrations found in the United States' groundwater that have caused dental fluorosis since the 1930's (Segreto et al., 1984; Dean, 1993). In the United States, it has been found that groundwater aquifers in five desert regions of southern California had high concentrations of fluoride, mainly the Coachella Valley (22%), the Colorado River basin (20%), the Mojave River area (10%), Owens Valley (3%), and the Antelope Valley (3%) (Dawson and Belitz, 2012). Groundwater can also be contaminated by fluoride through several anthropogenic industrial processes: cement and brick manufacturing, "coal fired power stations", electronics manufacturing, aluminum smelting and refining, beryllium abstraction plants, iron smelting and producing plants, etc. (Ramanathan, 2004; Nath and Dutta, 2015; Turner et al., 2005).

There are several current defluoridation methods. Defluoridation methods can be categorized into four main groups: 1) coagulation, 2) adsorption, 3) electrochemical methods, and 4) membrane processes. Coagulation processes involve using "chemical reagents such as lime, calcium, magnesium salts, poly aluminum chloride and alum" to form a precipitant with fluoride (Gupta and Ayoob, 2016). Adsorption involves using sorption media that is often packed in columns. Fluoride containing water is cycled through the columns, and the media can be regenerated, renewed or disposed. Some adsorption materials for defluoridation include: activated alumina, apophyllite, bauxite, bentonite, brushite, calcite, acidic clay, kaolinite clay, china clay, charfine and nirmali seeds, chitosan, clinoptilolite, "diatomaceous earth", "Fuller's earth", graphene, halloysite, hydroxyapatite, laterite, lignite, acid treated limestone powder, kaolinite, gibbsite, goethite, gypsum, magnesite, natrolite, "rare earth oxides", pumice stone, quartz, serpentine, aiken soil, alkaline soil, "Ando soil", stilbite, synthetic resins, vermiculite, and zeolite (Bower and Hatcher, 1967; Singano et al., 1997; Bhatnagar et al., 2011; Fawell et al., 2006; Fan et al., 2003; Turner et al., 2005; Murutu et al., 2012; Thole et al., 2012; Mourabet et al., 2011; Maiti et al., 2011; Asgari et al., 2012; Dutta et al., 2016). With adsorbents, higher removal is achieved with decreasing particle size of the adsorbent and increasing dosage (Srimurali et al., 1998), which we observed in our results. Electrochemical processes and membrane can be efficient but require power and expertise generally not available for on-site treatment in rural areas; hence, coagulation and adsorption processes are preferred. A detailed comparison is beyond our scope, but Nath and Dutta (2015) provide an extensive comparison.

In many areas of the world, treatment solutions are needed, especially in poor areas where drinking water treatment plants are not available. On-site or individual treatment alternatives can be attractive if constructed from common materials and if simple enough to be constructed and maintained by users with minimal training. A promising adsorbent is CaCO_3 . Calcium carbonate is a common chemical that can be found in rocks, such as dolomite, limestone and marble, and seashells, pearls and eggshells. 3.6% of the Earth's crust naturally contains calcium most of which is calcium carbonate (Lutgens and Tarbuck, 2000; Nath and Dutta, 2015). CaCO_3 has been shown to defluoridate water through precipitation and adsorption. Broeck et al. (2003) determined that using a column filled with granular CaCO_3 could be used as a post treatment for wastewater to remove fluoride from 8 mg/L down to 0.6 mg/L. Turner et al. (2005) showed that when F^- comes into contact with calcium carbonate, an instant F^- adsorption occurs and calcium

carbonate dissolves. Dissolution of calcium carbonate increases calcium concentration until saturation is reached and CaF_2 precipitation occurs. For our experiment, we believe the mechanism of F^- removal is ion exchange adsorption and is reversible. Precipitation of CaF_2 occurs only at higher concentrations, 10–20 mg/L F^- or more (Gupta and Ayoob, 2016; Nath and Dutta, 2015).

By comparing different adsorbents, such as commercial resins, seashells, eggshells and calcium pills, we have found that calcium carbonate has comparable fluoride removal abilities to the commercial resins that have been optimized for fluoride removal. Though there have been experiments performed already by other researchers on calcium containing materials (CaCl_2 , $\text{Ca}(\text{OH})_2$, $\text{Ca}(\text{NO}_3)_2$, $\text{Ca}_5(\text{PO}_4)_3$, CaO , CaCO_3), our objective is to develop a simple defluoridation system for potable water that requires no special operational expertise or the use of hazardous chemicals, and can be operated by the local people (Bhaumik et al., 2012; Nath and Dutta, 2015; Jimenez Reyes and Rios, 2010; Yang et al., 1999; Ben Nasr et al., 2011; Bhargava and Killedar, 1991; Christoffersen et al., 1984).

2. Materials and methods

2.1. Materials

Fluoride solutions were obtained by diluting a 1000 mg/L NaF ACS reagent grade standard solution from Ricca Chemical Company, USA. Powdered ACS reagent grade calcium carbonate was purchased from Thermo Fisher Scientific, USA and Sigma Aldrich, USA. Samples were measured in 50 mL plastic Falcon tubes that were obtained from Thermo Fisher Scientific, USA. SIR 900 and SBG2 synthetic resins were obtained from Resintech, USA, and Amberlite IRA 400 synthetic resins were obtained from Rohm and Haas, USA. Eggshells, calcium pills, and seashells were obtained from local stores. Eggshells and seashells were air dried and then ground to a powder using a blender. The approximate particle size of the calcium carbonates were 100 μm . DI water was used for all the experiments.

2.2. Instrumental analysis

Fluoride concentrations were determined using a Thermo Scientific Orion Versa Star Advanced Electrochemistry Meter and a Thermo Scientific Orion 9609 BNWP Ion Plus Sure-Flow Fluoride ion selective electrode. TISAB II from Thermo Scientific was used to maintain high, constant ionic strength, adjust the pH, and complex interfering species. Lab-line Instruments Inc. Environ-Shaker 3597 was used to shake the samples.

2.3. Methods of batch and isotherm experiments

Isotherm experimental conditions were chosen to match conditions needed to produce safe drinking water. We compared isotherms between different adsorbents: Resintech SIR 900, Resintech SBG2 Rohm and Haas Amberlite IRA 400, eggshell powder, seashell powder, calcium pill powder, and Sigma Aldrich calcium carbonate. We placed varying amounts of the adsorbents into 50 mL Falcon tubes and added 20 mL of 10 mg/L fluoride solution. We then shook the samples for 1 h at 100 rpm and measured the fluoride concentrations using the Thermo Scientific Electrochemistry Meter and Fluoride ion selective electrode. The amount of fluoride adsorbed by the adsorbents (x/m) was calculated and graphed with the corresponding concentration of fluoride. x/m (mg/g) values were calculated using the equation below:

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