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Design and development of sustainable remediation process for mitigation of fluoride contamination in ground water and field application for domestic use

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HIGHLIGHTS

- ChemoDefluoridation Unit (CDU) was designed for fluoride mitigation and evaluated in community of seventy five households.
- CDU has a maximum efficacy at initial fluoride concentration of 6 mg/L; however, it can be effective with maximum initial fluoride concentration of 14 mg/L without any affecting drinking water quality.
- The operational cost of CDU is estimated to be $0.002 per litre.
- Successful demonstration of CDU at a community level, its acceptability by the community and improvement in the health indicated by user satisfaction, makes this technology a sustainable option for fluoride mitigation in a marginalized community with limited means.

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ABSTRACT

Decentralised household chemo defluoridation unit (CDU) was developed and designed based on a combination of coagulation and sorption processes. Chemo-defluoridation process was optimised to reduce use of chemicals and increase acceptability among beneficiaries without affecting palatability of water. Chemical dose optimization undertaken in the laboratory using jar test revealed the optimum calcium salt to initial fluoride ratio of 60 for fluoride removal. Performance of CDU was evaluated in the laboratory for removal efficiency, water quality parameters, filter bed cleaning cycle and desorption of fluoride. CDU evaluation in the laboratory with spiked water (5 mg/L) and field water (~4.2 mg/L) revealed treated water fluoride concentration of less than 1 mg/L. Seventy five CDUs were installed in households at Sakhara Village, Yavatmal District in Maharashtra State of India. Monthly monitoring of CDUs for one year indicated reduction of the raw water fluoride concentration from around 4 mg/L to less than 1 mg/L. Post implementation survey after regular consumption of treated drinking water by the users for one year indicated user satisfaction and technological sustainability.

1. Introduction

According to estimates, groundwater is the source of domestic water for 80% of the rural and 50% of the urban areas in India (UNICEF et al., 2013). Naturally occurring fluoride concentrations in groundwater range from 0.5 to 48 mg/L in India depending on geological factors (Ayoob et al., 2008). Elevated concentrations are reported to be associated with leaching from the fluoride-bearing rocks like fluorspar, cryolite, fluorapatite and hydroxyapatite (Agarwal et al., 1997; Meenakshi et al., 2004), or with weathered formations of pyroxene amphibolites and pegmatites (Maliyekkal et al., 2008). Health effects caused by excess daily intake of fluoride, with drinking water as the major contributor, has affected people in 20 states of India (Maliyekkal et al., 2008). Depending on the daily fluoride intake and duration of exposure, fluorosis symptoms range from mild dental fluorosis to crippling skeletal fluorosis (Fawell et al., 2006; Hichour et al., 2000). Provision of appropriately treated drinking water is an important fluorosis mitigation measure.

There are many defluoridation options. Defluoridation technologies based on chemical separation are sorption on solid filter media (Azbar and Turkman, 2000), chemical precipitation (Nawlakhe and Paramasivam, 1993; Azbar and Turkman, 2000; Reardon and Wang, 2000) and coagulation (Pinon-Miramontes et al., 2003; Mameri et al., 1998). Physical separation processes for defluoridation include electro-dialysis (Tahaikt et al., 2006) reverse osmosis (Arora et al., 2004) and nano-filtration (Hu and Dickson, 2006). These physical

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separation processes are effective for defluoridation but they also remove beneficial ions in water and are expensive compared to other technologies. Additionally, the large quantity of reject water makes these processes less preferred in water scarce areas (Zuoa et al., 2008). In industrial countries, treatment using activated alumina is the option of choice. However in poor rural settings, most technologies have failed in the field due to high costs, non-availability of skilled operators, unpalatable taste of treated water and impractical operational requirements. A simple and affordable technology that is acceptable to users is the need of the hour.

Calcium phosphate-based adsorbents are effective and less expensive alternative to activated alumina for defluoridation (Turner et al., 2005). Bone charcoal or bone char, with the principal active component hydroxyapatite ($Ca_5(PO_4)_3OH$), is used as an inexpensive material for defluoridation in developing countries (Larsen et al., 1993; Lambert and Graham, 1995). The hydroxide ion can be exchanged for fluoride to form fluorapatite (Eq. (1)).

$$Ca_5(PO_4)_3OH + F^- \rightarrow Ca_5(PO_4)_3F + OH^-. \quad (1)$$

However in some communities, the use of animal bones is unacceptable due to religious beliefs. Alternative options are either the use of chemically produced hydroxyapatite for filtration or co-precipitation with hydroxyapatite produced by the addition of calcium and phosphate salts. There are several advantages of co-precipitation units over filter columns; ease of implementation, no requirement for filter material production and less need for monitoring and maintenance. Any calcium and phosphate compounds can be used to prepare the thermodynamically stable phase hydroxyapatite (Leamy et al., 1998), preferably the most economic and readily available ones. Fluoride is removed by either direct precipitation of fluorapatite and by the exchange of the hydroxide ion (Eq. (1)) in hydroxyapatite.

The co-precipitation method was first evaluated in the 1930s (Adler et al., 1938; Behrman and Gustafson, 1938). Various approaches for producing hydroxyapatite at low temperatures to co-precipitate fluoride were tested and a few have been used for household water treatment (Gao et al., 2009; Poinern et al., 2011; Wang et al., 2011; Yu et al., 2013).

The present research was performed to explore the integration and effectiveness of the chemical co-precipitation of fluoride with subsequent sand filtration in household water treatment units. The objective of the research was to develop a low cost, efficient and sustainable household treatment system that provides drinking water with fluoride concentrations < 1 mg/L with a potential of field implementation in fluoride affected rural areas in India. The paper details the design, chemical-dose optimization and the performance evaluation in the laboratory and field implementation of the Chemo-defluoridation unit (CDU).

2. Materials and methods

2.1. Chemicals and analytical procedures

Analytical grade chemicals and reagents were used without any further purification. A fluoride stock solution of 1000 mg/L was prepared by dissolving 2.21 g of oven dried sodium fluoride in 1 l distilled water. Spiked water was prepared by adding the required quantity of fluoride stock solution to tap water. Water was also collected from a fluoride-contaminated groundwater source (~4.2 mg/L) for laboratory experiments.

Analysis of water-quality parameters was carried out as per standard methods (APHA, 2012). Fluoride concentrations were measured potentiometrically using a standard ion selective electrode method (Orion Star A214).

All water samples were analysed for pH, electrical conductivity, fluoride, hardness, chloride and total phosphate. Field source water (2 bore wells) were additionally analysed for alkalinity, nitrate, sulphate, sodium and potassium.

2.2. Laboratory experiments

2.2.1. Jar test studies for dose optimization

Batch experiments (1 l) were performed at room temperature using the Jar test assembly (Phipps & Bird Stirrer Model # 7790-402). Calcium chloride dihydrate ($CaCl_2 \cdot 2H_2O$–Salt A) was added along with disodium hydrogen orthophosphate ($Na_2HPO_4 \cdot 2H_2O$–Salt B) in 1 l spiked water with an initial fluoride concentration of 5 mg/L. The concentration of Salt A was varied between 300 and 500 mg/L (2.7–4.5 mM) resulting in a salt to fluoride ratio of 60–100. The weight ratio of Salt A and Salt B was kept constant at 3:2 (Molar Ca: PO_4 ratio = 1.84). Experiments were conducted at a constant stirring rate of 100 rpm for 1 min followed by 20 rpm for 20 min. Chemical reaction between salts resulted in flock formation, which was allowed to settle for 20 min. The flock was not removed from the jar assembly in consecutive batch experiments.

Another set of dose optimization experiments was performed for different initial fluoride concentrations. Experiments were performed with Salt A to fluoride ratios ranging from 40 to 100. To achieve these ratios, spiked water concentrations were stepped from 5 to 20 mg/L with correspondingly varying Salt A concentrations from 200 to 1100 mg/L. The Salt A to Salt B weight ratio was kept constant at 3:2 during all experiments. Based on the process, a CDU was designed and the required chemical dose corresponding to initial fluoride concentrations was determined.

2.2.2. Chemo-defluoridation unit (CDU)

Fig. 1 shows a schematic representation of the CDU designed to treat 75 l of fluoride-contaminated water per batch. It comprises a batch reactor (100 l) for coagulation/flocculation and a sand filtration unit (100 l). Chemicals and contaminated water are added to the batch

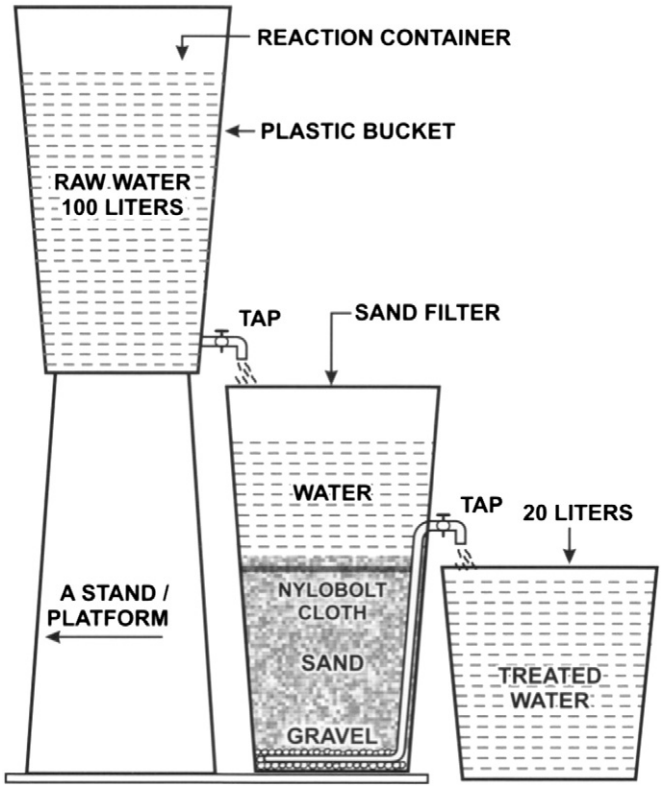

Fig. 1. Schematic representation of CDU design.

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