

Development of Household Defluoridation Unit Based on Crushed Burnt Clay Pot as Sorbent Medium: A Case of Keren Community, Eritrea

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Abstract

Fluoride in water in some parts of Eritrea is above the WHO guideline of 1.5 mg/L. One of the communities in Eritrea exposed to drinking water of high fluoride is Keren community and as a result, they suffer dental and skeletal fluorosis. A survey at 16 water sources in 13 villages was made and 87% of the samples exceeded the guideline, having fluoride levels 1.40-3.98 mg/L. Fluoride removal from synthetic water using crushed burnt clay pot as a sorbent medium was studied in a packed column. A preliminary experiment was carried out on a laboratory scale in mini column, with three different packed beds, 15, 20 and 25 cm depth. A flow rate of 2.5, 5, 10, and 15 ml/min having 5 mg/L fluoride was passed through each bed. The results indicated that the breakthrough volume and time increased with increasing bed depth of the column. On the other hand, an increase in flow rate reduced the treated volume at breakthrough and therefore decreased the service time. Ideal breakthrough occurred at 25 cm bed depth at a flow rate of 2.5 ml/min with breakthrough volume 7.3 L, resulted in reduction of fluoride from 5 to 1.48 mg/L. The result of the mini column was scaled up and tested in a pilot scale unit. The pilot scale managed to treat 324 L of water satisfying the WHO standards of fluoride concentration. The performance of the pilot column agreed with that obtained from the mini column and therefore, crushed burnt clay pot is suitable low cost adsorbent to remove fluoride from water.

Keywords: bed depth, breakthrough, crushed burnt clay pot, fluoride, fluorosis, Keren community

1. Introduction

Fluoride is one of the few chemicals that may be of public concern when present in excess in drinking water. Fluoride commonly occurs in the earth's crust making groundwater more susceptible to contamination (Manahan, 1994). Fluoride at elevated concentration is known to occur in a number of parts of the world and in such circumstances, can have adverse impact on public health and wellbeing (UNICEF, 1999).

It has been reported that there are numerous fluoride belts throughout the world where groundwater contain unsafe levels of fluoride. These belts span over 14 countries in Africa including Eritrea, 8 countries in Asia and 6 countries in America, all having water considered unsafe by the World Health Organization (Tekle-Haimanot, 1995). The part of the East Africa particularly the rift valley is susceptible to fluorosis as the fluoride concentration in drinking water fails to pass the current EU drinking water directives (Reimann et al., 2003).

Eritrea is situated in the horn of Africa and it shares part of the Rift valley. It has a total area of 124,320 km² and a population size more than 3.5 million (Srikanth et al., 2002; MLWE, 2007). Eritrea being part of arid and semiarid region of Africa is not adequately endowed with natural resources including water resource. Water is a scarce commodity in rural Eritrea and there are no perennial rivers except Setit River which is shared with Ethiopia. The majority of the population depends on groundwater for their consumption where the fluoride content of the water is high in some places of the country (Srikanth et al., 2002; Estifanos, 2005; Zerai, 1996). Fluorosis is a common phenomenon particularly in villages around Keren town and other parts on the Red Sea section of the country where groundwater is used for domestic purpose (Srikanth et al., 2002). Although not reported in scientific literatures, this is well known by local population and government offices.

Numerous myths and misconceptions of fluorosis are well spread in Keren community. Traditionally, they believe that occurrence of fluorosis, especially skeletal disorder is from an evil curse and they assume it is inheritable as well. Similar believes are common in some society such as in Kenya that eating hot potatoes or neglecting to brush teeth will lead to dental fluorosis while skeletal fluorosis is believed to come from carrying a child to field before the age of six months (Muller, 2007). However, this is not true and is purely a medical condition caused by taking high levels of fluoride in drinking water (WHO, 2006; UNICEF, 1999).

Fluoride intake to humans is necessary as long as it doesn't exceed the limits as it protects teeth decay. It has both beneficial and detrimental effects on human health in terms of the prevalence of dental caries, skeletal fluorosis and bone fractures. Studies by WHO indicated that there is a dose response relationship between the concentration of fluoride in drinking water and the prevalence of dental caries (WHO, 2006). According to WHO, maximum allowable limit from water uptake to humans is 1.5 mg/L (WHO, 1984) but depending on the climatic conditions such as temperature, it may range from 0.5 mg/L-1.0 mg/L (WHO, 1994; Dissanayake, 1991). When excessive exposure to fluoride occurs, it can give rise to a number of adverse effects ranging from mild dental fluorosis to crippling skeletal fluorosis (WHO, 2006). To minimize the prevalence and severity of tooth decay, the fluoride content in the water must be limited and if possible must be reduced to meet the WHO recommendation before use.

Defluoridation methods may be grouped based on removal mechanisms. They are contact precipitation, membrane techniques and adsorption/ion exchange methods. Precipitation requires addition of chemicals such as alkali, chlorine and aluminium sulphate or aluminium chloride which lead to the formation of fluoride salts. Precipitation method of fluoride removal like Nalgonda technique (Dahi et al., 1996) and limestone reactor (Reardon & Wang, 2000) are high in operational and maintenance cost. Specific cost estimation of operation and maintenance is difficult as cost varies from place to place because of the variation in cost of labour and material. However, they generally remain high compared to other methods. They also generate large volumes of sludge and may produce undesirable effects on water quality such as smell and taste (Tembhurkar & Dongre, 2006). Besides, the water may not achieve the desired water quality level of less than 1.5 mg/L.

Defluoridation device should be modest in investment, low maintenance cost, simple in design and operable at village level, and should meet acceptable water quality. Adsorption methods particularly in packed bed are preferred over the other defluoridation methods. This is mainly due to operation is easier, no daily sludge encountered, have reasonable running and investment cost, and the exhausted media could be replaced with virgin one at relatively longer period of time (McKay, 1995; Eyobel, 2006). However, not a single method meets the entire requirement and materials used as sorbent media may affect the water quality like its pH, turbidity, hardness and bacteriological contaminants.

Some efforts were made to develop fluoride sorbent materials elsewhere. In recent years a large number of cost effective adsorbents have been reported to possess fluoride removal capacity. Few are silica gel (Wang et al., 1995), natural soil (Mayabi et al., 2009), zeolites (Mayadevi, 1996), fired clay (Moges et al., 1996; Pranab, 2004), fly ash (Pranab, 2004). A wide range of material has been attempted for fluoride uptake and selection is based on removal capacity, design simplicity, local availability of materials and chemicals and user preference.

However, most of the adsorbent materials for defluoridation currently available are not cost effective and technically not feasible in rural areas (Ghorai & Pant, 2004). Besides, some of these materials such as bone char could produce undesirable taste in water and may be of low adsorption capacity if the charring process was not properly carried out (Sajidu et al., 2008). Therefore; it would be necessary to search for locally available defluoridation media that is safe and easy to use at household and community level.

Among the large number of cost effective adsorbents which have been reported to possess fluoride removal capacity, fired clay chips and natural soils (Mayabi et al., 2009; Mayadevi, 1996; Moges et al., 1996; Pranab, 2004) are cheap and are locally available in a large quantity. Fired clay chips as a sorbent medium for defluoridation could; hence, be employed as they have good fluoride removal capacity. Domestic clay column filters can be constructed by normally packing them using clay chips which could be obtained as waste of manufacturing brick, pottery or tile. The filter is based on up flow in order to allow for settling of suspended solids within the filter bed. The filter does not have a clean water reservoir and the filtration rate is controlled by slow withdrawal through the tap.

The design of defluoridation units requires a number of information. This could be obtained from series of batch, mini column and pilot plant studies. The latter is time consuming and expensive with regard to the amount of adsorbent and water usage. However, conducting a pilot scale column study is important to predict what would happen in a full scale column when various operating parameters are involved. These parameters include flow

rate, feed and product concentration, bed height, particle size, type of adsorbent, pH, temperature and viscosity (Ko Chun, 2002.).

Sometimes, it is difficult to apply small scale laboratory column studies data to design application as problems in large scale units could occur due to uneven flow patterns, selective flow paths and uneven packing (Ko Chun, 2002). Adsorbents provide a large surface area on which the contaminant molecules can adhere and are generally used in beds through which the wastewater passes for treatment continuously. The type of adsorbent is usually determined before sizing the column, as different kinds of adsorbent will have significant variation in adsorption capacity per unit amount of adsorbent used.

The important aspect of adsorption column design is the sizing of the column as its performance is subjected to different operating conditions. The shape of the breakthrough curve and the time taken for breakthrough are important characteristics for defining the dynamics and operational response of an adsorption column (Aksu & Gonen, 2004). The packed column design generally follows either scale up or Kinetic approach in which, a breakthrough curve from a test column, either laboratory or pilot scale is required. In scale up approach, the pilot test column is filled with the same adsorbent as to the mini-column to be used in full scale application. The linear flow rate and contact time should be kept constant during the scale up process for full scale application to obtain similar mass transfer characteristics (Eyobel, 2006).

Well established scientific study has not yet conducted to know the fluoride content of the groundwater nationwide in Eritrea. However, Srikanth et al. (2002) and Estifanos (2005) reported fluoride level of 3.73 mg/L and 3.27 mg/L respectively in some villages around Keren where fluorosis is salient. According to those studies, the fluoride level was in excess of the standard given by WHO and the incidental of dental fluorosis was over 50 % in children and about 20% among adults. In those villages, alternative water sources are not easily available and hence treatment of fluoride contaminated water is the most reasonable approach.

Despite the prevalent fluorosis symptoms in those areas, nothing has been done to help in the removal of fluoride in the community drinking water. As Eritrea is among the developing countries and is located among countries which are endemic to fluorosis (UNICEF, 1999), a study is required to exploit local fluoride adsorbent materials to remove fluoride at household levels from drinking water to meet the standards.

On this context, a defluoridation study was carried out using crushed burnt clay pot to develop household defluoridation. The adsorbent medium was selected because it is cheap and locally available. The medium was packed in a column and experiment was done to assess its performance under different operating parameters. The study attempted to evaluate the effect flow rate and of bed depth of the column on the performance of fluoride adsorption onto the medium for given fluoride concentration. A household defluoridation unit was designed and its application at household was checked on a pilot scale study.

2. Research Methodology

2.1 Study Area and Its Geological Characteristics

The study area is located in the midland to lowland along the river Anseba around Keren town in Eritrea. Geologically, the area is underlain by granotoid intrusive rich of biotite granite and granodiorite. Mica schists and amphibole schists of medium grade and chlorite schists of low grade metamorphism are the main lithological units recognized with pegmatite intrusions and aplitic dykes which are likely to contain high fluoride (Srikanth et al., 2002). The report indicated that the excess fluoride level in water is mainly attributed to the geological activities from the weathered rock.

2.2 Water Sample Collection and Analysis

A preliminary field survey to identify fluorosis prevalent areas was conducted and 16 sites identified. Key informants were contacted and interview was carried out for local people. Personal experiences and observations of the resident's conditions were also taken in to consideration. Randomly, water samples were taken from drinking water wells/boreholes and surface water in each village in 1000 ml plastic bottles and a total of 16 samples were collected. The bottles were first washed thoroughly with diluted HCl and distilled water before sampling. The samples were kept in a refrigerator until fluoride analysis was done to avoid any contamination. The SPADNS method was used to determine the fluoride concentration in water as recommended by American Public Health Association, American Water Works Association and American Dental Association (APHA, AWWA, WEF, 1995).

2.3 Adsorbent Collection and Preparation

Keren and its surrounding areas are well known for fired clay products in Eritrea. Fired clay pots were purchased

from local market and then crushed down manually into grains. After sun drying, the grains were sieved and graded using the US bureau of standards (Noriaki & Kameru, 1988). Particles passing through 2.36 mm and retained in 0.60 mm sieve were selected as a filter media to enhance the permeability and to decrease to a greater extent the turbidity and colour of the water (Beraki, 2014). The physical characteristic of the filter medium (particle size distribution, particle density, bulk density and porosity) was also determined using particle size distribution curve and water displacement method (Noriaki & Kameru, 1988).

2.4 Fixed Bed Continuous Flow Column

A mini column experiment was carried out to develop a household defluoridation unit for a pilot test. The crushed burnt clay pot medium was packed in a column as a defluoridating filter. Before packing, it was washed with tap water until clear water was obtained. This was to remove any fine particles which could clog the media and block the adsorption sites. The continuous flow experiment was carried out in a down flow mode in a mini column to correlate the flow rate and bed depth on the fluoride removal capacity before up scaling in to a household system. This was to select the ideal flow rate and bed depth that brings optimum fluoride removal.

The bed of the mini column was packed with crushed burnt clay pot and supported and closed by cotton pad and rubber stopper respectively to prevent flow of adsorbent together with the effluent. Moreover, the bed was rinsed with distilled water and left overnight to ensure a closely packed arrangement of particles.

A synthetic fluoride stock solution with 5 mg/L concentration was prepared for the experiment by dissolving anhydrous sodium fluoride, NaF, in distilled water. The solution was homogenized before the commencement of defluoridation using magnetic stirrer. The water was pumped in a down flow mode by a peristaltic pump. The pump was used to control flow rates at 2.5 ml/min, 5 ml/min, 10 ml/min and 15 ml/min in a fixed bed depth of 15 cm, 20 cm and 25 cm and maintained constant during each run.

Samples were withdrawn at the outlet of the mini column at 1 hour interval. Fluoride concentration was determined and a breakthrough curve was generated. The desired breakthrough concentration was determined at 30% of the initial concentration (5 mg/L) which was 1.5 mg/L. The experiment was carried out at room temperature (23 ± 1 °C). The pH of the inflow was maintained at 7.10 ± 0.10 throughout the experiment.

2.5 Scale up of Mini Column, Design and Construction of Household Defluoridation Unit

The breakthrough curve of the mini column was studied at different bed depth and flow rate to obtain optimum fluoride removal. The parameters of the mini column that showed ideal adsorption were scaled up for sizing of the household defluoridation unit. The linear flow rate of the mini column and pilot scale column was maintained the same to obtain similar mass transfer characteristics.

The Household Defluoridation Unit was designed on the assumption that 21 L of treated water was the daily requirement for cooking and drinking for a family having 5-7 persons each consuming 3-4 L/day. The unit was designed to serve for at least half month (15 days). This was chosen because of the practical limitation to obtain more adsorbent media, otherwise it could also be designed for more or less than that. Accordingly, the total volume of water to be defluoridated before reaching the breakthrough was assumed 315 L in the filter life, bringing fluoride level from 5 to 1.5 mg/L. The diameter of the pilot column was selected 10.3 cm as available from the market. From the direct scale up of the mini-column, the corresponding flow rate of the pilot column unit was worked out. The detail of the design calculation of the unit was done as in Table 3.

The Household Defluoridation Unit was constructed of PVC pipes of varying diameters and lengths. The Column was constructed using plumbing materials as available in most hardware stores. PVC pipe was used as the column bed and PVC female adapters were cemented to each end of the length of PVC column. The feed tank was placed over a wooden support at 90 cm above the ground to provide enough head and this was connected to the column through the PVC tubing. A circular perforated PVC plate with equal diameter to the column was placed at 5cm above the bottom of the column to support the adsorbent. A PVC tubing (external $\text{Ø}=2.5$ mm) was used to transfer water from the feed tank and passed through the center of the perforated circular plate to the bottom of the column and water was released at this point and water inside the column flowed upward. The flow rate was adjusted by placing a mark at the control valve so that the flow rate was 3 L/h and operated under this throughout the run. A 1/2-inch hole was drilled 10 cm below the top of the column and a tap was fixed to this point to collect the effluent in receiving tank.

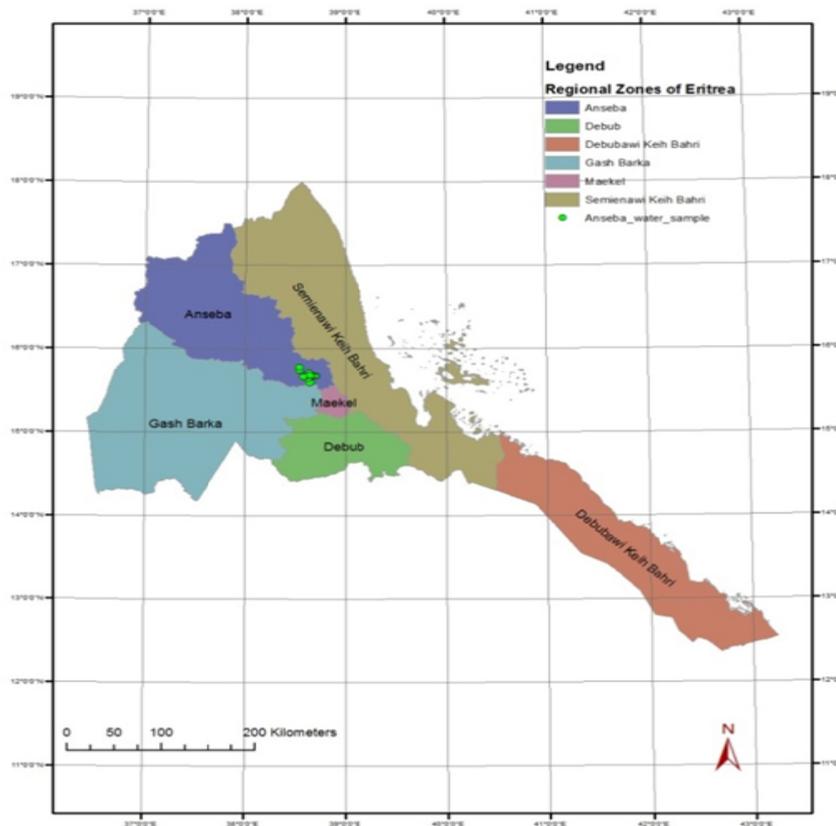
Unlike the mini column experiment, a synthetic fluoride stock solution was prepared from tap water for the pilot scale experiment. Moreover, up flow filter column was used in the pilot experiment so that suspended solids were allowed to settle within the filter bed. This was to reduce the turbidity of the water and to make it consumable for drinking. The performance of the household defluoridation unit was monitored at three hours

interval and operation was discontinued when the fluoride level reached 1.5 mg/L. The physico-chemical characteristic of the effluent water was also measured (pH, EC, turbidity and total hardness).

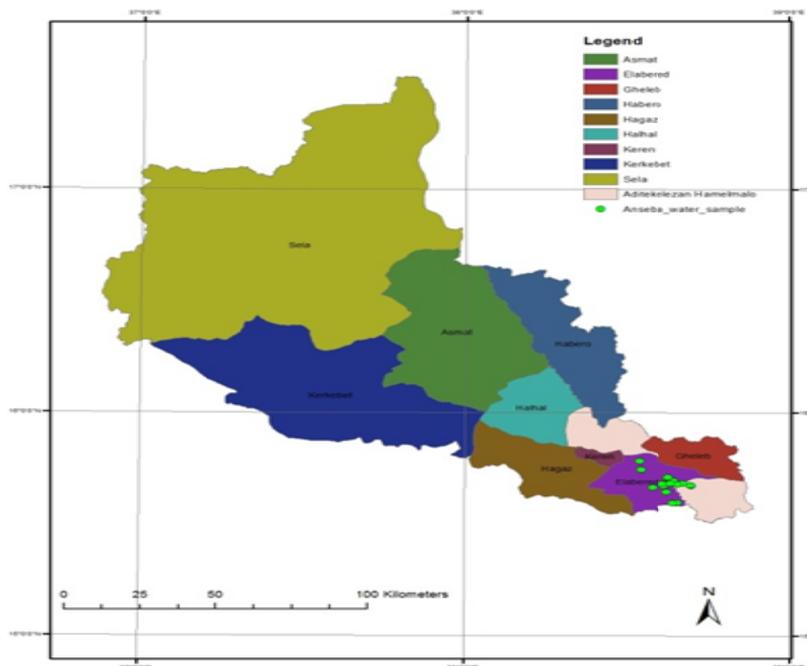
3. Results and Discussion

3.1 Introduction

The study has identified 13 villages that are endemic to fluorosis around Keren town. Some of the villages have two water sources and hence a total 16 water samples were collected for analysis. Figure 1 (a & b) shows the map of Eritrea in general and the specific villages in the study. The names of the specific villages are indicated in Figure 2.



a. Map of Eritrea and study area



b. Map of Anseba zone and the specific villages under study

Figure 1. Sampling area locational map

3.2 Fluoride Concentration and Occurrence of Fluorosis

Water samples were analysed for fluoride concentration and compared if they exceed the WHO standard as indicated by the horizontal line at 1.5 mg/L. The typical analysis of the fluoride level in villages around Keren is indicated in Figure 2. The analysis indicated that the maximum value of fluoride in water was 3.98 mg/L while the minimum was 1.40 mg/L. This was occurred in villages called Harkokia and Balwa 1 respectively.

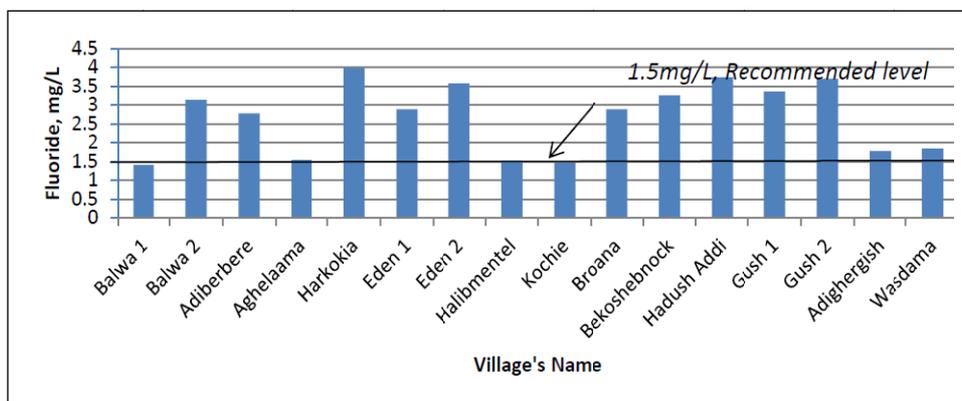


Figure 2. Fluoride level in villages around Keren

Figure 2 shows that 87% of the samples exceeded the WHO guideline. Moreover, the result of the study indicated that, except in two villages, Balwa 1 and Kochie, all water sources in other villages have fluoride levels ranging from 1.52 to 3.98 mg/L, thus exceeding the threshold value of 1.5 mg/L which is prescribed by WHO (1984). The horizontal line (fluoride recommended value by WHO at 1.5 mg/L) crossed the vertical lines of fluoride concentration of each village. Values above the line indicated fluoride levels that exceeded the WHO standards. Unpublished report by the Water Resource Department, (MLWE, 1999), Eritrea, indicated that dental fluorosis is not a wide spread problem in the country. Despite such reports; however, certain places of the

country are known to have high levels of fluoride in water. One of the areas with high fluoride content in drinking water is Keren and its surrounding (Srikanth et al., 2002; Estifanos, 2005; Zerai, 1996). The finding of this research also supported those researchers' conclusion.

A direct relationship between the level of fluoride in drinking water and magnitude and severity of fluorosis is evident as it manifested by mottling of the teeth of children and skeletal disorders of adults in those villages. A report by Srikanth et al. (2002) also indicated similar trends as 50% children and 20% adult are fluorosis infected at various levels.

3.3 Physical Characteristic of the Crushed Burnt Clay Pot

The Particle size distribution curve of the media is indicated in Figure 3 and its physical characteristics such as effective size, particle and bulk density, porosity and uniformity coefficient are presented in Table 1. The particle-size distribution curve shows not only the range of particle sizes present in the medium but also the type of distribution of various size particles. The results further indicated that the medium's bulk density lay within the typical mineral soils that have bulk densities that range from 1.0 to 1.6 g/cm³. Moreover, the porosity of the medium was 52% and hence this was believed to facilitate free water movement within the medium.

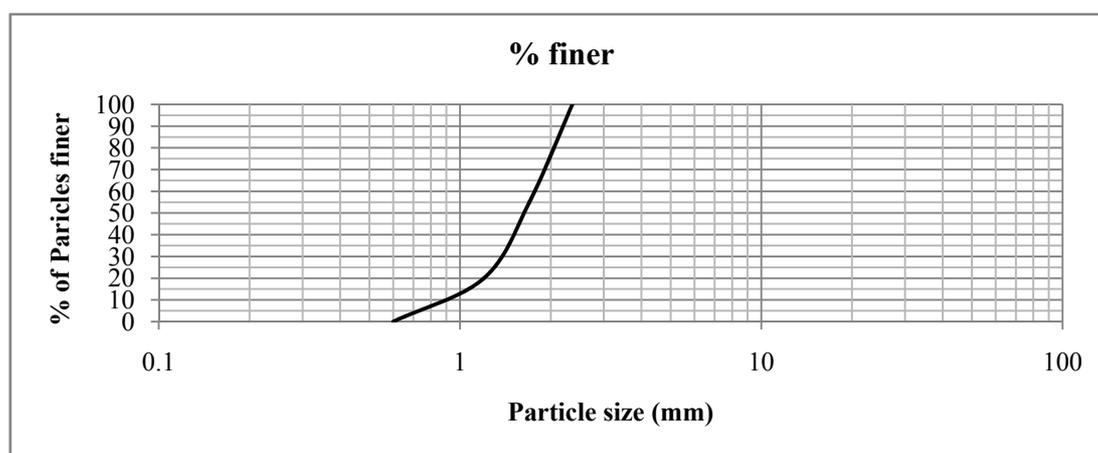


Figure 3. Particle size distribution curve

Table 1. Physical characteristics of crushed burnt clay pot

Physical Parameter	Corresponding Value
Grain Size (mm)	0.60-2.36
Grain effective size (mm)	0.90
Uniformity Coefficient	1.98
Coefficient of Gradation	1.40
Bulk Density (kg/L)	1.01
Particle Specific Gravity	2.10
Porosity (%)	52

3.4 Breakthrough Study of the Mini-Column

The typical adsorption of fluoride onto crushed burnt clay pot media are presented in the breakthrough curves (Figures 4-7). Figures 4, 5, 6 and 7 show the effect of bed depth and flow rate on the breakthrough curves of fluoride adsorption onto the crushed burnt clay pot at fixed bed depths of 15, 20, and 25 cm for a constant flow rate of 2.5, 5, 10 and 15 ml/min respectively. The breakthrough volume was taken at 30% of the initial fluoride concentration ($C_e/C_o = 0.3$) as indicated by the horizontal line in Figures 4-7 in which C_o and C_e are raw and treated water average fluoride concentration respectively. The horizontal line (recommended value by WHO as 1.5 mg/L for fluoride level) crossing each curve in each Figure indicated a point where the breakthrough occurred.

The breakthrough volume could be obtained by projecting the point of intersection vertically down for a given flow rate and the summary of the results of the Figures 4-7 are presented in Table 2.

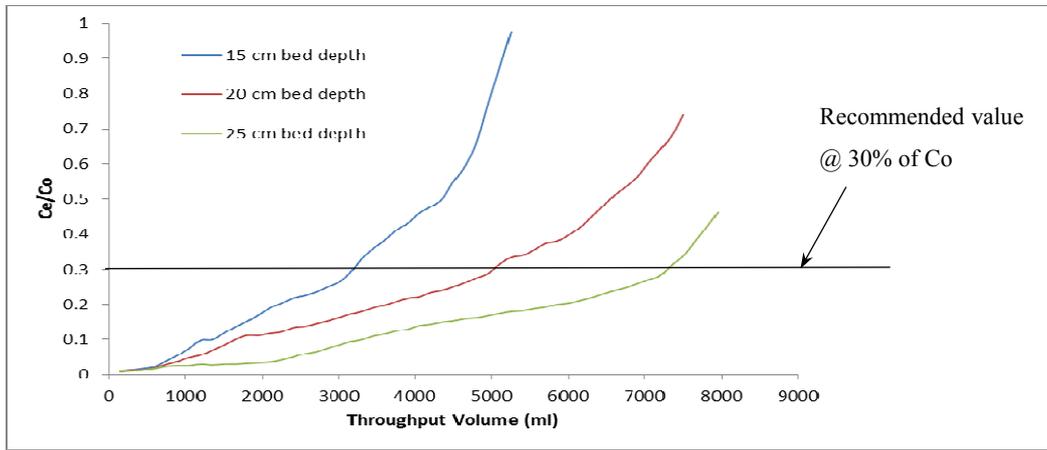


Figure 4. Effect of bed depth on breakthrough volume at a constant flow rate of 2.5 ml/min ($C_o = 5$ mg/L)

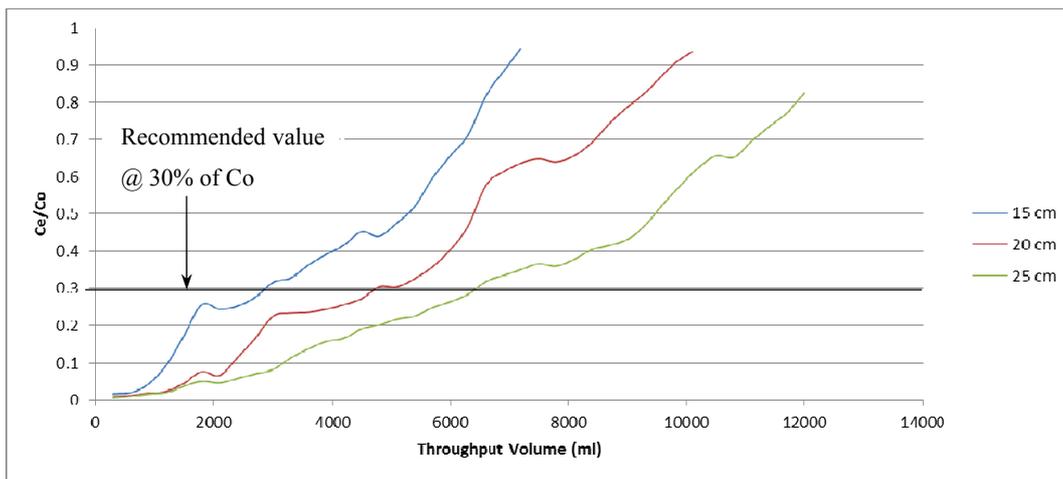


Figure 5. Effect of bed depth on breakthrough volume at a constant flow rate of 5 ml/min ($C_o = 5$ mg/L)

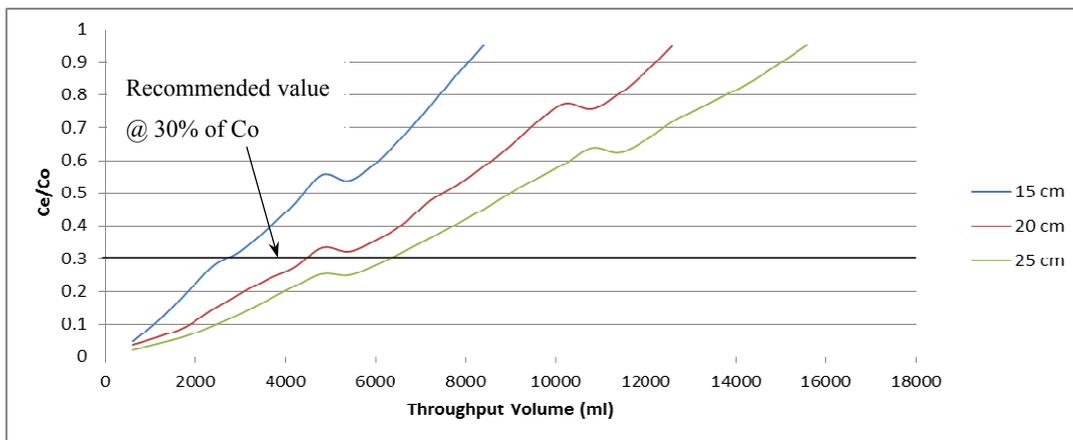


Figure 6. Effect of bed depth on breakthrough volume at a constant flow rate of 10 ml/min ($C_o = 5$ mg/L)

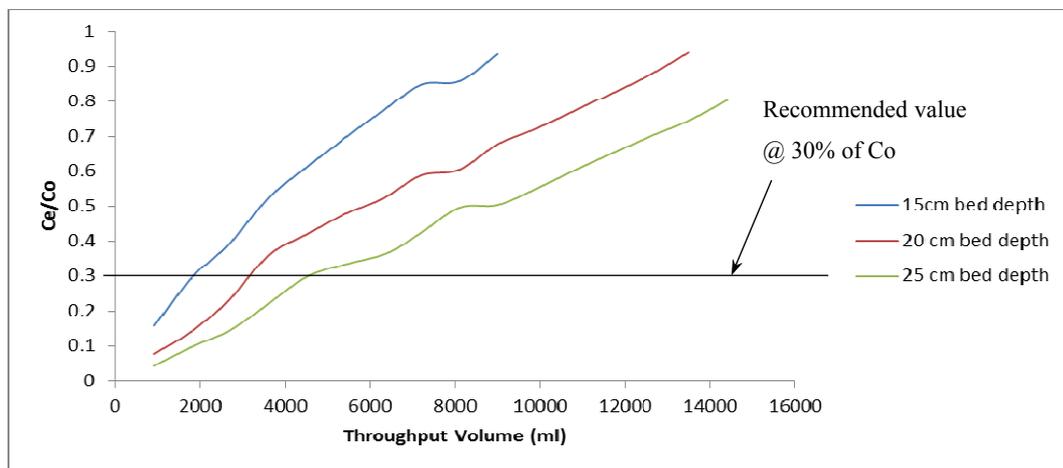


Figure 7. Effect of bed depth on breakthrough volume at a constant flow rate of 15 ml/min ($C_o = 5$ mg/L)

As can be seen from the Figures 4-7, most of the fluoride was initially adsorbed, so the concentration in the effluent was low. However, as the adsorption advanced, the effluent fluoride concentration increased slowly and exhausted gradually. A typical analysis on the effect of bed depth and flow rate on the breakthrough curve was carried out. The Results indicated that with increasing bed depth from 15 to 25 cm, it was observed that the breakthrough volume increased from 3.2 to 7.3 L at a flow rate of 2.5 ml/min and hence, the breakthrough time was increased proportionally from 21.33 to 48.67 hours (Figure 4 and Table 2). The analysis further shows that the breakthrough time was directly related to the bed depth of the column. This could be associated with an increased in bed depth, increased the adsorbent mass and hence provides greater adsorption sites which led to improve the adsorption performance. Moreover, an increased in bed depth could increase the contact time and hence improved the adsorption capacity.

Similar analysis was carried out on the effect of flow rate. An increased in flow rate from 2.5 to 15 ml/min resulted in a reduction of treated volume of water at the breakthrough associated with a decrease in service time of the bed. The typical result in Table 2 shows that 7.3 L and 4.5 L of water were treated at a flow rate of 2.5 ml/min and 15 ml/min respectively at a bed depth of 25 cm at a breakthrough point. It also shows that the breakthrough time was reduced from 48.67 to 5 hours for the same. However, at 15 cm bed depth, increasing the flow rate from 2.5 to 15 ml/min resulted in a decrease in a treated volume from 3.2 L to 1.8 L respectively and consequently the service time of the bed was reduced from 21.33 to 2 hours (Figures 4-7 and Table 2). Hence the bed depth and flow rate played the major role in the adsorption performance and thus the amount of water to be treated varied accordingly. This was mainly due to the decrease in contact time between the fluoride and the adsorbent at higher flow rates and shorter bed depths. Christian et al. (2005) has also stated that intra-particle diffusion controls the adsorption rate and hence, an early breakthrough occurred which led to a low bed adsorption capacity.

On the other hand, when the flow rate was decreased, the contact time in the column was longer, intra-particulate diffusion then became effective. Thus the adsorbate had more time to diffuse in to the particles of the adsorbent and a better adsorption capacity was resulted. Depending on the flow rate and bed depth used, the service time of the medium varied as indicated in Table 2 before reaching the WHO recommendations. Then after, regeneration or replacement of the medium would absolutely be necessary for the next run.

In an ideal adsorption system, the shape of the breakthrough curve has an S shape profile. The breakthrough curves indicated in Figures 5-7 show that at lower bed depths, the shape of the curve didn't show the characteristic S shape profile. However, at 25 cm column depth, the shape was approaching S shape curve for a flow rate of 2.5 ml/min (Figure 4) and hence an ideal adsorption is said to take place. Thus the parameter of this mini column that showed optimum adsorption and ideal breakthrough curve was selected for scaling up.

Table 2. Variable bed depth at a fixed flow rate in a fixed-bed mini column for the removal of 5 mg/L of fluoride by crushed burnt clay pot (mini-Column Internal diameter=2.3 cm)

Flow rate (ml/min)	Bed depth (cm)	Bed volume (cm ³)	Weight of adsorbent (g)	Breakthrough Volume (L)	Breakthrough Time (h)	Adsorbent exhaustion rate (g/L) (Col IV/col V)
2.5	15	62.35	60	3.20	21.33	18.75
	20	83.13	80	5.05	33.67	15.84
	25	103.91	100	7.30	48.67	13.70
5	15	62.35	60	2.93	9.75	20.48
	20	83.13	80	4.80	16	16.67
	25	103.91	100	6.39	21.167	15.65
10	15	62.35	60	2.605	4.33	23.03
	20	83.13	80	4.501	7.50	17.77
	25	103.91	100	6.15	10.25	16.26
15	15	62.35	60	1.80	2	33.33
	20	83.13	80	3.00	3.33	26.67
	25	103.91	100	4.50	5	22.22

The mini-column was operated for 6-9 hours and stopped for 15-18 hours and the on-off pump operation was continued until the complete exhaustion of the column. In some of the breakthrough profiles illustrated in Figures 4-7, effluent fluoride concentrations were lower than that of prior shutdown. This could be due to that the on-off-pump cycle allowed fluoride diffusion along the internal pore-surfaces and adsorption at binding sites resulted in a decrease in fluoride concentration in the stagnant layer around the particle. Thereafter, when the system was restarted, a higher concentration gradient was existed between liquid and solid surface fluoride concentrations. Those observations could lead to a suggestion that fluoride adsorption onto porous adsorbents are intra-particle diffusion limited. Similar suggestion was made by Christian et al. (2005), Yoseph (2007) and Ko Chun (2002).

3.5 Design of Household Defluoridation Unit (HhDU) for A Pilot Scale Experiment

Table 3 shows the details and summary of various parameters with their corresponding values and the design calculations, the household defluoridation unit used in the pilot scale experiment. The design was done for the worst condition (value) in which in this case the maximum value of fluoride was 3.98 mg/L (Figure 2). For safety purpose, the maximum design value of the fluoride was taken 5 mg/L and if the unit would be able to work on this condition, it would obviously be able to perform better for values less than the design value.

Design values were selected for practical consideration and from results of the mini column at which ideal adsorption were taken place. A daily water demand of 21 L was assumed and the unit was assumed to serve for at least half month (15 days) for a pilot run before reaching 1.5 mg/L fluoride concentration. Accordingly, in a period of 15 days, 315 L of water at 5 mg/L fluoride was to reduce to 1.5 mg/L and hence a total of 1102.5 mg of fluoride has to be removed in the operational period. At a sorption capacity of 0.26 mg/g of the medium (Beraki, 2014), the adsorbent mass required was 4240 g. The diameter of the pilot scale column was selected as available from the market to be 10.3 cm. The corresponding flow rate required at the pilot unit was 50 ml/min (3 L/hour) as it was worked out from the direct scale up. To accommodate 4240 g of adsorbent mass, 50 cm packed bed depth was required. But when an allowance was added at different sections of the column, the total height of the column was made 70 cm (Table 3).

Moreover, the summary of the scale up ratio of the mini column and pilot column is presented in Table 4. The scaling ratio was calculated by dividing corresponding values of the min-column to the pilot column. The schematic diagram of the pilot scale unit representing the Household Defluoridation Unit (HhDU) is also indicated in Figure 8.

Table 3. Design calculation of the Household Defluoridation Unit

Given Parameters	Description	Unit	Corresponding value
D	Daily personal water demand	L/(c d)	3-4
N	Number of users	N ^a	5-7
Vt	Total volume of water to be treated in a filter period	L	315
OSc	Operational sorption capacity (Fluoride/Adsorbent)	mg/g	0.26
pb	Bulk Density of Medium (loose)	Kg/L	1.01
Co	Raw water fluoride concentration	mg/L	5
Ce	Treated water average fluoride concentration	mg/L	1.5
q	linear flow rate (as selected from the mini column experiment)	cm/h	36
Derived Parameters			
Qd=D*N	Daily water treatment	L/d	21
F _t =Vt*(Co-Ce)	Total fluoride removal during a period	mg	1102.5
M=F _t /OSc	Amount of medium required for removal	g	4240
V _m =M/pb	Volume of the medium in the filter	L	4.198
Corresponding Dimensions			
Ø	Column diameter (selected as available)	cm	10.30
Q =q* Π*(Ø/2) ²	flow rate	L/h	3.0
Hw =Qd/Q	working hours per day	h/d	7
T ₁	Distance from bottom of the column to the bottom level of the packed media	cm	5
T ₂	Distance from top level of the packed media to the tap	cm	5
T ₃	Distance between the tap & top of the column	cm	10
hc=V _m /(Π*(Ø/2) ²	Total height of the packed column	cm	50
Htc	Total height of the column (T ₁ + T ₂ +T ₃ +hc)	cm	70

Table 4. Scale-up parameters and Scale-up ratios

Geometric Parameters	Mini column	Large column	Scale-up ratio
Cross sectional Area (cm ²)	4.15	83.32	0.05
Bed volume (cm ³)	103.87	4166.14	0.02
Operating Parameters			
Influent fluoride concentration (mg/L)	5	5	1
Flow rate (cm ³ /h)	150	3000	0.05
Filtration rate or Linear flow rate (Flow rate/Cross sectional Area, cm/hour)	36	36	1
Mass of adsorbent (g)	100	4240	0.02

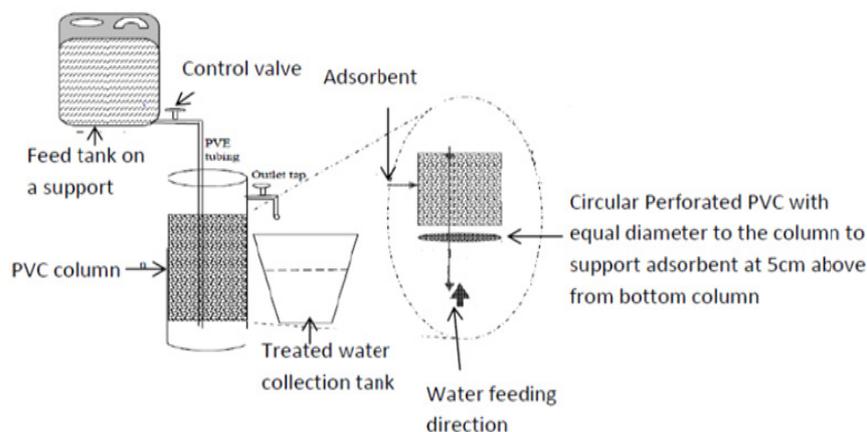


Figure 8. Detailed schematic diagram of the Household Defluoridation Unit

3.6 Performance of Household Defluoridation Unit (HhDU)

The designed household defluoridation unit (Table 3 and Figure 8) was run for a pilot experiment. The pilot scale unit has 50 cm packed bed depth and was packed with 4240 g adsorbent. Water containing 5 mg/L fluoride was passed through it at 50 ml/min (3 L/h) flow rate until breakthrough point was reached at 1.5 mg/L fluoride. The result of the pilot scale experiment on fluoride adsorption onto crushed burnt clay pot media is presented in a breakthrough curve where the concentration ratio C_e/C_0 is plotted versus treated volume (Figure 9). From typical analysis of the Figure, the total volume of treated water at the breakthrough point was 324 L and the corresponding breakthrough time was 108 hours. This was equivalent to 15.43 (108/7) days as the pilot scale was designed to run for 7 hours daily to treat 21 L of water at a flow rate of 3L/hour. This was slightly higher than the designed volume to be treated by 2.85%. Otherwise, the result was in coherence with the design value despite there was an increase in treated volume by 9 L than the anticipated design volume of 315 L (Table 5).

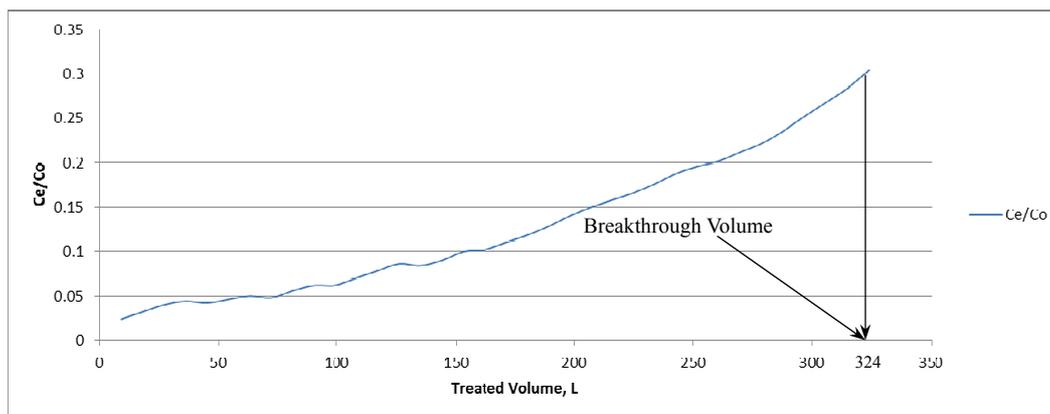


Figure 9. Treated Volume for the Pilot column experiment (flow rate=50 ml/min, $C_0 = 5$ mg/L, bed depth= 50 cm, packed mass = 4240 g)

A comparison (Table 5) was made between the design and working columns. The results of the pilot column were in coherence with the design value and hence, the household defluoridation unit was appropriate and efficient. The up-flow column unit used resembled a plug flow, where the lower parts of the packed bed become saturated at a time where the upper parts were still fresh. Then the saturation zone moved slowly towards the upper treated water point. Moreover, up-flow helped suspended particles to settle within the filter bed and thus reduced the turbidity of the water to a greater extent.

Table 5. Comparison of assumed and working columns

Parameter	Unit	Calculated/Theoretically assumed pilot column	Working pilot column
Treated water	L	315	324
Adsorption capacity	mg/g	0.26	0.27
Adsorbent exhaustion rate	g/L	13.70	13.10
Influent Fluoride concentration	mg/L	5	5
Effluent fluoride concentration	mg/L	1.50	1.52
Mass of adsorbent	g	4240	4240
Volumetric flow rate	ml/min	50	50
Linear flow rate	cm/h	36	36
Bed Internal Diameter	cm	10.30	10.30
Area	cm ²	83.36	83.36
Bed depth	cm	50	50
Bed volume	cm ³	4167.82	4167.82

Besides, the average adsorption capacity at breakthrough of the media when packed in mini column was 0.26 mg/g and the corresponding value was 0.27 mg/g in the pilot scale unit. The average adsorbent exhaustion rate was also compared in the mini and pilot column and the values were almost the same (Tables 2 and 5) which were 13.70 g/L and 13.10 g/L respectively. And hence it could be said that similar mass transfer was occurred during the adsorption process in both the mini column and pilot scale unit.

3.7 Treated Water Quality Monitoring

Water quality monitoring was carried out for selected physico-chemical characteristics of the treated water such as pH, turbidity, conductivity and total hardness. The conductivity, turbidity and total hardness of the treated water were slightly higher near the beginning but decreased soon. This was may be due to the release of fine and unstable soluble particle of the media. As time went, values were normalized and approached to the characteristics of the raw water. However, the effluent pH of the treated water was almost the same to the influent used. Hence, it could be concluded that there was no much deviation in values from the raw water. Moreover, the treated water was colourless, tasteless and there was no smell observed. This indicated that crushed burnt clay pot filter was efficient in its filtering activity of the raw water.

Table 6 shows the parameters used for the water quality test. The tests indicated that values were within the acceptable range despite there were slight increments. Hence, the Household Defluoridation Unit performed well and the rural communities are on the safe side if they employ this unit to reduce fluoride from 5 to 1.5 mg/L from drinking water.

Table 6. Physico-chemical analysis of treated water.

Water quality parameter	Range of values		WHO standards	Remark
	Influent	Effluent		
F ⁻ (mg/L)	4.90-5.10	≤1.52	Less than 1.50	Satisfactory
pH	6.90-7.20	7.0 -7.30	6.5-8.50	Good
EC (μS/cm)	190-210	230-260	Less than 500	Acceptable
Turbidity (NTU)	0.10-0.11	0.31-1.30	Less than 5	Acceptable
Total hardness as CaCO ₃ (mg/L)	240-260	276-309	-	Acceptable
Taste	Normal	Normal	-	Good
Smell	None	None	-	Good
Colour	Clear	Clear	-	Good

4. Conclusion

Fluorosis is a medical condition caused by high levels of fluoride such as in drinking water. The rural communities around Keren, Eritrea, suffer dental and skeletal fluorosis as result of consuming water containing excessive fluoride. A preliminary study was carried out to identify fluorosis endemic villages and 13 villages were identified in the study. The typical analysis of the fluoride concentration in the villages' drinking water indicated that the fluoride level of 87% of the water sources sampled was exceeded the WHO recommendation. The maximum fluoride level was 3.98 mg/L while the minimum was 1.40 mg/L. Defluoridation study was carried out to remove excess fluoride from water using crushed bunt clay pot as a sorbent media. Initially, the defluoridation study was conducted in a mini column unit to correlate the flow rate and bed depth for a given fluoride concentration of 5 mg/L. The result of the mini column indicated that optimum removal of fluoride was obtained at a bed depth of 25 cm and a flow rate of 2.5 ml/min, treating 7.3 L of water meeting the WHO standard.

The result of the mini column unit was scaled up and a household defluoridation unit was designed for a pilot scale experiment. The unit was developed based on local available materials and its performance was checked in a pilot scale run. The unit managed to defluoridate 324 L of water containing 5 mg/L of fluoride, satisfying the WHO fluoride recommendation. The performance of the pilot column agreed with that obtained from the mini column. The results of the pilot column were also in coherence with the design value and hence, the household defluoridation unit was appropriate and efficient. Moreover, the unit could easily be operated at village level and would be highly applicable for rural community who lack electricity, skilled operators and investment for using other defluoridation means. The unit also produced water of desirable quality and the exhausted media could be replaced with virgin one at relatively low cost. In addition, it would create job opportunities for the local community, involving them in constructing the unit. Therefore, the developed household defluoridation unit (HhDU) would be one of the best alternative defluoridation methods in fluorosis endemic rural Eritrea.

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References

- Aksu, Z., & Gönen, F. (2004). Biosorption of Phenol by Immobilized Activated Sludge in a Continuous Packed Bed: Prediction of Breakthrough Curves. *Process Biochem.*, 39, 599-613. [http://dx.doi.org/10.1016/S0032-9592\(03\)00132-8](http://dx.doi.org/10.1016/S0032-9592(03)00132-8)
- APHA., AWWA., & WEF. (1995). *Standard Methods for the Examination of Water and Wastewater* (19th ed.). Washington, DC: Public Health Association, American Water Works Association and American Dental Association.
- Beraki, B. M. (2014). *Investigation of Household Defluoridation of Water Using Local Materials as Sorbent Media: A case of Keren Community in Eritrea*. MSc Thesis, Department of Civil Engineering, Jomo Kenyatta University of Agriculture and Technology, Kenya.
- Christian, T. V., Henri, F., Catherine, P., & Yuh-Shan, H. (2005). Removal of lead (II) ions from synthetic and ream effluents using immobilized Pinus Sylvestris sawdust: Adsorption on a fixed bed column. *J. Hazardous Material*, 123, 135-144. <http://dx.doi.org/10.1016/j.jhazmat.2005.03.032>
- Dahi, E., Mtalo, F., Njau, B., & Bregnhj, H. (1996). *Defluoridation using Nalgonda Technique in Tanzania*. 22nd WEDC Confr., New Delhi.
- Dissanayake, C. B. (1991). The Fluoride Problem in the Groundwater of Srilanka -Environmental Management and Health. *Intl. J. Environ. Studies*, 19, 195-203. <http://dx.doi.org/10.1080/00207238208709990>
- Estifanos, H. (2005). *Groundwater Chemistry and Recharge Rate in Crystalline Rock: Case Study from the Eritrean Highlands*. M.Sc. Thesis, KTH Land and Water Resources Engineering, Sweden.
- Eyobel, M. D. (2006). *Removal of Fluoride from Water Using Granular Aluminium Hydroxide: Adsorption in a Fixed Bed Column*. M.Sc. Thesis, Environmental Science Program, Addis Ababa University, Ethiopia.
- Jolly, S. S., Prasad, S., Sharma, R., & Rai, B. (1971). Human Fluoride Intoxication in Punjab, India. *Fluoride*,

4(2), 64-69.

- Ko Chun, K. D. (2002). *Mass Transfer Modeling for Fixed-bed Sorption of Metal Ions on Bone Char*. PhD., Thesis, the Hong Kong University Science and Technology, Hong Kong
- Manahan, S. E. (1994). *Environmental chemistry* (6th ed.). USA: Lewis publishers.
- Mayabi, A. O., Mutothia, F. U., & Kamau, G. (2009). An Assessment of Soil Materials in Kenya as Sorbent Media for Defluoridation of Water. *Journal of Civil Engineering Research and Practice*, 6(2), 39-49.
- Mayadevi, S. (1996). Adsorbent for Removal of Fluoride from Water. *Ind. Chem. Eng.*, 38, 155-157.
- McKay, G. (1995). *Use of Adsorbents for the Removal of Pollutants from Wastewaters*. CRC Press, Boca Raton, New York, London and Tokyo.
- MLWE. (2007). *National Adaptation Programme of Action*. Ministry of Land, Water and Environment, Department of Environment (MWWE), Eritrea.
- Moges, G., Zwege, F., & Socher, M. (1996). Preliminary Investigations on the Defluoridation of Water Using Fired Clay Chips. *J. African Earth Sci.*, 21, 479-482. [http://dx.doi.org/10.1016/0899-5362\(96\)00030-9](http://dx.doi.org/10.1016/0899-5362(96)00030-9)
- Muller, K. (2007). *CDN's Experiences in Integrating Social Aspects in Fluorosis Mitigation*. Catholic Diocese of Nakuru, *Water Quality*. Reviewed by Peter Jacobsen, Kenya.
- Noriaki, A., & Kameru, J. G. (1988). *Soil Testing Manual*. Technical Cooperation of Jomo Kenyatta University College of Agriculture and Technology.
- Pranab, K. R. (2004). *Studies on Estimation of Fluoride and Defluoridation of Drinking Water*. Master Thesis. Department of Chemical Engineering, Indian Institute of Science, India.
- Reardon, E. J., & Wang, Y. (2000). A limestone Reactor for Fluoride Removal from Wastewater. *Environment Sci. Tech.*, 34(15), 3247-3253. <http://dx.doi.org/10.1021/es990542k>
- Reimann, C., Bjorvatnb, K., Frengtsada, B., Melakuc, Z., Tekle-Haimanot, R., & Siewersd, U. (2003). Drinking Water Quality in the Ethiopian Section of the East African Rift Valley I- data and health aspects. *Journal of the Science of the Total Environment*, 311, 65-80. [http://dx.doi.org/10.1016/S0048-9697\(03\)00137-2](http://dx.doi.org/10.1016/S0048-9697(03)00137-2)
- Sajidu, S. M. I., Masamba, W. R. L., Thole, B., & Mwatseteza, J. F. (2008). Groundwater Fluoride Levels in Villages of Southern Malawi and Removal Studies using Bauxite. *Intnl. J. of Physical Sci.*, 3(1), 1-11.
- Srikanth, R., Viswanatham, K. S., Kahsai, F., Fisahatsion, A., & Asmellash, M. (2002). Fluoride in Groundwater in Selected Villages in Eritrea. *Environmental Monitoring and Assessment*, 75(2), 169-177. <http://dx.doi.org/10.1023/A:1014491915537>
- Tekle-Haimanot, A. F., Bushera, B., & Mekonnen, Y. (1995). *Fluoride Levels in Water and Endemic Fluorosis in Ethiopian Rift Valley*. 1st International Workshop on Fluorosis Prevention and Defluoridation of Water, Ngurdoto, Tanzania, The International Society for Fluoride Research. Ethiopian Rift.
- Tembhurkar, A. R., & Dongre, S. (2006). Studies of Fluoride Removal Using Adsorption Process. *Journal of Environ. Sci. & Eng.*, 48(3), 151-156.
- UNICEF. (1999). *UNICEF's Position on Fluoridation*. A UNICEF Publication on Water, Environment, Sanitation and Hygiene. Issue 13.
- Wang, R., Li, H. N., & Wang, Y. (1995). Study of New Adsorbents for Fluoride Removal From Waters. *Water Qual. Res. J. Canada*, 30, 81-88.
- WHO. (1984). Guidelines for Drinking Water Quality. *World Health Organization, Geneva*, 1(2).
- WHO. (1994). *Expert Committee on Oral Health Status and Fluoride Use: Fluorides and oral health*. World Health Organization, Geneva, Technical Report Series, p. 846.
- WHO. (2006). *Fluoride in Drinking-Water*. Published by IWA Publishing, London, UK. World Health Organization.
- Yoseph, A. W. (2007). *Fluoride Removal From Water With Aluminium Oxide Hydroxide: A Pilot Study For Household Application*. M.Sc. Thesis, Environmental Science Program, Addis Ababa University, Ethiopia.
- Zerai, H. (1996). Groundwater and Geothermal Resources of Eritrea with the Emphasis on Their Chemical Quality. *Journal of African Earth Sciences*, 22(4), 415-421. [http://dx.doi.org/10.1016/0899-5362\(96\)00028-0](http://dx.doi.org/10.1016/0899-5362(96)00028-0)

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