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Preliminary Assessment of Low Cost Local Sorbent Materials for Water Defluoridation in Keren, Eritrea

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Abstract

Water in some parts of Eritrea has fluoride level above WHO guideline of 1.5 mg/L. One of the communities in Eritrea exposed to high fluoride in water is Keren community and as a result, they suffer dental and skeletal fluorosis. Fluoride sorbent local materials named crushed burnt clay pot, household ash, Keren and Adigerghish soil were studied in a batch defluoridation. The effect of amount of adsorbent, pH, contact time, particle size and fluoride concentration were examined. The pH of water played a major role and at pH 7, crushed burnt clay pot, Keren and Adigerghish soil performed at optimum but optimum pH for household ash was 4. However, contact time was similar and the equilibrium time was 120 minute. Particle size had no significant effect in case of crushed burnt clay pot, however, in the other three materials, fine particles performed better than coarse particles. In all adsorbents studied, similar increasing trend in adsorption were observed when fluoride concentration was increased. Fluoride adsorption comparisons were made and the study revealed that crushed burnt clay pot has superior capacity than the others. Its average fluoride removal capacity was about 0.26 mg fluoride/g medium. Others ranged from 0.08-0.1 mg/g for the same mass of 7 g and thus crushed burnt clay pot was selected for defluoridation.

Keywords: batchdefluoridation; Eritrea; fluoride; fluorosis; Keren; local sorbent

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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 [1]. A number part of the world contains fluoride at elevated concentration and thus can have adverse impact on public health and well-being [2]. Certain studies indicated that there are numerous fluoride belts throughout the world where groundwater contain unsafe levels of fluoride [3]. East Africa particularly the rift valley is among areas of the world which are susceptible to fluorosis as the fluoride concentration in drinking water fails to pass the current EU drinking water directives [4]. Eritrea being part of the rift valley, water in some part of its region is thus considered unsafe by the World Health Organization [5].

Water is a scarce commodity in rural parts of Eritrea and there are no perennial rivers except Setit River which is shared with Ethiopia. Groundwater is the main source of water for drinking and domestic purpose. However, according to certain studies, this groundwater contains high fluoride in some places of the country [6,7,8]. Fluorosis is thus a common phenomenon particularly in villages around Keren town and other parts in the Red Sea section of the country where groundwater is used for domestic purpose [6]. Although not reported in scientific literatures, this is well known by local population and government offices.

Certain studies carried out to estimate the fluoride content in groundwater in some parts of Rural Eritrea are a commendable one [6,7]. According to those reports, the fluoride level was above the WHO guideline for drinking water. Moreover, similar finding was reported elsewhere that fluoride levels in some of the fluorosis endemic villages around Keren town reach up to 4 mg/L [9]. The studies indicated elevated concentration of fluoride in the groundwater where dental fluorosis is prevalent. Because of this the communities suffered dental fluorosis from consuming excess fluoride in water. According to the study, the incidental of dental fluorosis in the affected villages was over 50% in children and about 20% among adults [6].

Fluoride intake 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 [10] when it exceeds the limits. WHO recommended 1.5 mg/L as a maximum allowable limit from water uptake to humans [5] but depending on the climatic conditions such as temperature, it may range from 0.5-1.0 mg/L [11,12]. 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 [10]. Therefore, to minimize the prevalence and severity of tooth decay, the fluoride content in water must be limited and if possible must be reduced to meet the WHO guideline before use.

Fluoride removal practice commonly known as defluoridation must be adopted in areas where there is excess fluoride in drinking water to protect fluorosis. However, the defluoridation device should be modest in investment, low maintenance cost, simple in design and operable at village level, and should meet acceptable water quality [9]. Among the various defluoridation methods available, 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 [13,14]. 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.

In recent years a large number of cost effective adsorbents have been reported to possess fluoride removal capacity. Few are silica gel [15], natural soil [16], zeolites [17], fired clay [18,19] and fly ash [19]. Among the wide range of adsorbent materials that have been attempted for fluoride uptake, selection is based on removal capacity, design simplicity, local availability of materials and chemicals and user preference [9].

However, most of the adsorbent materials for defluoridation currently available are not cost effective and technically not feasible in rural areas [20]. 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 [21]. Therefore; it would be necessary to search for locally available defluoridation media that is safe and easy to use at household and community level.

Fired clay chips and natural soils are among the large number of cost effective adsorbents which have been reported to possess fluoride removal capacity [16,17,18,19]. Those materials are cheap and are locally available in a large quantity. Clay filters have been used with some success in a number of regions, most notably in countries such as Sri Lanka [22]. Though there is significant variability in the fluoride uptake capacity, they could be potential alternatives in rural areas such as Eritrea as well.

The rural community around Keren town, Eritrea, has been suffering from dental and skeletal fluorosis as result of consuming excess fluoride from water. Despite reports that indicate fluoride level up to 4.0 mg/L in some of the villages around Keren where fluorosis is salient and the prevalent symptoms of fluorosis is common, yet nothing has been done to help in fluoride removal from the community's drinking water [6,7,9]. According to those studies, the fluoride level is in excess of the guideline for drinking water given by WHO [5] and thus defluoridation is indeed required to render the water potable.

In villages around Keren, alternative water sources with low fluoride level are not easily available and hence treatment of fluoride contaminated water is the most reasonable approach to prevent and control dental caries and skeletal fluorosis. Therefore, the study was carried out to assess, examine and compare the performance of locally available low cost adsorbent materials such as Fired clay, Keren soil, Adigerghish soil and Household Ash for fluoride up take. This is to help select the best medium that could be used for developing a household defluoridation unit in rural community of Eritrea such as Keren and its surrounding areas which are endemic to fluorosis.

2.Research Methodology

2.1. Adsorbent Collection and Preparation

Soil materials in their pristine state were collected from Keren and Adigerghish where the effect of fluorosis is salient. Stones and gravel were removed manually. A name was assigned for convenience according to the place of origin as Keren and Adigerghish mud soils. The soil materials are being exploited by the community for different purposes for making dish pot, coffee pot, and water pot. For this reason, Keren and its surrounding areas are well known for fired clay products in Eritrea. Fired clay pots were purchased from the market and then crushed down manually into grains and sieved. Household ash was also collected from households as by-product of burned firewood.

All the adsorbent materials were sun dried and the grains were sieved and graded using the US bureau of standards [23]. The photos presented in Fig. 1 (a, b, c and d) shows the different adsorbent material collected and tested for defluoridation study.

a. Adigerghish soil b. Burnt clay pot

c. Keren soil d. Household ash

Figure 1: Photos of Adsorbent materials before preparation

2.2. Batch Adsorption Test

A series of synthetic standard fluoride solution was prepared by dissolving 221 mg of anhydrous sodium fluoride, NaF in distilled water in a beaker. Magnetic stirrer was used to mix the solution and then diluted to 1000 ml. This is equivalent to 100 mg of fluoride ion (F^-) in a litre of distilled water. i.e. to say 100 mg/L of fluoride stock solution. 25 ml was taken from this fluoride stock solution using a pipette and diluted to 500 ml distilled water to prepare 5 mg/L F^- solution and defluoridation experiment was carried out. This prepared fluoride solution was used in all subsequent batch studies throughout the batch experiment.

In the batch experiment, six beakers containing 500 ml solution at 5 mg/L F^- concentration were employed of which one was a control (no adsorbent added). Adsorbents ranging from 3-10 g were added to the fluoride solution in the beakers. The beakers containing the mixture were placed in a jar tester and stirred at an agitation speed of 160 revolutions per minute for 2 hours. This was repeated in all subsequent batch experiments to test the different materials for their optimum capacity and examined the defluoridation performance of available adsorbents (crushed burnt clay pot, household ash, Keren and Adigerghish mud soil).

After two hours, samples were taken from the each beaker using a pipette of 10 ml. the samples were filtered using a Whatman® filter paper. Fluoride concentration in the solution was measured by the SPADNS using Hanna HI 83099 model (COD and Multiparameter) as outlined by the American Public Health Association, American Water Works Association and Water Environment Federation [24]. The effects of amount of adsorbent added, particle size, initial fluoride concentration, contact time, and pH (pH was adjusted by 0.1N HCL and 0.1N NaOH) on the fluoride removal capacity were examined.

Water quality test such as pH, electrical conductivity (EC) and turbidity were made to assess the side effects after adsorbents addition to the solution for fluoride removal from water. Comparisons for fluoride up take by the different sorbent materials were made and thus better sorbent media was selected based on the potential to remove fluoride for developing a household defluoridation unit to be used in a rural community.

2.3. Adsorption Capacity Determination

Batch adsorption experiment was carried out in order to investigate the fluoride adsorption capacities of the different materials collected. This was important to compare and select the material which has the highest capacity to remove fluoride from water. The fluoride adsorption capacity of the different materials and their efficiency to take up fluoride from the solution was known by measuring the residual fluoride concentration and was calculated using the equations(1) and (2).

$$\%\,of\ Adsorption = \frac{Co - Ct}{Co} * 100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

$$Adsorption\ capacity, X\left(\frac{mg}{g}\right) = \frac{Co - Ct}{M} * V \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots.. (2)$$

Where: Co = initial fluoride concentration

Ct = residual fluoride concentration at time t

M = mass of adsorbent

V = volume of the solution used in the batch

X = adsorption capacity (mg of fluoride removed/g of adsorbent used)

3. Results and Discussions

Preliminary adsorption batch studies were carried out using local materials (crushed burnt clay pot, Keren mud soil, Adigerghish mud soil and Household ash) and the effect of different parameters on fluoride removal capacity were studied.

3.1. Effect of Adsorbent Mass on Fluoride (F^-) Adsorption Efficiency

The percentage of F^- adsorbed for crushed burnt clay pot, Keren soil, Adigerghish soil and Household ash are presented in Fig.2. The results indicated that the adsorption capacities of the materials range from about 27.6% for Keren soil to 81.6% for crushed burnt clay pot at 10 g adsorbent mass. From the figure, crushed burnt clay pot has showed the highest removal capacity. This could be explained that crushed burnt clay pot was heat treated and hence adsorbent sites could possibly be activated while Keren and Adigerghish soil were in their pristine state and hence adsorbent sites might not be activated very well. The figure also shows that household ash had higher fluoride adsorption efficiency than Keren and Adigerghish soil next to crushed burnt clay pot. It is well documented that fluoride is adsorbed more to clay materials better when it is heat treated up to certain levels. Moreover, fluoride uptake is better in clays containing more Aluminum and Iron oxides and the results show that higher adsorption capacity could be associated to clay samples having higher Aluminum and Iron oxide [16,25].

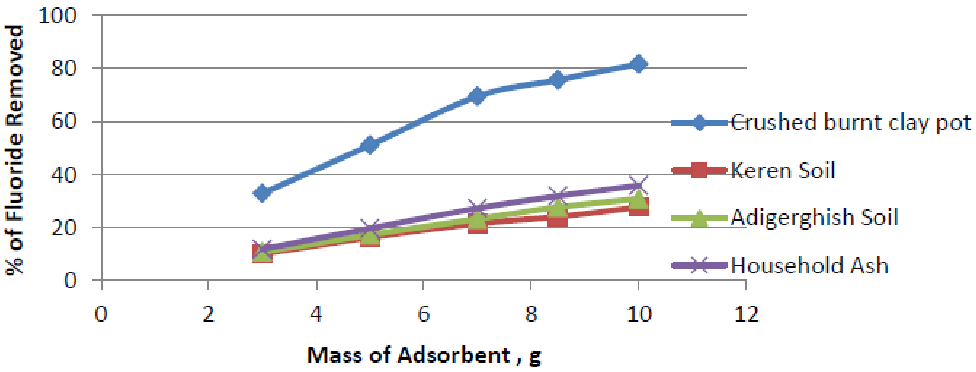

Figure 2: Effect of adsorbent mass on adsorption efficiency (Initial fluoride conc. 5 mg/L.)

From Fig. 2, it was observed that crushed burnt clay pot was found to have better removal efficiency than the others. When 7 g was added to the solution, the fluoride residue was reduced from 5 mg/L to almost 1.5 mg/L and at this level; the adsorption capacity of crushed burnt clay pot was 0.26 mg/g. However, the adsorption capacity of Keren soil, Adigerghish soil and household ash was 0.076 mg/g, 0.083 mg/g and 0.097 mg/g respectively at the same mass of 7 g. Hence, more adsorbent mass was required in those three adsorbents to bring fluoride to the same level as of 7 g of crushed burnt clay pot.

Fig. 3 shows the typical analysis of the fluoride residue in solution when different amount of crushed burnt clay pot doses were added to the solution. The figure shows that at 14 g/L dose, the fluoride residue was approaching to the WHO recommendation of 1.5 mg/L fluoride concentration. Moreover, the slop of the curve was falling rapidly before reaching 14 g/L adsorbent dose, however, it tend to flatten beyond that and the reduction in fluoride level was significantly low to the right. From this, it could be concluded that optimum mass required to bring a fluoride level from 5 mg/L to the WHO recommendation (1.5 mg/L) [5] was approximately 14 g/L in the case of crushed burnt clay pot. This dose was equivalent to 7 g of crushed burnet clay pot. Hence, in the subsequent experiments, 7g mass of adsorbent was employed for further testing.

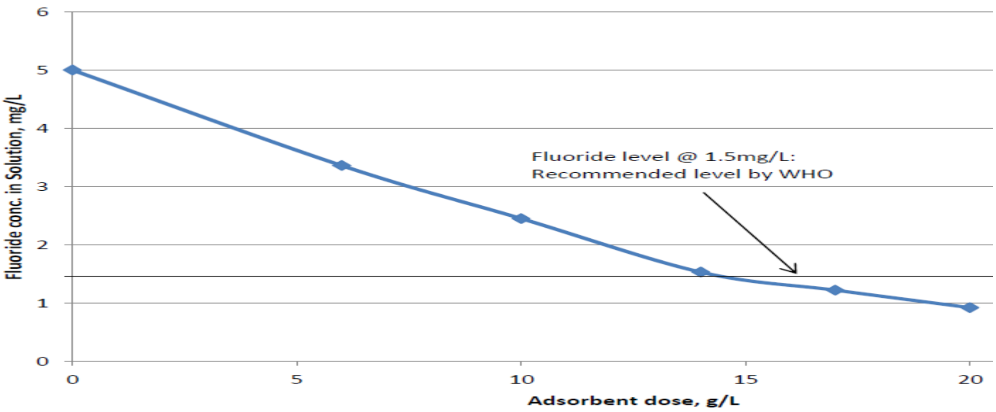

Figure 3: Fluoride residue in solution at different doses of crushed burnt clay pot

3.2. Effect of Contact Time on Fluoride Removal Efficiency

Investigation on the effect of contact time on the adsorption efficiency was studied. The typical analyses of the results were indicated in Fig. 4. The figure shows as the contact time increased, the rate of adsorption of fluoride increased rather rapidly in the first 50 min in the case of crushed burnt clay pot but then gradually approached a more or less constant value denoting attainment of equilibrium. The instantaneous sorption reaction in which fluoride ions adsorbed rapidly onto the surface of crushed burnt clay pot could be due to specific chemical interaction. The affinity and diffusive property could be contributing driving forces as well.

However, in the other three materials studied (Fig. 4), the increase in fluoride removal was gradual and this could be mainly the adsorption sites were few or not activated very well. Nevertheless an increase in fluoride removal at a retention time greater than 90 minutes was insignificant in all materials. At 120 minutes of contact time, equilibrium was attained as there was no further adsorption was observed to take place beyond that and thus 2 hour contact time was employed in all subsequent studies.

3.3. Effect of pH

Experiment was carried out to assess the effect of pH for the four adsorbent materials studied in a batch. The initial pH of water significantly played a major role and affected fluoride uptake capacity. Fig. 5 indicates the effect of pH and the optimum pH was 7 for all the materials except household ash. Higher adsorption capacity for household ash observed at lower pH values and the optimum pH was 4. The adsorption rapidly declined at pH greater than 7 for crushed burnt clay pot while for Keren and Adigerghish soils the decline was gradual. On the other hand, adsorption for household ash declined rapidly at pH greater than 4. Decrease in the adsorption efficiency might be due to the decrease in positive charges of the adsorbent and increase in competition of F^-. Similar remarks were made elsewhere [25,26] and the results of the experiment augmented those observations.

Nevertheless, adsorption is not just due to pH dependent but other mechanisms such as ligand exchange and complex-ion formation and precipitation may have played a role somewhere else. Hence this could affect the adsorption performances of the adsorbent to certain levels. This was in accordance to the observation made in certain studies [16].

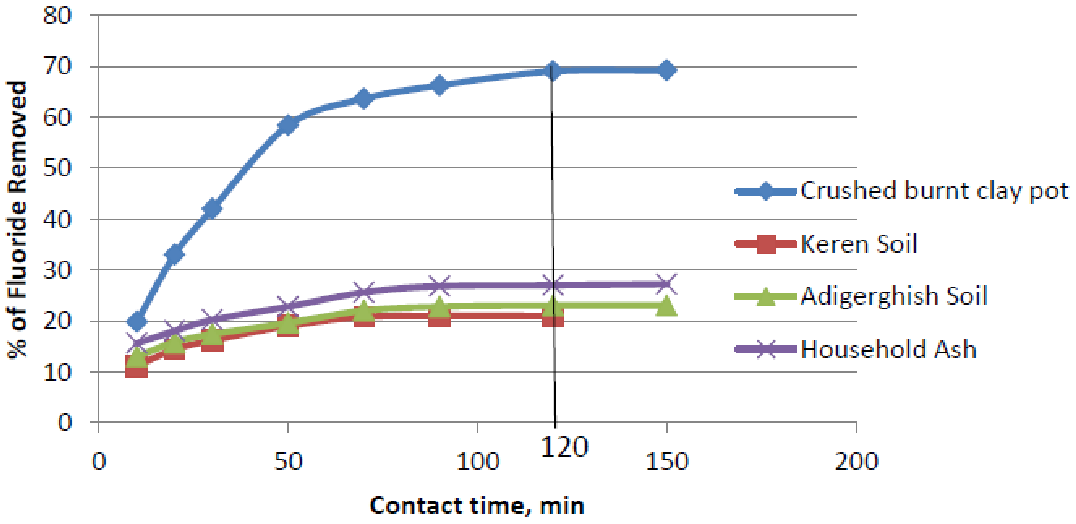

Figure 4: Effect of contact time on adsorption efficiency (Initial fluoride conc. 5.0 mg/L and adsorbent mass 7 g)

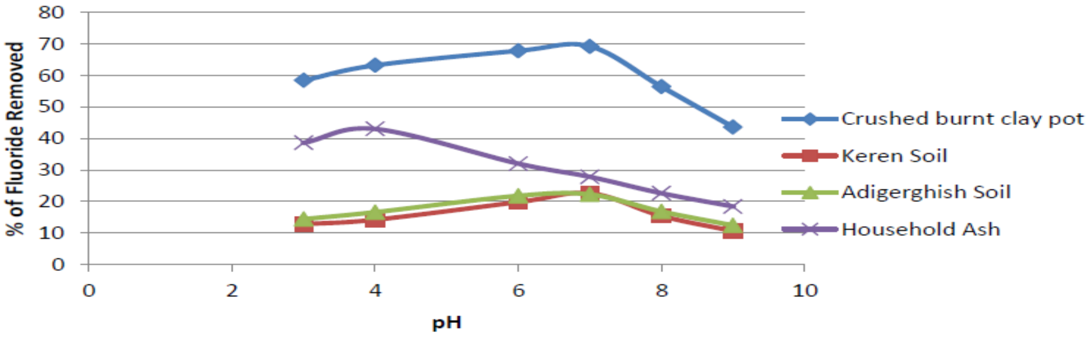

Figure 5: Effect of initial pH of raw water on fluoride removal (Initial fluoride conc. 5 mg/L & adsorbent mass 7g)

3.4. Effect of Particle Size

It is clearly shown in Table 1 that particle sizes ranging from 1.70 mm to 2.36 mm gave a lower adsorption capacity in the case of Keren and Adigerghish soils and more adsorption capacities was achieved for fine particle size (<0.15mm). However, adsorption capacity change was insignificant for crushed burnt clay pot with changing particle size. The table further shows that crushed burnt clay pot remained superior in removal capacity than the others. Effect of particle size for household ash was not studied as the household ash particles were very fine during sieve analysis and grading was not possible according to the sieve size analysis (0.15-2.36 mm) used in the study as indicated in Table 1.

Table 1: Effect of particle size on adsorption efficiency (Initial fluoride conc. 5 mg/L and adsorbent mass 7 g).

particle size	% Removal of fluoride		
(mm)	Crushed burnt clay pot	Keren Soil	Adigerghish Soil
<0.15	69.6	21.8	23.4
0.15-0.60	69.2	19.4	23
0.60-1.20	68.8	18.6	22.2
1.20-1.70	68.2	15.2	18
1.70-2.36	65.8	13.2	15.8

3.5. Effect of Initial Fluoride Concentration

A study was conducted to understand the effect of initial fluoride concentration on the adsorption efficiency. The experiment indicated that for a given 7 g mass of adsorbent, the fluoride removal efficiency increased gradually with increasing initial concentration. Fig. 6 indicates that the effect of initial fluoride and maximum removal efficiency was observed for crushed burnt clay pot followed by household ash. Increase in adsorption capacity with an increase in initial fluoride concentration could be mainly due to an increase in diffusion of fluoride to adsorption sites. Moreover, utilization of less accessible or less active sites of the adsorbent could occur when there was more fluoride in the solution to be taken up. Thus, this could increase the adsorption efficiency of the adsorbents. A similar study on different clay soils in Ethiopia was made [27] and thus this study supports the conclusion made by such reports.

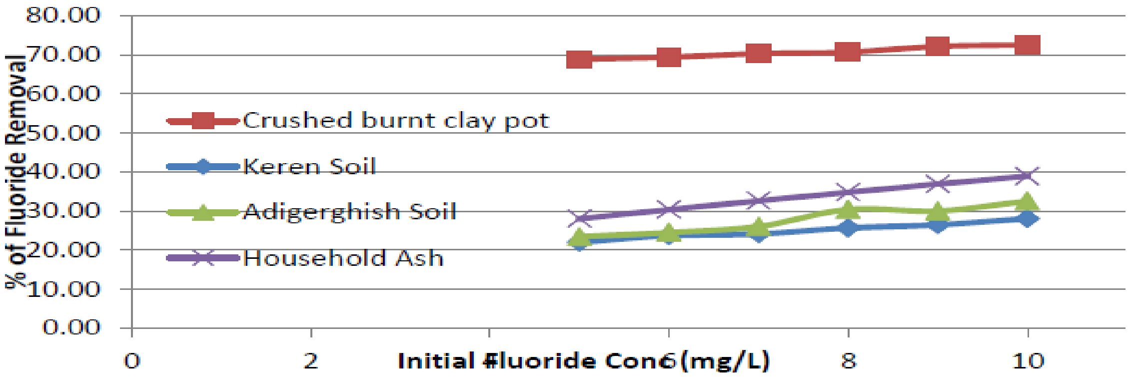

Figure 6: Effect of initial fluoride concentration on the adsorption efficiency (Adsorbent mass 7 g)

3.6. Selection of Adsorbent for Developing a Household Defluoridation Unit

The four local materials that have been studied in batch were compared by noting their adsorption capacities for fluoride uptake. As discussed before, the study indicated that crushed burnt clay pot had the highest removal capacity and could thus be selected for the development of household defluoridation unit. This is because crushed burnt clay pot as sorbent media could reduce the fluoride to an acceptable level with least adsorbent mass than the other three materials studied. However, there are other factors to be taken in to consideration before selection such as design simplicity, operation complexity of the unit and others. Moreover, the water quality of the effluent matters as well.

Water quality monitoring study was done to assess the effect of adsorbent addition to water for fluoride removal in the batch study. The water quality test was done for all the adsorbent media for selected water quality parameters such F-, pH, EC and turbidity and the results are presented in Table 2.

Table 2: Effect of adsorbent addition on water quality (adsorbent mass 10 g)

Type of adsorbent	Before adsorbent addition				After adsorbent addition			
	pH	EC	Turbidity	F^-	pH	EC	Turbidity	F^-
Crushed burnt clay pot	7.15	35 µS/cm	0.3 NTU	5 mg/L	7.25	45 µS/cm	90 NTU	< 1.5 mg/L
Kern soil	7.10	36 µS/cm	0.2 NTU	5 mg/L	7.28	48 µS/cm	105 NTU	> 1.5 mg/L
Adigerghish soil	7.20	38 µS/cm	0.1 NTU	5 mg/L	7.23	49 µS/cm	85N TU	> 1.5 mg/L
Household ash	7.16	35 µS/cm	0.2 NTU	5 mg/L	11.40	3.18 mS/cm	30 NTU	> 1.5 mg/L

It is evident from Table 2 that addition of adsorbents brought undesirable change in water quality and created more turbid water in all cases. Particularly a raise of pH and EC due to household ash was noted. However, fluoride levels were reduced to an acceptable level only in case of crushed burnt clay pot. Keeping other technical parameters the same for all, if selection of adsorbent was to be made based on the water quality of the effluent, none would be selected unless some means of improving the water quality was introduced. In spite of this, crushed burnt clay pot had shown a better removal capacity and EC and pH were within the permissible levels.

Nevertheless turbidity existed in all materials and hence the batch method of water defluoridation in this case could not be justified as the effluent water quality did not meet the WHO drinking water guideline. In spite of the development of turbid water, crushed burnt clay pot could be considered as a potential sorbent media for defluoridation because of its greater fluoride adsorption capacity, bringing fluoride from 5 mg/L to an acceptable level. Furthermore, the pH and EC of water remained unchanged before and after addition of crushed burnt clay pot and hence this could be an advantage over the other three materials which failed to bring the water fluoride level below 1.5 mg/L for the same mass of 10 g.

However, there could be many alternatives which could improve the turbidly of the water. A number of effective coagulants from plant origin have been identified elsewhere to enhance turbidity removal. Plant materials such as Nirmali, Okra, red bean, sugar and red maize [28], Moringa oleifera [29,30], cactus latifera, and seed powder of Prosopis juliflora [31] could remove turbidity to a greater extent and hence improve the water quality to an acceptable level.

Alternatively, the developed turbidity in water could be avoided by placing a filter media in a fixed bed. Therefore, taking all factors into consideration and the superiority of crushed burnt clay pot to remove fluoride, the media was thus selected for subsequent study for developing a household defluoridation unit based on a continuous flow adsorption column in a fixed bed depth. Water of desirable quality was then obtained satisfying the WHO guideline for drinking water and this was discussed in another paper elsewhere [9].

4. Conclusion

Batch Defluoridation study was carried out in fluorosis endemic places in Keren, Eritrea to assess low cost local adsorbents to take up fluoride from water. This study was done to find out fluoride sorbent materials which are easily available locally, cheap and able to reduce fluoride content to an acceptable level recommended by WHO. Four fluoride sorbent materials was identified and named crushed burnt clay pot, Keren soil, Adigerghish soil and household ash according to the place of origin. The optimum performances of the four adsorbent to take up fluoride were tested for the effect of amount of adsorbent, initial pH, contact time, particle size, and initial fluoride concentration. The initial pH of water played a major role and at pH of 7, crushed burnt clay pot, Adigerghish and Keren soil performed at optimum but the optimum pH for household ash was 4. However the contact time was similar for all and the equilibrium time was 120 minute (2 hours). Particle size had no significant effect in the case of crushed burnt clay pot, however, in other three materials, fine particles performed better than coarse particles. In all adsorbents studied, similar increasing trend in adsorption were observed when the initial fluoride concentration was increased. However, crushed burnt clay pot had the upper hand.

Fluoride adsorption comparisons were made and the study revealed that crushed burnt clay pot has superior capacity than the others. Its average fluoride removal capacity was about 0.26 mg fluoride/g medium. Others ranged from 0.08-0.1 mg/g for the same mass of 7 g and thus crushed burnt clay pot was selected for defluoridation.

Eritrea possibly has a number of places which are endemic to fluorosis. A thorough study to assess the fluoride levels of water at fluorosis endemic places is indeed needed. To put indigenous solution in such places, many locally available fluoride sorbent materials should be tested for water defluoridation so that rural community would be able to make choices based on their preferences, accessibility and affordability.

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