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REGENERATION OF FLUORIDE-SATURATED BONE CHAR BY MEANS OF WOOD ASH AND HEAT

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ABSTRACT: The regeneration of fluoride ion (F)-saturated bone char with different concentrations of wood ash in distilled water and by heating was investigated. Samples of 100 g of regenerated bone char were added into plastic containers containing 250 mL of natural water with 6.5 mg F/L collected from a borehole. The water was sampled at 30 min intervals for F analysis using an ion selective fluoride meter. The results indicated that the highest F removal efficiencies with the 2, 4, 6, and 8% wood ash concentrations were approximately 83, 84, 86, and 87%, respectively. The F removal efficiencies of the bone char when it was regenerated by heating for 3, 4, and 5 hr were 86, 89, and 89%, respectively. It is suggested that regenerating bone char with wood ash may be a better choice than regeneration with heating because the wood ash is locally available, easy to use in a household, and it does not demand an energy input. In contrast, regeneration by heating might encourage the cutting down of trees. We found that 2% wood ash, with a fluoride removal efficiency of 83%, is the best means for regenerating bone char because it can deliver an effluent with acceptable pH values for human consumption.

Keywords: Bone char regeneration; Defluoridation; Heat regeneration; Wood ash regeneration.

INTRODUCTION

High levels of the fluoride ion (F) in drinking water may cause fluorosis with serious health effects, including those on teeth and bones.[1,2] High concentrations of the F have been detected in the underground water in many countries, e.g., Tanzania and Kenya, and finding water that is safe for human consumption may be difficult.[3] In Tanzania, extremely high concentrations of up to 45 mg F/L have been reported in the Rift Valley area and the most affected regions are Mwanza, Mara, Shinyanga, Arusha, Kilimanjaro, and Singida.[4] Endemic skeletal and dental fluorosis are widespread in the East African Rift Valley regions, including Arusha, due to the high levels of F in the water sources.[5,6] Although the World Health Organization set, in 1984 and reaffirmed in 1993, a guideline of 1.5 mg F/L (1.5 ppm) as a “desirable” upper limit, it also allows countries to set Country Standards, their own national standards or local guidelines.[7] The limit of 1.5 mg F/L has been seen to be unsuitable in some countries and lower Country Standards have been set of 1 mg/L in India and 0.6 mg/L in Senegal, West Africa.[8,9] A rider to the Indian limit is “lesser the fluoride the better, as fluoride is injurious to health.”[8] However, the Tanzania Bureau of Standards (TBS) recommends for F in potable water a lower limit of 1.5 mg/L and a tolerable level 4.0 mg/L.[10] The reason in adopting such a high tolerable value in Tanzania is based on the difficulties complying with the WHO guideline in a country with regionally high F concentrations and problems with water scarcity.[11]

F from drinking water can be removed by various treatment technologies, based on the principles of the ion-exchange/adsorption process, including by coagulation

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and precipitation,[12,13] adsorption,[14] and the addition of chemicals to cause precipitation.[15] Among these methods, adsorption is preferable to the other F removal techniques because of its low cost, flexibility and simplicity of design, and its ease of operation and maintenance.[16] Adsorption technology using bone char at the household level has been demonstrated to be able to provide safe water in fluorotic areas in developing countries.[17] The adsorption method with bone char is both economical and practicable because the media required can be obtained locally and are easy to use at both household and community levels, while for the other technologies the necessary defluoridation media are expensive and technically non-feasible.[18] Although the bone char material gets exhausted when used continuously, studies indicate that it can be regenerated before it is dumped.[19,20] The objective of the present study was to investigate whether a cheap, simple, suitable, and environmentally friendly method for regenerating F-saturated bone char for the effective defluoridation of water supplies to acceptable standards could be developed with the use of wood ash suspensions or heating.

METHODS

Preparation of the bone char: The fresh brown-gray bone char used for the community and household filters commercially available at the Ngurdoto Defluoridation Research Station (NDRS) was utilized. The collected cattle bones were heated in a local kiln with limited supplies of oxygen to high temperatures of 500–550ºC using wood charcoal. By this means, the heating makes the bone char hygienically permissible for the defluoridation of water. Moreover, it ensures the removal of the organic part of the bone that adds taste and color to the water but does not take part in the F release. Therefore, producing high quality bone char depends on closely following the guidelines for the temperature and oxygen content in the furnace.

Preparation of the wood ash: The wood ash left after combustion of charcoal and wood in a home fireplace was used. Its chemical composition was not investigated in this study. Fine wood ash particles, 0.063 mm or less in size, were used for the preparation of the wood ash suspensions.

Preparation of the wood ash suspension: Different concentrations of wood ash suspension were prepared by adding 20, 40, 60, and 80 g of wood ash, respectively, to each beaker in four sets of four 1000 mL plastic beakers, each of which contained 1000 mL of natural water from the Nganana borehole, and then constantly stirring the beakers. Plastic, rather than glass, beakers were chosen to reduce possible F adsorption by the beakers. The apparatus was cleaned with distilled water before and after use. Thus 4L of each wood ash suspension concentration, 2, 4, 6, and 8%, respectively, were prepared.

Regeneration of the F-saturated bone char with wood ash: A sample of 5 kg of F-saturated bone char, with particle sizes ranging from 0.5 to 3 mm in diameter, from the bone char filter was transferred into a 7 L plastic bucket and rinsed with clean water. Then, 1000 mL of water was poured into the plastic bucket with the F-saturated bone char and shaken to get rid of all the free fluoride ions. Lastly, the rinsed F-saturated bone char from the plastic bucket was dried on a sheet ready for regeneration. After being dried, the F-saturated bone char was put in a clean 7 L plastic bucket. Then, 3000 mL of wood ash suspension was added to the 7 L plastic bucket with the bone char media. The mixture was stirred/shaken periodically for two days, then cleaned with safe water, turned out of the bucket, and left to be dried. The bone char was then re-packed in the filter ready for the defluoridation process.

Regeneration of the F-saturated bone char with heat: A sample of 5 kg of F-saturated bone char from the bone char filter was transferred into a 7 L plastic bucket and rinsed with clean water. Then, 1000 mL of water was poured into the 7 L plastic bucket with the F-saturated bone char and shaken to get rid of all the free fluoride ions. Lastly, the rinsed F-saturated bone char from the plastic bucket was dried on the sheet ready for regeneration. After being dried, the 5 kg of F-saturated bone char was put into a pot, 1000 mL of clean water was added, and the mixture was heated for 3, 4, or 5 hr while being stirred/shaken. The mixture was then washed with distilled water, removed from the pot, and left to dry. The bone char was then re-packed in the filter ready for the defluoridation presses.

Determination of the F concentrations: The concentration of fluoride ions in the solutions was established using the Mettler Toledo Seven Compact pH ion meter and reference fluoride electrode. Reference standards with 1 and 10 mg F/L were prepared from the appropriate dilutions of a stock solution of sodium fluoride (NaF) with 100 mg F/L and Total Ion Strength Adjusting Buffer (TISAB) solution. A water sample of 5 mL was pipetted and transferred into a 25 mL plastic beaker. The pipette was rinsed with distilled water and 5 mL of TISAB were then pipetted and transferred to the plastic beaker containing the water sample. The magnetic stirrer and the fluoride electrodes were immersed into the mixed samples and stirred slowly. The Mettler Toledo Seven Compact pH ion meter was switched on in order to read the millivolts when a steady state was reached. The TISAB buffer was added before the measurement to achieve a constant pH and to break up fluoride complexes. The measurement of the pH value was done with a HI 99121 pH meter with a HI1292 electrode.

F removal efficiency: 100 g bone char samples regenerated by 2, 4, 6, and 8% wood ash suspensions and by heating, respectively, were put in different plastic containers. A sample of 250 mL of borehole water from Nganana with 6.5 mg F/L was added to each container and the mixture left for a contact time of 30 min. A water sample from each container was analyzed for F until a concentration of <1.5 mg F/L was reached before adding more borehole water for treatment. The volume of water able to be treated to 1.5 mg/L was computed for all samples.

The defluoridation capacity of the regenerated bone char was computed by the initial and residual F concentrations of the water and the mass of the bone char used by the equation:

$$DC_{FC} = \frac{S_0 - S_t}{X_{fc}}$$

where:

DC_{FC} = Defluoridation capacity of the regenerated bone char (mg F/g)
S_0 = Initial fluoride concentration (mg F/L)
S_t = Residual fluoride concentration after contact time t (mg F/L)
X_{fc} = Regenerated bone char media (g/L)

The F removal efficiency of the regenerated bone char was calculated from the equation:

$$Q_t = \frac{S_0 - S_t}{S_0} \times 100$$

where:

Q_t = Fluoride removal efficiency of the regenerated bone char (%)
S_0 = Initial fluoride concentration (mg F/L)
S_t = Residual fluoride concentration after contact time t (mg F/L)

RESULTS

Regeneration of F-saturated bone char using wood ash: The regeneration of the F-saturated bone char, by releasing F from it, by using the 2, 4, 6, and 8% wood ash suspensions is shown in the Figure 1. The wood ash suspension was changed every hour. The baseline F values for the bore water (6.5 mg F/L) and the wood ash suspension (0.47 mg F/L) are not included in the results shown in Figure 1.

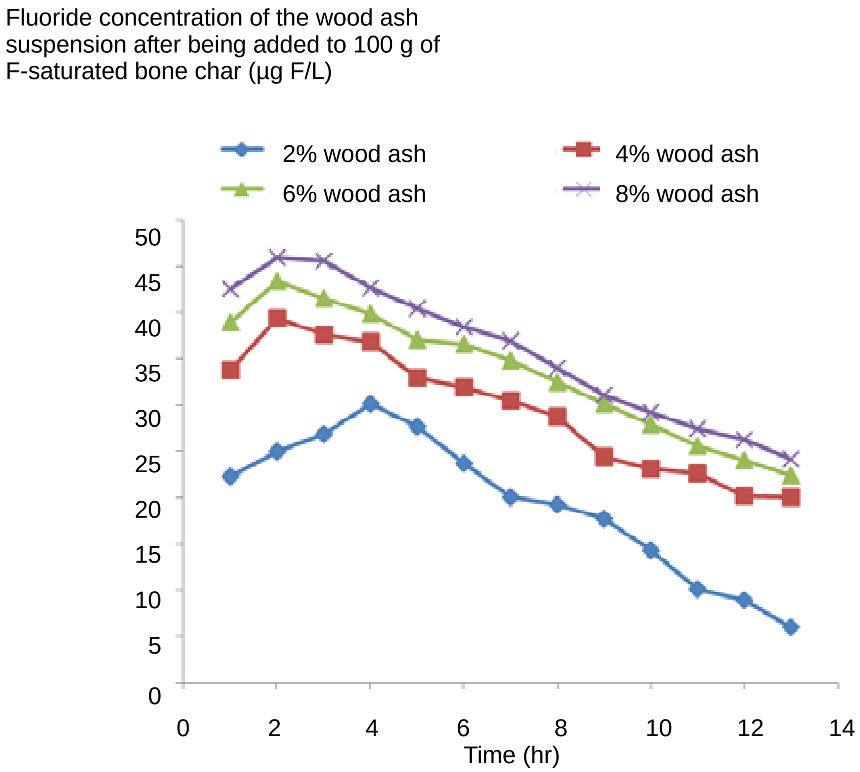

Figure 1. The fluoride concentrations (µg F/L) in the 100 mL of ash suspensions of different strengths after being added to 100 g of F-saturated bone char.

It is noted that the F released from the F-saturated bone char increased with increasing concentrations of wood ash suspension. The rate of F release increased rapidly initially and then gradually began to fall. The F concentrations in the wood ash suspension during the 2nd hr were 24.95, 39.4, 43.4, and 49.01 µg/L for the 2, 4, 6, and 8% wood ash suspensions, respectively, while for the 13th hr they were 6.05, 20.08, 22.42, and 24.15 µg/L, respectively. The level of F in the wood ash suspension prior to adding the F-saturated bone char was 6.97 mg F/L, with the contribution from the borehole water being 6.5 mg F/L and that from the wood ash being 0.47 mg F/L.

Effect of temperature on the regeneration of F-saturated bone char using wood ash: The effect of temperature, in the range of 20–100°C, on the regeneration of F-saturated bone char with wood ash is illustrated in Figure 2.

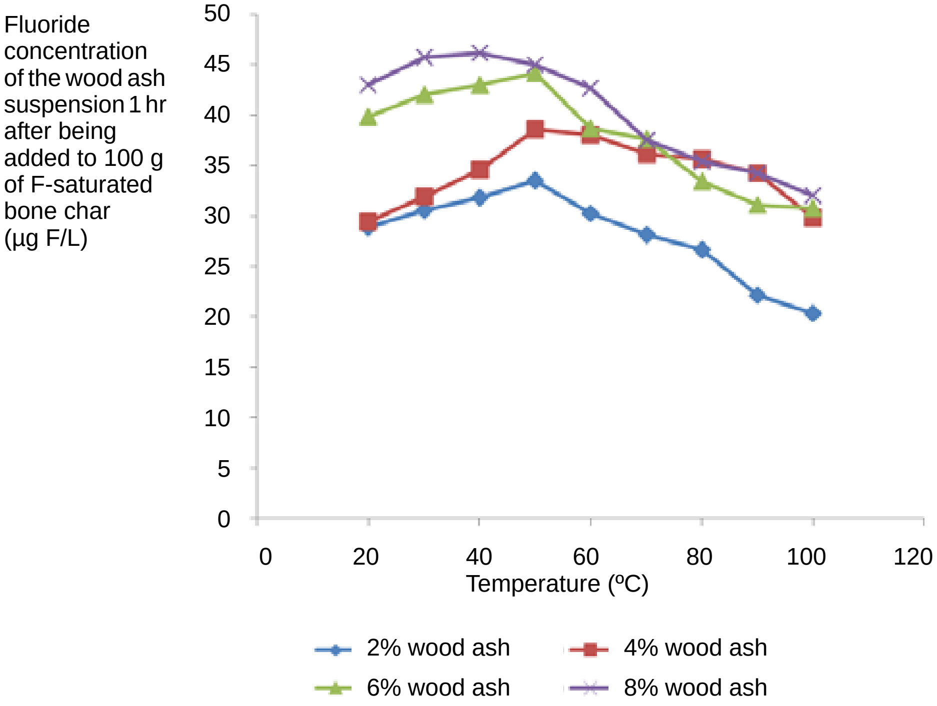

Figure 2. The effect of temperature on the fluoride concentrations (µg F/L) in 100 mL of ash suspensions of different strengths after 1 hr of contact with 100 g of F-saturated bone char.

The maximal F release from the F-saturated bone char was found to occur at 50°C with the maximum concentrations of F in the wood ash suspension being was 33.47, 38.55, 44.12, and 46.08 µg/L for the 2, 4, 6, and 8% wood ash suspensions, respectively. A marked drop in F removal was observed at 100°C with the concentrations of F in the wood ash suspension being 20.33, 29.8, 30.76, and 32 µg/L for the 2, 4, 6, and 8% wood ash suspensions, respectively. However, the study showed no significant difference in the F released with the different concentrations of wood ash suspension used at the specified temperatures ($p > 0.05$).

Effect of the duration of heating on the regeneration of F-saturated bone char with heat: The effect of the duration of heating, in the range of 3–5 hr, on the regeneration of F-saturated bone char with heat is illustrated in Figure 3.

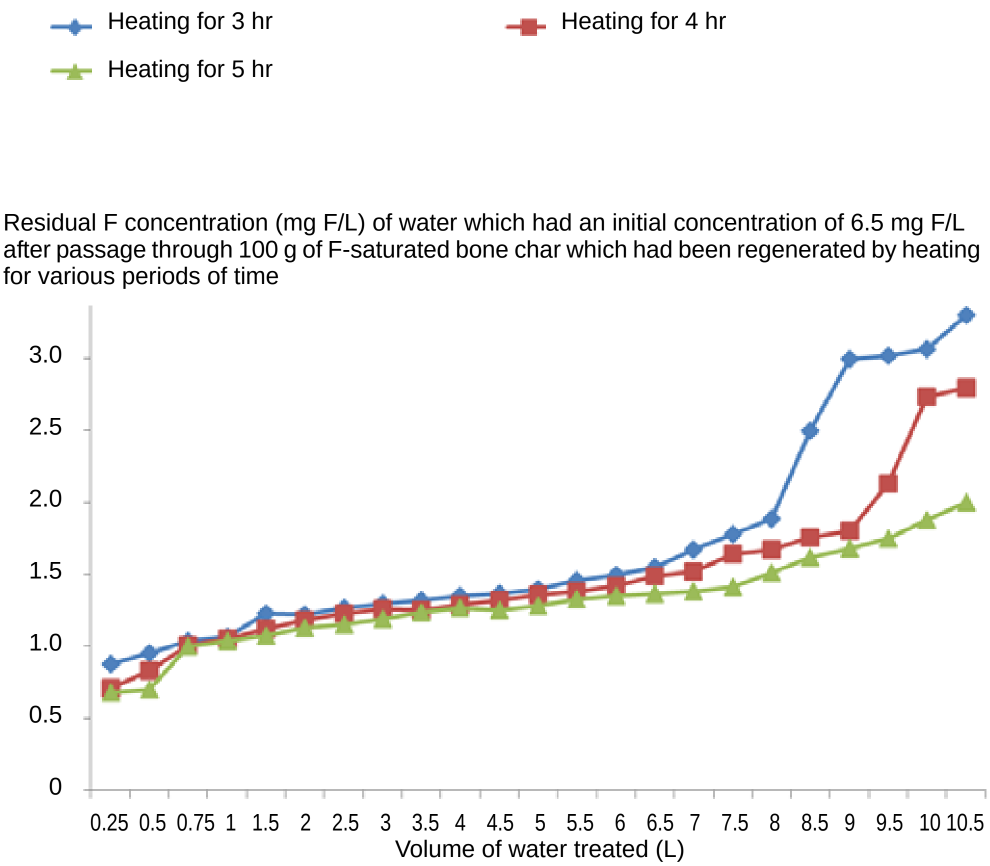
Residual F concentration (mg F/L) of water which had an initial concentration of 6.5 mg F/L after passage through 100 g of F-saturated bone char which had been regenerated by heating for various periods of time

Figure 3. The residual F concentration (mg F/L) of water which had an initial concentration of 6.5 mg F/L after passage through 100 g of F-saturated bone char which had been regenerated by heating for 3, 4, and 5 hr.

The degree of regeneration of the F-saturated bone char from heating increased as the heating time increased. The highest defluoridation capacities were 0.562, 0,0579, and 0.0582 mg F/g bone char when 250 mL of borehole water were treated with regenerated bone char that had been heated for 3, 4, and 5 hr respectively. The corresponding reductions in the borehole water from the initial concentration of 6.5 mg F/L, were to 0.88, 0.71, and 0.68 mg F/L, respectively. However, the differences in the defluoridation abilities of the bone char regenerated with heat for 3, 4, and 5 hours were not significant ($p > 0.05$).

F removal efficiency of F-saturated bone char regenerated with heat: Heating affected the physical properties and F binding of the F-saturated bone char with the F removal efficiency increasing as the duration of the heating increased. The highest F removal efficiencies were 86.4, 89.0, and 89.54%, for F-saturated bone char regenerated by heating for 3, 4, and 5 hr, respectively. The maximum volumes of water able to be treated from an initial F concentration of 6.5 mg F/L to a residual F concentration of 1.5 mg F/L with bone char regenerated by heating for 3, 4, and 5 hr were 5500, 6500, and 7500 mL, respectively.

Defluoridation ability of F-saturated bone char regenerated with wood ash: The fluoride removal ability of the F-saturated bone char regenerated with wood ash suspensions increased as the concentrations of wood ash increased (Figure 4).

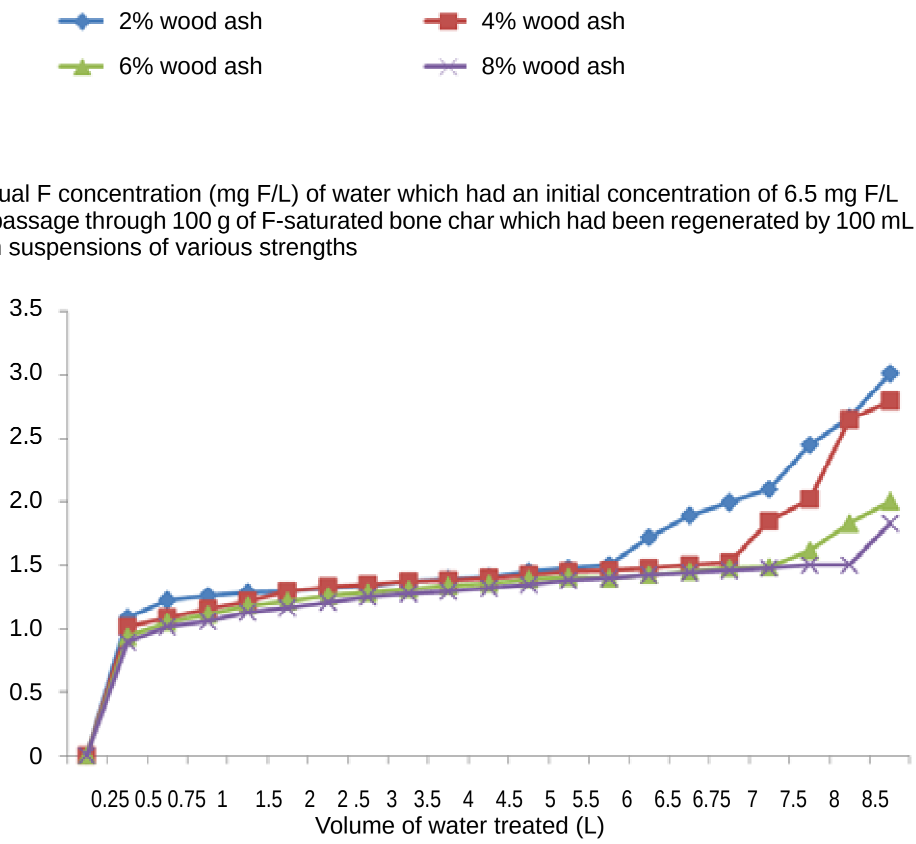
Residual F concentration (mg F/L) of water which had an initial concentration of 6.5 mg F/L after passage through 100 g of F-saturated bone char which had been regenerated by 100 mL of ash suspensions of various strengths

Figure 4. The residual F concentration (mg F/L) of water which had an initial concentration of 6.5 mg F/L after passage through 100 g of F-saturated bone char which had been regenerated by 100 mL of ash suspensions of various strengths.

As the volume of water being treated by the 100 g of regenerated bone char increased, the F level to which the borehole water, with an initial F concentration of 6.5 mg/L, was lowered increased. The highest defluoridation capacities were 0.541, 0,0548, 0.056, and 0.0556 mg F/g bone char when 250 mL of borehole water were treated with regenerated bone char that had been treated with 2, 4, 6, and 8% wood ash suspensions, respectively. The corresponding reductions in the F level in the borehole water, from the initial concentration of 6.5 mg F/L, were to 1.09, 1.02, 0.94, and 0.90 mg F/L, respectively. The maximum volumes of water able to be treated from an initial F concentration of 6.5 mg F/L to a residual F concentration of 1.5 mg F/L with the 2, 4, 6, and 8% wood ash suspensions were

5500, 6500, 7000, and 7500 mL, respectively. The defluoridation capacities corresponding to treating these maximal volumes were 0.05, 0,05, 0.0501, and 0.05 mg F/g bone char for the 2, 4, 6, and 8% wood ash suspensions, respectively. The reason for the rapid declines in the F removal capacity with lower wood ash suspension concentrations, as depicted in Figure 4, is that the number of vacant sites offered by the reconstituted bone char for the sorption of F increases with the use of higher wood ash suspension concentrations. Thus, the 8% wood ash suspension would provide more of the hydroxyl/carbonate ions required for the exchange with fluoride ions on saturated bone char compared to those provided by the 6%, 4%, and 2% wood ash suspensions.

F removal efficiency of F-saturated bone char regenerated with wood ash: The efficiency the of wood ash in regenerating the F-saturated bone char increased with increases in wood ash concentrations and decreased with decreases in the contact time. The maximum F removal efficiencies were 83.23, 84.31, 85.54, and 86.15% for the 2, 4, 6, and 8% wood ash suspensions, respectively. The lowest F removal efficiencies were 76.92, 76.92, 77.07, and 76.92%, for the 2, 4, 6, and 8% wood ash suspensions, respectively. The reduction in the maximal F removal efficiency with lower concentrations of wood ash suspension can be considered to be due to fewer hydroxyl/carbonate ions being released for exchange with fluoride ions attached to the bone char.

DISCUSSION

The fluoride ion is known to exchange with hydroxyl, carbonate, hydrogen carbonate, and phosphate ions of bone char during F removal from water using bone char.[9] When fluoride or carbonate ions are in contact with apatite mineral, like the hydroxyapatite minerals of bone and enamel, they can substitute for the hydroxyl ion and form a more acid resistant structure, fluoroapatite.[4]

$$Ca_{10}(PO_4)_6(OH)_2 + 2F^- \rightarrow Ca_{10}(PO_4)_6F_2 + 2OH^-$$

With fluoroapatite being more resistant to acid, it offers a strong protective layer to the tooth enamel against the development of dental caries compared to hydroxyapatite. However, an excessive F intake may result in the reaction going beyond the replacement of hydroxide, and lead to the formation of calcium decafluoride which is a very hard, brittle material and not appropriately suited for the functions of the skeletal structure.[21]

$$Ca_5(PO_4)_3F + 9F^- \rightarrow Ca_5F_{10} + (PO_4)^{3-}$$

The findings of the present study show that the F removal efficiencies and the F removal capacities of bone char regenerated with wood ash at room temperature and other temperatures, and regenerated with heat were not significantly different. However, the release of F from F-saturated bone char is dependent on the concentration of the wood ash suspension, the temperature, and the duration of the contact.

The rate of releasing F from the F-saturated bone char using wood ash suspension was more rapid in the initial stages of application and then decreased significantly. This may be due to instantaneous sorption reactions in which hydroxyl/carbonate ions were adsorbed/ exchanged rapidly on to the surface of the F-saturated bone char. The hydroxyl/carbonate ions required to exchange with fluoride ions from the F-saturated bone char were much more available during the initial stages compared to the last hours and this may be due to a decrease in the number of sorption sites on the bone char.

At a 95% confidence interval level, the differences in the regenerated bone char at room temperature for the 2 vs. 6%, 2 vs 8%, and 4 vs 8% wood ash suspension concentrations were statistically significant ($p<0.05$) while the differences for the 2 vs. 4% and 6% vs. 8% wood ash suspension concentrations were not ($p>0.05$). This may be due to an increased availability and diffusion of hydroxyl/carbonate ions from wood ash as the concentration increases. These ions release fluoride ions from the F-saturated bone char by an ion-exchange mechanism and physical adsorption on to the activated carbon present on the bone char. The maximum F releases ranged from 30–46 µg/L and the lowest was from 6–24 µg/L as depicted in Figure 1. Therefore, the option of using wood ash to regenerate F-saturated bone char in bone char filter in both households and the community may be worth further study.

At the optimum temperature, more hydroxyl/ carbonate ions were released from the wood ash and diffused faster into the bone char structure by replacing fluoride ions attached to the bone char by an ion-exchange mechanism or by adsorption on to the surface of the bone char. A similar observation was reported by Kanyora et al.[22] At higher than optimal temperatures, the release of F from the bone char was probably lowered as the bone char was starting to attain equilibrium. In addition, there may have been a lower availability of the hydroxyl/carbonate ions required for the substitution with fluoride ions in the bone char. The highest level of F release from the F-saturated bone char was with the 8% wood ash suspension and ranged from 33–46 µg/L while the lowest level of F release was the 20–32 µg/L obtained with the 2% wood ash suspension (Figure 2). However, no significant differences were present at the 95% confidence interval, at the varying temperatures, when comparing bone char regenerated with the 2 vs 4%, 2 vs 6%, and 2 vs 8% wood ash suspension concentrations.

The principal reactions in the F-saturated bone char during its regeneration with heat treatment were:

$$Ca_{10}(PO_4)_6F_2 + 2OH^- \xrightarrow{heat} Ca_{10}(PO_4)_6 + 2HF\uparrow + O_2\uparrow$$

$$Ca_{10}(PO_4)_6F_2 + 2OH^- \rightarrow Ca_{10}(PO_4)_6(OH)_2 + 2F^-$$

Heat had effects on the physical structural and binding process of F of the F-saturated bone char. Heating might result in fewer sorption binding sites, and thus limit the F uptake by the bone char material. Regeneration of F-saturated bone char by heating at higher temperatures may damage the hydroxyapatite structures. Similar outcomes were published by Kaseva[19] and Naohito et al.[23] Thus, more investigation is required on how heat can best regenerate F-saturated bone char.

CONCLUSION

The Nganana borehole, in a case study area, has 6.5 mg F/L which is higher than the tolerable level for potable water recommended by the Tanzania Bureau of Standards of 4.0 mg/L This high level adversely affects the health of the villagers using the water. The present study has shown that the use of a 2% wood ash suspension is a very effective and cheap method for F removal by regenerating F-saturated bone char. F-saturated bone char regenerated with a 2% wood ash suspension has the capacity to reduce the F concentration in water from 6.4 mg/L to 1.09 mg/L with a F removal efficiency of 83%, which is well within the acceptable standards for domestic water use. Wood ash is produced daily in every household at Meru district, which shows that it can be a cheap method for the regeneration of F-saturated bone char for treating the high F water in the study area.

In addition, this study gives hope for the attempts to use wood ash for the regeneration of F-saturated bone char, particularly to the villagers in the Meru district community. Additional communities all over the world affected by comparable problems could evidently also benefit. Hopefully, this method for the regeneration of F-saturated bone char may be advanced further based on the results provided in this study. A household bone char filter, when repacked with regenerated bone char, should be used for three months, as described by the manufacturer, and for only one bucket (20 L) of borehole water per day. The water to be defluoridated should have a maximum F content of 6.5 mg/L.

The utilization of wood ash as a means of regenerating F-saturated bone char appears to be a very hopeful method. There is a need for further research on various chemical compositions to improve the problems associated with chemical residues that impact on the quality of water used for human consumptions.

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