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Full Length Research Paper

Practical Approach for Removal of Natural Organic Matter and Defluoridation of Maji ya Chai River Water: Use of Acid Pre-treated Bone Char and Coagulants

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ABSTRACT

The removal of natural organic matter (NOM) and fluoride from Maji ya Chai River water (MCRW) using bone char (BC) and coagulants was investigated. Fine BC particles ranging from 0.045 to 0.2 mm were used in a 1 litre half-filled container, stirred at 300 rpm for 60 min followed by addition of a commercial coagulant and allowed to settle for 24 hours to remove NOM. MCRW practicable and affordable defluoridation method for removal of NOM through defluoridation with BC pre-treated with 0.3 N HCl followed by coagulation with Poly-aluminium chloride (PACl) is proposed. Reduction of fluoride from 21 mg/L to 1.7 mg/L was achieved by use of 5 g/L of 0.3 N HCl acid pre-treated BC and 66 mL of 1% PACl per litre of MCRW. The acid pre-treated BC was found to double defluoridation capacity of BC, while adjustment of pH of MCRW with HCl acid to pH 3 has tripled defluoridation capacity of BC. The 0.3 N HCl acid pre-treated BC is preferred to avoid impractical working with acid solution for pH adjustment in households and communities. The treated water pH was within the acceptable portable water range of 6.5 to 8.5. PACl is a preferable coagulant for removal of NOM from MCRW since it is more effective and safer compared to use of alum and zetafloc 703.

Keywords: *Coagulants, Defluoridation, Fluoride removal capacity, Maji ya chai, NOM removal, Pre-treated bone char.*

INTRODUCTION

Maji ya Chai River (MCR) is located in Arumeru district which is among the areas where most of water sources contain fluoride above the acceptable limit (URT-Ministry of Water, 2013; Chen and Schäfer, 2015). The features and characteristics of Maji ya Chai River ("tea river" in Swahili language) have been detailed by Aschermann *et al*. (2016). The Maji ya Chai River water (MCRW) has natural organic matter (NOM) throughout the year, which provides its tea like colour (Aschermann *et al*., 2016). The characteristics and seasonal variation of the MCRW was studied by Aschermann *et al*. (2016) while Chen and Schäfer (2015) investigated NOM, which was measured as total organic carbon (TOC). The concentration of TOC observed for the MCRW was 35 mg C/L (Chen and Schäfer, 2015) which is quite high compared to the EPA guideline of 2 mg C/L of water. The MCRW measured fluoride content was 20.9 mg/L, which is high compared to the WHO drinking water guideline of 1.5 mg F/L and 4 mg F/L, respectively.

NOM can effectively be removed by membrane filtration (Chen and Schäfer, 2015) or coagulation (O'Melia *et al*., 1999; Cheng and Chi, 2002; Liu *et al*., 2009; Matilainen *et al*., 2010). Membrane filtration removes NOM in water by allowing the small molecules of water to pass through the membrane while excluding humic and fulvic acids large molecules which are the main constituents of NOM. Depending on the type of membrane (nanofiltration, ultrafiltration, or osmotic), other water contaminant species can be removed as well depending on their size. The use of membrane filters necessity to have a pumping power, to overcome transmembrane pressures (Aschermann *et al*., 2016), hence high cost of membrane filters and use of pump will be practically a challenge for the technology to be applied in the rural areas. Table 1 shows capabilities of separation of different types of membranes and operation pressure (van Rijn, 2004).

Table 1: Pressure driven membrane process, their properties and application (van Rijn, 2004)

Process	Pore size (nm)	Pressure (MPa)	Separation capability/Application
Microfiltration	50-5000	0-0.3	Retention of bacteria, colloids, protozoa. Used for water treatment, beverage clarification
Ultrafiltration	5-100	0.05-0.5	Retention of viruses, bacteria and dissolved substances. Used in water purification, pharmaceutical industries and food industries.
Nanofiltration	1 -10	0.5-2.5	Separation of low MW substances (200-300 Da) and divalent salts
Molecular sieving	0.3 - 1	1.5-10	Retention of all dissolved ions. Used for gas separation

For removal of both NOM and fluoride from MCRW nanofiltration (NF) or reverse osmosis (RO) will be required as studied by Shen and Schäfer (2015). This approach will be costly since membranes are not easily available. For a sustainable technology for rural communities of developing countries such as Tanzania, consumables to be used should be affordable and easily available. A practical approach is therefore, provision of a technology which can be used in the rural communities without requiring highly trained personnel at affordable costs.

The Use of Bone Char and Coagulants for Defluoridation and NOM Removal

Bone char (BC) has been shown to defluoridate water sufficiently and its standard preparation procedure have been developed (Mbabaye *et al*., 2017). However, BC does not remove NOM, thus its use has to be in combination with other methods for removal of NOM. An overview of different coagulants which can be used for coagulation of NOM have been discussed (Matilainen *et al*., 2010). The mostly used coagulants are alum and poly-aluminium chloride (PACl) (Wang *et al*., 2004). Alum has the advantage of being relatively cheaper than PACl, but it has a disadvantage of having high coagulant residuals in purified water which can cause Alzheimer's disease (Matilainen *et al*., 2010). On the other hand, PACl has the advantage of being less sensitive to temperature and pH compared to alum, better NOM removal capacity and less sludge is produced (Matilainen *et al*., 2010). Table 2 gives comparison of the functional parameters of alum and PACl coagulant reported by Zouboulis *et al*. (2008).

Table 2: Comparison of functional parameters of alum and PACl coagulants (Zouboulis *et al*., 2008)

Parameter	Alum	PACl
Temperature	Alum hydrolysis and production of the positively charged hydroxyl complexes, responsible for the coagulating colloidal impurities of natural waters, are affected by temperature.	PACl is less affected with temperature since it contains pre-polymerised forms of aluminium.
pH	PH range controls the types of hydroxyl species of aluminium produced.	pH has less impact on the species produced as it contains pre-polymerized forms of aluminium.
Aluminium species	Mostly the aluminium monomeric hydroxyl species produced have cationic charge ranging from +1 to +3.	Both polymeric and monomeric species are formed. Al_{13} species (7+) is formed in relatively high concentration.
Kinetics	Coagulation kinetics are slower	Coagulation kinetics are faster.

Defluoridation of Water for Households and Communities

The Ngurdoto Defluoridation Research Station (NDRS) has been involved in development and distribution of Community Defluoridation Unit (CDU) and Household Defluoridation Unit (HDU) since 1990s (URT, 2013[a]; Dahi, 2013). Up to 2009, 160 HDU units were supplied by NDRS to NGOs for use in Kimosoni and Nyamakata villages in Meru district and by year 2013, 14 HDU units were being monitored by the NDRS (URT-Ministry of Water, 2013). The latest HDUs are based on a bucket and a BC column (Figure 1). The developed HDUs are used for a period determined by the amount of BC loaded in the unit and other parameters as defined by Equation (1).

$$C = \frac{10^3 HI}{BDG} \qquad (1)$$

Where B is water demand per person per day (L/(person/day)), C is a period of operation in days, D is the number of users (persons), G is the amount of fluoride removed (g/L), H is BC fluoride sorption capacity (g/kg), and I is the mass of BC required (kg). The HDU bucket type developed unit has a BC loading capacity of 4 kg.

Alternative Approach for Household Defluoridation and NOM Removal

The objective of this study was to illustrate a possibility of using an acid pre-treated bone char in combination with PACL, a commercially available water treatment coagulant to remove both fluoride and NOM in MCRW by a simple method of mixing and settling. This approach is practical in the sense that, one will only need a bucket filled with water to which the pre-treated BC will be stirred in and later addition of a coagulant, PACl, then allowing the water to settle to get clarified water free of fluoride and NOM. In this case a household will not be required to possess or procure an HDU.

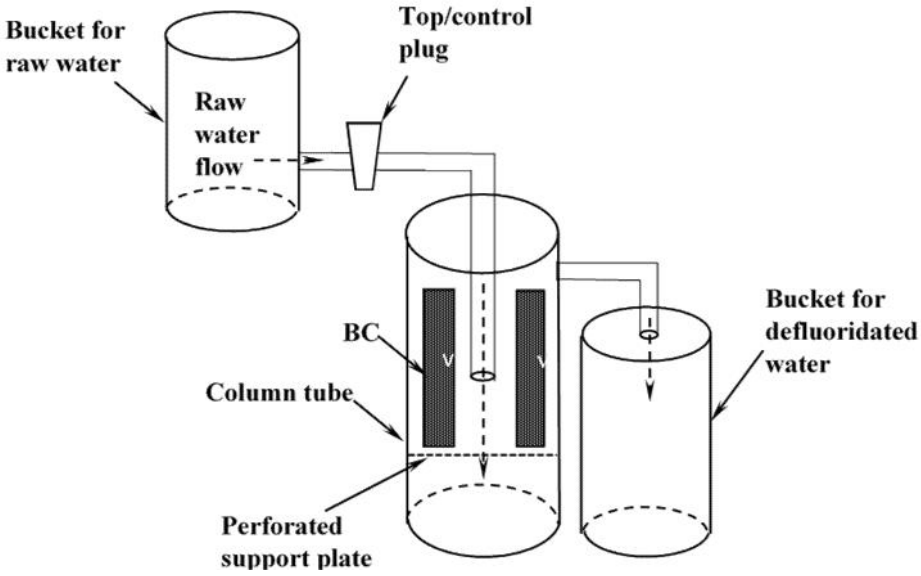

Figure 1: A schematic HDU made of buckets and a column (adopted from URT-Ministry of Water, 2013a)

METHODS AND MATERIALS

Bone Char Preparation

Bone char (BC) used was calcinated at 400^{o}C using the procedure developed by Mbabaye *et al*. (2017). BC particle size used were 45 µm < d < 200 µm, which is not suitable for utilization when defluoridation is done in a parked column, such as HDU, as they create high resistance to flow leading to column clogging (Mbabaye *et al.*, 2017). However, particles of this size were found to settle quickly when stirred in a defluoridation container. Bone char particles of less than 45 µm were sieved out as they showed slow settling rate. The use of particle size (45 µm < d < 200 µm) in this work is meant to utilize the BC waste resulting from screening of crushed calcined BC to obtain particle size relevant for HDU and CDU columns developed by the NDRS. Figure 2 shows the distribution of the BC particles sizes used as measured based on the percentage cumulative by volume using Malvern Master Sizer 2000.

Bone Char Acid Pre-treatment and MCRW pH Adjustment

Acid treated BC was prepared by soaking and mixing BC in solution of 0.01 N, 0.1 N, 0.3 N and 0.5 N HCl at a ratio of 1:1.5 (BC: HCl acid solution) weight ratio for 24 hours. MCRW pH adjustment to pH 3 or pH 7 was achieved by using a 0.1 N HCl while monitoring the pH with Sartorius PT-15 pH meter.

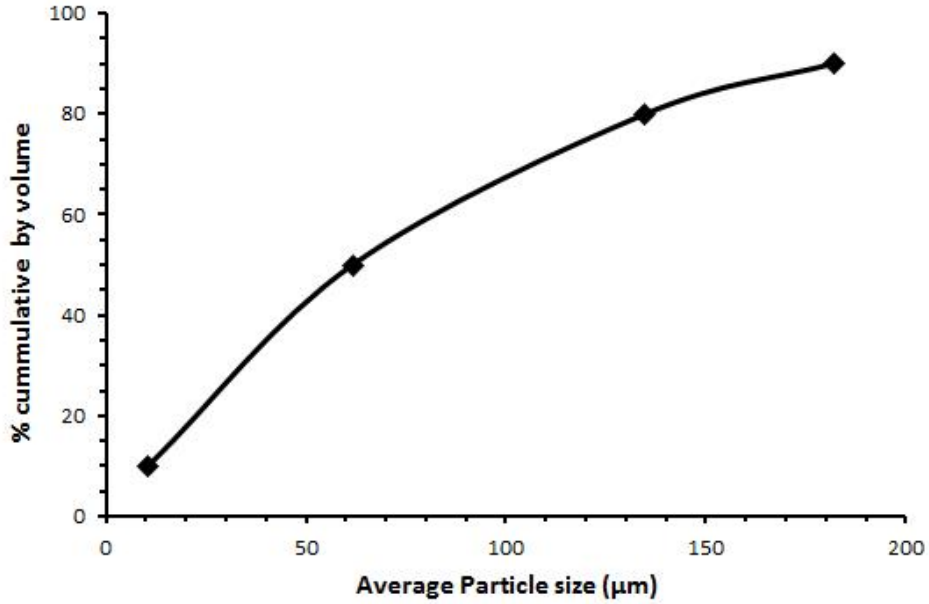

Figure 2: BC cumulative particle size distribution as determined by Malvern Master Sizer Instrument

Collection of Water Samples

Maji ya Chai River water (MCRW) sample was collected along the Maji ya Chai River (MCR) at Maji ya Chai township in Arumeru district (geo coordinates -3.37111 N, 36.89639 E and altitude 1162.23 m) in November 24, 2016 after a short rain. The Maji ya Chai River catchment map has been documented by Aschermann *et al*. (2016). The population of Maji ya Chai ward in Meru district, Arusha region was 29,313 (URT, 2013b) with an average household population of 4 persons. The river flows along different villages and is used as source of water for activities, such as, washing and gardening excluding household cooking and drinking because of high presence of fluoride and NOM. Three plastic buckets of 20 litres were used to collect the MCRW at once.

Defluoridation with Bone Char and Coagulation of NOM with Coagulants

Defluoridation experiments were carried out using either acid pre-treated or untreated BC by stirring rapidly (300 rpm) for 5 min, known weight (2 – 8 g BC/L of water) of fine BC particles ranging from 0.045 to 0.2 mm followed by slow stirring at 40 rpm for 60 min in a half-filled 1 litre container. The MCRW used was either raw or pH regulated. Since BC does not remove NOM, coagulation process was used to remove NOM using commercial coagulants (PACl, alum, and Zetafloc 703), which were obtained from Junaco Group of Companies in Dar es Salaam. Coagulation tests for removal of NOM were conducted using jar tests. A volume of 66 ml of 1% coagulant solution was used. The removal of NOM was measured by comparing the reduction of the initial colour of MCRW compared to deionized water. The targeted residual fluoride concentration in water was less than 2 mg/L, which is the maximum permissible fluoride content limit in drinking water in accordance with International Reference Centre (IRC) which has been adopted by NDRS (URT-Ministry of Water, 2013a).

Measurement of Residual NOM (Colour) and Fluoride

The reduction of colour of MCRW after removal of NOM was measured using

Labtronics-Digital spectrophotometer (model: LT-31) by measuring light transmittance at 430 nm wavelength in accordance with Babcock and Singer, (1979). Deionized water was used as a reference for clarity of water. Its transmittance (T) was set at 100 T, that is, having full light transmittance. The raw MCRW transmittance was found to be 88 T. Therefore % NOM removal was obtained as per Equation (2).

$$\%\ increase\ in\ transmittance = \frac{T_m - T_o}{100 - T_o} * 100 \qquad (2)$$

Where T_m is measured transmittance and T_o is transmittance of raw MCRW (88 T). Fluoride concentration measurements were carried out using Metrohm 913 pH meter with fluoride electrodes in accordance with Mbabaye *et al*. (2017).

RESULTS AND DISCUSSION

The physical properties of MCRW are shown in Table 3 as compared to deionised water which was used as a reference on NOM/colour removal with transmittance (T) of 100 T and Uhai bottled water (packed in various bottles ranging from 0.5 to 12 L) with transmittance of 99.1, which is the mostly consumed water in volume-wise by a wide population in Dar es Salaam, Tanzania. The raw MCRW had a transmittance of 88 T, pH 8.16 and a fluoride content of 21.9 mg/L of water.

Table 3: Properties of MCRW collected on Nov 24, 2016 at Maji ya Chai township

	TDS (mg/L)	EC (µS/cm)	Fluoride content F^- (mg/L)	pH	Turbidity (NTU)	Transmittance at 430 nm (T)
MCRW	625	1045	20.9	8.16	0.73	88
Deionised water (Ref)	2.94	5.10	-	9.7	0.16	100
Uhai bottled water	112.5	187.5	0.67	7.52	0.34	99.1

Effect of Adjusting MCRW pH Before Defluoridation

Figure 3 shows the residual fluoride when MCRW with original fluoride concentration of 20.9 mg/L and pH of 8.16 was defluorinated at different pH conditions. Raw MCRW was used as a reference. Lowering of the initial pH of the MCRW was found to improve fluoride removal for a given amount of BC used (2 – 8 g BC/L of water). The residual pH of the MCRW after treatment were 9.48, 8.54, and 7.31 for unadjusted, adjusted to pH 7 and adjusted to pH 3, respectively. The results were similar to those obtained by Medellin-Castillo *et al*. (2007) who studied the removal of fluoride in water using BC with water pH ranging from 2 to 12. Their finding showed that, adsorption capacity depended on the physical-chemical properties of BC surface and the pH of solution. Lowering pH of solution developed high concentration of H^+ ions, which because of their small size, hence faster mobility, they quickly adsorb to the BC surface and influence the surface of the BC to become more positively charged and become more capable of attracting and accumulating/adsorbing more F^- ion as also observed by Medellin-Castillo *et al*. (2007).

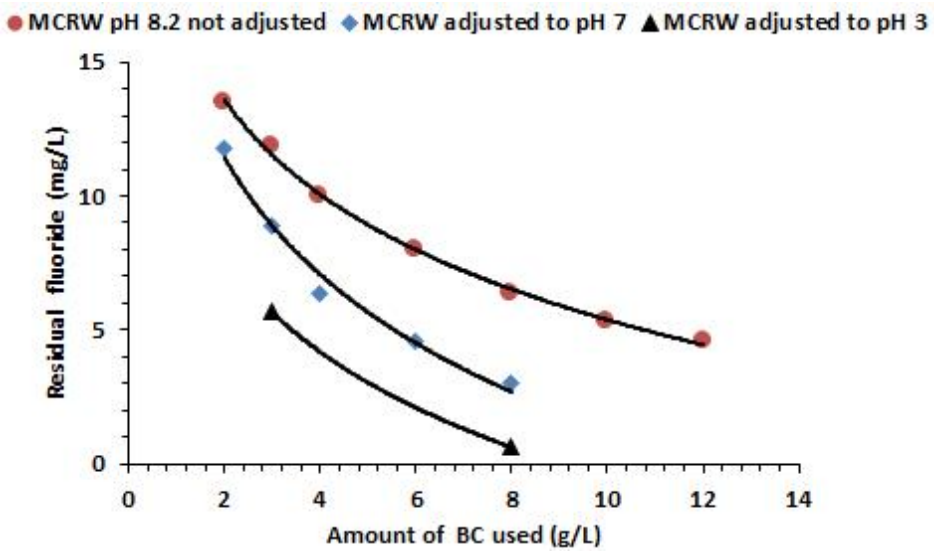

Figure 3: Effect of pH of MCRW on fluoride removal

Effect of Pre-treatment of BC with Acid on Fluoride Removal

Bone char treated with acid of various concentration as shown in Figure 4 showed improved defluoridation as the concentration HCl acid was increased from 0.01 N to 0.5 N. Residual fluoride of 4 – 6 mg/L from the original concentration of 20.9 mg/L required 8 g/L of BC treated with HCl solution of 0.1 N, while 5 g/L of BC treated with 0.3 N HCL achieved about the same results. Result of defluoridation with BC treated with 0.5 N HCl using 5 g/L of BC loading showed little difference with BC treated with 0.3 N HCl solution. That means, there is no necessity of using higher acid concentrations than 0.3 N HCl for pre-treating BC to improve its adsorption capacity or in other words to saturate its surface charge with hydrogen ions. Studies by Mahramanlioglu *et al.* (2002) have shown increased adsorption of fluoride from aqueous solution due to the increase of surface charge of the acid pre-treated adsorbent. Thus, pre-treating BC with 0.3 N HCl in this study was sufficient to improve the surface charge of the BC similar to pre-treatment with a 0.5 N HCl. The comparison of the effect of acid pre-treated BC to untreated BC is shown in Figure 5.

Comparison of Adjusting MCRW pH and Treatment of BC with Acid Before use on Defluoridation

Figure 5 combines the results shown in Figures 3 and 4. In Case 1; untreated BC was used in raw MCRW without adjusting water pH. In Case 2 and Case 3, MCRW was adjusted to pH 7 and pH 3 with HCl before adding untreated BC, respectively. In Case 4 and Case 5, BC treated with 0.3 N and 0.5 N HCl, respectively were used. The results showed that, adjustment of MCRW to pH 3 had best results. Although this was a good improvement, pre-treated BC will be easy to use in the rural household communities as it can be safely distributed and handled with people with different levels of knowledge rather than to have acid solution in households or communities. The acid pre-treated BC Case 4 (0.3 N HCl) showed improved defluoridation similar to the Case 2 where the pH of MCRW was adjusted to 7. Result of defluoridation with BC pre-treated with 0.5 N HCl (Case 5) at 5 g/L BC loading showed little difference with BC pre-treated with 0.3 N HCl solution. Previous studies by Mahramanlioglu *et al.* (2002) showed that, defluoridation of water decreased with increasing pH of water from 3.5 to 8. At low pH the adsorbent becomes positively charged,

hence attracting more negatively charged fluoride ions. Thus, acid pre-treatment of BC in this study had made the adsorbent to have more positively charged surface compared to untreated BC. The finding in this case indicated that defluoridation of MCRW with pH reduced to below 7 lead to results similar to using acid pre-treated BC.

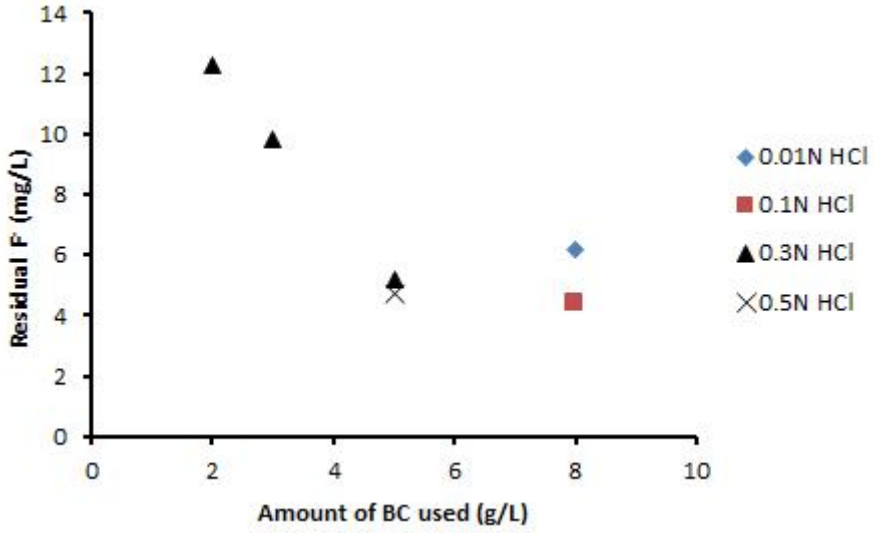

Figure 4: Effect of pre-treatment of BC with different concentrations of HCl acid on F-1 adsorption

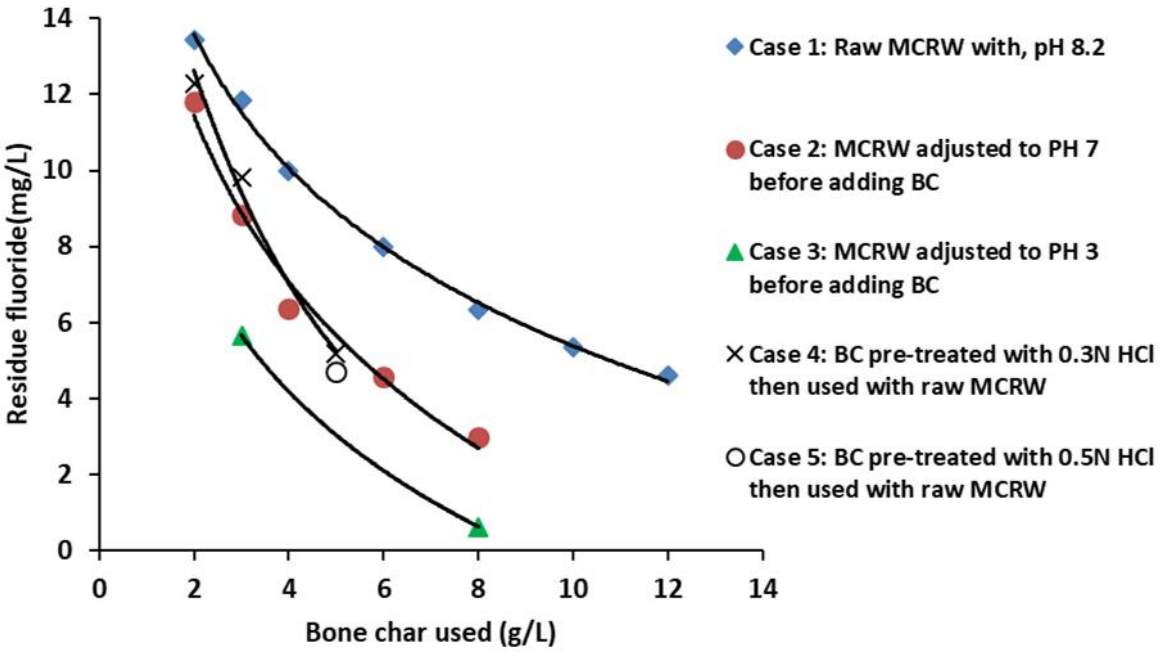

Figure 5: Effect of MCRW pH adjustment or BC pre-treatment on residual fluoride

Figure 6, compares the percentage reduction of fluoride in MCRW if 5 g/L of BC are used for the five cases in Figure 5. Using Case 1 as a reference, Case 2 showed 16% increase in fluoride removal obtained by adjusting the pH of MCRW to

7, while Case 3 showed 25% improved reduction of fluoride when pH of MCRW was adjusted to 3. Cases 4 and 5 showed improved fluoride removal using 0.3 and 0.5 N HCl acid pre-treated BC in which improved fluoride removal of 18% and 20% was obtained, respectively. The 18% defluoridation improvement with 0.3 N HCl acid pre-treated BC is around the same as pre-adjusting MCRW pH to 7 or using BC acid pre-treated with 0.5 N HCl. Thus, there is no need of pre-treating BC using acid with concentration higher than 0.3 N HCL. The BC used in all these experiments was of same batch with particle size distribution as shown in Figure 2 and the MCRW was collected once with physical properties as shown in Table 3.

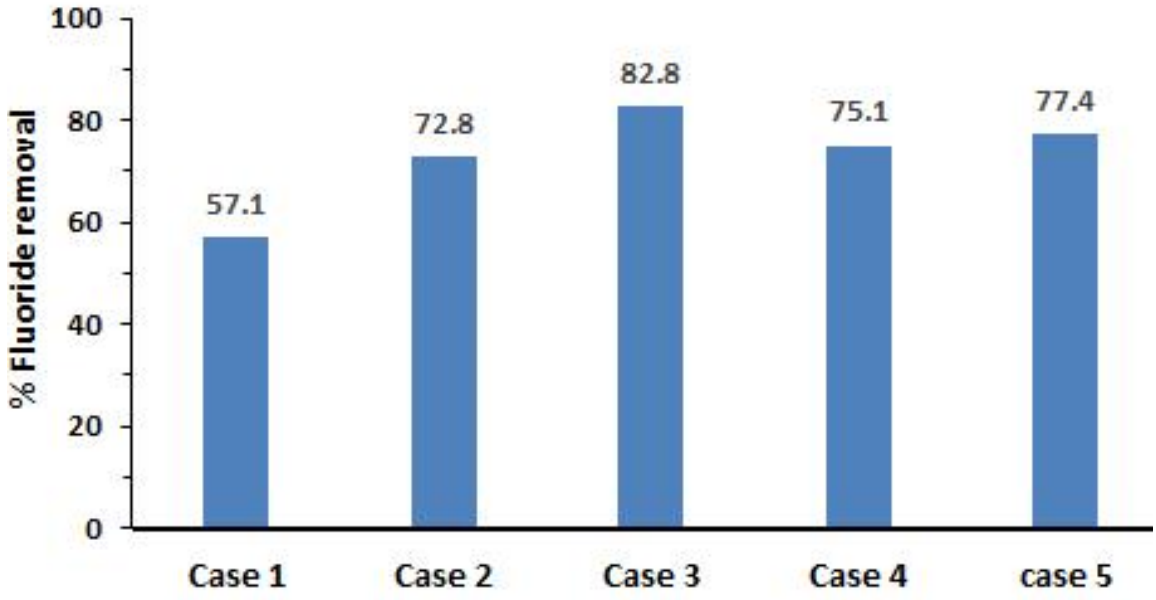

Figure 6: Percentage Fluoride reduction from 20.8 mg/L in MCRW using 5 g/L BC (description of the Cases is as in Figure 5)

The capacity of adsorbents to adsorb a solute from a solution is expressed in grams of solute adsorbed per gram of adsorbent (Wei *et al*, 2016). For this study, BC fluoride removal capacity was estimated by considering a fixed amount of mg of fluoride adsorbed. By taking a residual fluoride content of 5.6 mg/L in all case, the amount of fluoride removed is 15.3 mg F^-/L (Table 4). By drawing a straight horizontal line from the y-axis, coordinates (0, 5.6) in Figure 5, that is, removal of same amount of fluoride for the different modes, as expressed for the 4 cases, the amount of BC required was read in the x-axis for all the four Cases as tabulated in Table 4. The BC removal capacity (mg F^-/g BC) under the different conditions was computed by taking the ratio of fluoride removed to the amount of BC used. The results showed that by pre-treating BC with 0.3 N HCl (Case 4) the removal capacity of Fluoride was doubled compared with Case 1 in which untreated BC was used. Adjusting the MCRW pH to 7, Case 2, had almost doubled fluoride removal capacity compared to Case 1. Adjusting the pH of MCRW to pH 3 before defluoridation with BC (Case 3), showed threefold increase in fluoride removal compared to Case 1. Dahi (2015) indicated a significant improved effect in defluoridation capacity obtained when BC was pre-washed with acid but the strength of acid used was not given.

Table 4: BC F^- removal capacity with MCRW pH adjusted or BC pre-treated at a given residual fluoride

	BC used (g/L)	mg F^-/L removed	BC Removal capacity (mg F^-/g BC)
Case 1 in Figure 6	9.33	15.3	1.64
Case 2 in Figure 6	5.2	15.3	2.94
Case 3 in Figure 6	3.0	15.3	5.10
Case 4 in Figure 6	4.67	15.3	3.28
This is considering a case of residual fluoride of 5.6 mg/L from initial concentration of 21.9 mg/L for all cases			

Removal of NOM and Residual Fluoride to Acceptable Level

The recommended fluoride concentration for drinking water is 1.5 and 4 mg/L in accordance with WHO guideline and TBS guideline, respectively. For this reason, this work aimed at developing a practical (easy to use in rural community and affordable) approach of reducing the fluoride concentration to 2 mg/L or less and removal of NOM to acceptable level. Table 5 shows some of the results obtained when defluoridation was attempted with either 8 g/L or 5 g/L of various BC per litre of MCRW followed by coagulation with 66 ml/L of 1% PACl. Residual fluoride achieved using 8 g/L BC when pH was adjusted to 7 and 3 (case A and B) were 2.99 mg/L and 0.63 g/L, respectively, compared to 6.33 mg/L using raw MCRW. Pre-treatment of BC with 0.1N HCl (Case C) and using 8 g/L BC lead to residual fluoride of 4.41 mg/L.

The study aimed at reducing the amount of BC used while achieving target residual fluoride of not more than 2 mg/L after removal of NOM which was monitored by increased transmittance towards that of deionized water set as 100 T. Table 3 shows the residual fluoride after removal of NOM with PACl and the transmittance of the resulting water. The best result for fluoride removal is when 8 g/L BC was used **(A)** (0.3 mg F^- /L) with MCRW pH adjusted to 7 before defluoridation. Otherwise defluoridation using raw MCRW and 8 g/L BC followed with NOM removal with PACl **(Ref)**, showed similar results to defluoridation with 5 g/L BC treated with 0.3N HCl **(B)** or 0.5N HCl **(C)** followed with NOM removal with PACl. The NOM removal indicated by the transmittance of the resulting water was almost the same 98.2 -98.7 which was similar to Uhai bottled water, Table 3 for the four cases **Ref, A, B and C**. The best NOM removal was achieved by using a 5 kDa ultrafiltration membrane **(D)** which attained transmittance of 99.8 about the same as deionized water. Thus, pre-treated BC with 0.3 N HCl acid solution followed by the addition of 66 ml of 1% PACl for coagulation of NOM lead to removal of both fluoride and NOM of the MCRW to acceptable level.

Table 5: Fluoride and NOM removal in MCRW using BC and coagulant or ultra-filtration membrane

Water Sample or BC used	**(Ref)** Raw MCRW	**(A)** MCRW Modified to pH 7	**(B)** BC pre-treated with 0.3N HCl	**(C)** BC pre-treated with 0.5 N HCl	**(D)** MCRW filtered with ultrafiltration membrane
BC used (g/L)	8	8	5	5	0
Residual F^{-*} (mg/L)	6.33	2.99	5.17	4.71	20.4
Residual F^{-**} (mg/L)	1.66	0.3	1.66	1.51	-
Transmittance (T)	98.7	98.4	98.2	98.5	99.7
Residual F^{-*}after adsorption with BC for 60 min. Residual F^{-**} after treatment with 66 ml/L 1% PACl coagulant for removal of NOM					

Effectiveness of Coagulants Used for NOM Removal

The effectiveness of removal of NOM and residual fluoride using same amount of BC and coagulant is as shown in Table 6. The residual fluoride and percentage increased transmittance were best when PACl was used. Transmittance was almost 91% with PACl compared to 55% and 44% for Alum and ZetaFloc 703, respectively. The residual fluoride was 1.24 mg/L with PACl, compared to 1.49 mg/L and 2.2 mg/L for alum and ZetaFloc 703, respectively. The electroconductivity and pH of the treated MCRW was within the same range after treatment with the three coagulants. Thus, PACl appeared to be the preferable coagulant for NOM removal form MCRW. An overview on removal of NOM using coagulants by different researchers indicated that PACl compared to alum salts has less temperature and pH dependent, better NOM removal capacity and lower dose requirement (Matilainen *et al*., 2010). Wang *et al*. (2004) studied speciation of PACl and concluded that, PACls with high OH/Al ratio exhibit quite stable speciation under various conditions. This is also supported by the review by Matilainen *et al*. (2010), who indicated that PACl Al-species are considered to be the most efficient in coagulating flocs due to their larger size and higher positive charges. The finding in this study demonstrated similar phenomena as the percentage increase in transmittance achieved was 91 for PACl compared to 55 for alum. ZetaFloc 703 was the least in both fluoride and NOM removal.

Although 5 kDa ultrafiltration (UF) membrane with pores of about 1.9 nm (Aschermann *et al*., 2016) showed excellent result on NOM removal, that is, with increased transmittance of 98% (Table 6), UF membranes have a disadvantage that, it is not easily available and it is not affordable. In addition, filtration process has to overcome trans-membrane pressure (Aschermann *et al*., 2016), which means a pump is required and UF membrane don't remove fluoride. Membrane filtration also has a problem of membrane fouling (Matilainen *et al*., 2010), which leads to decrease in flux. Therefore, application of UF membranes in rural communities will have more challenge compared to coagulants.

Table 6: Effect of Different coagulants on properties of clarified MCRW

Means of NOM removal	66 mL 1% Alum	66 mL 1% PACl	66 mL 1% ZetaFloc 703	MCRW filtered with 5 kDa UF membrane	Raw MCRW
BC used (g/L)	10	10	10	0	0
Residual F^- (mg/L)	1.49	1.24	2.20	20.4	20.9
Turbidity (NTU)	0.93	0.73	0.22	1.05	0.73
Transmittance (T)	94.6	98.9	93.3	99.8	88
% Increase in transmittance	55	90.8	44.2	98.3	0
EC (µS/cm)	1077	1163	1070	1050	1045
pH	8.3	8.5	8.6	10	8.2

CONCLUSIONS

The study has shown that, a practical and affordable approach for defluoridation and removal of NOM from MCRW was achieved. The MCRW can be defluoridated with BC pre-treated with 0.3 N HCl followed by coagulation with PACl to reach acceptable level of fluoride content of less than 2.0 mg F^-/L. That was achieved by use of 5 g/L of acid pre-treated BC and 66 mL of 1% PACl per litre of MCRW. Bone char pre-treated with 0.3N HCl solution was found to double defluoridation capacity, that is, amount of fluoride removed per mass of

BC used (mg/g) compared to un-treated BC used with raw MCRW.

Although adjustment of pH of MCRW with acid to pH 3 appeared to have 3 times defluoridation capacity improvement compared to untreated BC, use of 0.3N HCl acid pre-treated BC is preferred to avoid impractical working with acid solution for pH adjustment during deployment of the technology in rural households' communities.

Poly-aluminium chloride (PACl) is a preferable coagulant for removal of NOM from MCRW since it produces more clear water (higher transmittance) compared to use of alum and zetafloc 703. It is also preferable health wise compared to alum and relatively easy to apply compared to the use of 5 kDa UF membrane which produced clearer water compared to PACl. The BC particle size range of 45 µm <d< 250 µm was found to be suitable for defluoridation by mixing with MCRW in a container followed by settling and decantation of the clear water. BC particles of 45 µm< d< 250 µm, are not suitable in defluoridation column units as they lead to column fouling.

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REFERENCES

Aschermann G., Jeihanipour A., Shen J., Mkongo G., Dramas L., Croué J.P., and Schäfer A. (2016). Seasonal variation of organic matter concentration and characteristics in the Maji ya Chai River (Tanzania): Impact on treatability by ultrafiltration, Water Research, 101: 370-381. DOI: 10.1016/j.watres.2016.05.022

Babcock D.B. and Singer P.C. (1979). Chlorination and coagulation of humic and fluvic acids. Journal American Water Works Association, 71(3): 149-152. https://doi.org/10.1002/j.1551-8833.1979.tb04318.x

Chen J. and Schäfer A. (2015). Factors affecting fluoride and natural organic matter (NOM) removal from natural waters in Tanzania by nanofiltration/reverse osmosis. Science of the Total Environment, 527-528: 520-529. DOI: 10.1016/j.scitotenv.2015.04.037

Cheng W.P. and Chi F.H. (2002). A study of coagulation mechanisms of polyferric sulfate reacting with humic acid using a fluorescence-quenching method. Water Research, 36: 4583-4591. DOI: 10.1016/s0043-1354(02)00189-6

Dahi E. (2015). Optimisation of bone char production using the standard defluoridation capacity procedure. Research report. Fluoride, 48 (1): 29-36.

Dahi E. (2013). 25 Years of Defluoridation Technology Project. Keynote paper: XXXIst International Society of Fluoride Research, Tehran, 10: 18-21.

Liu H., Hu C., Zhao H. and Qu J. (2009). Coagulation of humic acid by PACl with high content of Al13: The role of aluminium speciation. Separation and Purification Technology, 70(2): 225-230. DOI: 10.1016/j.seppur.2009.09.020

Mahramanlioglu M., Kizilcikli I. and Bicer I.O. (2002). Adsorption of fluoride from aqueous solution by acid treated spent bleaching earth. Journal of Fluorine Chemistry, 115(1): 41-47. DOI: 10.1016/S0022-1139(02)00003-9

Matilainen A., Vepsäläinen M. and Sillanpää M. (2010). Natural organic matter removal by coagulation during drinking water treatment: a review. Advances in Colloid and Interface Science, 159: 189-197. DOI: 10.1016/j.cis.2010.06.007

Mbabaye G.K., Mtalo F., Minja R.J.A. and Legonda I. (2017). Standardizing defluoridation of community waters

using bone char, Journal of Water Supply: Research and Technology-AQUA, 66(2): 131-139. https://doi.org/10.2166/aqua.2017.056

Medellin-Castillo N.A., Leyva-Ramos R., Ocampo-Perez R., Garcia dela Cruz R.F., Aragon-Pina A., Martinez-Rosales J.M., Guerrero-Coronado R.M. and Fuentes-Rubio L. (2007). Adsorption of fluoride from solution on bone char, Industrial & Engineering Chemistry Research, 46(26): 9205-9212. https://doi.org/10.1021/ie070023n

O'Melia C.R., Becker W.C. and Au K.K. (1999). Removal of humic substances by coagulation. Water Science and Technology, 40(9): 47-54. https://doi.org/10.2166/wst.1999.0440

United Republic of Tanzania, URT (2013a). Strategy for scaling up defluoridation activities and piloting research findings in fluoride belt. Report, Ministry of Water.

United Republic of Tanzania, URT (2013b). Population and Housing Census: Population Distribution by Administrative Areas. National Bureau of Statistics, Ministry of Finance, Dar es Salaam and Office of Chief Government Statistician, President's Office, Finance, Economy and Development Planning, Zanzibar.

van Rijn C.J.M. (2004). Membrane Science and Technology Series,10: Nano and Micro engineered membrane technology. Elsevier.

Wang D., Sun W., Xu Y., Tang H. and Gregory J. (2004). Speciation stability of inorganic flocculant – PACl. Colloids and Surfaces A: Physicochemical Engineering Aspects. 243 (1-3), 1-10. https://doi.org/10.1016/j.colsurfa.2004.04.073

Wei S., Niu P., Wang R., Fang D., Yang X. and Dan, Y. (2016). Binary systems of kaolinite and hematite: Preparation, characterization and adsorption characteristics for fluoride. Materials Transactions, 7(57): 1171-1176. https://doi.org/10.2320/matertrans.M2015448

Zouboulis A., Traskas G. and Samaras P. (2008). Comparison of efficiency between poly-aluminium chloride and aluminium sulphate coagulants during full-scale experiments in a drinking water treatment plant. Separation Science and Technology, 43(6): 1507–1519. https://doi.org/10.1080/01496390801940903