Development of a Point-of-Use Drinking Water Purifier Using Aluminum Oxide-Based Flocculent-Disinfectant Composite

Patricia Namate[1,2], Feleke Zewge[2], and Eyobel Mulugeta[3*]

[1]*Basic Sciences Department, Lilongwe University of Agriculture and Natural Resources, P.O. Box 219, Lilongwe, Malawi*

[2]*African Centre of Excellence for Water Management, Addis Ababa University, P.O. Box 1176, Addis Ababa, Ethiopia*

[3]*Bio and Emerging Technology Institute, Ministry of Innovation and Technology, P.O. Box 5954, Addis Ababa, Ethiopia*

*** Corresponding author:***

email: eyoya2005@gmail.com

Received: November 8, 2022
Accepted: March 10, 2023

DOI: 10.22146/ijc.79024

Abstract: *One in every three people in the world still lacks access to clean drinking water. Aside from microbiological pollution, high fluoride content in drinking water is one of the most serious problems in African countries. This study aimed to contribute to the availability of clean water by developing a point-of-use drinking water purifier using an aluminum oxide-based flocculent-disinfectant composite. Batch experiments were conducted to determine factors affecting fluoride removal efficiency (FRE) and* E. coli *log reduction efficiency. AO of 75 mg/L, 800 mg/L alum, lime (35% alum dose), and 1.5 mg/L* $Ca(OCl)_2$ *doses achieved 95% FRE and 5* $\log_{10}$ *reductions of* E. coli *using 15 mg/L as initial fluoride concentration (*$[F^-]_0$*), and* 10^5 *CFU/100 mL* E. coli *concentration.* $[F^-]_0$ *affected FRE but showed no effect on E. coli log reduction. The optimum pH of the solution for both FRE and* E. coli *log reduction was found to be in the range of 4–8. Three prototypes in powder form were developed. The prototypes were tested on real water samples from the Ethiopian Rift Valley, and the results were found to be within the drinking water standards, thus indicating the capability of the developed products to purify contaminated water for human consumption.*

Keywords: *fluoride; aluminum oxide; water purifier;* E. coli

■ INTRODUCTION

While access to safe drinking water remains critical to human survival, the majority of the world population lacks access to it [1]. For example, estimates in 2017 indicated that 785 million people worldwide lacked access to basic drinking water supply and sanitation, with around 144 million of these individuals depending on contaminated water resources. Duke et al. [2] also pointed out that even people with "improved" water sources, such as household connections, public standpipes, and boreholes, are still at risk of consuming microbiologically and chemically contaminated water. In Africa, the spread of deadly waterborne diseases such as dental and skeletal fluorosis, diarrhea, dysentery, typhoid, and cholera are linked to a lack of access to clean water and poor sanitation [3]. Naik [4] pointed out that less than 50% of the rural communities in Africa have access to both safe water and sanitation. The authors further acknowledged that the continent continues to suffer from the problem of acute water scarcity, clean water deficit, and crises.

Countries within the African Great Rift Valley Region are at high risk of consuming contaminated water. For example, studies have indicated that fluoride concentration is exceptionally high in countries within the East African Great Rift Valley Region [5]. In some countries, such as Malawi [6], Ethiopia [7], and Tanzania [8], the hydrochemical characteristics of groundwater are highly associated with high fluoride concentration above the recommended level. In Ethiopia, about eight million people consume water containing fluoride concentration above 1.5 mg/L, that is permissible limits established by the WHO [9]. Zewge [9] reported that the daily intake of fluoride concentration above 1.5 mg/L is linked to dental and skeletal fluorosis. Similar cases are reported in countries such as Sudan, Nigeria, Kenya, Tanzania, and Malawi, with a great occurrence of dental and skeletal fluorosis in the majority of populations [10-12].

The most prevalent household water treatment processes include chlorination, filtration, solar disinfection, boiling, coagulation, and flocculation [3]. These are effective at reducing microbial contamination. Various technologies have also been developed worldwide, including in Ethiopia, to treat water sources with high fluoride concentration [8,13]. The coagulation, precipitation on $Al_2(SO_4)_3$, CaO, and $CaCl_2$, adsorption on activated alumina, aluminum-based adsorbents, bentonite, ion exchange with synthetic resins, and bone char are the most extensively employed fluoride removal techniques in developing countries [14-15]. In rural Ethiopia, some of these techniques have proven to be inefficient in removing exceptionally high fluoride levels in the Central Rift Valley region [10]. Therefore, activated alumina and bone char have been proposed in the region.

Coagulant/disinfection products (CDPs) have the distinct benefit of offering microbial quality improvement, turbidity reductions, and a post-treatment free chlorine residual (FCR) among the various available point of use (POU) techniques (e.g., boiling, household chlorination, and ceramic filtration). These are readily available sachets that contain at least two main active ingredients (typically in powdered form), namely a coagulant (e.g., aluminum or ferric salt) and a disinfectant (i.e., chlorine variant). Examples include the PUR product, which is a small sachet containing powdered ferric sulfate (a flocculent) and calcium hypochlorite (a disinfectant), Bishan Gari as well as Aquatabs products [16]. The Bishan Gari water purifier has the following advantages, according to Bogale [16]: a longer shelf life than liquid chlorine, a small volume that makes it simpler to carry and store, it is locally made, and inexpensive. The downside of the Bishan Gari water purifier include that it does not reduce fluoride levels to below 1.5 mg/L for water sources with high fluoride concentration, and people do not like the smell of the treated water.

The combination of aluminum sulfate and lime in water purifiers also reduces fluoride levels in drinking water. The insoluble aluminum hydroxide flocs formed during this process are responsible for the co-precipitation of the fluoride ions, a technique called Nalgonda [14]. However, this method is not suitable for treating water with high total dissolved solids (TDS) and raw water fluoride concentration exceeding 10 mg/L. In addition, the aforementioned water purifiers generate a lot more sludge after treatment since the technologies consume more aluminum sulfate dose to reduce the fluoride to its permissible level, and some of the water purifiers do not effectively treat turbid water. Hence, there is a need to look for a solution to minimize the amount of sludge produced after treatment and also to effectively treat turbid water.

Therefore, POU water treatment technologies with a combination of coagulants and disinfectants are among the technologies used to empower people and communities which use unimproved water sources and water sources with high fluoride concentration to improve water quality by treating it at home [12,17-18]. Research conducted by Mulugeta et al. [19] compared the performance of commercially available activated alumina (AA) and aluminum oxide (AO) synthesized in the laboratory. It showed that the fluoride removal capacity of AO synthesized in the lab was twice that of the commercially available aluminium oxide, such as activated alumina and pseudo-boehmite. These defluoridation technologies occur at the source, and therefore during transportation and storage of drinking water, there might be a high risk of microbial contamination together with fluoride contamination. Therefore, there is a need to look for technologies to remove both fluoride and microbes. Therefore, the objectives of this study were (a) to conduct batch fluoride and bacterial removal experiments using the synthesized AO, aluminum sulfate, calcium hypochlorite, and lime under optimized conditions, (b) to test the composite on actual water samples from the Ethiopian Rift Valley region, and (c) to develop a prototype household water purifier using aluminum oxide-based flocculent-disinfectant composite.

■ EXPERIMENTAL SECTION

Materials

The reagents and chemicals utilized were received without any treatment. The materials used in this study were anhydrous sodium fluoride (99.0% NaF, BDH Chemicals Ltd, Poole, England); aluminium sulfate ($Al_2(SO_4)_3 \cdot 14H_2O$, which was purchased from Awash Melkasa Aluminium Sulfate and Sulfuric Acid Factory, Ethiopia); sodium hydroxide, calcium hypochlorite, and sodium thiosulfate (99% purity, Merck, Germany); the lime was purchased from Senkele Lime Factory, Ethiopia. Total ionic strength adjustment buffer (TISAB) was prepared following the recommended procedure, except for the ethylenediaminetetraacetic acid (EDTA) that was substituted with cyclohexanediaminetetraacetic acid (CDTA). The Ethiopian Health and Nutrition Research Institute (EHNRI) provided a permanent *E. coli* stock culture. *E. coli* cells used in this study were prepared from the permanent culture.

Instrumentation

The instruments used in the present study were A pH/ISE meter (Orion Model EA 940 Expandable Ion Analyzer, USA) fitted with a combination ion selective fluoride electrode (Orion Model 96-09, USA) used to measure the concentration of fluoride; muffle furnace (Carbolite, ELF Model, UK); turbidity meter (Thermo Scientific Orion AQ4500 Turbidimeter, USA); UV-vis spectrophotometer (ThermoFisher Scientific, USA) and portable conductivity/TDS meter (HACH, HQ1140, USA).

Procedure

E. coli *enumeration*

Before and after treatment, the concentration of viable *E. coli* bacteria was determined using the pour plate method, as described in standard method 9215B [20]. The following formula in Eq. (1) was applied to the *E. coli* count.

$$\frac{\text{CFU}}{100}\text{mL} = \frac{\text{Average count}}{\text{Volume of the sample}} \times 100 \qquad (1)$$

where CFU is the Colony Forming Unit

Analysis of fluoride

The fluoride stock solution was prepared by dissolving 2.21 g of anhydrous sodium fluoride in deionized water and filling up to 1000 mL with deionized water. The fluoride solutions used during the batch process were made by diluting the stock solution. The pH/ISE meter was calibrated using standard solutions containing fluoride concentration of 0.5, 1.0, 5.0, 10.0, and 20.0 mg/L before each trial. An equal volume of TISAB was added to each sample before measuring the fluoride concentration. All samples were filtered through a 0.2 μm filter paper for fluoride analysis. All tests were carried out at room temperature.

Chlorine measurement

Calcium hypochlorite was used to produce a 1% chlorine stock solution. The chlorine solutions used during the batch process were made by diluting the stock solution. The chlorine and residual chlorine concentrations were measured by the *N*,*N*-diethyl-*p*-phenylenediamine (DPD) method [21].

Synthesis of aluminum oxide (AO)

AO was synthesized using the standard method developed by Mulugeta and his research colleagues [15]. The precursor material was aluminum sulfate (alum). AO was prepared by mixing 100 g of $Al_2(SO_4)_3 \cdot 14H_2O$ in 500 mL of distilled water while stirring with a magnetic stirrer until complete dissolution. NaOH (2 M) was used to adjust the pH. The precipitate (AO) was dried by exposure to sunlight. The AO was activated by heating it at 300 °C in a furnace for 1 h. It was then thoroughly washed with deionized water to remove excess Na_2SO_4. The AO material was sun-dried again and crashed into powder with a mortar and pestle.

Batch experiments

All batch tests were performed in a 500 mL beaker containing 500 mL of fluoride-spiked distilled water and *E. coli* under constant mixing conditions on a magnetic shaker at room temperature. The sample solution was regularly removed from the beaker. The residual fluoride concentration was determined immediately after adding an equal amount of TISAB to a 10 mL sample solution. Chlorination was stopped with a quencher of 50 μL of 10% sodium thiosulfate. All tests were conducted in triplicate, and the mean values were used. The fluoride removal efficiency (FRE) was determined using Eq. (2).

$$\%\ \mathrm{FRE} = \left(\frac{C_0 - C_t}{C_0}\right) \times 100 \tag{2}$$

where C_0 and C_t are the initial and at any time (t) fluoride concentration in solution (mg/L), respectively. The removal efficiency of *E. coli* was expressed as the $\log_{10}$ reduction value of bacteria using Eq. (3).

$$\mathrm{LRV} = \log_{10} \frac{N_t}{N_0} \tag{3}$$

where, LRV = log reduction value of bacteria counts at time t; N_0 = initial bacterial concentration at time 0, N_t = final bacterial concentration after a treatment time t.

Optimization of operation parameters

Batch operation parameters were optimized by varying one parameter at a time and allowing the rest to remain constant. The effect of dose on fluoride removal efficiency and *E. coli* log reduction was studied by varying doses of aluminum sulfate (700, 800, 900, 1000, and 1100 mg/L), AO (45, 75, 105, 135 and 165 mg/L), lime (35% alum dose), and calcium hypochlorite (0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5 and 4.0 mg/L) at constant initial fluoride concentration of 15 mg/L and approximately 10^5 CFU/100 mL *E. coli* concentration.

The contact time effect was studied by changing time (10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 110, and 120 min). The effect of initial fluoride concentration ($[F^-]_0$) was studied by varying the fluoride concentration (5, 10, 15, 20, 25, and 30 mg/L). The pH effect was examined by changing the initial pH of the solution (3, 4, 5, 6, 7, 8, 9, and 10). The effect of initial *E. coli* concentration was investigated by spiking the water with different *E. coli* concentrations (10^4, 10^5, 10^7, and 10^8 CFU/100 mL).

Testing the composite with actual water samples

Actual water samples were collected from six different sites within the Ethiopian Rift Valley, namely the Bora district (Tube, Tejitu, Dolessa), the Adami Tulu Jido Kombolcha (ATJK) district (Germama, Anano), and the Dugda district (Serity) in 30 L plastic Jerri cans. The samples for *E. coli* analysis were collected in sterilized bottles. The Jerri cans were labeled for identification before being adequately stored and transferred to the laboratory. The optimized dose from the batch experiments was tested on these water samples. Water samples were analyzed for different parameters before and after treatment. Methods of water quality analysis were done according to standard methods for examining water and wastewater specified by the American Public Health Association [20]. The water samples were analyzed for the following parameters: turbidity, pH, electrical conductivity (EC), fluoride, *E. coli*, alkalinity, sulfate, aluminum, residual chlorine, and chloride. The obtained values were compared against the WHO drinking water standards to evaluate their suitability for drinking purposes.

Development of a prototype household water purifier

Batch experiments were conducted on actual water samples to optimize a combination of four materials that can effectively treat 10 L of water samples. The water purifier prototype was developed based on batch experiments conducted on actual water samples. Three prototypes were developed as high, medium, and low dose for treating 10 L water samples with 5, 10, and 15 mg/L fluoride concentration, respectively. A dose of 75 mg/L AO, 800 mg/L alum, 15% lime, and 2 mg/L $Ca(OCl)_2$ was used to develop a low-dose prototype to treat water with fluoride concentration ≤ 5 mg/L. A dose of 85 mg/L AO, 850 mg/L alum, 15% lime, and 2 mg/L $Ca(OCl)_2$, was used to develop a medium dose prototype to treat water that contains fluoride concentration ≤ 10 mg/L. A dose of 95 mg/L AO, 900 mg/L alum, 15% lime, and 2 mg/L $Ca(OCl)_2$ was used to develop a high-dose prototype to treat water that contains fluoride concentration ≤ 15 mg/L. The high, medium and low doses weigh 11.9, 10.6, and 9.9 g, respectively. Finally, the materials were packed in a single sachet for POU water treatment. The developed prototypes were tested on water samples from the Ethiopian Rift Valley. The treated water samples were analyzed for turbidity, pH, electrical conductivity, fluoride, *E. coli*, alkalinity, sulfate, aluminum, residual chlorine, and chloride.

■ RESULTS AND DISCUSSION

Optimization of Process Parameters

The effect of dose

Fluoride removal efficiency increased with an increase in dosage from 83.2 to 95.7%, as shown in Table 1. This is attributed to the increased production of higher amounts of precipitates and aluminum hydroxides, which increased the fluoride removal efficiency [22]. There was no improvement in the percent fluoride removal at higher dosages than 75 mg/L AO, 280 mg/L lime, and 800 mg/L alum due to the presence of excess precipitates compared to fluoride ions, assuming that the amount of aluminum complex per fluoride ion stays constant. Higher alum/AO doses could lead to the wastage of chemicals without significantly increasing the amount of fluoride removed.

This dosage of 75 mg/L AO, 800 mg/L alum, and 280 mg/L lime is adequate to lower the fluoride levels in drinking water to 1.5 mg/L. Similar results were obtained by Shimelis et al. [23] on the removal of fluoride from water using aluminum hydroxide. After a dose of 1.6 g/L, there was no significant change in fluoride removal; hence that dose was considered the optimum dose. Various combinations of AO and alum did not produce efficient *E. coli* log reduction, as shown in Table 2. The various combinations only reduced *E. coli* in the range of 0.0021 to 0.0091 log, corresponding to 2 to 9% *E. coli* removal. As such, a dose of 75 mg/L AO and 800 mg/L alum was selected for further dose optimization with calcium hypochlorite. The dose of 75 mg/L AO, 800 mg/L alum, and 280 mg/L lime was combined with different concentrations of calcium hypochlorite in the range of 0.5 to 4 mg/L. There was a slight increase in fluoride removal after adding calcium hypochlorite, as shown in Table 3. The reaction in Eq. (4) occurs when calcium hypochlorite is added to water.

$$Ca(OCl)_2 + 2H_2O \rightarrow 2HOCl_{(aq)} + Ca^{2+}_{(aq)} + 2OH^-_{(aq)} \qquad (4)$$

The calcium ion in the solution reacts with fluoride to form calcium fluoride (CaF_2), according to Eq. (5).

$$Ca^{2+} + 2F^- \rightarrow CaF_2 \qquad (5)$$

Calcium fluoride is insoluble and is left like a precipitate. This contributes to fluoride removal via precipitation [24]. Calcium hypochlorite disinfects water through an inactivation process that causes damage to the cell membrane and cell wall of *E. coli* [25].

After adding calcium hypochlorite to water, hypochlorous acid ($HOCl^-$) is formed. Proteins and peptidoglycan bound to the cell wall and the cell membrane are the initial points of attack for chlorine disinfection [26]. After the cell wall and membrane damage, $HOCl^-$ can penetrate the cell wall and reach the

Table 1. Fluoride removal efficiency (%) for various combinations of alum, AO, and 280 mg/L lime

AO (mg/L)	Alum (mg/L)				
	700	800	900	1000	1100
45	83.2 ± 0.71	85.7 ± 0.81	90.1 ± 0.08	91.2 ± 0.07	92.1 ± 0.45
75	85.8 ± 0.43	92.1 ± 0.51	93.2 ± 0.56	93.9 ± 0.73	94.1 ± 0.18
105	87.1 ± 0.63	92.5 ± 0.54	92.9 ± 0.54	94.1 ± 1.12	94.9 ± 0.38
135	89.7 ± 0.71	93.1 ± 0.87	93.7 ± 0.76	94.5 ± 0.34	95.2 ± 0.41
165	90.7 ± 0.98	94.7 ± 0.53	94.8 ± 0.34	95.1 ± 0.28	95.7 ± 0.37

Table 2. log_{10} reduction (10^2 CFU/100 mL) of *E. coli* for various combinations of alum, AO, and 280 mg/L lime

AO (mg/L)	Alum (mg/L)				
	700	800	900	1000	1100
45	0.21 ± 0.05	0.24 ± 0.01	0.29 ± 0.01	0.33 ± 0.04	0.40 ± 0.05
75	0.31 ± 0.05	0.41 ± 0.05	0.51 ± 0.04	0.68 ± 0.04	0.60 ± 0.10
105	0.35 ± 0.10	0.52 ± 0.04	0.56 ± 0.01	0.59 ± 0.01	0.61 ± 0.05
135	0.40 ± 0.10	0.67 ± 0.04	0.72 ± 0.04	0.83 ± 0.04	0.87 ± 0.01
165	0.60 ± 0.01	0.73 ± 0.02	0.08 ± 0.05	0.90 ± 0.05	0.91 ± 0.05

cytoplasm, followed by reactions with purine and pyrimidine bases of the nucleic acids, whereby thymidine deoxyguanosine and uridine monophosphate are preferred targets. There was an increase in log reduction for *E. coli* with an addition of calcium hypochlorite (Table 3). An increase in the dose of calcium hypochlorite led to an increase in $\log_{10}$ reduction of *E. coli.* A low concentration of calcium hypochlorite produces low $HOCl^-$ concentrations. As a result, at low concentrations, microbial inactivation is low due to its inability to reach the cytoplasm and cause damage to the DNA [27]. A 0.5 mg/L of $Ca(OCl)_2$ only achieved 1 $\log_{10}$ reduction (about 90%) of *E. coli*. In contrast, higher chlorine concentrations cause a larger impairment of the cell wall and membrane, enabling HOCl to penetrate the cell wall and react with nucleic acids. This results in high-level DNA damage to the bacterial cells, impeding DNA amplification, even for short amplicons [26]. This is why 4 mg/L of $Ca(OCl)_2$ achieved a 6.7 $\log_{10}$ (99.9999%) reduction of *E. coli*. These results agree with a previous study by Owoseni et al. [21] on the survival of *E. coli* collected from secondary effluents of two wastewater treatment facilities in the Eastern Cape Province of South Africa with different chlorine concentrations. The bacteria log reduction ranged from 1 to 5 at low calcium hypochlorite concentrations (0.5 to 1.5 mg/L). At higher calcium hypochlorite concentrations (2 to 4 mg/L), the bacteria log reduction ranged from 6.0 to 6.7.

The residual chlorine concentration was in the range of 0.09 to 1.72 mg/L. Residual chlorine is defined as the concentration of chlorine species present in water after the oxidant demand has been met [28]. The report from the Centers for Disease Control and Prevention indicates that the presence of residual chlorine in drinking water indirectly implies the absence of disease-causing organisms, secondly, it indicates that the water is protected against recontamination during transportation and storage at home [29]. The free chlorine level in drinking water correlates with the absence of most disease-causing organisms and hence serves as a measure of water potability. The WHO recommends that free chlorine residual range of 0.2–0.5 mg/L is maintained at all points in the water supply for locations with little risk of cholera or associated epidemics [30]. Since 1.5 mg/L of calcium hypochlorite produced 5 log reduction for *E. coli,* which is a good performance. The residual chlorine concentration was 0.48 mg/L, which is enough to protect water from recontamination and is within the WHO permissible limits. The dose of 1.5 mg/L $Ca(OCl)_2$, 75 mg/L AO, 800 mg/L alum, and 280 mg/L lime was selected as an optimum dose for further experiments.

Effect of contact time

Fig. 1(a) shows that fluoride removal was fast within the first 15 min. Fluoride removal attains almost an equilibrium condition after 60 min. This indicates that all active sites for binding fluoride ions are occupied quickly [31]. A contact time of 30 min is enough to remove 90% of fluoride in a solution with the dose of 75 mg/L AO, 800 mg/L alum, 35% lime, and 1.5 mg/L calcium hypochlorite. This shows that a contact time more than 30 min does not affect fluoride removal. Similar

Table 3. Fluoride removal efficiency (%) and *E. coli* log reduction with 75 mg/L AO, 800 mg/L alum, 280 mg/L lime, and different concentrations of calcium hypochlorite at 15 mg/L of fluoride and 10^5 CFU/100 mL

Concentration of $Ca(OCl)_2$ (mg/L)	Fluoride removal efficiency (%)	Log_{10} reduction for *E. coli*	Residual chlorine
0.5	94.31 ± 0.06	1.0 ± 0.07	0.09 ± 0.04
1.0	95.11 ± 0.09	2.7 ± 0.05	0.12 ± 0.03
1.5	95.32 ± 0.27	5.0 ± 0.50	0.48 ± 0.10
2.0	96.56 ± 0.13	6.0 ± 0.05	0.60 ± 0.06
2.5	96.71 ± 0.11	6.1 ± 0.10	0.92 ± 0.07
3.0	97.22 ± 0.24	6.2 ± 0.70	1.21 ± 0.01
3.5	97.31 ± 0.56	6.4 ± 0.90	1.50 ± 0.02
4.0	97.89 ± 0.70	6.7 ± 0.40	1.72 ± 0.09

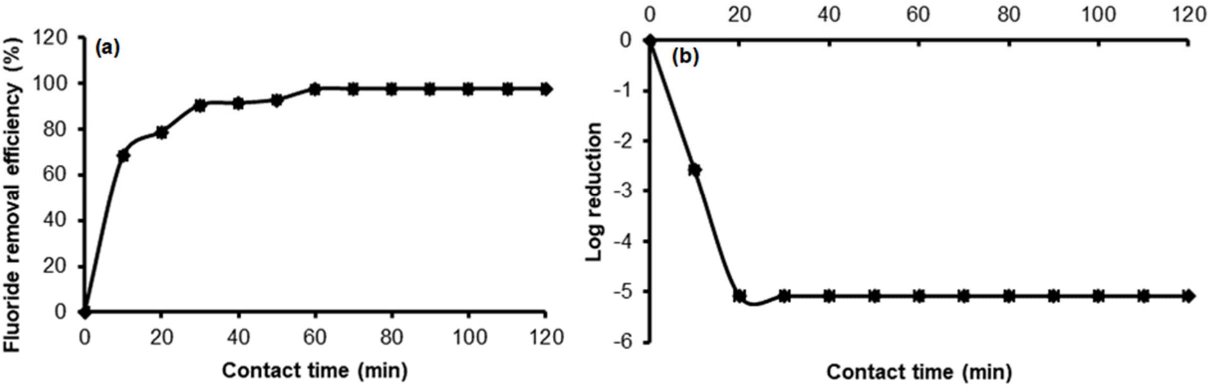

Fig 1. The effect of contact time on (a) fluoride removal and (b) *E. coli* log reduction ($[F^-] = 15$ mg/L, pH = 6.7, *E. coli* = 10^5 CFU/100 mL, AO = 75 mg/L, alum = 800 mg/L, lime = 280 mg/L and $Ca(OCl)_2 = 1.5$ mg/L)

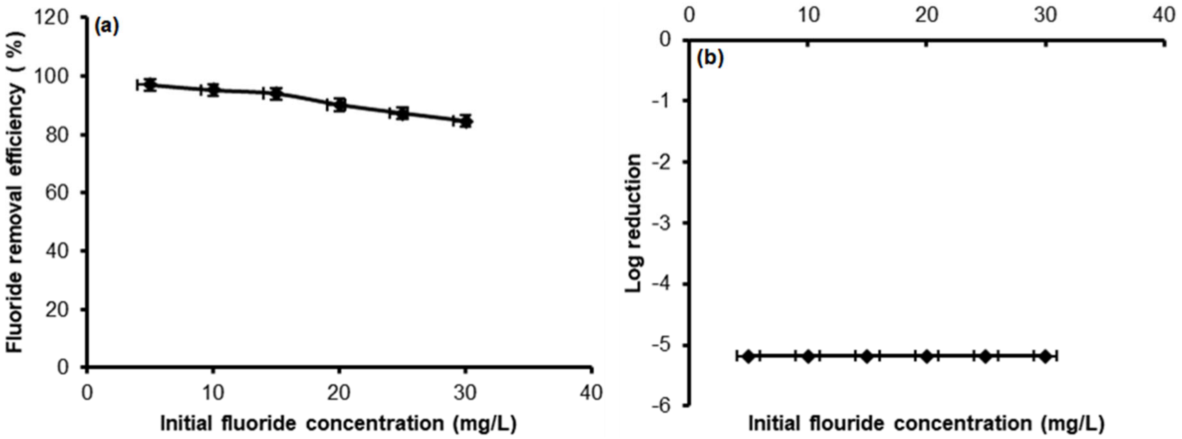

Fig 2. The effect of $[F^-]_0$ on (a) F^- removal efficiency, (b) *E. coli* log reduction (contact time = 30 min, pH = 6.7, *E. coli* = 10^5 CFU/100 mL, AO = 75 mg/L, alum = 800 mg/L, lime = 280 mg/L and $Ca(OCl)_2 = 1.5$ mg/L)

results were observed by Zewge [9] for the removal of fluoride from water using a combined aluminum sulfate/hydroxide process. Fig. 1(b) shows a 2.5 $\log_{10}$ reduction of *E. coli* bacteria after 10 min of exposure. *E. coli* reductions of 5 $\log_{10}$ were achieved within 20 min. After 20 min, there was no significant change in *E. coli* log reduction. The results agree with previous research findings by Owoseni et al. [21] on the inactivation of *E. coli* using calcium hypochlorite. The results showed that 5 $\log_{10}$ *E. coli* reduction was achieved within 20 min and that after 20 min, there was no significant difference in *E. coli* log reduction. This shows that more than 20 min of contact time does not affect *E. coli* inactivation. Because the reactions involved *E. coli* log reduction and fluoride removal, 30 min was enough to reduce the fluoride concentration to 1.5 mg/L and achieve 5 *E. coli* log reduction. Therefore, a contact time of 30 min was selected as the optimum contact time for further experiments.

Effect of initial fluoride concentration

The findings were plotted as fluoride removal efficiency versus initial concentration of fluoride (Fig. 2(a)) and *E. coli* log reduction versus $[F^-]_0$ (Fig. 2(b)). Maximum fluoride removal occurred at lower fluoride concentration, as shown in Fig. 2(a). For a given amount of coagulant level, fluoride removal is high at low fluoride concentration because there are more aluminum oxide complexes for fluoride ions [32]. As fluoride concentration increases, the binding potential of aluminum oxide reaches saturation, resulting in a decline in fluoride removal. This is attributed to the intensive use of all active sites on the complex's surface, and the incoming fluoride ion from the water being repelled by repulsive forces [33]. These results are in line with Hussein and Vegi [31], who found that the fluoride removal efficiency reduced from 93 to 78% with the increase in $[F^-]_0$ from 8 to 25 mg/L. Initial fluoride concentration did not significantly affect *E. coli* log reduction, as shown in Fig. 2(b). Fluoride is toxic to bacteria, fungi, plants, and animals at high concentrations [34]. To overcome this problem of fluoride toxicity, *E. coli* bacteria developed fluoride resistance mechanisms. *E. coli* has a class of regulatory RNAs, or riboswitches, that bind to fluoride and regulate gene expression in response to this anion. Riboswitches are metabolite or ion-sensing structured RNA motifs typically found in the noncoding regions of specific mRNAs. They regulate the expression of neighboring protein-coding regions via various mechanisms, including transcription termination, translation blockade, and alternative splicing [35]. This explains why fluoride concentration did not affect *E. coli* log reduction.

Effect of pH

The fluoride removal process, which involves the hydrolysis of alum and the preferential adsorption of fluoride ions onto aluminum oxide undergoing precipitation, is highly dependent on the pH of the solution.

The percentage of fluoride removal increases as the pH of the solution increases from 3 to 8, and the maximum fluoride removal is observed at pH 6 to 7 (Fig. 3(a)). $Al(OH)_3$ is responsible for fluoride removal. The $Al(OH)_3$ floc is believed to adsorb F^- strongly, and the formation of this precipitate is optimal in a pH range of 6 to 7 [36]. Below the pH of 5, there is insufficient aluminum hydroxide to precipitate $Al(OH)_3$ since aluminum species such as Al^{3+}, $Al(OH)^{2+}$ and $Al(OH)_2^+$ are prevalent [37]. When the pH was raised to 10, the percentage of fluoride removal gradually decreased because soluble $Al(OH)_4^-$ is the predominant species that repel the fluoride ions. This is consistent with the findings of [38], who also found that the maximum fluoride removal using aluminum oxide was achieved with a pH range of 6 to 7. The populations of *E. coli* were reduced by about 1 and 5 $\log_{10}$ CFU/100 mL in a pH range of 3 to 10, with a maximum log reduction achieved at 5–6 (Fig. 3(b)). pH is an essential factor that influences the efficiency of disinfection. When calcium hypochlorite is added to water, Eq. (4) occurs:

Hypochlorous acid dissociates to produce the hypochlorite ion, as shown in Eq. (6).

$$HOCl \rightarrow H^+ + OCl^- \qquad (6)$$

Hypochlorous acid is more reactive than hypochlorite ion and a more powerful disinfectant and oxidant [27]. A higher pH allows for more hypochlorite ions, resulting in less hypochlorous acid in the water [39]. Low pH allows for the formation of fewer hypochlorite

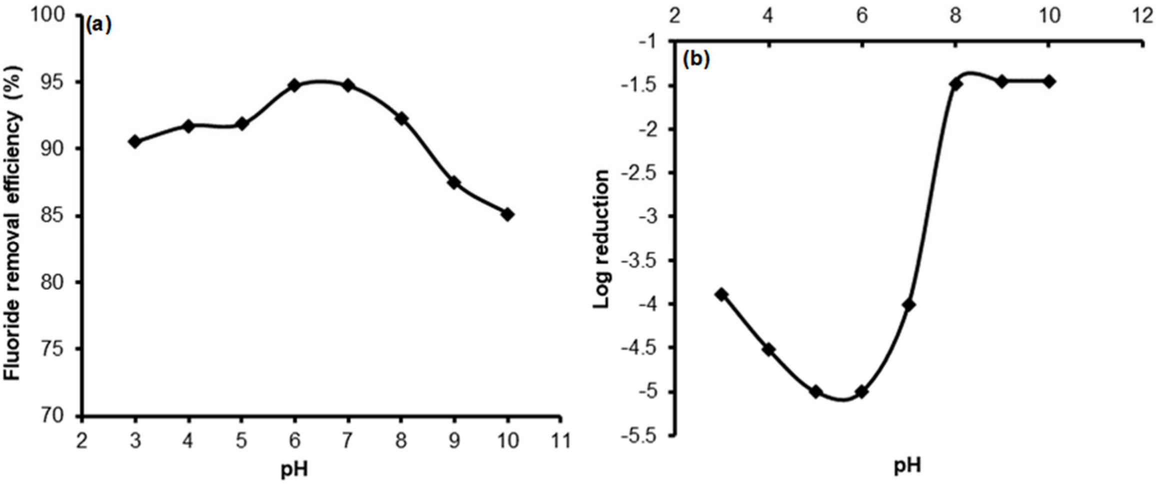

Fig 3. The pH effect on (a) fluoride removal and (b) *E. coli* log reduction ($[F^-]$ = 15 mg/L, contact time = 15 min, *E. coli* = 10^5 CFU/100 mL, AO = 75 mg/L, alum = 800 mg/L, lime = 280 mg/L and $Ca(OCl)_2$ = 1.5 mg/L)

ions and more hypochlorous acid. Hypochlorous acid is the most effective type of free chlorine residual, i.e., chlorine that is accessible to kill microbes in water, whereas hypochlorite ions are far less effective disinfectants. As a result, disinfection is more efficient at low pH (with high amounts of hypochlorous acid in water) than at high pH (with high amounts of hypochlorite ions in water) [40]. This explains why *E. coli* concentrations were lower in the acidic medium compared to the primary media. These findings are consistent with Owoseni et al. [21], who showed that maximal log reduction of *E. coli* with calcium hypochlorite was attained in pH solutions ranging from 3 to 7.

Effect of initial E. coli concentration

The initial *E. coli* concentration did not affect fluoride removal (Fig. 4(a)). This might be attributed to the fact that there is no competition between *E. coli* and fluoride ions for the binding site. The log reduction rate decreased as *E. coli* concentration increased (Fig. 4(b)). At initial concentrations of 10^4 and 10^5 CFU/100 mL, 5 and 6 $\log_{10}$ reduction was achieved. At initial concentrations of 10^7 and 10^8 CFU/100 mL, the $\log_{10}$ reduction was reduced to 2.

More calcium hypochlorite or more extended treatment periods will be needed to destroy the same proportion of *E. coli* cells at very high cell concentrations (10^8 CFU/100 mL and more). The *E. coli* concentration effect seen here may be attributed to the concentration of molecules available to interact with *E. coli* cells [27]. For the inactivation of *E. coli*, there is a need for direct interaction between a given concentration of hypochlorite in a solution and bacterial cell membranes [21]. This suggests that a saturation point is reached at high *E. coli* concentrations where enough bioactive molecules are required to associate with prominent receptor locations within the cell. The concentration of hypochlorite ions available to kill *E. coli* cells becomes very limited at high *E. coli* concentrations, and hence a higher reduction rate is not achieved [27]. These results are in line with those of Liang et al. [41], who argued that the initial *E. coli* concentration affects the log inactivation of *E. coli*.

Testing the Composite with Actual Water Samples

The concentrations of Fluoride and *E. coli* before and after treatment are shown in Table 4. The FRE was in the range of 77.5 to 89.1%. Treated water samples from Serity, Tejitu, Germama, and Anano had fluoride concentration that were above the acceptable WHO drinking water standard. The reduced fluoride removal efficiency could be attributed to the alkalinity of the water samples. Alkalinity includes hydroxides, carbonates, and bicarbonate. Carbonate has a high affinity for $Al(OH)_3$. The observed decrease in fluoride removal may be attributed to the rivalry for aluminum hydroxide complexes between the carbonate and fluoride anion [36].

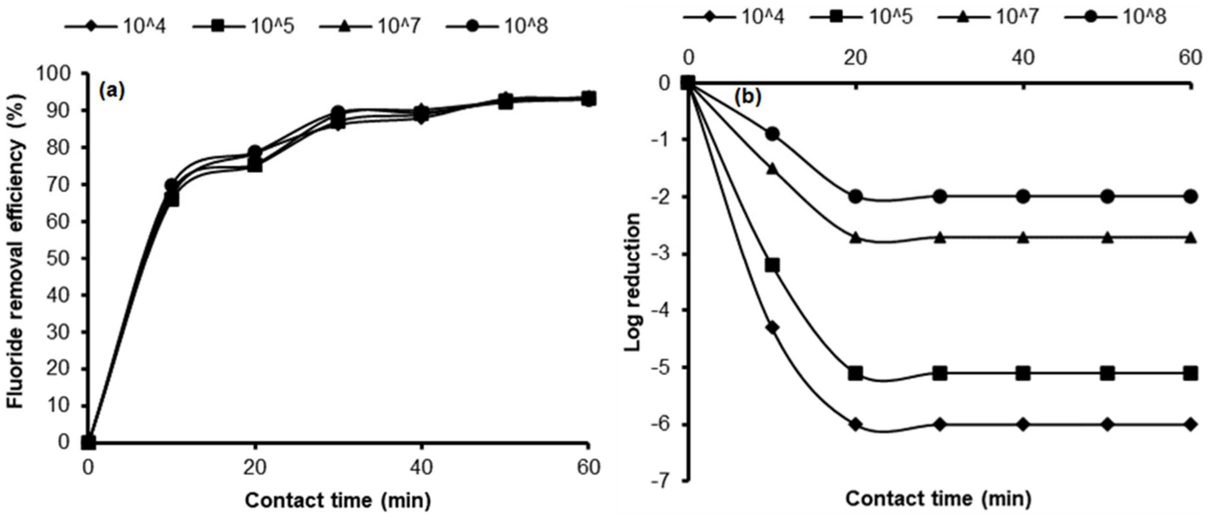

Fig 4. The effect of initial *E. coli* concentration on (a) fluoride removal and (b) *E. coli* log reduction

Table 4. Concentrations of different water quality parameters for actual water samples

Drinking water quality parameters	Tube		Serity		Dollessa	
	RW	TW	RW	TW	RW	TW
Fluoride (mg/L)	3.67 ± 0.10	0.4 ± 0.01	12.3 ± 0.34	1.6 ± 0.08	5.55 ± 0.23	0.97 ± 0.12
pH	7.55 ± 0.12	7.20± 0.10	8.10 ± 0.34	7.24 ± 0.3	8.19 ± 0.40	6.62 ± 0.20
EC (µs/cm)	410 ± 0.79	546 ± 0.50	770 ± 0.90	836 ± 1.50	530 ± 1.03	767 ± 1.00
TDS (mg/L)	205 ± 1.22	273 ± 1.50	385 ± 1.72	458.5 ± 0.50	265 ± 2.33	383.5 ± 2.16
E. coli (CFU/100 mL)	1.9×10^5 ± 13.0	19 ± 2.12	1.6×10^5 ± 10	0	1.77×10^5 ± 21	0
Turbidity (NTU)	58 ± 0.17	5 ± 0.09	3.3 ± 0.01	<1 ± 0.01	<1 ± 0.03	<1 ± 0.01
Alkalinity as $CaCO_3$ (mg/L)	739 ± 0.81	-	821 ± 0.91	-	856 ± 0.12	-
Aluminum (mg/L)	0.01 ± 0.01	0.14 ± 0.01	0.01 ± 0.001	0.15 ± 0.01	0.01 ± 0.001	0.04 ± 0.005
Chloride (mg/L)	1.19 ± 0.01	2.35 ± 0.10	1.20 ± 0.12	3.10 ± 0.05	0.40 ± 0.03	1.27 ± 0.20
Sulfate (mg/L)	87 ± 0.89	135 ± 1.00	98 ± 0.91	140 ± 1.50	50 ± 0.70	117 ± 1.50
Residual Chlorine (mg/L)	-	0.08 ± 0.01	-	0.39 ± 0.04	-	0.33 ± 0.01
Drinking water quality parameters	Tejitu		Germama		Anano	
	RW	TW	RW	TW	RW	TW
Fluoride (mg/L)	9.63 ± 0.31	1.63 ± 0.03	15.1 ± 0.09	2.8 ± 0.03	10.5 ± 0.24	2.36 ± 0.67
pH	8.10 ± 0.12	7.32 ± 0.01	8.01 ± 0.45	7.16 ± 0.01	7.66 ± 0.82	6.90 ± 0.01
EC (µs/cm)	700 ± 1.33	836 ± 2.00	1115 ± 1.61	1335 ± 1.50	1101 ± 1.23	1348 ± 1.50
TDS (mg/L)	350 ± 1.21	418 ± 1.50	557.5 ± 1.83	667 ± 1.50	550.5 ± 0.21	674 ± 2.00
E. coli (CFU/100 mL)	1.98×10^5 ± 23	0	1.08×10^5 ± 17	87 ± 4	1.3×10^4 ± 12	0
Turbidity (NTU)	<1 ± 0.01	<1 ± 0.02	59 ± 0.12	7 ± 0.02	<1 ± 0.01	<1 ± 0.01
Alkalinity as $CaCO_3$ (mg/L)	809 ± 0.21	-	791 ± 0.21	-	784 ± 0.01	-
Aluminum (mg/L)	0.01 ± 0.002	0.06 ± 0.005	0.02 ± 0.002	0.06 ± 0.001	0.01 ± 0.002	0.18 ± 0.005
Chloride (mg/L)	0.90 ± 0.21	2.10 ± 0.20	1.43 ± 0.02	3.97 ± 0.08	0.82 ± 0.02	2.70 ± 0.10
Sulfate (mg/L)	67±0.45	127 ± 1.00	56 ± 2.12	97 ± 1.50	30±0.21	73 ± 0.50
Residual Chlorine (mg/L)	-	0.44 ± 0.02	-	0.12 ± 0.01	-	0.42 ± 0.01

Note: RW: Raw water. TW: Treated water

A similar interfering role of alkalinity due to carbonate ions on fluoride removal by activated carbon was also reported [42].

Treated water from Tube and Germama had *E. coli* concentration that was above the WHO drinking water standard. The residual chlorine of these water samples was 0.08 and 0.12 mg/L, respectively, which is below WHO drinking water standards. This could be attributed to the turbidity of the water samples. The turbidity of these water samples was 58 and 59 NTU for Tube and Germama, respectively. Turbidity contains organic compounds [43]. The organic compounds in turbidity are known to react with hypochlorite, increasing the chlorine demand and thereby reducing *E. coli* log reduction. In addition, organic and inorganic particles contained in turbidity protect microorganisms from free chlorine disinfection, a phenomenon recognized as 'particle association' [39]. The stabilization of cell membranes protects by restricting access to critical components for cellular inactivation and microbial inactivation [44]. These results align with the findings of Léziart et al. [45], where *E. coli* log reduction reduced from 4 log reduction to 1 log due to turbidity. It was discovered that total organic carbon interferes with preserving free chlorine residual. To preserve adequate chlorine disinfection performance, the turbidity of the raw water is suggested to be 1 to 5 NTU or a high dose of coagulant and chlorine is required. The results show that the actual water samples require a high dose of calcium hypochlorite to meet the drinking water standard for a turbid water sample. On the other hand, aluminum, chloride, sulfate, pH, alkalinity, and total dissolved solids were within the permissible limits. To meet the drinking water standards, batch experiments were conducted to find the dose that could effectively treat actual water samples with high fluoride concentration and high turbidity. The results showed that the water samples were alkaline, and hence the lime dose was reduced and $Ca(OCl)_2$ concentration increased from 1.5 to 2.0 mg/L to effectively reduce *E. coli* concentration. A dose of 95 mg/L AO, 900 mg/L alum, 15% lime, and 2 mg/L calcium hypochlorite effectively treated the actual water samples with up to 15 mg/L fluoride and 58 NTU turbidity.

Development of a Prototype

The prototypes were developed to be used as follows: (1) Mix contents in 10 L of water, (2) Stir the well for 5 min and let the water stand for 25 min, (3) Use a clean cloth to filter the treated water and dispose of the filtered floc, (4) The water is ready to use.

The developed prototypes were tested on actual water samples to assess their suitability for drinking purposes. The water quality parameters for treated water were within the WHO drinking water standards, as shown in Tables 5, 6 and 7). Residual chlorine concentrations were 0.29 to 0.46 mg/L, that lies within the recommended concentration. This means that the treated water can be protected against recontamination.

■ CONCLUSION

The results showed that the dose of 75 mg/L AO, 800 mg/L alum, 35% lime, and 1.5 mg/L calcium hypochlorite effectively achieved 95% fluoride removal and 5 $\log_{10}$ reduction of *E. coli*. A contact time of 30 min

Table 5. Concentrations of raw and treated water by the high dose developed POU prototype

Drinking water quality parameters	Germama		Serity	
	RW	TW	RW	TW
Fluoride (mg/L)	15.1 ± 0.01	1.45 ± 0.12	12.3 ± 0.12	1.03 ± 0.9
pH	8.01 ± 0.16	7.12 ± 0.05	8.1 ± 0.23	7.11 ± 0.05
Electrical conductivity (μs/cm)	1115 ± 0.23	1410 ± 0.70	770 ± 0.26	920 ± 1.20
Total dissolved solids (mg/L)	558 ± 0.76	705 ± 1.40	385 ± 0.23	460 ± 1.20
E. coli (CFU/ 100 mL)	1.08×10^5 ± 12	0	1.6×10^5 ± 21	0
Turbidity (NTU)	59 ± 1.20	<1 ± 0.01	3.3 ± 0.01	<1 ± 0.01
Alkalinity as $CaCO_3$ (mg/L)	841 ± 1.23	-	831 ± 0.47	-
Aluminum (mg/L)	0.02 ± 0.01	0.1 ± 0.01	0.01 ± 0.001	0.18 ± 0.02
Sulfate (mg/L)	56 ± 0.92	107 ± 0.15	98 ± 0.72	133 ± 0.93
Chloride (mg/L)	1.43 ± 0.54	4.83 ± 0.0	1.2 ± 0.24	3.4 ± 0.07
Residual chlorine (mg/L)	-	0.40 ± 0.01	-	0.42 ± 0.01

Table 6. Concentrations of raw and water treated by the medium dose developed POU prototype

Drinking water quality parameters	Anano		Tejitu	
	RW	TW	RW	TW
Fluoride (mg/L)	10.5 ± 0.13	1.07 ± 0.50	9.63 ± 0.02	0.79 ± 0.05
pH	8.2 ± 0.12	6.89 ± 0.50	8.1 ± 0.30	7.12 ± 0.01
Electrical conductivity (μs/cm)	1040 ± 0.23	1270 ± 1.20	700 ± 0.19	911 ± 1.50
Total dissolved solids (mg/L)	520 ± 0.34	635 ± 1.31	350 ± 0.45	455.5 ± 1.50
E. coli (CFU/100 mL)	1.3×10^4 ± 1	0	1.98×10^5 ± 10	0
Turbidity (NTU)	<1 ± 0.01	<1 ± 0.01	<1 ± 0.01	<1 ± 0.01
Alkalinity as $CaCO_3$ (mg/L)	784 ± 0.021	-	809 ± 0.21	-
Aluminum (mg/L)	0.01 ± 0.001	0.2 ± 0.003	0.01 ± 0.001	0.1 ± 0.005
Sulfate (mg/L)	30 ± 0.21	97 ± 1.3	67 ± 0.45	109 ± 1.2
Chloride (mg/L)	0.82 ± 0.02	3 ± 0.02	0.9 ± 0.21	2.58 ± 0.01
Residual chlorine (mg/L)	-	0.44 ± 0.02	-	0.46 ± 0.01

Table 7. Concentrations of raw and water treated by the low dose developed POU prototype

Drinking water quality parameters	Tube	
	RW	TW
Fluoride (mg/L)	3.67 ± 0.10	0.39 ± 0.01
pH	7.55 ± 0.12	6.56 ± 0.50
Electrical conductivity (µs/cm)	410 ± 0.79	765 ± 1.50
Total dissolved solids (mg/L)	205 ± 1.22	382 ± 1.50
E. coli (CFU/100 mL)	1.9×10^5 ± 13	0
Turbidity (NTU)	58 ± 0.43	2 ± 0.01
Alkalinity as $CaCO_3$ (mg/L)	739 ± 0.81	-
Aluminum (mg/L)	0.01 ± 0.001	0.16 ± 0.003
Sulfate (mg/L)	87 ± 0.89	147 ± 1.50
Chloride (mg/L)	1.19 ± 0.01	2.77 ± 0.20
Residual chlorine (mg/L)	-	0.29 ± 0.01

was enough to achieve 90% fluoride removal and 5 log reduction of *E. coli*. Fluoride concentration showed an impact on fluoride removal as high fluoride concentration resulted in reduced fluoride removal efficiency. On the other hand, fluoride concentration did not significantly affect *E. coli* log reduction. The pH of raw water showed an impact on both fluoride removal and *E. coli* log reduction. Maximum fluoride removal was achieved at pH 6 to 7. Maximum *E. coli* log reduction was achieved at pH 6 to 7. Acidic pH values also achieved high log reduction. Initial *E. coli* concentration did not show any effect of fluoride removal, but it impacted *E. coli* log reduction. Water samples from the Ethiopian Rift Valley required a high dose of alum, AO, calcium hypochlorite, and a low dose of lime because of the alkalinity and organic matter associated with turbidity. Three prototypes were developed for treating water samples with a fluoride concentration ≤ 15 mg/L. The POU prototypes were tested on water samples from Rift Valley. The treated water was analyzed for fluoride, pH, EC, TDS, *E. coli*, alkalinity, aluminum, sulfate, turbidity, residual chlorine, and chloride. The obtained values were compared with the WHO drinking water standards. All parameters were within the permissible limits. The developed products effectively treat water with fluoride concentration above 10 mg/L and turbidity above 5 NTU, thus indicating the capability of the developed products to purify contaminated water for human consumption.

ACKNOWLEDGMENTS

We would like to sincerely thank the World Bank Group for providing research fund through the Africa Centre of Excellence for Water Management (ACEWM), Addis Ababa University, for awarding the scholarship for Patricia Namate to pursue her MSc studies. We would also like to thank the communities around the sampling areas for their dedicated support during the data collection.

AUTHOR CONTRIBUTIONS

All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.

REFERENCES

[1] Immurana, M., Iddrisu, A.A., Mohammed, Z., and Toby Joseph Mathew, K.K., 2022, Access to basic drinking water and sanitation in Africa: Does financial inclusion matter?, *Cogent Social Sci.*, 8 (1), 2057057.

[2] Duke, W.F., Nordin, R.N., Baker, D., and Mazumder, A., 2016, The use and performance of BioSand filters in the Artibonite Valley of Haiti: a field study of 107 households, *Rural Remote Health*, 6 (3), 570.

[3] Admasie, A., Abera, K., and Feleke, F.W., 2022, Household water treatment practice and associated factors in rural households of Sodo Zuria District, Southern Ethiopia: Community-based cross-sectional, *Environ. Health Insights*, 16, 11786302221095036.

[4] Naik, P.K., 2016, The water crisis in Africa: Myth or reality?, *Int. J. Water Resour. Dev.*, 33 (2), 326–339.

[5] Kut, K.M.K., Sarswat, A., Srivastava, A., Pittman, C.U., and Mohan, D., 2016, A review of fluoride in African groundwater and local remediation methods, *Groundwater Sustainable Dev.*, 2-3, 190–212.

[6] Mapoma, T.W.H., Xie, X., Zhang, L., Nyirenda, M.T., Maliro, A., and Chimutu, D., 2016, Hydrochemical characteristics of rural community groundwater supply in Blantyre, southern Malawi, *J. Afr. Earth. Sci.*, 114, 192–202.

[7] Tekle-Haimanot, R., Melaku, Z., Kloos, H., Reimann, C., Fantaye, W., Zerihun, L., and Bjorvatn, K., 2006, The geographic distribution of fluoride in surface and groundwater in Ethiopia with an emphasis on the Rift Valley, *Sci. Total Environ.*, 367 (1), 182–190.

[8] Nyangi, M.J., Chebude, Y., and Kilulya, K.F., 2020, Fluoride removal efficiencies of Al-EC and Fe-EC reactors: Process optimization using Box–Behnken design of the surface response methodology, *Appl. Water Sci.*, 10 (9), 214.

[9] Zewge, F., 2016, Combined aluminum sulfate/hydroxide process for fluoride removal from drinking water, *Bull. Chem. Soc. Ethiop.*, 30 (3), 391–401.

[10] Malago, J., Makoba, E., and Mazuka, A.N.N., 2017, Fluoride levels in surface and groundwater in Africa: A review, *Am. J. Water Sci. Eng.*, 3 (1), 1–17.

[11] Dey, S., and Giri, B., 2016, Fluoride fact on human health and health problems: A review, *Med. Clin. Rev.*, 2 (1), 2.

[12] Demelash, H., Beyene, A., Abebe, Z., and Melese, A., 2019, Fluoride concentration in groundwater and prevalence of dental fluorosis in Ethiopian Rift Valley: Systematic review and meta-analysis, *BMC Public Health*, 19 (1), 1298.

[13] Ayalew, A.A., 2023, Comparative adsorptive performance of adsorbents developed from kaolin clay and limestone for de-fluoridation of groundwater, *S. Afr. J. Chem. Eng.*, 44, 1–13.

[14] Dubey, S., Agrawal, M., and Gupta, A.B., 2018, Advances in coagulation technique for the treatment of fluoride-contaminated water: A critical review, *Rev. Chem. Eng.*, 35 (1), 109–137.

[15] Mulugeta, E., Zewge, F., Johnson, A.C., and Chandravanshi, B.S., 2013, A high-capacity aluminum hydroxide-based adsorbent for water defluoridation, *Desalin. Water Treat.*, 52 (28-30), 5422–5429.

[16] Bogale, G.G., 2020, Hotspots of unimproved sources of drinking water in Ethiopia: Mapping and spatial analysis of Ethiopia demographic and health survey data 2016, *BMC Public Health*, 20 (1), 878.

[17] Holtslag, H., and Mekonta, L., 2014, HWTS in Ethiopia: The State of Art and Ideas to Scale Up, *Internal Report*, Millenium Water Alliance, Addis Ababa, Ethiopia.

[18] Sharma, S., and Bhattacharya, A., 2017, Drinking water contamination and treatment techniques, *Appl. Water Sci.,* 7 (3), 1043–1067.

[19] Mulugeta, E., Zewge, F., Johnson, C.A., and Chandravanshi, B.S., 2014, Aluminum hydro(oxide)-based (AO) adsorbent for defluoridation of drinking water: Optimisation, performance comparison, and field testing, *Water SA*, 41 (1), 121–128.

[20] Bridgewater, L., American Public Health Association, American Water Works Association, and Water Environment Federation, 2012, *Standard Methods for the Examination of Water and Wastewater*, 22nd Ed., Eds. Rice, E.W., and Bridgewater, L., American Public Health Association, Washington DC, USA.

[21] Owoseni, M.C., Olaniran, A.O., and Okoh, A.I., 2017, Chlorine tolerance and inactivation of *Escherichia coli* recovered from wastewater treatment plants in the Eastern Cape, South Africa, *Appl. Sci.*, 7 (8), 810.

[22] Dargahi, A., Atafar, Z., Mohammadi, M., Azizi, A., Almasi, A., and Ahagh, M.M., 2016, Study of the efficiency of alum coagulant in fluoride removal from drinking water, *Int. J. Pharm. Technol.*, 8 (3), 16772–16778.

[23] Shimelis, B., Zewge, F., and Chandravanshi, B.S., 2006, Removal of excess fluoride from water by aluminum hydroxide, *Bull. Chem. Soc. Ethiop.*, 20 (1), 17–34.

[24] Wang, L., Zhang Y., Sun N., Sun W., Hu Y., and Tang H., 2019, Precipitation methods using calcium-containing ores for fluoride removal in wastewater, *Minerals*, 9 (9), 511.

[25] Mizozoe, M., Otaki, M., and Aikawa, K., 2019, The mechanism of chlorine damage using enhanced green fluorescent protein-expressing *Escherichia coli*, *Water*, 11 (10), 2156.

[26] Dodd, M.C., 2012, Potential impacts of disinfection processes on elimination and deactivation of antibiotic resistance genes during water and wastewater treatment, *J. Environ. Monit.*, 14 (7), 1754–1771.

[27] Erkmen, O., 2010, Antimicrobial effects of hypochlorite on *Escherichia coli* in water and selected vegetables, *Foodborne Pathog. Dis.*, 7 (8), 953–958.

[28] Vargas, T.F., Baía, C.C., Machado, T.L.S., Dórea, C.C., and Bastos, W.R., 2021, Decay of free residual chlorine in wells water of northern Brazil, *Water*, 13 (7), 992.

[29] CDC, 2012, *Global WASH-Related Diseases and Contaminants*, Center for Diseases Control and Prevention, Atlanta, Georgia, USA.

[30] WHO, 2019, *Results of Round Two of the WHO International Scheme to Evaluate Household Water Treatment Technologies,* World Health Organization, Geneva, Switzerland.

[31] Hussein, I.A., and Vegi, M.R., 2020, Defluoridation of drinking water using coalesced and un-coalesced mica, *Appl. Water Sci.*, 10 (2), 64.

[32] Alemu, S., Mulugeta, E., Zewge, F., and Chandravanshi, B.S., 2014, Water defluoridation by aluminium oxide–manganese oxide composite material, *Environ. Technol.*, 35 (15), 1893–1903.

[33] Getachew, T., Hussein, A., and Rao, V.M., 2015, Defluoridation of water by activated carbon prepared from banana (*Musa paradisiaca*) peel and coffee (*Coffea arabica*) husk, *Int. J. Environ. Sci. Technol.*, 12 (6), 1857–1866.

[34] Li, S., Smith, K.D., Davis, J.H., Gordon, P.B., Breaker, R.R., and Strobel, S.A., 2013, Eukaryotic resistance to fluoride toxicity mediated by a widespread family of fluoride export proteins, *Proc. Natl. Acad. Sci.*, 110 (47), 19018–19023.

[35] Breaker, R.R., 2009, Riboswitches: From ancient gene-control systems to modern drug targets, *Future Microbiol.*, 4 (7), 771–773.

[36] Emamjomeh, M.M., and Farzadkia, M.F., 2016, *Fluoride Removal by Electrocoagulation (EC) Process: Aluminium Chemistry and Speciation*, Proceedings of 47th The IRES International Conference, St. Petersburg, Russia.

[37] Hu, C.Y., Lo, S.L. and Kuan, W.H., 2003, Effects of co-existing anions on fluoride removal in electrocoagulation (EC) process using aluminum electrodes, *Water Res.*, 37 (18), 4513–4523.

[38] Waghmare, S.S., and Arfin, T., 2015, Fluoride removal from water by aluminium based adsorption: A review, *J. Biol. Chem. Chron.*, 2 (1), 1–11.

[39] Chahal, C., van den Akker, B., Young, F., Franco, C., Blackbeard, J., and Monis, P., 2016, "Chapter Two - Pathogen and Particle Associations in Wastewater: Significance and Implications for Treatment and Disinfection Processes" in *Advances in Applied Microbiology*, Volume 97, Eds., Sariaslani, S., and Michael Gadd, G., Academic Press, Cambridge, USA, 63–119.

[40] US Environmental Protection Agency, 2009, *Drinking Water Contaminants, National Primary Drinking Water Regulations,* U.S Government Publishing Office, Washington DC., USA.

[41] Liang, Z., Chan, W.L., Tian, X., Lai, A.C.K., Lee, P.K.H., and Chan, C.K., 2022, Inactivation of *Escherichia coli* in droplets at different ambient relative humidities: Effects of phase transition, solute and cell concentrations, *Atmos. Environ.*, 280, 119066.

[42] Sano, H., Omine, K., Prabhakaran, M., Darchen, A., and Sivasankar, V., 2018, Groundwater fluoride removal using modified mesoporous dung carbon and the impact of hydrogen-carbonate in borehole samples, *Ecotoxicol. Environ. Saf.*, 165, 232–242.

[43] Stange, C., Sidhu, J.P.S., Toze, S., and Tiehm, A., 2019, Comparative removal of antibiotic resistance genes during chlorination, ozonation, and UV treatment, *Int. J. Hyg. Environ. Health*, 222 (3), 541–548.

[44] Li, Y., Song, Y., Huang, Z., Mei, L., Jiang, M., Wang, D., and Wei, Q., 2023, Screening of *Staphylococcus aureus* for disinfection evaluation and transcriptome analysis of high tolerance to chlorine-containing disinfectants, *Microorganisms*, 11 (2), 475.

[45] Léziart, T., Dutheil de la Rochere, P.M., Cheswick, R., Jarvis, P., and Nocker, A., 2019, Effect of turbidity on water disinfection by chlorination with the emphasis on humic acids and chalk, *Environ. Technol.*, 40 (13), 1734–1743.