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# Fluoride Removal with Augmented Bentonite in Drinking Water and Community Perceptions in Arusha, Tanzania

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FLUORIDE REMOVAL WITH AUGMENTED BENTONITE IN DRINKING WATER

AND COMMUNITY PERCEPTIONS IN ARUSHA TANZANIA


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By

Kara Okular



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ABSTRACT


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Thesis supervised by Dr. David M. Kahler

High fluoride concentrations in drinking water affect millions of people around the world. The World Health Organization recommended a fluoride concentration in drinking water of $\leq 1.5$ mg/L; fluoride above this concentration can cause long-term dental and skeletal fluorosis. Fluoride imparts no taste to water; therefore, people do not realize that they are ingesting high fluoridated water. Researchers sought to develop a material to remove fluoride from drinking water through sorption. Aluminum was used to form a double ionic layer on the bentonite clay surface. Once mixed in contaminated water, a coagulant, ferric sulfate, was used to separate the bentonite by coagulation and flocculation. For use in a point-of-use format, calcium hypochlorite was tested to reduce potential pathogens. Fluoride reduction was tested in the laboratory with deionized water or synthetic groundwater with sodium fluoride added, and in the field with natural

fluoride-rich water from Tanzania, by an ion selective electrode.  The augmented

bentonite could also be used in public water treatment.  Ultimately, researchers hope to

recommend a centralized treatment technology that will reduce fluoride and potential

pathogens.

The community of Olkokola, in the Arusha region of Tanzania, was investigated

to gain a greater understanding of residential life.  Researchers investigated water use and

perceptions through household surveys and interviews with the local water board.  The

results indicated significant problems with consistent access to water proximal to

residents.  Olkokola residents experience electrical interruptions, water collection issues,

a lack of water treatment, hygiene problems, and erosion issues.

DEDICATION

I would like to dedicate this thesis to my boyfriend, roommates, and family for

their constant support and encouragement through the past two years of my research.

# ACKNOWLEDGEMENT

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$Al_2(SO_4)_3$- Aluminum Sulfate

c- Concentration

$CaCO_3$- Calcium Carbonate

$Ca(ClO)_2$- Calcium Hypochlorite

°C- Degrees Celsius

DI- Deionized, deionized water

EPA- Environmental Protection Agency, United States

$Fe_2(SO_4)_3$- Ferric Sulfate

FNU- Formazin nephelometric unit

g- Gram

HCl- Hydrochloric Acid

IRB- Institutional Review Board

ISE- Ion Selective Electrode

ISO- International Organization for Standardization

kPa- Kilopascal

l- Liter

mg- Milligram

mm- millimeter

$MgSO_4$- Magnesium Sulfate

NaF- Sodium Fluoride

NaOH- Sodium Hydroxide

POU- Point-Of-Use

rpm- Rotations Per Minute

TISAB- Total Ionic Strength Adjusted Buffer

USD- United States Dollar

V- Volume

WHO- World Health Organization

$W/m^2$- Watt per square meter

XRPD- Powder X-ray Diffractometer (PANalytical X'Pert Pro MPD)

# 1. Introduction

The United States and other countries have been artificially adding fluoride to their

drinking water for over 50 years (Browne et al., 2005). The World Health Organization

(WHO) recommends that the safe level of fluoride in drinking water is ≤1.5 mg/L (World

Health Organization, 2017); the American Dental Association and the United States

Public Health Service concluded that the optimal amount of fluoride in drinking water for

children under the age of six is 0.7 mg/L (U.S. Department of Health Education and

Welfare, 1962; Wright et al., 2014) to help protect tooth enamel and prevent decay.

According to the Tanzania Bureau of Standards (2018), National Environmental

Standards Compendium, the preferred limit for fluoride in drinking water is 1.5 mg/L and

the absolute maximum limit is 4.0 mg/L. Water above the 1.5 mg/L WHO (2017)

standards is considered to have high fluoride. The World Health Organization (1994)

corrected their fluoride consumption standard to match climatic conditions and

recommended that national standards be set to incorporate varying temperatures.

Fantong et al. (2010) used the Galagan & Vermillion (1957) equation for an average

annual temperature of 28.7° Celsius and found that the optimal fluoride level should be

0.6 mg/l if residents are drinking >3 liters of water per day.

Fluoride occurs naturally in volcanic rock and can enter groundwater (Edmunds &

Smedley, 2013). Fluoride ions are introduced into waterways by either natural conditions

or from anthropogenic sources (Mohapatra et al., 2009). Locations of geothermal

hotspots such as volcanoes typically contain more fluoride. The prevailing theory is that

various types of volcanic rocks are rich in fluoride and can deposit it into the passing water. Basalt, granite, shale, and syenite are common volcanic rocks that contain fluoride (Edmunds & Smedley, 2013). Cryolite, fluorapatite, fluorite, topaz, and villiaumite are minerals in volcanic rocks that contain higher levels of fluorine compared to other minerals (Nanyaro et al., 1984). High concentrations of calcium and magnesium levels are common in bodies of water that have low fluoride (Nanyaro et al., 1984). Edmunds & Smedley (2013) reported a correlation between high calcium and magnesium levels and low fluoride levels.

Edmunds & Smedley (2013) hypothesized that fluoride concentrations are higher in arid climates since there is more contact time between the groundwater and rock; furthermore, tropical areas with high rainfall levels have diluted groundwater fluoride concentrations. There are many unknown factors that control fluoride in water. One theory is that deep groundwater is expected to contain higher fluoride levels because of the high residence time in aquifers as the sediments reach equilibrium, while another is that shallow groundwater near volcanos are affected by hydrothermal inputs, which causes the potential of high fluoride levels (Edmunds & Smedley, 2013). Unfortunately, drinking from shallow groundwater increases the risk of pathogens; shallow groundwater is exposed to the elements, humans, and animals, which increases the chances of pathogens in the water (Islam et al., 2011).

Ingestion of high levels of fluoride is a concern for children under the age of five since they are undergoing major developmental growth, which makes them susceptible to

fluoride effects, known as fluorosis (Edmunds & Smedley, 2013). The degree of severity of the deformities depends on the concentration of fluoride consumed. Fluorosis typically has dental and skeletal clinical presentations. Dental fluorosis is the hypomineralization of the enamel which causes higher than normal tooth porosity (Burt & Eklund, 1992). It also causes mottled teeth (Figure 1) which consists of white patches, pitting, or enamel staining (Browne et al., 2005). Dental fluorosis reduces the strength of the enamel which causes weakening and potential tooth loss (Edmunds & Smedley, 2013). Evans & Darvell (1995) determined that children are most susceptible to dental fluorosis on the edge, front, and middle of the maxillary central incisors during an eight-month period centered around the ages of 19 to 20 months for males and 25 to 26 months for females. Skeletal fluorosis often presents itself with similar symptoms as arthritis and is characterized by the deformation of long bones, such as bowed legs and, in extreme cases, neurological difficulties (Reddy, 2009). People with kidney diseases are thought to be more prone to skeletal fluorosis since they are unable to efficiently filter and excrete fluoride (Reddy, 2009; Xiong et al., 2007). Malin et al. (2019) found that plasma fluoride level changes are associated with changes in kidney and liver parameters. The study went on to say that kidney or liver abnormalities could affect fluoride adsorption. Fluorosis is irreversible (Edmunds & Smedley, 2013). Ishii and Nakagaki (cited by Evans & Darvell, 1995) showed that children who had almost fully developed maxillary central incisors when they began ingestion of water with high fluoride levels had less severe dental fluorosis compared to the younger children who ingested water with high fluoride, who's teeth were less developed.

**Figure 1: Severe dental fluorosis (Division of Oral Health National Center for Chronic Disease Prevention and Health Promotion, 2019).**

Small levels of fluoride in water can benefit oral hygiene but when paired with fluoridated products, the chance of fluorosis is increased. There are other avenues besides drinking water for children to ingest high levels of fluoride. Children could consume excessive amounts of fluoride from consumption of fluoridated water and dental products. Fluoridated toothpaste and mouthwash entered the European market in the 1970s and now comprises 95% of the toothpastes available for purchase (Browne et al., 2005). In areas with naturally high fluoride levels in water, using fluoridated toothpastes could cause children to ingest fluoride beyond the recommended dose. Brushing children's teeth at too young of an age and with too much fluoridated toothpaste can cause children to ingest higher levels of fluoride than intended. Children under the age of six should brush their teeth with no more than a pea sized amount of fluoridated toothpaste because they lack the ability to properly spit (Wright et al., 2014). The use of infant formula with high fluoride water, in children under a year, can cause excess fluoride consumption during major developmental growth (Browne et al., 2005; Osujp et al., 1988).

In the United States, dentists often apply fluoride treatments in the form of gel or paste on children's teeth to help protect them from further decay. This practice is to ensure that children's teeth are receiving enough fluoride for the tooth enamel to prevent caries (Larsen et al., 1985). In recent studies, fluoride was found to have antibacterial properties (Clarkson & McLoughlin, 2000). It increases the acid resistance of the enamel and reduces the acid tolerance of plaque causing bacteria (Marquis, 1990).

Removing fluoride from water is difficult and typically expensive. Researchers have experimented with multiple removal methods. There are three main categories for removal: membrane, thermal distillation, and adsorption techniques (Mohapatra et al., 2009). The techniques for membrane removal are reverse osmosis, nanofiltration, dialysis, and electro-dialysis. Reverse osmosis and nanofiltration use a membrane to obtain solutes while water passes through it, but nanofiltration is slightly different having less resistance to the passage of solvents and has larger pores (Mohapatra et al., 2009). Reverse osmosis creates highly pure water but is very expensive and needs a significant amount of energy, while nanofiltration requires less energy (Mohapatra et al., 2009). Both techniques produce brine waste that is unsafe for consumption and is difficult to dispose of safely (Meenakshi & Maheshwari, 2006). Dialysis separates solutes by transporting solutes through a membrane and electro-dialysis uses ion-exchange membranes with an electric field to remove ions (Mohapatra et al., 2009). All of these techniques are expensive, require a trained professional, and need a significant amount of consistent energy.

The second category for fluoride removal is thermal distillation. Unfortunately, fluoride cannot be boiled out of water. Fluoride, $F^-$, is an ion which is impossible to remove from water via boiling. However, it is possible to use distillation to purify water of fluoride. Distillation is the act of separating mixtures by using heat (Kiss, 2013). Fluoride removal using distillation is a multi-step process. It starts with bringing the water to a boil, collecting the evaporated water, and finally condensing the water back to liquid form. Theoretically, the condensed water will contain less fluoride than the starting water since fluoride cannot evaporate. Thermal distillation is not commonly used and not widely studied as a form of fluoride removal. The lack of thermal distillation use is because the evaporated water needs collected and chilled. Households often do not have the equipment needed and likely do not have the time to conduct the procedure.

The other category for fluoride removal, adsorption, is often researched more than membranes because it is typically more accessible and less expensive. Each sorbent needs to be evaluated properly for sorption capacity, pH effect and dependence, removal time, stability of substance, regeneration, cost, and any other anion or cation interference (Mohapatra et al., 2009). Most adsorption substances are point-of-use (POU) but some have been modified for treatment systems of small communities. Some common adsorbents are alumina and aluminum modified materials, clays and soils, calcium-based minerals, carbon-based minerals, and synthetic compounds, while other materials used are ion-exchange resins, layered double hydroxides, and zeolites (Mohapatra et al., 2009). These adsorbents have been experimented with in molecular sieves and filters.

Bone char is a common sorbent that is widely used and locally made across Africa. A study done by Dahi (2015) explains that the bone char color plays a role in defluoridation, color, taste, and total dissolved solids in the water. To create black bone char, the ideal color for the highest defluoridation, bones are fired at 550° Celsius with a small amount of oxygen when cooling. To create a white bone char, the amount of oxygen is increased during the cooling phase (Dahi, 2015). The bone char is then crushed into a fine powder that can be added to drinking water. Although bone char removes a large amounts of fluoride at a 4 mg/l F$^-$ initial concentration, it is controversial because the act of burning animal bones is culturally unacceptable by some populations, also there can be an unpalatable taste, dark color, and high total dissolved solids (Dahi, 2015; Medellin-Castillo et al., 2007).

Molecular sieves augmented with aluminum hydroxide is a technique for fluoride ion removal. An experiment conducted by Du et al. (2016) used molecular sieves with amended natural zeolites. Unfortunately, the aluminum (hydr)oxide amended molecular sieve lost its sorption capacity with more reuses, likely because of aluminum loss (Du et al., 2016). Another study by Du et al. (2017) examined pure aluminum (hydr)oxide, AlOOH, and AlOOH-amended sodalite in column filtration. The material lasted until 2,000 bed volumes when fluoride removal was no longer adequate, and aluminum was detected in the water.

Farrah & Pickering (1986) found that the adsorption by the amorphous gibbsite (Al(OH)$_3$) or alumina (Al$_2$O$_3$) were pH dependent. Maliyekkal et al. (2006) determined

that manganese oxide coated alumina had greater fluoride removal than activated alumina alone and that the process is pH dependent. Activated alumina filters are common among POU defluoridation techniques. Venkobachar et al. (1997) demonstrated a two-chamber unit with the top unit containing a microfilter with activated alumina and a lower unit for water collection. The filter needed regeneration every 1.5 to 3 months; that is, the sorption sites were cleared of their previous sorbates. Filter limitations often include low adsorption capacity over time and frequent replacements (Meenakshi & Maheshwari, 2006).

Kau et al. (1998) showed that bentonite clay has a higher fluoride sorption capacity than other clays, like kalinite. Bentonite is a swelling clay which indicates that it has a high cation exchange capacity (Weil & Brady, 2008). Due to its near-neutral buoyancy in water, powdered bentonite swells in water and remains suspended for a long period, which results in high turbidity, commonly observed as cloudy water. Turbidity is an important water quality parameter for drinking water acceptability and a coagulant is generally used to settle particles. Ferric sulfate, $Fe_2(SO_4)_3$, is commonly used in water treatment facilities to settle particles (Masters & Ela, 2008). Turbidity with and without ferric sulfate was measured to determine how well it settles the bentonite. For acceptability and to remove suspended solids, which can harbor pathogens, the bentonite needs to be settled in a reasonable amount of time for people to be willing to use the product in their household.

Our approach to an augmented bentonite clay was to create a substance that could potentially be incorporated into a water treatment plant. The Olkokola survey responses revealed that many households know that they should boil their water but simply choose to not, which is consistent with previous studies (Edokpayi et al., 2018; Kahler et al., 2016). The low adherence to point-of-use (POU) water treatment and also that there is no effective, practical POU system for fluoride removal suggests that a centralized system is needed. The WHO recommends the goal of water treatment should be a safely managed water supply, which requires centralized action (World Health Organization, 2017).

Deionized water and aluminum sulfate washed bentonite uses an adsorption mechanism to attract fluoride ions. Washing the bentonite twice creates a double ionic layer that allows fluoride ions to attach to the surface. The augmented bentonite can be used to remove fluoride from fluoride-rich groundwater and in conjunction with disinfectants, make it safe for consumption. A POU system can be created by having a single-use packet that contains the augmented bentonite, coagulant, disinfectant, and pH buffer. Households would be able to mix the packet into a 20-Liter bucket and after letting it settle, consume the treated water. In a community water treatment system, the augmented bentonite can be incorporated by adding an additional step before coagulation and flocculation.

We hypothesize that we can create an augmented bentonite that can remove fluoride from groundwater through cation exchange. We sought to investigate materials to generate the double ionic layer to enhance cation exchange potential. The use of a coagulant, ferric

sulfate, has the potential to remove small amounts of fluoride.  Our goal is to create a

fluoride removing material that can be implemented with either a POU mechanism or

water treatment facility.

## 2. Methods

### 2.1 Bentonite Augmentation

Bentonite clay is a swelling clay that is often used in landfill liners because of its low hydraulic conductivity (Shackelford et al., 2000). Bentonite is widely available and relatively inexpensive. The surface of bentonite is negatively charged and has a high surface-to-volume ratio compared to other aluminum-silicate clays (Weil & Brady, 2008). Using bentonite as a fluoride removal substance is not a new practice. In a study by (Srimurali et al., 1998), various clays were compared against each other to rank their fluoride removal capacity. One hundred mg of the clays were added to 50 ml of 5.0 mg/l fluoride solution and mixed for 5 hours. The study found that at optimal conditions, bentonite had a 46% fluoride removal rate compared to the 18.2% removal rate of kaolinite clay (Srimurali et al., 1998).

Fluoride is a negatively charged ion and the bentonite surface layer is also negative, therefore they will repel each other. In order to create an attraction, a double ionic layer on the bentonite is necessary (Figure 2). Introducing positive ions to the negative bentonite forms a new surface layer, which can now attract fluoride ions (Weil & Brady, 2008). To achieve the double ionic layer, the clay is washed with positive solutions (Sposito, 2004).

To expose the negative bentonite surface layer, the clay was washed with either sodium hydroxide (NaOH), deionized (DI, 18 MΩ·cm) water, or hydrochloric acid (HCl).  An initial wash strips any existing positive ions attached to the bentonite.  Washing it is important because there is a possibility that there are multiple ions already sorbed to the clay's surface which could reduce the optimized double ionic layer.  DI water was found to be the most effective and inexpensive wash.

Since both the bentonite surface layer and fluoride ions are negatively charged, there needs to be an intermediate positive ion to attract the fluoride.  We tested various solutions aluminum sulfate ($Al_2(SO_4)_3$), calcium carbonate ($CaCO_3$), and magnesium sulfate ($MgSO_4$), to see which one removed the most fluoride.  We chose to focus our efforts on aluminum sulfate because, washing the clay a second time with $Al_2(SO_4)_3$ allows $Al^{3+}$ ions to sorb via cation exchange to the negative surface layer.  Ideally, the bentonite surface is completely saturated with $Al^{3+}$ ions (Sposito, 2004) where fluoride ions may attach.
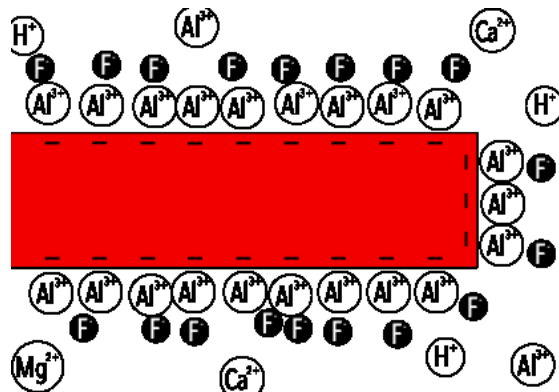


**Figure 2: Ideal configuration of augmented bentonite double ionic layer.  Image credit: Kathleen Glancey, Duquesne University.**

Aluminum-augmented bentonite has been investigated previously in filters for fluoride removal. Creating a point of use filter seems practical for households but they often lack the ability to handle large quantities of water at once. Filters containing aluminum typically need frequent replacements, making them impractical for household use.

The bentonite clay was washed with either NaOH (Fisher Scientific, Fair Lawn, NJ), DI water, or HCl (Fisher Scientific). Approximately 35 ml of the appropriate solution and 5 ml of bentonite were added to 50 ml conical tubes and placed on an orbital shaker for 45 minutes at 250 rotations per minute (rpm). The tubes were then placed in a centrifuge for 10 minutes at 8,000 rpm and then the supernatant was removed, tested, and discarded. The clay was put on an aluminum foil tray and placed in a drying oven at 100° Celsius for over 48 hours. After drying, the clay was crushed with a mortar and pestle and passed through a #100 sieve (nominal opening $1.50 \times 10^{-4}$ m). The same methods were used to wash the clay a second time with either aluminum sulfate, $Al_2(SO_4)_3$, (Alfa Aesar), calcium carbonate, $CaCO_3$, (Fisher Scientific), ferric sulfate, $Fe_2(SO_4)_3$, (Alfa Aesar), or magnesium sulfate, $MgSO_4$, (Fisher Scientific).

Each secondary wash solution was created near saturation. The aluminum wash solution was of a concentration near the saturation of $Al_2(SO_4)_3$, which consisted of 85 g $Al_2(SO_4)_3$ in 100 ml of DI water and prepared at 27° Celsius, room temperature. The calcium carbonate wash was prepared near saturation, with 0.005 g in 100 ml of DI water. The ferric sulfate wash was prepared near saturation, with 26 g in 100 ml of DI

water. The magnesium sulfate wash was prepared near saturation, consisting of 0.01 g

$MgSO_4$ in 50 ml of DI water.

After the DI wash, the dried, crushed bentonite was added to an Al solution of, 170 g

$Al_2(SO_4)_3$ in 180 ml DI water, 24.137 g/l Al. The solution was used to wash 155 g of DI

washed bentonite in two batch reactors. After 45 minutes at 250 rpm on an orbital

shaker, the bentonite was settled by centrifuge at 3,000 rpm for 50 minutes. To

determine the sorption of Al to the surface of bentonite, both the initial solution and post-

treatment supernatants were analyzed for Al concentration with a photometer (YSI 9300,

Yellow Springs, OH, USA), which used an Eriochrome Cyanine R indicator and were

conducted in accordance with the manufacturer instructions. The photometer is

measurement range is 0-0.5 mg/l Al; a limit of detection was not provided. The

aluminum solution was diluted by a factor of $10^{-7}$ in three steps. The supernatant was

diluted by a factor of $10^{-6}$ in two steps.

The equilibrium sorption of aluminum on bentonite was determined by the analysis of

aluminum sorption with time. Three batch reactors (50 ml conical tubes) were set up

with 4.3 g of DI washed bentonite and 20 ml of the aluminum solution. At each timestep,

the bentonite and solution were put on an orbital shaker at 250 rpm for their allotted time,

1, 2, or 3 hours. The tubes were then placed in a centrifuge for 14 minutes at 6,000 rpm.

The supernatant was compared to the control; all measurements were conducted with the

photometer method outlined above.

To confirm the substrate as bentonite and compare with other aluminum augmented materials, the substrate was tested with X-ray powder diffraction (XRPD, PANalytical X'Pert Pro MPD powder X-ray diffractometer). PANalytical X'Pert Pro MPD powder X-ray diffractometer operating in Bragg-Brentano geometry using $CuK_\alpha$ radiation with an average wavelength of 1.5418 Å. The voltage was 45 kV and the current was 40 mA. The structural characteristics of raw bentonite, NaOH, NaOH and $MgSO_4$, NaOH and $Al_2(SO_4)_3$, DI, DI and $MgSO_4$, and DI and $Al_2(SO_4)_3$ washed bentonite was measured. The angle was from 2 degrees to 71 degrees. The step size was 0.0167113 degrees. Each step time was 100.330 with the total time of an hour.

## 2.2 Fluoride Equilibrium and Sorption Testing

To measure the equilibrium sorption of fluoride on bentonite, six batch reactors (50 ml conical tubes) were set up with 0.5 g of NaOH and $Al_2(SO_4)_3$ washed bentonite in 10 mg/l $F^-$ solution of sodium fluoride, NaF (Fisher Scientific), and DI water. The reactors were shaken on an orbital shaker at 100 rpm for an allotted time. The allotted times were 15, 30 minutes, 1, 2, 3, and 8 hours. After shaking, the tubes were put in a centrifuge for 10 minutes at 10,000 rpm. Fluoride measurements were performed according to EPA method 9214. Per the methods, each measurement consisted of the combination of 10 ml of the solution that was added to 10 ml total ionic strength adjuster buffer (TISABII, Orion, Thermo Scientific, Barnstable, MA, USA) and combined for at least one hour. The purpose of TISABII is to stabilize the fluoride solution by maintaining the ionic

strength and keeping the pH constant. Fluoride measurements were completed with an ion selective electrode (ISE, Orion, Thermo Fisher Scientific).

To test fluoride removal of the augmented bentonite, three test waters were used: DI water with NaF, synthetic groundwater with NaF, and spring water with naturally occurring fluoride. Synthetic groundwater was made referencing the EPA's standardization of synthetic groundwater (EPA, 2002). Fluoride sorption was tested with various amounts of augmented bentonite in a fluoride rich solution and were placed in duplicate 50 ml batch reactors. Fluoride solutions were prepared by adding NaF to DI water or synthetic groundwater; the spring water had naturally occurring fluoride. The reactors were placed on an orbital shaker for 3 hours at 100 rpm. The tubes were then put in a centrifuge for 10 minutes at 8,000 rpm. If the coagulant, ferric sulfate, $Fe_2(SO_4)_3$, was added to the tubes before shaking, they were not placed in the centrifuge. Fluoride measurements were made with a fluoride ISE under that same method described in this section.

**2.3 Turbidity**

In order to settle the large bentonite particles, coagulation and flocculation was used. These mainly electrostatic processes are commonly used in water treatment systems. Coagulation is a reaction that makes particles combine together to create a large mass that can then either settle out or be filtered (Ismail et al., 2019). Flocculation is the process of agitation to encourage particles to create a large mass that can be filtered out (Ismail et

al., 2019).  Ferric sulfate is used as a coagulant to adhere the bentonite particles together so that after flocculation they will settle and can be filtered out.

The experiment was conducted in duplicate batch reactors with 0.25 g of raw bentonite and 50 ml DI water.  One set of reactors only had bentonite and another set had bentonite and 0.05 g of ferric sulfate.  The tubes were agitated by hand for 15 minutes and left to settle for various times.  Turbidity was measured with a turbidimeter (2100q IS, Hach Company, Loveland, CO, USA), following the method ISO 7027:1999 (E), at a control, 10, 45 minutes, 1, 1.25, 1.5, 2, and 3 hours.

**2.4 pH Testing**

The World Health Organization (2017) recommends for safe drinking water to set the pH range at 6.5 to 8.5.  The addition of the coagulant and other proposed materials may alter the pH of the drinking water.  The pH of synthetic groundwater and spring water was compared to the control and monitored after the addition of various amendments to determine individual and combined pH changes.  To counteract the pH decrease, sodium oxocalcium hydroxide (soda lime, $CaHNaO_2$, Alfa Aesar) was used to increase the pH (Islam et al., 2011).

The pH was measured using a pH meter (Thermo Scientific) and temperature electrode (Thermo Scientific).  Batch reactors were used to test the pH changes for the multiple amendments (Table 1).  Each test was performed in 100 ml synthetic groundwater and in

duplicate. To obtain the small amount of calcium hypochlorite, $Ca(ClO)_2$, a stock

solution of 0.2 g of $Ca(ClO)_2$ and 50 ml of DI water was created, and 0.1 ml of the stock

was transferred to the required beakers. The pH concentration was measured right after

adding the specified substances, at 60 minutes, and at 180 minutes.

| | Raw Bentonite | $Fe_2(SO_4)_3$ | $Ca(ClO)_2$ |
|---|---|---|---|
| Control | - | - | - |
| Test 1 | 0.5 g | - | - |
| Test 2 | - | 0.1 g | - |
| Test 3 | - | - | 0.0004 g |
| Test 4 | 0.5 g | 0.1 g | - |
| Test 5 | 0.5 g | 0.1 g | 0.0004 g |

**Table 1: Substance measurements for pH tests in 100 ml synthetic groundwater.**

The pH change based on the synthetic groundwater (EPA, 2002) and the amendments

was modeled with Visual MINTEQ version 3 (Gustafsson, 2012). The modeling was

used to estimate the pH of the synthetic groundwater with the proposed POU packet. In

Visual MINTEQ, the ionic strength and pH were set to be calculated from mass and

charge balance. The model inputs were based on the components of synthetic

groundwater and the materials proposed for the POU combination (Table 1), except for

raw bentonite.

**2.5 POU Prototype**

The most widespread health issue in water quality is pathogens. Point-of-use water

treatment has been reported by World Health Organization (2017) as a critical step to

deliver safer water to those without centralized safely managed water.  Previously, a combination of coagulants and disinfectants have been used for a POU treatment by P&G and Islam et al. (2011).  To both reduce pathogens and fluoride in a single POU technology, we combined the augmented bentonite, coagulant, pH adjuster, and a disinfectant into one cohesive packet.  Combining a fluoride removal substance with a disinfectant allows for one multipurpose solution to treat contaminated drinking water.

Fluoride, pH, turbidity, microbial, and aluminum tests were completed during the POU prototype experiments.  Sodium fluoride was combined with water from a nearby river and placed in batch reactors with the proposed mixture.  To measure bacterial disinfection, untreated Monongahela River, Pittsburgh, PA, water was used.  The experiment consisted of six 20-liter buckets filled with river water.  The control was one bucket and contained 8-liters of river water and an added fluoride concentration of 20 mg/l; the Monongahela does not contain naturally high fluoride levels.  Two buckets consisted of 8-liters of river water, a 10 mg/l fluoride concentration, 38.8 g of DI water and aluminum sulfate washed bentonite, 8 g of ferric sulfate, 0.032 g of calcium hypochlorite, and 3.2 g of soda lime.  Two other buckets consisted of 8-liters of river water, a 10 mg/l fluoride concentration, 38.8 g of DI water and aluminum sulfate washed bentonite, and 8 g of ferric sulfate.  Each bucket was stirred for 30 seconds and left to sit for one hour.

To test for fluoride, 10 ml from each bucket was taken and added to 10 ml of TISABII, in 50 ml conical tubes, and let sit for at least an hour, by method 9214.  Fluoride

measurements were taken again at 24 hours and 48 hours later, following the same procedure mentioned above. All measurements were conducted by the method described in Section 2.2

To test for pH, pH and temperature probes were placed directly into each bucket for measurement. All measurements were conducted following the same procedure as mentioned in Section 2.4.

To test the turbidity, samples were taken in sterile bags at the same time and stored in a refrigerator. Before testing the samples, the bags were agitated and brought back to room temperature. The turbidimeter measurements were conducted according to the method ISO 7027:1999.

Bacteria was enumerated by membrane filtration according to EPA method 10029 (EPA, 1999). Per the methods, the sample was filtered through a sterile funnel and a sterile pad and then was transferred to an agar plate for incubation. The method was amended for field testing procedure, including a hot water bath for funnel sterilization and a candle for tweezer sterilization. After each use, the funnels were placed in the hot water bath for at least 15 minutes and the tweezers were sterilized by flame after each use. After testing the samples, they were placed in a 35°C incubator for 24 hours; the bacterial colonies were then counted.

Aluminum leaching from the augmented bentonite into the treated water is a concern. The World Health Organization (2017) states that the standard of aluminum in drinking water is 0.1-0.2 mg/l. The concentration of 0.1 mg/l of aluminum is achievable for large water treatment facilities, but smaller treatment facilities are required to keep aluminum levels below 0.2 mg/l. The potential for aluminum to remain in the water after treatment is a concern for implementation of this technique. Aluminum concentrations were measured at approximately the same time and had a $10^{-3}$ dilution. The aluminum concentration was also tested 24 hours later, following the same procedure in Section 2.1.

## 3. Results and Discussion

### 3.1 Bentonite Augmentation

From the 24.137 g of Al that was calculated to be in the pre-treatment solution, only 0.774 g Al (standard deviation, $\sigma$ = X, $n$ = 3) remained in the liquid phase, based on the measurement of the supernatant. An average of 97% of the aluminum was sorbed to the bentonite surface; therefore, the raw bentonite washed with DI water allows $Al^{3+}$ to sorb to the surface.

Aluminum equilibrium on bentonite was determined over a three-hour period (Figure 3). The sorbed material ratio, $C^*$, is computed by (1) and (2). The difference in aluminum concentration is small and there is a wide range of measurements by the photometer.

$$m_{i,f} = C_{i,f} \mathbb{V} \tag{1}$$

$$C^* = \frac{(m_i - m_f)}{m_s} \tag{2}$$

where $C_i$ is the initial sorbate concentration in initial solution, $C_f$ is the final sorbate concentration in the post-treatment solution, $\mathbb{V}$ is the volume of the solution, and $m_i$, $m_f$, and $m_s$ are the mass of the sorbate in the initial solution, the mass of the sorbate in the post-treatment supernatant, and the mass of the sorbent, respectively.
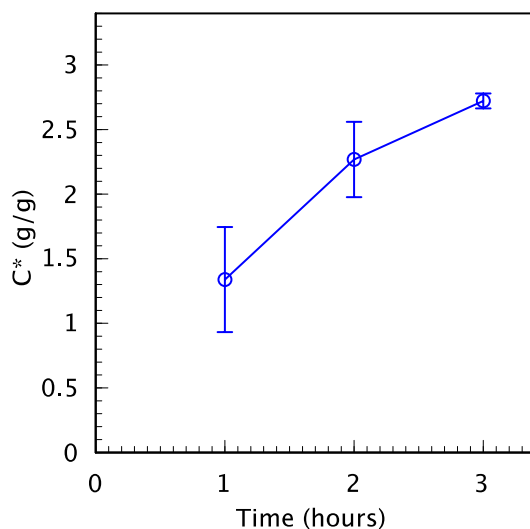
**Figure 3: Aluminum concentrations in the supernatant after various times. Error bars show the standard error.**

The aluminum bentonite sorption system does not appear to reach equilibrium within three hours. To produce the augmented bentonite, a treatment time of one hour was used. Equilibrium of one hour was used as the most feasible method for the experiments.
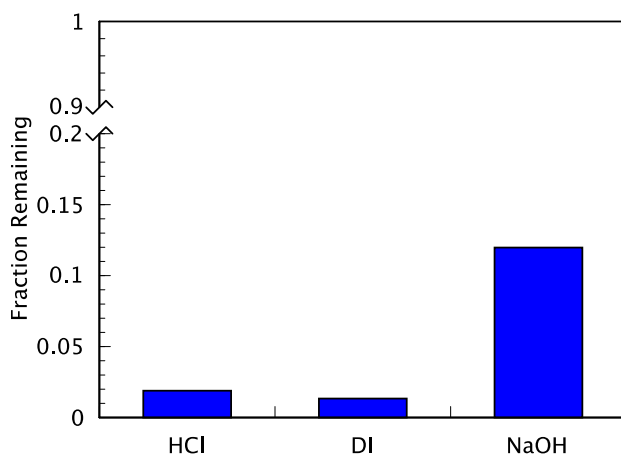


**Figure 4: HCl, DI water, and NaOH fluoride reduction.**

Sodium hydroxide, deionized water, and hydrochloric acid were tested for removal of existing ions on the clay. To test for the best initial wash, they were all washed a second

time with aluminum sulfate. The washed bentonites were tested for their fluoride

sorption ability in a deionized water and NaF solution with consist shaking and centrifuge

times. NaOH and $Al_2(SO_4)_3$ washed bentonite only removed 20% of the fluoride, while

HCl and $Al_2(SO_4)_3$ washed bentonite removed 98.2% and DI water and $Al_2(SO_4)_3$ washed

bentonite removed 98.7% (Figure 4). Hydrochloric acid and DI water were the best

initial washes because they created a clean bentonite surface for aluminum to attach. DI

water and $Al_2(SO_4)_3$ washed bentonite was ultimately chosen as the preferred augmented

substance because DI water is safer to use than hydrochloric acid.

The XRPD results of the raw bentonite, DI washed bentonite, and the DI plus $Al_2(SO_4)_3$

washed bentonite (Figure 5) were consistent with previous studies (Caglar et al., 2009).

The results indicate that the peaks of our raw bentonite follow other previously tested raw

bentonite.
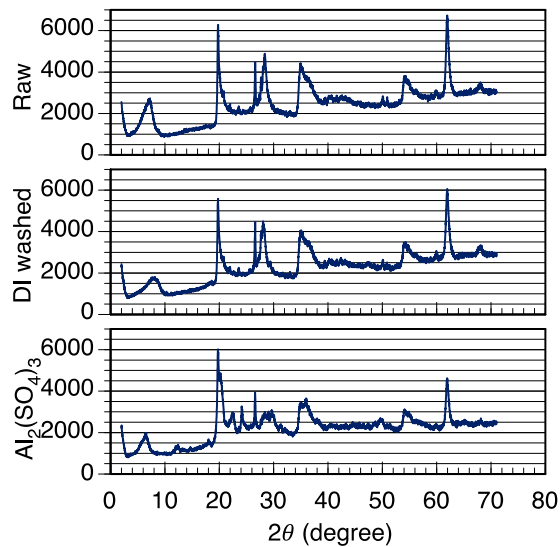


**Figure 5: XRPD of various augmented bentonites. The graph shows intensity over angle. The peaks were analyzed with previous studies to determine if the appropriate match. The XRPD experiment was done with raw bentonite, DI washed bentonite, and NaOH and $Al_2(SO_4)_3$ washed bentonite.**

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## 3.2 Fluoride Equilibrium and Sorption Testing

Sorption of fluoride to augmented bentonite was measured over an eight-hour period. Equilibrium was determined based on relative change in residual fluoride concentration of less than 2%. There was an apparent maximum sorption of fluoride to the augmented bentonite at 10 minutes. The sorbed fluoride concentration plateaued at hour three (Figure 6) through eight. These results indicate that at three hours the augmented bentonite has reached fluoride sorption equilibrium.
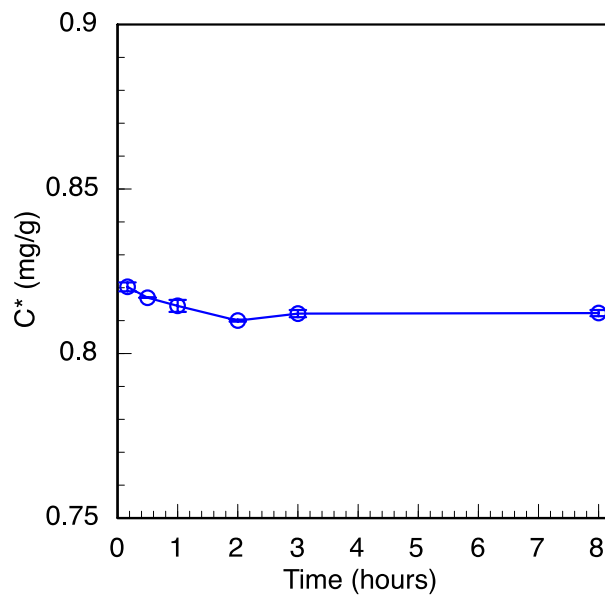


**Figure 6: Equilibrium of the sorbed amount of fluoride at various times with aluminum augmented bentonite. Equilibrium was determined to be at hour 3. Experiments were conducted at 3 hours on the orbital shaker. Error bars show standard error.**

The fluoride sorption to bentonite data were plotted on logarithmic axes of sorbed material ratio (2) to equilibrium concentration, $C$ (Figure 7). The Freundlich sorption isotherm (Fetter, 1999) is the relationship between the ratio of the sorbed mass to the sorbate mass (2) and the concentration (3) with linearization (4). These data support the

initial results that the DI washed, aluminum-augmented bentonite (hereafter, each will be

referred to by the wash) exhibits the strongest sorption as the $C*$ for the washed bentonite

is consistently greater than the other two washes. There are two outliers noted in Figure

7. The outliers are likely from settling issues in the centrifuge at the time of the

experiment.

$$C^* = k\ C^N \tag{3}$$
$$\log C^* = \log k + N\ \log C \tag{4}$$

where $N$ is the exponent that describes the nonlinearity of the system and $k$ is the rate at

which equilibrium sorption increases with concentration.
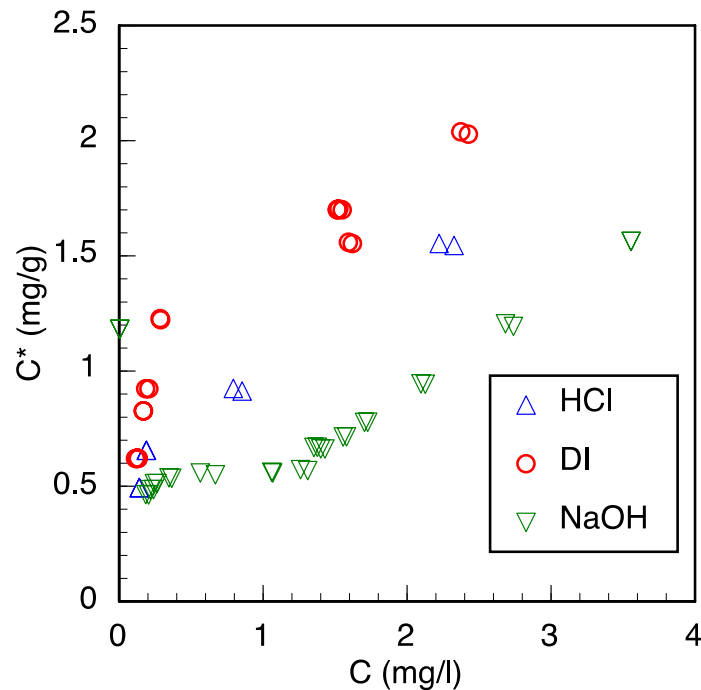


**Figure 7: Sorption isotherm. Shows the bentonite sorption isotherms that were conducted with 0.25-1 g of the listed initial washes and aluminum sulfate secondary wash. The Freundlich isotherm (3) is shown.**

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Langmuir (1916, 1917, 1918) developed a kinetic and equilibrium sorption model initially based on the concept of a single layer (5) with a finite sorption capacity. The sorption data were plotted to compare to the Langmuir model (Figure 8), the washed bentonite data revealed stronger sorption than NaOH and HCl washes; more interestingly, the NaOH washed bentonite exhibited a pattern consistent with a dual-site Langmuir isotherm (Fetter, 2008). The dual-site Langmuir model was extended to consider multiple sorption sites (6) (Swenson & Stadie, 2019). The dual-site Langmuir sorption isotherm was developed in recognition of the reaction of a material to the partial pressure of the sorbate (Fetter Jr., 1977); here, we observe that the higher concentration resulted in a stronger sorption, which suggests that there might be a two-surface sorption mechanism.

$$\frac{C}{C^*} = \frac{1}{\alpha\,\beta} + \frac{C}{\beta} \tag{5}$$

$$C^* = \frac{\alpha_1\beta_1 C}{1 + \alpha_1 C} + \frac{\alpha_2\beta_2 C}{1 + \alpha_2 C} \tag{6}$$

where $\alpha_i$ and $\beta_I$ are empirical coefficients related to the sorption affinity and maximum sorption capacity, respectively.
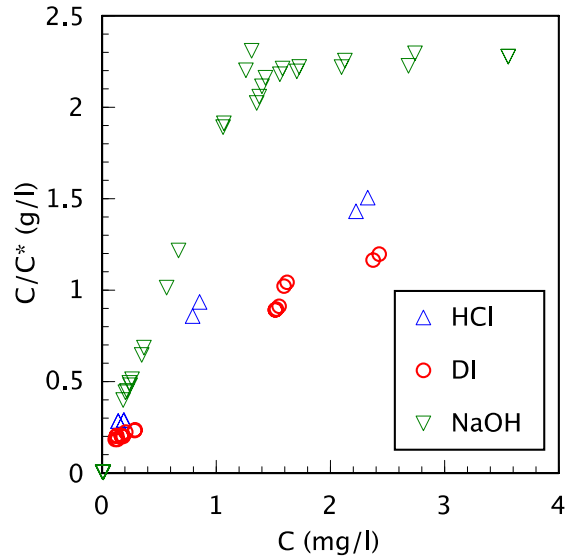
**Figure 8: Langmuir sorption isotherm.  Data in this figure is the same data shown in Figure 7.  Shows the bentonite sorption isotherms that were conducted with 0.25-1 g of the listed initial washes and aluminum sulfate secondary wash.**

Both sorption isotherm models were used to determine the mechanism of the fluoride sorption.  The use of both models indicate that the fluoride sorption data is following the appropriate sorption trend to demonstrate the adsorption technique of cation exchange.

The total fluoride reduction was tested at 0.75 g bentonite in 50 ml of a 10-16.5 mg/l range of fluoridated water (Figure 9). Initially, DI water washed bentonite was tested but removed no fluoride because there were theoretically few sorption sites, that is, positively charged surfaces, available for the fluoride sorption.  Magnesium sulfate proved an insufficient fluoride removal substance because magnesium has only a 2+ charge and did not sufficiently attract fluoride.  Bentonite washed with DI water and calcium carbonate showed to have a reasonable amount of fluoride removal, 30%.  The most notable fluoride reduction, 98%, was bentonite washed with DI water and aluminum sulfate with

ferric sulfate added before shaking.  Aluminum has a 3+ charge which will attract the most fluoride out of the other tested solutions.
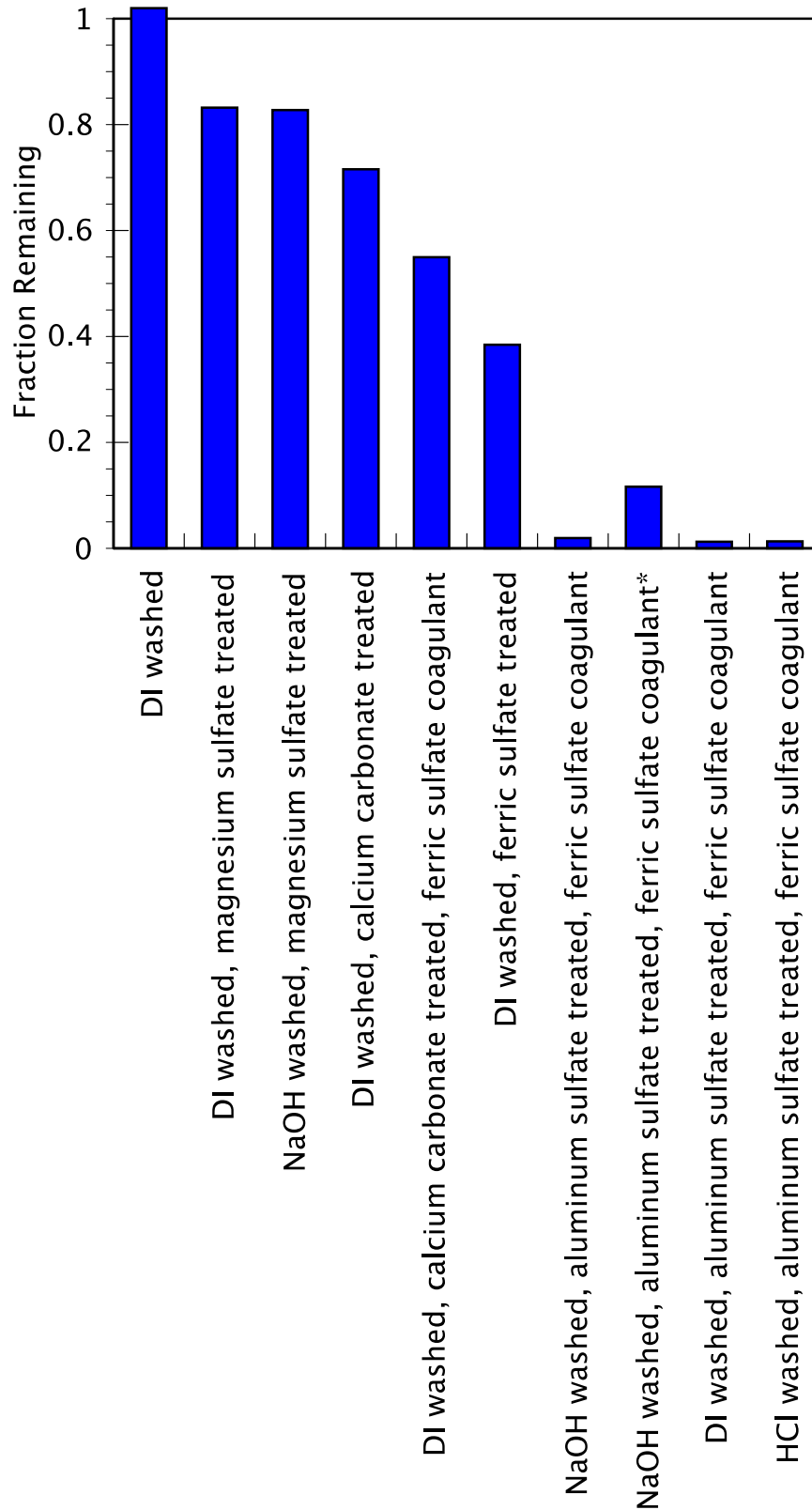
**Figure 9: Fluoride reduction for 0.75 g of the appropriate bentonite. Shows how much fluoride the augmented bentonite is removing compared to the starting fluoride concentration. *Ferric sulfate added after shaking.**

## 3.3 Turbidity

Traditionally, turbidity is an operational standard, as low turbidity demonstrates proper treatment operations. It is also an acceptability standard as consumers tend to avoid cloudy water. The addition of the bentonite causes an unacceptable amount of suspended particles; furthermore, these particles have fluoride sorbed to them. Settling the bentonite particles will also prevent ingestion of the fluoride saturated clay. The use of ferric sulfate as a coagulant removed the bentonite through flocculation. The use of the coagulant allowed the suspended bentonite particles to settle out of the batch reactors while the control, reactors with only bentonite, never reached the measurable turbidity limit of the instrument (Figure 10).
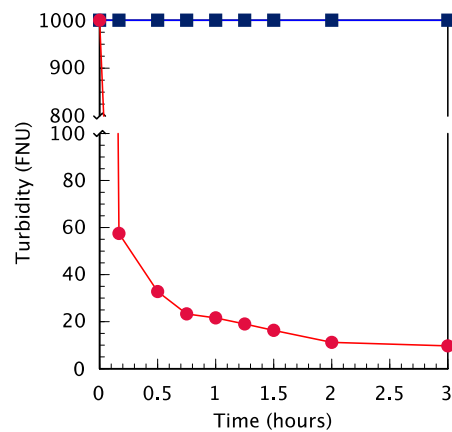


**Figure 10: Turbidity versus time for synthetic samples of bentonite with (red) and without (blue) the coagulant, ferric sulfate in DI water.**

**3.4 pH Testing**

Maintaining the appropriate pH level in drinking water is critical. The pH testing results for the control, synthetic groundwater, was pH = 8.065 initially and 7.85 after 180 minutes. These results are in line with the drinking water standards for pH of 6.5 to 8.5 (World Health Organization, 2017). The second set of beakers, 0.5 g bentonite and synthetic groundwater, initially measured a 9.33 pH and the final pH was 9.3. The set of beakers with 0.1 g ferric sulfate and synthetic groundwater, measured an initial pH of 6.45 and a final measurement of 4.85. The $Fe^{3+}$ in Ferric sulfate attracted suspended solids, which are usually negatively charged (Masters & Ela, 2008). This left the sulfate, which may have formed sulfuric acid, which may have lowered the pH and makes the water unfit for drinking. Ferric sulfate cannot be used in drinking water without a pH buffer to make the pH slightly more basic. The set of beakers with 0.004 g $Ca(ClO)_2$ and synthetic groundwater an initial pH of 8.24 and had a final measurement of 8.33. The calcium hypochlorite is basic and in such a small amount made the solution slightly more basic. The set of beakers with 0.5 g bentonite, 0.1 g ferric sulfate, and synthetic groundwater, initially measured a pH of 6.2 and had a final pH measurement of 4.65. This measurement is too acidic for safe drinking water. There must be an addition of a pH buffer to make it more basic to create a final neutral drinking water pH. The final set of beakers with 0.5 g bentonite, 0.1 g ferric sulfate, 0.004 g $Ca(ClO)_2$, and synthetic groundwater, measured an initial pH of 6.39 and a final pH of 5.55. The combination of all components, with a relatively large amount of acidic ferric sulfate, cause the solution to be more acidic.

The addition of a buffer or basic material is necessary when treating drinking water with augmented bentonite, ferric sulfate, and calcium hypochlorite. The pH after 3 hours is 5.55, which is below the acceptable pH level for drinking water. Soda lime, a base, is necessary to increase the solutions pH for it to be acceptable for drinking water standards.

The synthetic groundwater calculations from Visual MINTEQ computed the pH as 8.119 which is comparable to the expected pH listed in EPA (2002). Once the coagulant, ferric sulfate, was added, the pH dropped to 3.202. Ferric sulfate is an acidic compound that decreases the pH of the water. The disinfectant, calcium hypochlorite, was added and the pH slightly increased to 3.211. Calcium hypochlorite has a pH of 10.8. The final addition of the pH buffer, soda lime, the pH increased to 7.901, which is an acceptable level for drinking water. The addition of a pH buffer is necessary for the treated water to be safe for consumption.

**3.5 POU Prototype**

The average fluoride concentration reduction measurements for the prototype tests are listed in Table 2. On average, the buckets reduced the fluoride concentration by 86% compared to the stock. In this larger setting, the results show that fluoride sorption is comparable to our other data from smaller tests. The fluoride was tested after 24 and 48 hours had passed to determine if there was any fluoride reentering the water (Table 2). The measurements increased slightly after a day of sitting but this could be from a lack of

precise initial measurements.  However, the 24 and 48-hour measurements show that

fluoride is not steadily reentering the water after adsorption to the bentonite clay.

| | Initial (mg/l) | 24 hours (mg/l) | 48 hours (mg/l) |
|---|---|---|---|
| **Control** | 9.46 | 9.77 | 10.6 |
| **Bucket 1** | 1.63 | 1.73 | 1.02 |
| **Bucket 2** | 1.31 | 1.45 | 0.508 |
| **Bucket 3** | 1.66 | 1.80 | 1.77 |
| **Bucket 4** | 0.87 | 1.33 | 1.48 |

**Table 2: The initial, 24 hours, and 48 hours fluoride measurements of each bucket.  All measurements are around the 1.5 mg/L World Health Organization (2017) standards.**

For the prototype tests the pH of the control was 7.804.  The measured river water pH is

typical for freshwater rivers.  All of the bucket's pH values measured in a range from

4.164 - 4.212.  The buckets had similar pH values, which are too acidic for drinking

water.  The likely reason from the low pH is that not enough soda lime was added to the

mixture.  If there was more pH buffer or less ferric sulfate, the pH will be able to measure

within the acceptable drinking water level.

Turbidity of the samples was taken to ensure that the coagulant settled the bentonite

particles.  The control reading was at 2.13 FNU, which is acceptable for drinking water.

All the buckets measured within a range of 38 to 58 FNU.  The potential reason behind

slightly higher turbidity readings is that the measurement was taken right after agitation.

Flocculate might have been stirred up after agitation and be present in the samples,

creating higher turbidity.  Letting the water sit and allowing all particles to settle can

ensure that the final drinking water will be clear.  A way to combat the higher turbidity is

to use a fabric filter to remove the remaining particles after letting it sit for the allotted time.  This would ensure low turbidity when the household went to use the water hours after it settled.

The samples were observed for the number of bacterial colonies that grew after 24 hours. The control was the only sample that contained 18 *Escherichia coli* colonies and no other bacterial colonies.  The remaining buckets measurements are as follows, bucket 1 consisted of 19 bacterial colonies, bucket 2 had 16 bacterial colonies, bucket 3 had 37 bacterial colonies, and bucket 4 consisted of 45 bacterial colonies.  Buckets 1 and 2 had less colonies present on the agar plate which is feasible because they contained calcium hypochlorite.  An explanation for the lack of E.coli in bucket 3 and 4 could be from faulty microbiological testing reagents.  However, the results indicate that calcium hypochlorite is sufficient in the removal of bacteria and is acceptable to use in POU water treatment.

The aluminum in the control bucket of Monongahela River water measured 0.02 mg/l Al. Bucket 1 was tested before dilution and thus tested above the measurable limit of the photometer.  The remaining three buckets followed the appropriate dilution.  Bucket 2 measured 2.9 mg/l Al, bucket 3 measured 2.5 mg/l Al, and bucket 4 measured 3.1 mg/l Al, after the appropriate calculations to correct for dilution.  After letting the buckets sit for 24 hours the aluminum was tested again at the same dilution standard.  Control measured 0.00 mg/l Al, bucket 1 measured 2.3 mg/l Al, bucket 2 measured 3.9 mg/l Al, bucket 3 measured 2.6 mg/l Al, and bucket 4 measured 2.5 mg/l Al, after the appropriate

calculations to correct for dilution. The Al concentration only slightly changed

overnight, which could be from small procedural differences in the photometer.

The high aluminum concentrations in the buckets is expected to be from poorly attached

aluminum on the bentonite clay. The acceptable aluminum limits in drinking water is 0.1

to 0.2 mg/L (World Health Organization, 2017). The excess aluminum on the bentonite

could be from the low centrifuge speed of 3,000 rpm. The low centrifuge speed could

cause the extra aluminum to not separate from the clay, but instead stay weakly attached.

If there is aluminum weakly attached to the bentonite, putting it in water and agitating it

could cause it to detach and linger in the water. Centrifuging the clay, during the second

wash, at a higher speed could potentially remove the excess aluminum on the bentonite

clay.

## 4. Olkokola

The many rural communities North of Arusha have unreliable drinking water and lack a centralized water treatment facility. Preliminary testing results indicate high levels, 4-12 mg/L, of fluoride in drinking water. Fluoride is tasteless and colorless in water (Vithanage & Bhattacharya, 2015). Furthermore, residents frequently consume large amounts of tea, which can contain high amounts of fluoride. Tea can absorb fluoride from fluoride-rich soil or water (Das et al., 2017). Unfortunately, preliminary studies also confirmed the prevalence of potentially pathogenic bacteria (Okular et al., 2019).

The community of Olkokola is located in the Arusha region, approximately 20 kilometers north of the city of Arusha, Tanzania. It is a near the East African Rift Valley, which is a continental rift zone that created trenches and volcanos. Olkokola is divided into four subcommunities, Engorika, Olama, Shaudo, and Stesheni. Olkokola spans throughout hills and valleys (Figure 11), west of Mount Meru, which is from where the community currently receives their water. The water is taken from a spring, then piped to the division box (Figure 12 (a),(b)) located in the sub village Engorika, and distributed to two subcommunities, Engorika and Shaudo. The other two areas, Olama and Stesheni, receive their water from a separate division box, although they appear to originate from the same or nearby source. Throughout the water pipe system, there are various branching points for the water to reach the far ends of the community. Unfortunately, there is no known map of the distribution system.

**Figure 11: Overlooking the valley of subcommunity Olama from a road to the division box in Engorika.**



(a)



(b)
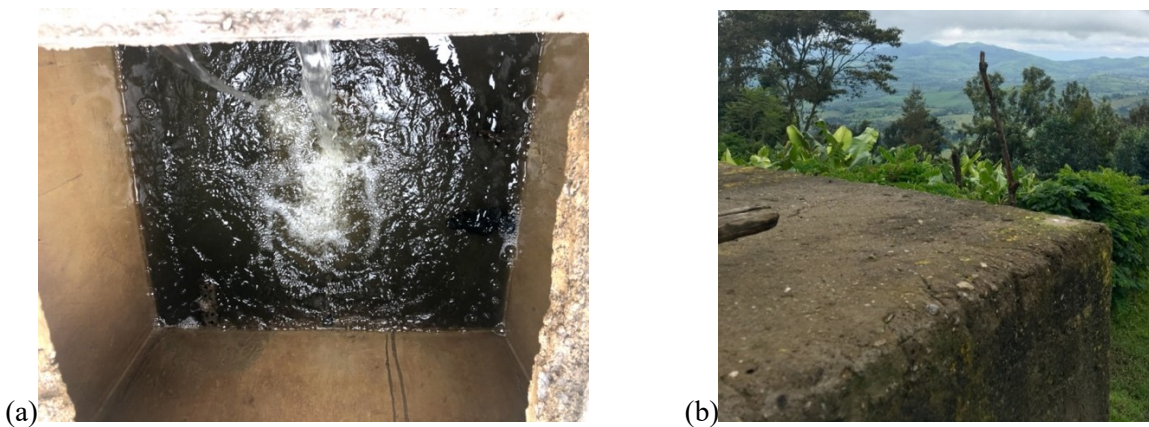
**Figure 12: Division box in Engorika. (a) Inside the division box. (b) Exterior view of the division box.**

The community of Olkokola is connected to Duquesne University through the Spiritan community and a university student organization, Pure Thirst.  Pure Thirst connects Spiritans, students, and community leaders together to discover ways for better water access.  The organization teaches students how to create positive change in the world.

While in the community, Pure Thirst conducted a household survey. Based on the survey data, all households collect water from public taps (Figure 13). Based on our work with the community leadership, who provided the locations of all of the taps, there are no other sources of water available throughout the year. Households collected water in various forms of plastic buckets. A common bucket used was a yellow jerry can (Figure 13). Many of these public taps were observed to be gated (Figure 14), to keep livestock and wildlife from disturbing them. Pipe disturbance is an issue in the community either from exposed pipes or tap damage (Figure 15). If a water tap is broken, community members often do not have the supplies or the means to travel to gather the supplies to fix the problem (Figure 16); therefore, repairs are slow to come.



**Figure 13: Public water tap with jerry cans.**

**Figure 14: Gated public water tap.**


**Figure 15: Exposed and damaged water pipe.**

**Figure 16: Broken public water tap.**

The community has two primary schools adjacent to each other located in the Engorika subcommunity, named Engorika and Olkokola.  Each school goes from nursery to form seven.  At the time of visit (May 2019) Olkokola primary school had 1,219 students and 13 teachers, while Engorika primary school had 923 students and 14 present teachers with an additional teacher away at college.  Parents decide which school their child attends.  There are plans to build a secondary school in between the elementary schools because the closest secondary school is over seven kilometers away and there is no routine public transportation.  Many children do not continue to secondary school because of the distance of the secondary school, financial hardship, or they are needed at the house.  Families are responsible for buying uniforms and other supplies.  Families often do not have a steady income and might not be able to pay for supplies for multiple children.  Many families need children to help out with daily household and agricultural tasks which could prevent them from attending school.  Based on discussions with the

school principals, male students leave school earlier at a higher rate to perform manual labor.

Olkokola has one health clinic in Stesheni.  It lacks internal plumbing for water and at the time of visit had one working tap.  The clinic collects rainwater in addition to the community piped spring water.  Also, at the time of visit, the latrine system had failed because of erosion and soil compaction.  The health clinic conducts baby checkups once a week on a set day.  During these visits, the babies are weighed and given any needed vaccines.  It is the practice for the family to keep the child's booklet (Figure 17 (a),(b)) of health information and bring it to each visit.  Inside the booklet is a graph to track the child's weight at each visit to indicate the child's growth in reference to the WHO growth standards (Figure 17 (b)).  It also shows basic information like name, birthdate, gender, next visit date, etc.  Children under the age of five are meant to consistently visit the clinic for health checks.  Children often do not visit the clinic after the age of two, usually from the mother getting pregnant again and not being able to take the other children.  Expectant mothers also visit the clinic routinely for prenatal checkups on a separate day.
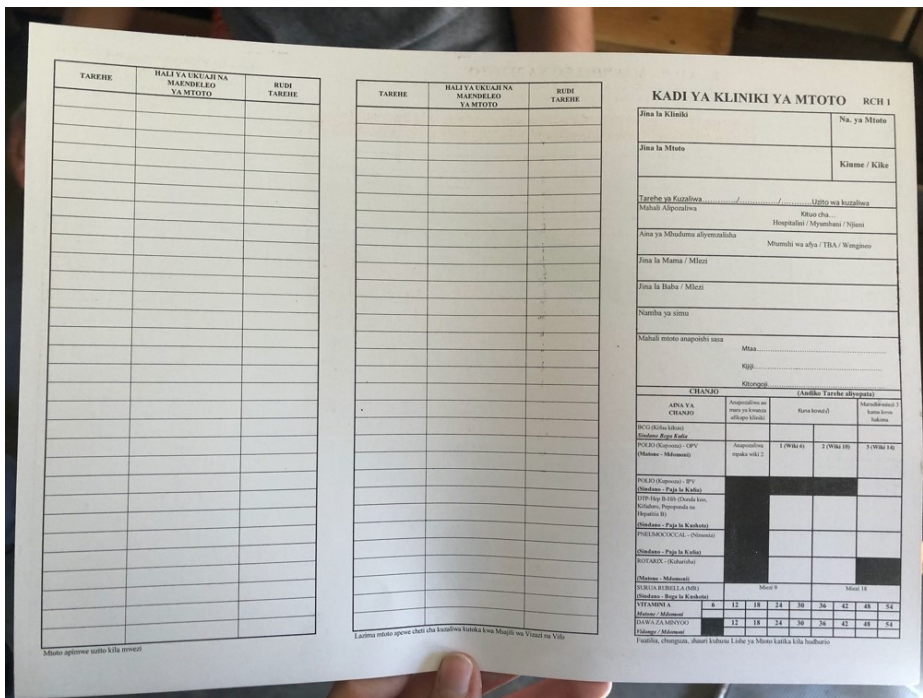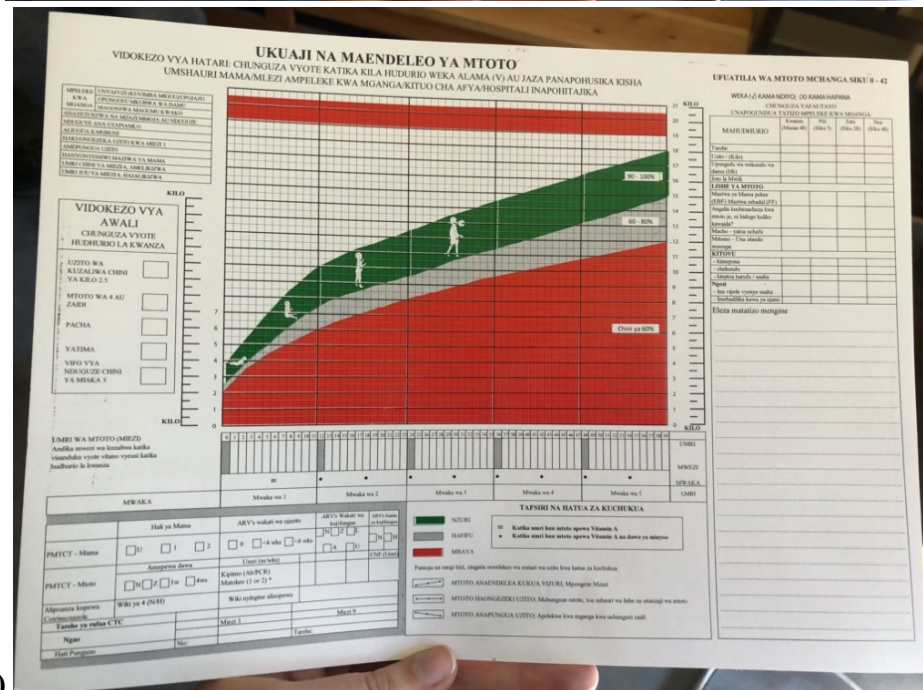
**Figure 17: Children's health booklet. (a) Front of booklet. (b) Back of booklet with growth chart.**

Olkokola experiences large amounts of erosion. Roadways are continuously in danger of being covered by hillside landsides. While visiting the community, we observed people

removing loose dirt from the hillside to put into bags and place them back on the hillside.

These sandbags were used to prevent hillside erosion onto the roadways. Olkokola's

volcanic soil is nutrient rich and ideal for farming. Erosion is also destroying agricultural

land and washing away nutrient rich soil. Many families rely on farming and livestock

for food and income. Another roadway issue is erosion along the treads of the road

(Figure 18). The ditches are created during the wet season when rain is rushing through

the ditches at high speeds. A concern of community members is a roadway that is

experiencing massive erosion that connects Engorika and Shaudo to the other

subcommunities and Arusha. Community leaders described wanting a bridge to replace

the road. The concern of the road is that during the wet season it is frequently flooded,

and people do not cross it, preventing them from going to school or the health clinic.



**Figure 18: Road erosion in Olkokola. Road to the division box in Engorika.**

Olkokola lacks stable and reliable precipitation data that is necessary for the overall water

resources information for the benefit of the community. Precipitation information for the

region can be helpful for farmers to know when to plant crops, it also can help with erosion control. The community also can benefit by understanding how much water is potentially infiltrating their drinking water supply. Understanding the relationship between rainfall and fluoride is an ongoing task and a weather station will allow researchers to explore potential correlations between excess rainfall and decreased/increased fluoride concentrations.

To monitor precipitation, a weather station (Figure 19) was built at a resident's house that will ensure accurate data and protection to the device. Weather station (Meter Group, Pullman, WA, USA), data are collected every fifteen minutes for solar radiation ($W/m^2$), air temperature (°C), wind and gusting wind speed (m/s), wind direction (degrees), vapor pressure (kPa), precipitation (mm), lightning activity (counts), atmospheric pressure (kPa), and vapor pressure deficit (kPa).



**Figure** 19**: Weather station, featuring Abbey Whitewood.**

## 4.1 Methods

While visiting Olkokola with Pure Thirst, an organization from Duquesne University, we conducted a household survey, approved by Duquesne Institutional Review Board (IRB), Protocol 2018/12/2. The survey's function was to collect information about family hygiene, sanitation, and drinking water habits. The household survey was conducted throughout each Olkokola subcommunity. There were 48 surveys completed by two teams consisting of two researchers and a translator. The researchers chose the households to ensure a random selection. The survey questions were asked to the main caretaker. The caretaker was read and given a consent form in their given language to establish the nature of the questions. The caretaker was also informed that they could withdraw at any point during the survey.

The survey started with asking the age, sex, and highest level of schooling for each household member. It went on to ask various questions surrounding income, hygiene, electricity, cooking facilities, water collection and treatment, and tea consumption. If there was a child under the age of five, the height/length and weight were recorded, and if the child experienced diarrhea in the past two weeks.

## 4.2 Results and Discussion

Out of the 48 surveys started, two were withdrawn upon the request of the caretaker, creating a total of 46 surveys completed. Out of the 45 annual income responses, 64% said that they had an annual income between 1-1,700,000 TZS. There were no responses for greater than 6,000,000 TZS annually, which is approximately 2,580 USD. Some households gave an explanation of income by saying they rely on livestock or that there was a family member working in Nairobi, Kenya.

When caretakers were asked about the household electricity, 76% said they have solar power with a battery and 22% said they have no electricity. A frequent comment was that if there was no sun then the power would go out because there is no backup power storage or source. Interruptions lasted over 24 hours for 28% of respondents. Cooking facilities for all surveys were open fire with only two households have one chimney. The majority of households cooked under a roof, typically separate from the main house.

We asked caretakers what type of toilet household members use and how many households share that toilet. Most responses, 76%, said they did not share their toilet with any other households, 6% said they share with one other household, 9% said they share with two, and 9% said they share with three. Sharing a toilet with other households increases the usage which could shorten the life of the latrine. Using an unimproved pit latrine was the most common response at 93% of the 46 responses. An unimproved pit latrine means that there is either nothing or a piece of wood covering the pit and no

ventilation.  Concerns about unimproved pit latrines are that there is no mechanism for erosion control, meaning that the ground could soften and the pit caves in on itself.  If there is not a sturdy structure to stand on over the pit, small children are at risk of falling into the pit.

Researchers also asked questions relating to hand washing hygiene.  When asked if there was a place to wash your hands near the toilet, 63% said yes and 37% said no.  Another question regarding if they had soap available and how often, 63% said they always have soap available, 28% said they sometimes do, and 9% said they never have soap available.  The majority of responses, 89%, said they use commercial soap.  It is important that proper hand washing techniques are utilized in the household to decrease the spread of pathogens.  Around 96% of caretakers said that they wash their hands before preparing food.  Preparing food with clean hands is extremely important for reducing the spread of bacteria.  Limiting the amount of bacterial exposure can help to reduce the amount of illness, especially in young children.  Children under the age of five are susceptible to bacterial diseases because they have a tendency to not have proper hygiene practices.  Having a reliable market for hygiene products would enable caretakers to have access to soap and therefor allow them to ensure that proper hygienic practices can always be followed in the household.

The majority, 96%, of households have an adult woman (over the age of 15) gather the water.  There was one household that said a female under the age of 15 gathered the water.  Gathering the water from a primary source took anywhere from one minute to 60

minutes (Figure 20).  Collecting water from a secondary source took anywhere from six

minutes to 120 minutes (Figure 21).  There were three responses that said they never have

to go to a secondary source for water.  Uninterrupted water service would shorten the trip

for the water collector.
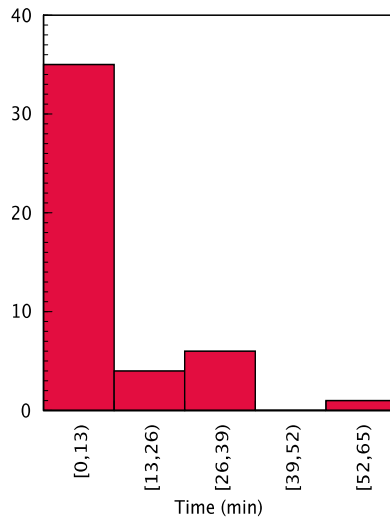
**Figure 20: The amount of time to collect water from the primary source as the travel time to and from the tap.**
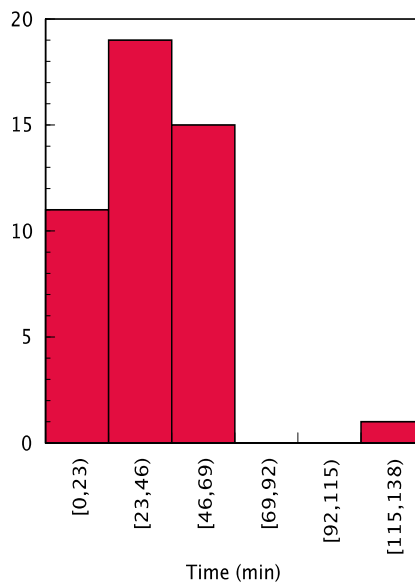
**Figure 21: Time to fetch water from secondary source as the travel time to and from the tap.**

Since Olkokola does not have a water treatment facility, households are responsible for treating their own water. When asked what type of water treatment they used 26% said they boil the water while 74% said they do not treat the water. Drinking untreated water exposes the drinker to a variety of coliforms that have the potential to be pathogenic. Another way to potentially contaminate drinking water is by how one retrieves the water from the container. Responses about how they retrieved the water were that 60% said they pour directly from the container, 38% said the use a cup that has a handle, and 2% said they use a cup with no handle. This question was out of 45 responses, one household declined to answer. Reaching into the container to collect water can transfer bacteria from the hand into the water.

Olkokola residents tend to drink more tea than water each day. Out of the 46 survey responses, many households said they did not drink much water every day. Table 3 shows how many liters of water per day and how many households responded. Households drink much more tea than water every day (Table 4).

| Liters of Water per Day | Number of Households |
|---|---|
| 0-1 | 18 |
| 2-3 | 17 |
| 4-5 | 5 |
| 6-7 | 1 |
| 8-9 | 0 |
| 10-20 | 4 |

**Table 3: How many liters of water each household drinks in a day.**

| Cups of Tea per Day | Number of Households |
|---|---|
| 1-2 | 2 |
| 3-4 | 9 |
| 5-6 | 9 |
| 7-8 | 11 |
| 9-10 | 6 |
| 11-12 | 5 |
| 13-14 | 1 |
| 15-16 | 1 |
| 20-25 | 1 |

**Table 4: How many cups of tea each household drinks in a day.**

Drinking tea appears beneficial because the water is being boiled for a long time before consumption. Even though drinking tea is favorable, people are not drinking enough water that their bodies need. A health issue that can arise from lack of water is kidney disease. Kidneys need a lot of water to function properly and if someone does not drink enough water for years, kidney damage is possible. Drinking water needs to be emphasized to all ages but especially the young and old who are the most susceptible to organ damage.

**5. Conclusion**

Using augmented bentonite as a fluoride removal method is viable. We found that bentonite clay washed with deionized water and aluminum sulfate uses an adsorption mechanism to remove fluoride from groundwater. The best bentonite wash was deionized water and aluminum sulfate, with the addition of ferric sulfate before shaking. The bentonite wash showed a fluoride reduction of 98%. Emphasizing the double ionic layer of the bentonite clay and the aluminum creates a perfect surface for fluoride sorption. Through the process of cation exchange, the bentonite is able to create a double ionic layer with aluminum ions. The research also shows that the augmented bentonite can be used in conjunction with disinfectants to treat water for consumption.

The components involved in augmenting the bentonite are practical and accessible to more people than other previously mentioned fluoride removal techniques. The research presented is the groundwork for a community scale water treatment plant with fluoride removal. The goal of this research is for the augmented bentonite to be implemented into a community water treatment plant where high levels of fluoride in their drinking water source are natural. For communities experiencing high levels of fluoride, over 10 mg/l, augmented bentonite with aluminum is a solution that can decrease the fluoride to acceptable concentrations.

The limitations of the augmented bentonite are the production costs and the potential aluminum remaining in the treated water. The cost of the packet is 4.50USD. The price

of creating the POU product is too expensive to expect households to purchase multiple packets. Some ways to lower the price are to reuse the aluminum wash solution, since aluminum sulfate is the most expensive material. Another way is to reduce the strength of the aluminum sulfate wash. Reducing the strength will lessen the amount of aluminum needed for the material production. The second limiting factor for the material is the high amounts of leftover aluminum in the water. Ways to curtail this is to spin the centrifuge faster when washing the bentonite. More revolutions per minute could shake any poorly attached aluminum off the clay, therefor preventing aluminum from entering the water.

Future research should focus on adding to the Langmuir two-sorption isotherm. The NaOH and aluminum washed bentonite data points showed a two-surface Langmuir isotherm (Figure 22). The isotherm needs more points to substantiate that the double ionic layer is indeed occurring on DI and aluminum sulfate washed bentonite. Other future research could be to test other coagulants beside ferric sulfate. Newer coagulants are being used in water treatment facilities that might work better with the augmented bentonite. More future work could consist of delving into secondary washes with calcium carbonate instead of aluminum sulfate. Calcium carbonate washed bentonite worked in the same manner as aluminum sulfate except it removed less fluoride. In areas where the natural fluoride is not as high as 10 mg/l, a calcium wash might be sufficient. More research should be done on calcium carbonate washed bentonite to determine if it can be used as a POU water treatment technique.
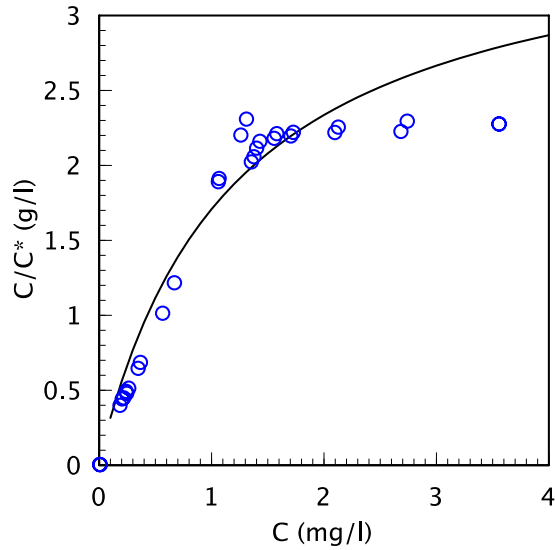
**Figure 22: Langmuir two-surface sorption isotherm with model. The data points fluoride measurements from NaOH and aluminum sulfate washed bentonite. The smooth line is the sorption isotherm model.**

Olkokola in the Arusha region of Tanzania requires a water treatment facility to provide residents with a continuous and clean water supply. Clean drinking water would benefit all consumers, human and animal. Our augmented bentonite has the potential to be implemented into a community wide water treatment plant. Historically, the community does not favor POU methods for water treatment, therefor a treatment facility would be the most beneficial. Having one facility that would reliably disinfect and defluoridate the water would allow the community to have access to safe drinking water.

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