



## Household-level Fluoride reduction from drinking water using crushed fired clay – proof of concept

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### ABSTRACT

Residents of Bunyangabu District in Uganda have reportedly suffered from cases of dental fluorosis due to consumption of water with high levels of Fluoride from Ntabago Stream. A household-level sand filter incorporating crushed fired clay as an adsorbent was designed for a household to reduce the Fluoride concentration in their drinking water from the natural raw water levels (2-3 mgF/L) to permissible, healthy levels (0.5-1.0 mg/L). Pieces of fired clay bricks were crushed, and particles of 150  $\mu$ m to 300  $\mu$ m in size were selectively obtained by sieving. Stream water was filtered through replicate model layered filter columns of the prepared clay, sand (fine sand of 150  $\mu$ m-2 mm; coarse sand of 2-5 mm), and gravel (6-15 mm). It was found out that fired clay layers of 2.5 and 5 cm thick reduce the concentration of Fluoride in water by up to 74%, to less than 1.0 mg/L. This was deduced by evaluating Fluoride concentration in both the clay and water before and after filtration, using the SPADNS Colorimetric method. Use of crushed fired clay for reduction in Fluoride concentration in drinking water is the focus of this paper. Additional work will optimise filter design to improve overall water quality.

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**Keywords:** Defluoridation, filter column, household-level water treatment.

### INTRODUCTION

Fluoride is found in all natural waters at varying concentrations (Djossou et al., 2015; Hisseien et al., 2015). Seawater generally contains about 1.0 mg/L, while rivers and lakes mostly exhibit concentrations of less than 0.5 mg/L. In groundwater however, low and high concentration of Fluorides can occur, depending on the nature of the rocks and the occurrence of Fluoride-bearing rocks (Djossou et al., 2015). Many of the lakes of the African Rift Valley especially

the soda lakes, have extremely high Fluoride concentration.

Geographical areas around the world that have concentrations of Fluoride include Syria, Jordan, Egypt, Libya, Algeria and Morocco. It also includes the rift valley area in Africa from Sudan through Kenya to Tanzania and Malawi. Another geographical area extends from Turkey through Iraq, Iran and Afghanistan to India, northern Thailand and China. The most affected areas by Fluoride include India, China and Rift Valley

countries in Africa (Demelash et al., 2019; Mosonik, 2015).

In Uganda, 6.45% of the water sources exceed the standard value of Fluoride in drinking water. The maximum value of Fluoride in these waters is reported to be 3.31 mg/L and the most affected areas include volcanic areas of Elgon, Mbale, Moroto and the Rift Valley of western Uganda (Malago et al., 2017).

Preliminary tests carried out in October 2018 on water from Ntabago Stream in Western Uganda showed concentrations of Fluoride between 2 mg/L to 3 mg/L, which has led to fluorosis and discolouration of teeth for the residents consuming the water (Figure 1). According to the Ugandan Standard (UNBS, 2008), this range is greater than the permissible levels (0.5 mg/L to 1.0 mg/L for potable treated water, and 1.5 mg/L for potable untreated water). This has made the water unsafe for human consumption due to high concentration of Fluoride in water (Das and Mondal, 2016).

The National Water and Sewerage Corporation (NWSC) has provided piped water, but because of the cost attached, the

residents prefer free water from this stream to piped water, which makes them susceptible to health risks associated with Fluoride contamination (Table 1). 34% of the population in Rwimi Town Council have access to safe and clean water (MWE, 2018). This implies that the bigger percentage (66%) of the population have no access to safe and clean water. The focus of this study is mainly on the potential for defluoridation of water from Ntabago Stream to permissible levels using crushed fired clay as an adsorbent.

Crushed fired clay was chosen as it is readily available to households in Uganda where small scale industries involved in firing of clay bricks for construction, firing of pottery, and charcoal stove linings are common. Other adsorbents have been studied for fluoride reduction including bone char, ceramics, and oxides (Djousse Kanouo et al., 2020; Yadav et al., 2018; Habuda-Stanić et al., 2014; Madhukar et al., 2014; Loganathan et al., 2013), but these may not be available at household level.



**Figure 1** Affected residents of the study area showing teeth discolouration.

**Table 1:** Health impacts of Fluoride in drinking water.

Fluoride (mg/L)	Health Impact
< 0.5	Dental caries
0.5–1.5	Optimum dental health, works against dental caries
1.5–3	Dental fluorosis, blackening and pitting of enamel and teeth from long-term exposure, mottled enamel, Roentgenographic bone changes, polydipsia
3–8	Skeletal fluorosis, damages foetus, increase in F-concentration in milk, infant mortality due to calcification of blood vessels, lack of intelligence quotient in children, osteosclerosis, renal diseases, elevated serum alkaline phosphatase, stiffness of knees and hips, increased bone mineral density, bone and joint pains.
10–100	Gastroenteritis, skin irritation, deformation of bones and other skeletal abnormalities, thyroid changes, growth retardation, kidney damage, crippling fluorosis.

Compiled from Madhukar et al. (2014)

## MATERIALS AND METHODS

### Raw water quality

Grab samples of stream water were collected from three locations along the Ntabago Stream (Figure 2) for raw water Fluoride concentration analysis.

### Defluoridation model filters

Model filters were prepared for defluoridation filter test runs as follows:

Pieces of fired clay bricks were crushed and sieved to obtain the particles that pass through a 300  $\mu\text{m}$  sieve and are retained on a 75  $\mu\text{m}$  sieve. After sieving, the clay was washed to eliminate very fine particles (dust of less than 75  $\mu\text{m}$ ). Clays reportedly have a Fluoride adsorption capacity of 84-95 mg/g; and over 85% adsorption for clays with high Al and Fe content. Heat treatment (firing) increases adsorption capacity (Madhukar et al., 2014).

Sand was washed using clean water until clean water from it was observed. The sand was sun dried for two days, and fine sand and coarse sand portions were obtained after sieving. Coarse sand and fine sand of 2 mm to 5 mm, and 150  $\mu\text{m}$  to 2 mm, respectively, were used.

The gravel to be used was thoroughly washed with clean water to eliminate dust and then sun dried, after which gravel of size 6 mm to 15 mm obtained by sieving.

The model filter layers were arranged with the gravel at the bottom, followed by coarse sand, then fine sand, and crushed fired clay at the top. The gravel and sand layers had a constant thicknesses (5 cm) while the crushed fired clay had varying layer thickness (2.5 cm, 5 cm, 7.5 cm) in each of the three model filters.

A 3 mgF/L solution comprised of distilled water and Sodium Fluoride (NaF) was prepared for the model filter test runs. After stock preparation, the solution was analyzed to confirm the concentration of Fluoride ions using SPADNS Colorimetric Method (APHA/AWWA/WEF, 1999). 750 mL of the 3 mgF/L solution was passed through the model filter in each test run, and the filtrate was analysed for Fluoride concentration.

The SPADNS Colorimetric Method was used to determine the concentration of fluoride in the water before and after filtration. This Method is based on the reaction between fluoride and a zirconium-dye

and relies on the fact that when Fluoride reacts with certain zirconium dyes, a colourless complex anion and a dye are formed. The complex, which is proportional to the fluoride concentration, tends to bleach the dye which therefore becomes progressively lighter as the Fluoride concentration increases. In the case of the Fluoride ion reaction with Zr-SPADNS (sodium 2-(parasulphophenylazo)-1,8-dihydroxy-3,6-naphthalene disulphonate), the resulting coloured complex is measured in a spectrophotometer at 570 nm (APHA/AWWA/WEF, 1999; HP, 2000).

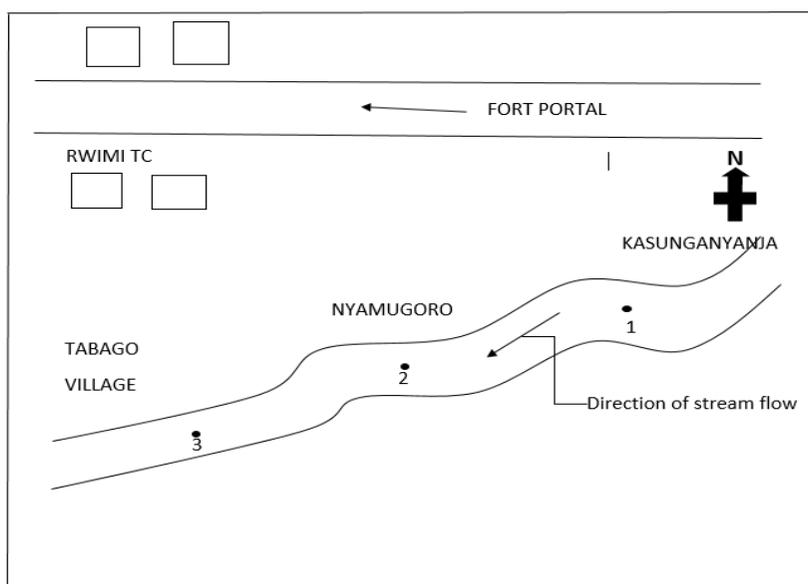
To determine the clay-adsorbed portion of Fluoride, the clay was removed from the filter and a homogenized sample washed using 250 mL of 1 M of hydrochloric acid for 1-hour in order to extract Fluoride ions in a solution. The acid enabled the detaching of the Fluoride ions from clay particles into the washing solution (Moon et al., 2015).

After one hour, the suspension was filtered using a 0.45 µm micropore filter and the filtrate was then analysed for Fluoride concentration according to the SPADNS Colorimetric Method as described above.

### Adsorption capacity and effect of adsorbent layer thickness

Fluoride reduction with tests with model filters were carried to determine layer thickness of the adsorbent and the initial Fluoride ion concentration in the solution was determined at ambient temperature (~25 °C). The Fluoride concentration in the filtrate and fired crushed clay from the filter was determined using SPADNS Colorimetric Method. Three model filter setup experiments were used, and three test runs carried out on each.

The effect of the adsorbent layer was investigated by varying the crushed fired clay layer thicknesses, and spiking the water to be filtered with 3 mgF/L. The fired clay thickness in the filters was varied from between 2 cm to 10 cm (2.5 cm, 5 cm, and 7.5 cm). The concentrations of Fluoride in the filtrate were determined after each test (Motora and Tesema, 2017). The concentration of Fluoride retained in the clay layer was determined in one of the three test runs.



**Figure 2:** A sketch map showing location of sampling points along Ntabago Stream.

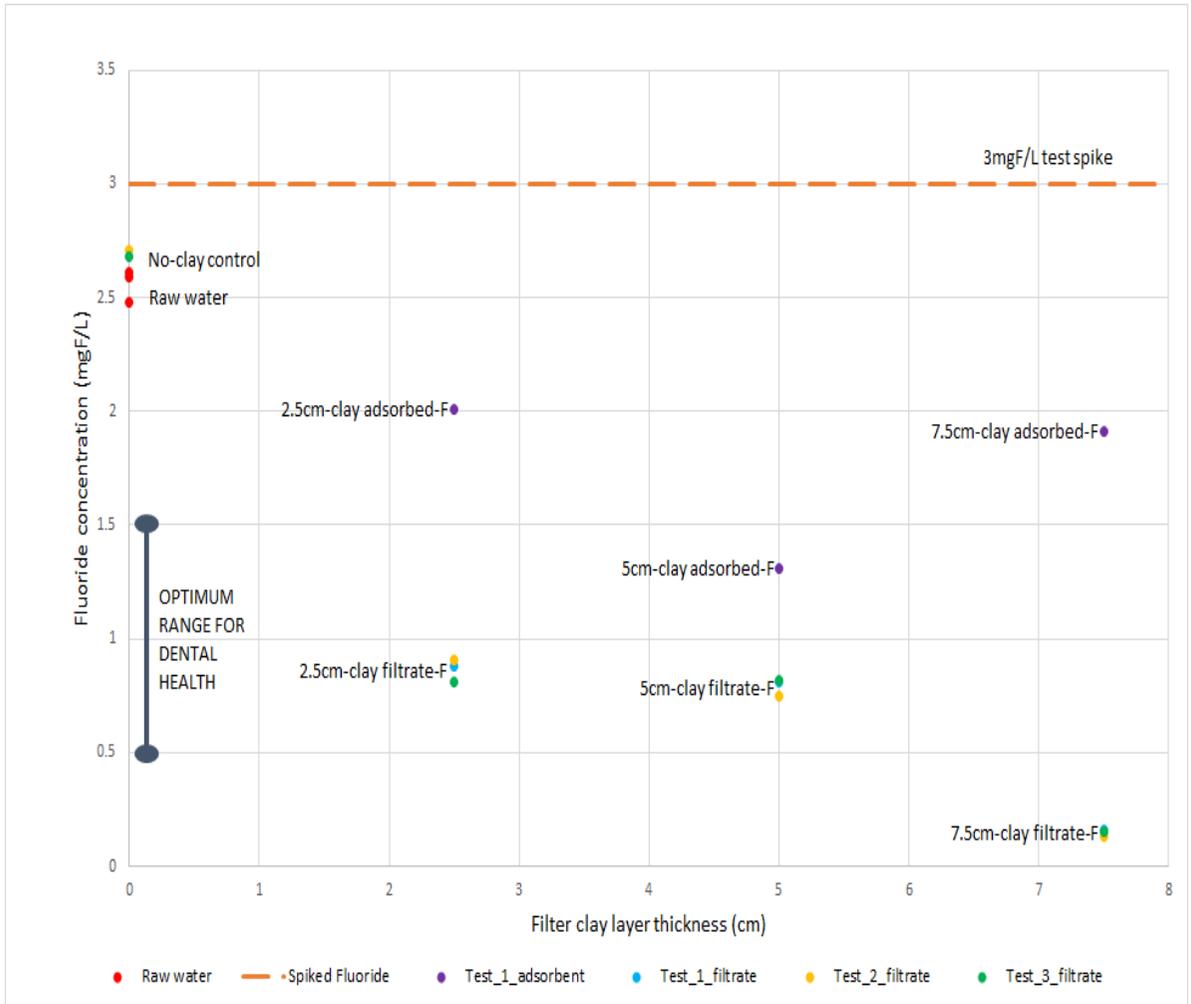
## RESULTS

### Defluoridation test runs

The 2.5 cm and 5 cm crushed fired clay layer filters were able to reduce to the Fluoride concentration in the filtered water by 71% and 74% respectively to within the optimum range for dental health (0.5-1.5 mgF/L), which meets the Uganda National Standard maximum limit of 1.5 mgF/L (Figure 3). Other studies with fired clay show

similar reduction levels (Mottora and Tesema, 2017).

The 7.5 cm clay layer filter reduced the Fluoride levels by 95% to below 0.5 mgF/L (Figure 3), which is a risk factor for dental caries. Reduction in the clay-free control was about 10%. Therefore, the 5 cm clay layer filter was chosen as a conservative option for further study.



**Figure 3:** Defluoridation test runs show reduction of Fluoride to within and below the optimum range for dental health in filtrate from model filters with a crushed fired clay layer receiving water spiked with 3 mgF/L.

## DISCUSSION

With the 7.5 cm clay layer, 95% of the Fluoride could be accounted for in the filtered water and HCl acid-washed clay, which compares favourably with the findings of Moon et al., (2015). However, with the 5 cm and 7.5 cm clay layers only about 70% of the spiked Fluoride was accounted for at the end of the test run. It is therefore important to carry out more tests to account for the remaining 30% in order to understand the defluoridation process better, and to determine how long the clay layer can remain effective before needing to be replaced.

### Filtered water quality

Raw water quality improvement in other parameters of interest was therefore

checked with a 5 cm clay layer test filter (Table 2).

While the 5 cm clay layer model filter reduces Fluoride levels to optimum, and improves other physicochemical parameters (Electrical Conductivity), it produces water which needs still improvement in Apparent colour, Turbidity and Total Suspended Solids levels (Table 2). Further optimization of the sand filter layer thickness is needed for overall physicochemical and biological water quality improvement for household level drinking water treatment. An adaptation of the HACCP (Hazard Analysis Critical Control Point) methodology outlined by Gokpeya et al. (2019) for household level clay water filters shall form the basis for this assessment.

**Table 2:** Raw water quality before and after filtration.

Parameters	Units	Initial quality	Filtrate quality	Uganda National Standards for potable water	Filtered water compliance
pH		7.11	7.32	6.5 – 8.5	OK
Electrical Conductivity	μS/cm	980	348	≤ 1500	OK
Colour (apparent)	PtCo	207	<b>74</b>	≤ 15	<b>High</b>
Turbidity	NTU	39	<b>9</b>	≤ 5.0	<b>High</b>
Total Suspended Solids	mg/L	44	<b>9</b>	0.0	<b>High</b>

## Conclusion

Ntabago Stream has between 2-3 mg F/L, which is above the National Standard and above the optimum range for dental health (0.5-1.5 mgF/L). A crushed fired clay layer of 5 cm thickness in a model filter was adequate for reducing Fluoride by 74% to the permissible levels. The filter system was also effective at reducing water Turbidity, Electrical Conductivity, Apparent colour and Suspended Solids.

Further studies will be done on optimization of the defluoridation model filter and process. This would aid in determining the contact time for optimum Fluoride reduction, the period after which the adsorption sites are fully saturated, potential for adsorbent regeneration, maintenance best practice for the filter system, and the filter layer requirements for achieving overall safe drinking water quality at household level.

### COMPETING INTERESTS

The authors declare that they have no competing interests.

### AUTHORS' CONTRIBUTIONS

All authors contributed to the conception and design, and analysis. EW was the principal investigator. BN and BK contributed to material preparation and data collection. EW led the manuscript writing and made revisions.

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