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## **Highlights**

- Borehole water contained higher fluoride concentration than water from springs
- Some water sources (14% of samples) contained fluoride levels higher than 1.5 mg/L, the WHO guideline value for drinking water
- High fluoride levels correspond to high TDS values at near-neutral pH

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 A modified Galagan Equation was used to calculate the optimum fluoride in water for Sukulu Hills, and was found to be 0.4 mg/L. All water samples contained fluoride>0.4 mg/L

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# **Fluoride Contamination and Its Optimum Upper Limit in Groundwater from Sukulu Hills, Tororo District, Uganda**

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#### **Abstract**

This study was carried out to assess fluoride (F) concentration and to determine its upper permissible limit in groundwater from Sukulu Hills, a phosphate mining area in Tororo District, Uganda, where groundwater is the main source of drinking water. Water samples were collected from boreholes and protected springs within a three-kilometre radius from the foot of the Hills. The physico-chemical parameters and F concentration were analysed using potentiometric methods. The water samples collected had a pH range of 6.0-7.2, electrical conductivity of 148-750  $\mu$ S/cm, and TDS values of 75-378 mg/L; and these parameters were within the WHO normal range for drinking water. F concentration in groundwater from boreholes ranged from 0.4 to 3 mg/L, whereas in springs it was from 0.2-2.4 mg/L. High Flevels corresponded with higher TDS values at near-neutral pH. The WHO and Uganda National Bureau of Standards (UNBS) guideline value of 1.5 mg/L F was exceeded by groundwater from spring S3 and boreholes B3 and B7 (14% of samples) in the study. Given the five-year average weather conditions of Tororo, a modified Galagan equation was applied to calculate the recommended  $F$  of the area, and was found to be 0.4 mg/L. All water sources studied contained average F levels higher than 0.4 mg/L. These findings imply a possible risk to the local population which depends on this water being exposed to dangers of high F intake.

**Key words:** Fluoride, Groundwater, Sukulu Hills, Tororo, Uganda

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## **1 Introduction**

Fluoride ion (F) in low quantities ( $\leq 0.7$  mg/L) is an essential component of normal mineralisation of bones and formation of dental enamel [1, 2]. However, excess  $F$  (> 1.5) mg/L) when ingested directly or indirectly, causes adverse physiological effects, leading to slow, progressive crippling of bones and teeth, a condition known as fluorosis, usually manifested as permanent brown tooth stains [3, 4]. Table 1 shows the different F levels in water with associated effects of consumption of such water.

$F$ Concentration (mg/L)	<b>Health Impact</b>
$0.0 - 0.5$	Limited growth and fertility, dental caries [2]
$0.5 - 1.5$	Promotes dental health, prevents tooth decay [6]
$1.5 - 4.0$	Dental fluorosis (mottling of teeth) [2, 7]
$4.0 - 10$	Dental fluorosis, skeletal fluorosis (pain in back and neck bones) [7]
>10.0	Crippling fluorosis [7]

**Table 1 Fluoride Levels in Drinking Water and Effects on Human Health** [5]

The recognition of the benefits of F in prevention of tooth decay in the early 1900s led many governments to introduce policies that required mandatory addition of artificial F of up to 1 mg/L as sodium fluoride or hexafluorosilicic acid to public water supplies. The World Health Organisation (WHO) adopted water fluoridation as an effective oral health intervention in 1994 [2]. However, given the recent understanding that the role of  $F$  in caries prevention is topical rather than systemic [8], public water fluoridation regulations have since been revised or reversed in most western countries. Fluorosis is endemic in over 25 countries around the world, and is most prevalent in India, China, and parts of Africa in the Rift Valley areas of Ethiopia, Kenya, Tanzania, Western Uganda [9]. Fluorosis has been associated with consumption of groundwater with excess  $F$  [4].

Other health problems associated with excessive intake of F have also been reported worldwide, including fibre (tendons and ligaments) degeneration, low haemoglobin levels, deformities in red blood cells, skin rashes, depression, gastro intestinal problems, urinary tract malfunctioning and bone resorption [10]. F also reduces the effectiveness of some enzymes [11]. As a result, the WHO has set a guideline value of  $F$  in drinking water of 1.5

mg/L [12]. The U.S EPA recommends a  $F$  concentration between 0.6 and 0.9 mg/L in drinking water, but has set the enforceable limit at 4 mg/L, while the Uganda National Bureau of Standards (UNBS) specifies that the maximum desirable limit of F in drinking water is 1-1.5 mg/L [13].

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However, dental fluorosis has been reported in areas with water F levels lower than 1 mg/L. Mφller et al [14] examined the prevalence of dental fluorosis in low F areas in Uganda and reported a prevalence of dental fluorosis between 3.4-42.5% in villages consuming water with F levels between 0.17-0.30 mg/L. Another study by Rwenyonyi et al [15] revealed up to 45% prevalence of fluorosis in a Ugandan district with average of  $0.5 \text{ mg/L F}$  in water. Manji, et al. [16] reported dental fluorosis in 78% of the children from a Kenyan village with a F level between 0.1-0.46 mg/L in potable water. Similar findings have also been reported from Tanzania [17]. Smith and associates reported a case where 61% of the boys living in a Sudanese village with only 0.65 mg/L in drinking water developed dental fluorosis [18].

The not-so-direct relationship between dental fluorosis prevalence and F content of drinking water as highlighted in the aforementioned studies may be explained by: (1) other sources of F intake than drinking water might have contributed to the F total daily intake; (2) other factors such as age, quality of nutrition, altitude and daily temperature influence the prevalence of fluorosis at any given level of F intake. The role of these factors in fluorosis prevalence has been investigated and proved relevant [19, 20]. Fluorosis is more prevalent in hotter regions, even when F in drinking water is within the WHO acceptable levels. This is because hot conditions lead to higher consumption of water, hence higher F intake [20]. The effect of altitude on the prevalence of fluorosis was found to be significant, with high fluorosis prevalence at higher altitude [15]. The reason for this observation is still unclear, but there appears to be high  $F$  retention in body tissues at high altitude. On the basis of these findings, the WHO generally recommends that when setting the standards for a country or region, factors such as climatic conditions, volumes of water intake, and intake of F from other sources be considered.

Galagan and Vermillion [21] presented an equation to determine the optimum drinking water F level, taking into account the average daily temperature and daily water intake.

Parts per million of 
$$
F = 0.34/E
$$
 (1)

The figure 0.34 (optimum water consumption) was calculated from data for an area where the optimum F concentration is known. E is the estimated average daily water intake for children through 10 years of age in ounces per pound of body weight. The value of **E** may be calculated from the estimation equation;

$$
E = -0.038 + 0.0062 \times temperature \tag{2}
$$

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When applying this equation, the temperature value should be at least a 5-year average for the figure to be truly representative, and is taken as mean maximum temperature in degrees Fahrenheit. If the original metric units used by Galagan and Vermillion are converted to SI units, the equation to calculate the optimal upper level of F- in potable water (in mg/L) becomes:

Optimal upper level of F concentration (in mg/L) = 
$$
\frac{0.022}{0.0104 + 0.000724 \times AMMT \text{ °C}}
$$
(3)

AMMT is the Annual Mean Maximum Temperature over a 5-year period.

Equation (3) has been used to calculate optimum water F levels in different countries [21-23] and for Uganda, the value was found to be about 0.67 mg/L [24]. The equation estimated the daily water intake under different temperature conditions in United States during the late 1950s, where children were presumed to take 44% of their total fluid intake as milk with negligible F levels.

In Tororo, milk is expensive and not readily available, and even when children are given milk, it is diluted with water. The true water intake by children in Tororo may be similar to that in Sudan due to relative similarity in weather conditions and the standards of living [23]. Therefore, a factor of 0.56 was applied to Equation (3), which allows for the fact that under current conditions in Tororo where high malnutrition rates have been reported [25], and nearly all the children's fluid intake is water. The modified equation used to calculate the optimal upper level of  $F$  in potable water in Chile and Sudan was:

Optimal upper level of F concentration = 
$$
\frac{0.022 \times 0.56}{(0.0104 + 0.000724 \times AMMT \text{ °C})}
$$
(4)

Equation  $(4)$  was used in this study to calculate the optimum  $F$  in drinking water in Tororo.

F in groundwater comes mainly from leaching of fluoride-containing rocks. Phosphate minerals containing F, such as fluorapatite  $(Ca_{10}(PO_4)_6F_2)$  and francolite  $(Ca_{10-x}$  $yNa_xMg_y(PO_4)_{6-}z(CO_3)_{7}F_{0.47}F_2$  have been reported in Sukulu Hills in Tororo [26] and this

$$
(2)
$$

coincides with several anecdotal reports of the high prevalence of fluorosis in the area. It has also been shown that F accumulates in certain food crops grown in fluoride-rich soils [27], another route to the food chain. This route further increases the total daily intake of F beyond recommended 0.7 mg per day from all sources for children of one to four years.

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The characteristics of groundwater are influenced by geochemical processes such as weathering of minerals, precipitation, dissolution, ion exchange, oxidation, reduction, and residence time (and flow rate) of groundwater [28]. F levels in groundwater has been shown to be greater in dry seasons than wet seasons as a result of evaporative concentration of water [29]. Factors that influence the solubility of fluoride-containing minerals, including pH, calcium concentration, organic matter content among others. Chaïrat, et al. [30] demonstrated that fluorapatite dissolution is pH dependent and the mechanism of this dissolution was shown to be initiated by the relatively rapid removal of F then  $Ca^{2+}$  at the surface, via protons for  $Ca^{2+}$  exchange reactions. Dissolution rates are controlled by the destruction of this F and  $Ca^{2+}$  depleted surface layer by breaking of Ca-O bonds, facilitated by two distinct parallel reactions: (a) the adsorption/penetration of protons, which dominates at acid conditions and (b) the hydration of the  $\sim$ CaOH<sub>2</sub><sup>+</sup> complex (at the surface) in neutral and basic conditions.

In dissolved form, F concentrations in natural waters have been found to depend on a number of factors including temperature, pH, presence or absence of complexing and precipitating ions, solubility of fluoride-bearing minerals, anion exchange capacity of aquifer materials (OH<sup>-</sup> for F), type of geological formations traversed by water and the amount of time that water is in contact with a certain geological formations [31].

Dissolution of these minerals is enhanced in acid medium as shown in Equation (5).  $Ca_{10}(PO_4)_6F_2 + 12 H^+ \longrightarrow 10 Ca^{2+} + 6 H_2PO_4 + 2 F$ (5)

However, in neutral to basic conditions, fluorapatite dissolves through exchange of F with hydroxide and the subsequent hydration of the  $\sim$ CaOH $_2$ <sup>+</sup> complex [30]. Fluorite minerals also dissolve in basic media through a similar mechanism as shown in Equation (6), enhancing F availability.

$$
\text{CaF}_2 + 2\text{HCO}_3 \rightarrow \text{CaCO}_3 + 2\text{F}^+ + \text{H}_2\text{O} + \text{CO}_2 \tag{6}
$$

The released F may remain as free ions in water, adsorb on particulates or combine with cationic organic and inorganic species in water or soil. Ingestion of such water is the main direct route through which F enters the food chain [12]. The physicochemical parameters namely pH, EC and TDS were therefore determined in an attempt to infer the factors facilitating fluorapatite dissolution in Sukulu Hills.

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A significant number of people born and raised from Sukulu Hills area have aesthetically objectionable brown teeth stains. This intrinsic tooth discolouration is a cosmetic, and a functional problem [32] affecting self-esteem of affected individuals, especially adolescents. Recognising the risk faced by the local population as highlighted above, and given the scanty information available on the level of  $F$  in groundwater in Uganda and Sukulu Hills, this study was aimed at documenting the extent of Fcontamination of groundwater in the area and to establish the relationship between F levels and the physico-chemical properties of the water. We also sought to use climate data of the area to determine the optimum F concentration in drinking water.

#### **2 Materials and methods**

#### **2.1 Study Area**

The Sukulu Hills are located in Osukuru Sub-County, Tororo District of South-Eastern Uganda. They are a group of hills which rise about 200 m above the surrounding plains. The Sukulu phosphate deposits are located between  $0^{\circ}$  36' N -  $0^{\circ}$  39' N and 34 $^{\circ}$  07' - 34 $^{\circ}$ 11' E, about 5 km SSW of Tororo Municipality. The region is served by a good road system, the road from Tororo to Mbale, Tororo-Busia and Malaba-Kampala highway. Tororo is an important rail junction, linking with Kenya, Mbale and Soroti and westwards, with Kampala.

The average weather conditions of Tororo for 5 consecutive years is presented in Table 2 below:





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Outside Prior

Sukulu Hills is an area rich in minerals including carbonatite phosphate rocks, rare earth oxides, mica, Pyrochlore, zircon, baddeleyite among others. Phosphate mining is done there and cement manufacture is done in the nearby Tororo Cement factory. The area was chosen because many people who were born and raised there usually show signs of dental fluorosis, and geological study of the rocks revealed the presence of fluoride-containing rocks [35].

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## **2.2 Sampling**

Systematic sampling was used to cover nearly all groundwater sources in the area. Samples of groundwater were collected (in duplicates) once a month for 12 months, (Jun 2015-May 2016) from all boreholes and springs (17 boreholes and five springs) within a radius of 3 km from foot of the Sukulu Hills, a fluorosis endemic area in Tororo District.

Polypropylene sampling bottles were washed thoroughly with distilled water and rinsed twice with the water to be sampled. Two composite water samples were collected from each water source by picking three grab samples and mixing them in a container before the final composite (about 120 mL) was taken. This ensured the samples taken were representative of the average characteristics of the water source.

Sample bottles were labelled as B1 to B17 (for samples of borehole water) and S1 to S5 (for water samples from Springs). Table 3 shows the location of the water sources sampled which can be located in Figure 1. Electrical Conductivity (EC), Total Dissolved

Solids (TDS) and pH were determined on-site. F concentration was determined within 12 hours of sampling.

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#### **2.3 Experimental**

Standards and reagent solutions were purchased from Thermo Scientific Orion, MA, USA. A handheld multiparameter meter (HANNA 991300) was used to measure pH, EC and TDS on-site. One-point calibration for pH and EC measurement was done with the Hanna Quick Cal calibration solution as instructed on the user manual.

The concentration of F- was measured by potentiometric method (Ion Selective Electrode) following the EPA Method 9124 [36] using a F Ion Selective Electrode (Orion 9609BNWP, Thermo Fisher Scientific Inc., MA, USA) with an ion meter (Orion Star A214 meter, Thermo Scientific Inc., MA, USA).

The F - Ion Selective Electrode (FISE) was filled with the *Optimum Results A* filling solution (Cat. No. 900061, Thermo Fisher Scientific Inc., MA, USA) and connected to the ion meter. The Orion stirrer (Cat. No. 096019) and Automatic Temperature Compensation (ATC) probes were also connected and the ion meter set to the ISE mode as instructed in the user manual. The electrode was calibrated by measuring 20 mL of F standard solution (1) mg/L) into a 50 mL polypropylene beaker and 20 mL of Total Ionic Strength Adjustment Buffer (TISAB) II was added to it. The FISE, Automatic Temperature Compensation (ATC) and stirrer probes were inserted, and with uniform stirring, a stable reading in the auto-read mode was recorded. The probes were rinsed with distilled water and blotted dry before use in the next standards. This procedure was repeated with  $F$  standards of 2 and 10 mg/L to complete the calibration.

To determine F concentration in water samples, 20 mL of sample was measured into a 50 mL plastic beaker, 20 mL of TISAB II added. FISE, ATC and stirrer probes were run to obtain a stable reading. The procedure was repeated to measure  $F$  in all the water samples.

### **3 Results and discussion**

### **3.1 Physico-chemical parameters (pH, TDS, EC)**

Five springs and 17 boreholes were located and their coordinates and elevation shown in Table 3. The pH recorded in borehole water was generally in the normal range of 6.5-8.5

for natural waters (Table 3). However, low pH was observed in all spring water, except S3 which had average of pH 7. Acidity of natural water usually arises from hydrolysis of polyvalent cations dissolved in it. The observed near-neutral pH of borehole water enhances the dissolution of F containing minerals through a mechanism described by Equation (6).



### **Table 3 Water quality parameters of groundwater from Sukulu Hills**



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No health-based guideline value for TDS are proposed in the 1993 WHO Guidelines, as reliable data on possible health effects associated with the ingestion of TDS in drinkingwater were not available. However, the presence of high levels of TDS in drinking-water (greater than 1200 mg/L) may be objectionable to consumers [37]. Water with extremely low TDS may also be unacceptable because of its flat, insipid taste. Our findings revealed TDS values ranging between 74-378 mg/L, which is within normal range for drinking water (< 800 mg/L) by the WHO 1993 guideline values. High TDS value also enhances the ionic strength and thus increases the solubility of F- minerals such as fluorite [38].

In this study, EC values of the samples were found to be in the range of 100 to 823 μS/cm. It indicates high mineralization in the area as shown by some samples with  $EC > 500$ µS/cm. Cations infer acidic character that enhances F availability according to equation (5). There was a positive correlation between high EC/TDS and F concentration (Figure 2). This is consistent with the findings of Rango et al [39] who found a similar relationship between F and EC/TDS in Ethiopian Rift Valley. Elevated EC and TDS is attributed to the geology of the area, known to contain ores of phosphorus, iron, calcium among others, whose dissolution increases mineralisation of groundwater.

## **3.2 Fluoride Distribution in Groundwater**

## *3.2.1 Spring Water*

F levels varied from 0.3-2.41 mg/L in spring water. Most springs contained F levels within the WHO and UNBS permissible range for drinking water, except S3 whose values were consistently high throughout the year (Table 4), with the annual average  $F$  of 1.7 mg/L, higher than the WHO and UNBS limits of 1.5 mg/L for drinking water. This spring directly drains water from the old phosphate mines, reaching deep excavations with mine wastewater that has interacted with fluoride-rich rocks over long periods. The long rock-water interaction times and the observed high salinity promote dissolution of F containing minerals, consistent

with reports from the study done by Rao and Prasad [40]. F levels exceeded the calculated guideline value of 0.4 mg/L reported in this study.

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#### *3.2.2 Borehole Water*

Borehole water generally contained higher F levels (0.4-3.0 mg/L) compared to spring water (Table 4), with mean annual F concentration in all borehole water generally higher as indicated in Figure 2. Boreholes B2, B3 and B7 recorded average F levels higher than the WHO guideline value of 1.5 mg/L. With reference to the calculated limit of 0.4 mg/L, water from all boreholes contained excess F.

The 1971 WHO international standards recommended control limits for F in drinking water for various ranges of the annual average of maximum daily air temperatures; control limits ranged from 0.6-0.8 mg/L for temperatures of 26.3-32.6 °C and 0.9-1.7 mg/L for temperatures of 10-12 °C [8]. Tororo, being a relatively warm area (26-32.6°C category) would require a maximum F rage of 0.6-0.8 mg/L. This is also supported by the findings and recommendations of Møller et al [14] who calculated a F guideline value of 0.6 mg/L of drinking-water for Uganda. By this value, about 70% of borehole water sampled contained excess F.

There is an apparent positive correlation between F levels and TDS Figure 2A and pH (Figure 2B). High F values seemed to be associated with near neutral pH. Borehole water registered generally higher amounts of F compared to spring water (Figure 2).



**Figure 2 Variation of Fluoride with TDS (A) and pH (B) in Groundwater from Sukulu Hills**

This difference could be attributed to water residence time on rocks; water from springs having higher flow rates with less residence time on rocks. Borehole water is extracted from deeper water aquifers, probably with more soluble fluoride-containing rocks. These findings are consistent with those from the study done by Rao & Prasad [40] and reports by Sivasankar & Darchen [31] which indicated a positive correlation between F concentration in water and depth of the springs and residence time of water on rocks. Table 4 shows the variation of F in water throughout the study period. It is noticeable that spring water generally registered lower F levels throughout the year.

# **Table 4 Monthly Concentration of Fluoride in Water from Springs and Boreholes in Sukulu Hills**

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- Missing results due to breakdown of boreholes at the time of sampling.

Figure 3 was used to established the relationship between average monthly precipitation and F in selected water sources. Wet months of September-November 2015 recorded higher levels of F in spring S3 as compared to the low-rainfall months (June-

August). The trend is similar but less regular for water from B3. The observation is inconclusive due to the shorter survey period, but higher levels of  $F<sup>-</sup>$  during rainy months is attributed to higher dissolution rate of F containing rocks as rain water seeps through soil.

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**Figure 3 Relationship between fluoride and precipitation in selected water sources**

## **3.3 Optimum fluoride level for Tororo District**

When equation (4) was applied using the weather data for Tororo where the AMMT is about 31 $\degree$ C (Table 2), the optimal upper level of F concentration of 0.4 mg/L was obtained. This figure should be taken as the F guideline value for drinking water in Tororo, as it takes into account the actual weather conditions and thus an estimate of water intake of people in the area. If this value is taken as a reference, nearly all the water sources sampled contained excess F (Figure 2).

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## **4 Conclusion**

From the results and discussion presented in Section 3, most drinking water sources in Sukulu Hills meet the physicochemical parameter (pH, EC and TDS) requirements for drinking water as prescribed by WHO and US-EPA.

From the weather data of Tororo, average temperature over an five-year period was found to be  $31^{\circ}$ C. Using this temperature and the modified Galagan equation, the calculated optimum upper level of F for drinking water in Tororo is 0.4 mg/L. This should be taken as the F guideline value for Tororo, and drinking water containing F levels higher than this value are considered unsafe. All water sources sampled contained average  $F > 0.4$  mg/L. This implies that the local population is exposed to the toxic effects of excess F. It is therefore essential for the relevant authorities to sensitise the local people about the dangers associated with consumption of water with excess F, consider alternative sources domestic water, and adopt water treatment technologies to remove excess F.

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**6 Declarations of Interest:** None.

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