

Application of soil composition for inferring fluoride variability in volcanic areas of Mt. Meru, Tanzania

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Abstract: Predicting fluoride levels in water within fluoride endemic areas is an issue of high significance. As a result several methods including mathematical models have been reported to suit the task. However, most of these methods have limited practicality to low income communities. This study presents the potentials of employing soil characteristics to predict the level of fluoride in groundwater. The study is based at the areas around Mount Meru in Northern Tanzania. The volcanic sediments around this mountain had been segregated by geological studies into various lithologies. In this study water and soil samples were collected at springs in volcanic sediments categorized as main cone group, mantling ash, Tengeru lahar, Ongadongishu lahar and Ngarenanyuki lahar. Fluoride levels in water were then correlated to elemental composition of the soil. Water samples showed that fluoride was low in the main cone group, mantling ash and Tengeru lahar whereby the median concentration was 1mg/l but it was high in Ngarenanyuki and Ongadongishu lahars whereby the median concentrations were 4mg/l and 9mg/l respectively. Soil analyses indicated that high levels of aluminium do coincide along with low sodium levels, and vice versa. In addition high levels of sodium in soil are accompanied by high levels of calcium. Correlation studies indicated a strong negative relationship between aluminium in soil and fluoride in spring water with $r^2 = 0.847$. On the other hand, a positive correlation was obtained between calcium in soil and fluoride in water with correlation coefficient, $r^2 = 0.765$. Likewise, sodium indicated a positive correlation with fluoride in water ($r^2 = 0.458$). So long as high levels of Na and Ca in soil or water normally result to formation of salts on the banks of water sources after prolonged evaporation during dry seasons, the correlation established between fluoride and such elements in soil can enable people within volcanic areas to identify water sources with unacceptable levels of fluoride in their areas hence reducing the risks of fluorosis.

Keywords: Fluoride, Soil, Mantling Ash, Lahar, Volcanic Rock, Mount Meru, Tanzania

1. Introduction

Fluorine is halogen gas, the most reactive and the most electronegative of all. Fluorine has the ability to form both covalent and electrovalent bonds, hence it is capable of attacking almost all other elements, with the exception of oxygen and nitrogen [1]. When in contact with apatite mineral like the hydroxyapatite ($\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$) minerals of bone and enamel, fluoride can substitute the hydroxyl ion [2]. By so doing, it strengthens the bone or enamel to prevent dental caries when taken in appropriate amount [3] or makes it brittle or mottling of teeth when in excessive [4].

Individuals are mainly exposed to fluoride by ingestion through being drinking water [4] and rarely from food

additives [5]. The recommended fluoride dietary value is based on age groups such that the value is 0.1 to 1mg per day and 1.5mg to 4mg of per day for infants and adults respectively [6]. Excessive dietary intake of fluoride causes different types of fluorosis, primarily dental fluorosis, skeletal fluorosis and crippling fluorosis depending on the level and period of exposure. The World Health Organization (WHO) recommends a fluoride concentration of 1.5mg/l in drinking water sources, assuming that a person who takes 2 litres of drinking water per day ingests 3mg of fluoride per day [4]. The health implications due to prolonged drinking water containing fluoride levels between 1.5-3mg/l is mottling of teeth, alias dental fluorosis, while water with fluoride between 4-8mg/l causes malformation of the bones and movement difficulties, a condition known as skeletal fluorosis. Beyond

the concentration of 8mg/l, fluoride causes crippling fluorosis, a syndrome which causes the bone functions to grow together hence leads to immobility [4].

Fluoride availability in environment is mainly attributed to the nature of the parent rock [7], weathering of volcanic rocks [8], volcanic eruptions [9], industrial and mining activities [10] as well as coal burning [11]. The mechanism for fluoride availability in the environment can include mineral carbonation and ion exchange [12]. In both cases the concentration is controlled by the pH value as well as the type of cations present in surrounding soil or waters. Fluoride adsorbs on metal cations in soil at acidic medium by forming complex compounds with Fe or Al; with the maximum adsorption of takes place at pH 5.5 [13] but dissolves in water to give free fluoride ion or hydrated spheres at the pH 8 and beyond [14]. The soil with pH below 7.2 is regarded as acidic soil while that above 7.2 is alkaline soil [8]. The alkaline soils exhibit a positive correlation with fluoride concentration.

Tanzania is one of the fluoride endemic countries [9] like other countries in the East African Rift system. The East African Rift system has been associated with high fluoride in water. The rift system starts around the Jordan and extends down through Sudan, Ethiopia, Uganda, Kenya and the United Republic of Tanzania. Many of the lakes of the Rift Valley System especially the soda lakes, have extremely high fluoride concentrations; for instance, fluoride concentration in the Kenyan Lakes Naivasha, Magadi, Nakuru, Elementaita, Bogoria, and Baringo were 2.4, 84, 344, 463, 738, and 5.4mg/l respectively [15].

Fluoride problem in Tanzania was officially documented in 1974 after the study which disclosed that about one third of the country recorded high fluoride concentrations in its water sources [16]. Further analysis indicated that the maximum fluoride levels in Dodoma was 92mg/l, Arusha 78mg/l, Singida 67mg/l, Kilimanjaro 25mg/l, Coast region 20.4mg/l, Tanga 20mg/l, Mwanza 18mg/l and Shinyanga 14mg/l [16]. Imputed by lack of no alternative water sources in certain parts of the country, the allowable upper concentration limit for fluoride in drinking water was designated to be 8mg/l [17]. This guideline remaining in application up to the year 2008 when the upper concentration limit of fluoride in drinking water was revised and a new value of 4mg/l was the recommended [18].

Mount Meru, a volcanic mountain in Northern Tanzania is focal point for fluoride in Arusha Region. The mountain hosts a number of perennial springs and hence it is the sole source of water in the area. In attempt to predict fluoride variability in the area, a study carried out in the North Eastern part of the mountain reported that low fluoride springs occurs at high elevation characterised with unaltered lavas while high fluoride occurs in low elevation within the lahars [19]. Earlier geological work had pointed out the volcanic eruption in Meru happened in a series of events leading to segregation of volcanic sediments into various lithologies [20]. Based on such information, it was envisaged that each lithology had different influence on fluoride variability. Therefore, the general objective of this study was establishing geochemical

characteristics of areas with or without high fluoride around Mount Meru, in Northern Tanzania whereas the specific objective was to compare elemental composition of soil to fluoride levels in water within selected major lithological divides which host water springs around the slope of Mt. Meru. So long as high level of Na and Ca in soil or water normally result to encrustation of salts [5] in following high rates of evaporation during dry seasons, the correlation established between fluoride and such elements in soil can help local people, with little training, in identifying water sources with unacceptable levels of fluoride within their areas hence reducing the risks of fluorosis.

2. Materials and Methods

2.1. Study Area

This study was conducted between July, 2013 and July, 2014 around Mount Meru (4565m a.s.l) in Northern Tanzania. The volcanism in Meru is attributed to the main rift faulting known to have taken place between 1.15 and 1.2 million years BP [20] with the series of volcanic eruptions. The lithology pattern of the volcanic sediments of mount Meru is dominated by the main cone group, mantling ash, laharic deposits, and fluvio-volcanic sequences coupled with small petrographic range (olivine-poor to nephelinite to phonolite). In this study, the main group and the mantling ash formation were considered together simply because the two formations intersect and form the mantling ash which does not obscure the underlying formation. The lahars that were included in this study are Tengeru, Ongadongishu and Ngarenanyuki, Figure 1.

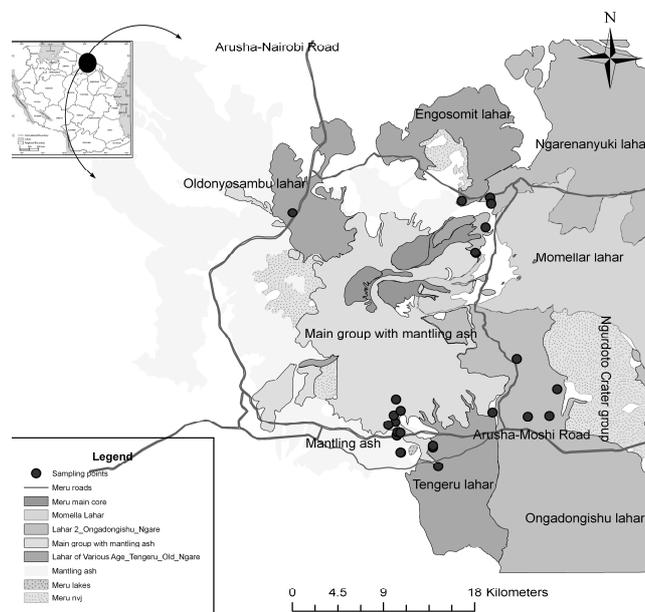


Figure 1. Sampling locations

The climatic condition of Mount Meru is oceanic rainfall with continental temperatures. Rainfall data indicate that the southern slopes of mount Meru receives much higher rainfall

of up to 1000 mm than the south eastern slope which receives 950 mm rainfall per year. The mean annual temperature decreases linearly upslope with a lapse rate of 0.56°C per 100 m starting at the foothills and the maximum and minimum temperature on the lower slopes (settlement areas) ranges are 15 - 30°C and 12 - 17°C, respectively [21] [22]. Water Supply in Meru relies on surface drainage, boreholes and perennial springs [20].

2.2. Sample Collection and Preparation

2.2.1. Water Samples

Water samples were collected at perennial springs in plastic bottles. Analyses of physical chemical parameters such as pH, electric conductivity, and temperature were carried out in situ using a portable pH-conductivity meter (HI 981305 HANNA Instruments). A total of 28 samples were collected and analysed. The analysis of fluoride was conducted in laboratory at the Ngurdoto Defluoridation Research Station in Arusha using platinum series fluoride selective electrode model 51928 and ISE/pH meter.

2.2.2. Soil Samples

Soil samples were collected near water springs at a depth ranging 0-20cm using a knife and shovel whereby 11 samples were analysed. The samples were stored in plastic bags for transport to the laboratory. The sample was oven dried at 105°C to constant weight, gently disaggregated in a porcelain pestle and mortar and passed through a plastic sieve of mesh size 2 mm. 12g of powdered sub-sample were mixed with 2.7g of a binder and pulverized at 180 rpm for ten minutes using a Fritsch Pulverisette®. The pulverized sample was pressed at 15 bar pressure to produce pellet with the diameter of 32mm using pellet presser machine Retsch® PP 25. The similar procedure was followed for the standard reference material (SRM) soil Montana®. The samples and soil Montana were analysed using the XRF and the result were interpreted using XRF software program called SPECTRO X-LAB® during sample measurement.

Matrix correction was performed based on the standard reference materials and the certificate value of the particular element as provided by the National Institute for Standard and Technology (NIST). The measured values (observed values) were corrected against the readings on reference standards and the certificate values of the respective element based on equations 1-2, below.

$$d_x = (SRM - CV_x) / CV_x \quad (1)$$

$$Element_x = OV_x - (OV_x * d_x) \quad (2)$$

Where: d_x = deviation of standard reference material (SRM) from certificate value (CV) of element x , OV_x = is the observed value of element x , and $Element_x$ = is the soil level of the particular element x . Samples were grouped according to their lithological formation for analysis purpose.

2.2.3. Geographic Information

A geological map of Arusha [20] was obtained from the

geological survey of Tanzania. The map was digitized in ArcGIS 10.1. To locate the sampling points, a global position system (GPS, Garmin – HTC 350) was used to capture the location during the sampling process. All the points were projected in the World Geodetic System 1984 (WGS 84).

3. Result and Discussion

3.1. Distribution of Fluoride

The distribution of fluoride in springs found along the Southern and Southern Eastern slopes of Mount Meru have been presented in Table 1. The results show that the high values of fluoride occur in springs of the Ongadongishu and the Ngarenanyuki lahars while the springs in the Tengeru lahars and the main group record low fluoride concentrations, Figure 2.

The average fluoride concentration in the main core and mantling ash group is 1.09mg/l while that of and the Tengeru lahar was of 1.25mg/l. In a different scenario; the Ongadongishu and the Ngarenanyuki lahars recorded high fluoride levels in springs. The mean fluoride concentrations in these lahars were 4mg/l and 10mg/l for the Ngarenanyuki and Ongadongishu lahars respectively.

Low level of fluoride observed in the Tengeru lahar might be associated with the fact that most of the springs studied in this area occur at the interface of the lahars and the main core or the mantling ash group. Perhaps, the lahar receives waters which had a reasonable contact time with the main core and the mantling ash group which is characterised with low fluoride in spring water mainly due to high aluminium content in soil, explained later. Besides, the springs in the lahar emerges from basaltic lavas which have been reported to cause minimal fluoride content to interacting water [23].

The fluoride values in water were compared to the 2008 Tanzania guideline for the upper concentration limit of fluoride in drinking water (4mg/l) and the 1984 World Health Organisation guideline (1.5mg/l). Most of the sources in the Tengeru lahar, the main core and mantling ash group could meet both the Tanzanian and the WHO guidelines. However, all water sources in both Ongadongishu and Ngarenanyuki lahars do not meet the WHO guideline, with few of them being within the Tanzania fluoride guideline, Figure 3. At the same time, high level of fluoride in the Ngarenanyuki lahars are comparable to previous study [23]. Fluoride levels in the lahars has been linked to long residence time of water in the volcanic ash and sedimentary soils [23], the fact that might hold for the high fluoride in the Ongadongishu as well. Likewise, pH values in springs of the main core, mantling ash group as well as in Tengeru lahars ranged from 5.95 to 6.79 while it ranged from 6.9 to 7.8 for springs in Ongadongishu and Ngarenanyuki lahars.

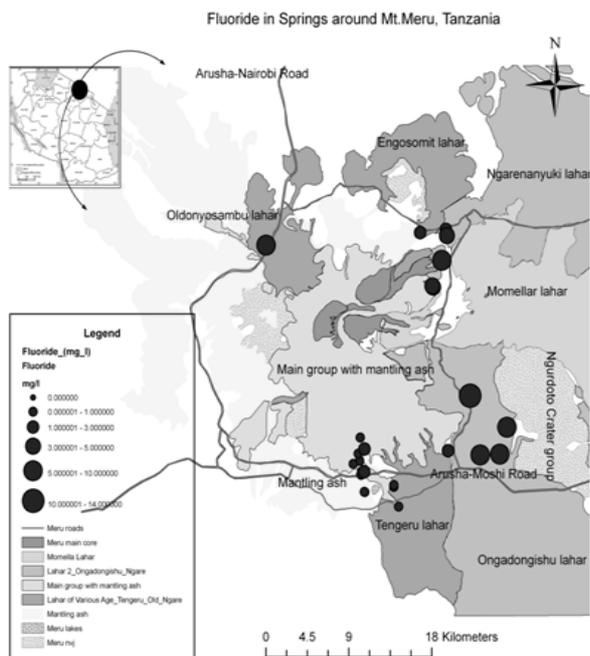


Figure 2. Fluoride levels at springs in various volcanic sediments of mount Meru.

The pH values of water had a direct implication to the fluoride levels. The values were mild acidic in areas with low fluoride while those with high fluoride appeared to be somehow alkaline. Table 1, indicates the pH values along with the fluoride levels in springs. The lowest value observed was 5.95 at AKH 2 within the main ash group. Generally, this sediment group recorded the lowest pH values which ranged from 5.95 to 6.59. The pH values for the springs within the main cone group laid between 6.37 to 6.79 while within the tengeru lahar, USA 1 appeared to have exceptionally high pH value of 7.63, the rest had the values below 7. The highest pH values were obtained from both the Ngarenanyuki lahar and Ongadongishu lahar. Within the ongadongishu and ngarenanyuki lahars the pH values ranged from 7.11 to 7.75 and 6.93 to 7.79 respectively. NGARE 5 recorded the lowest pH and fluoride values within the lahars. The general observation with regard to pH values is that sources high fluoride values occur along high pH and vice versa for sources within the same volcanic sediment group.

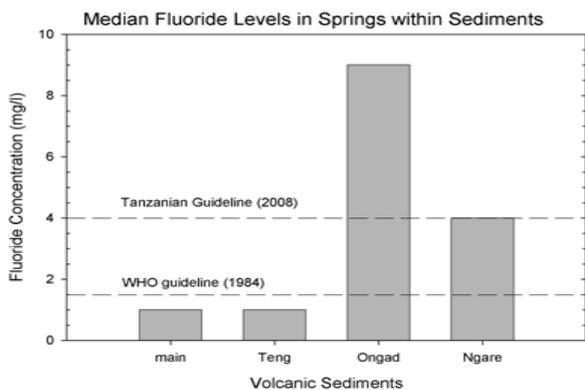


Figure 3. Comparison of fluoride levels to WHO and local guideline

3.2. Soil Content

The results for XRF analysis for soil composition, Table 2, show that apart from silicon, aluminium had the highest concentration of all the elements. The percentage of aluminium in soil ranged from 13% at CHA 4 to 30% at AKH 4 points found within Ongadongishu lahar and the mantling ash respectively. The other element which showed high proportional level was iron. The concentration of iron ranged from 0.9% (at AKH 2) to 18% (AKH 9) within the mantling ash and the main cone group respectively.

The element which showed the lowest proportional concentration among the major element was magnesium. The level of magnesium was less than 1% at AKH 2, AKH 4, AKH 12 and AKH 14. On the other hand, all the points within the Ngarenanyuki and Ongadongishu had the levels of magnesium greater than one percent. At least every point contained phosphorous and titanium to the level of 1% and beyond. The highest phosphorus level at all points was 2% while that of titanium was 3% which was recorded at AKH 9 within the main cone group. Generally, the titanium contents correlated somehow to iron content in the soil. The other elements of interest with regard to soil composition were calcium and sodium. The concentration of calcium ranged from 3% at AKH 12 to 8% at AKH 12 while the concentration of sodium was less than 1% at AKH 4 but it was 13% at CHA 4 within the Ongadongishu lahar.

Generally, from the variability of individual element within the soil, it can be established that certain elements are antagonistic in nature. It has been observed that high levels of aluminium do coincide along with low sodium levels and vice versa. For instance, the highest level of aluminium recorded in this study was 30% while the lowest level of sodium was less than 1% both values were recorded at AKH 4. Likewise, the lowest content of aluminium was 13% at CHA 4, the point at which sodium had its highest level of 13%. In addition high levels of sodium in soil are accompanied by high levels of calcium.

The elemental levels in soil were also analysed based on their lithological background. It was observed that average levels of aluminium content in soils were 265 within the main core and mantling group, 22% in Tengeru lahar, 18.5% in Ngarenanyuki and 15.5% in the Ongadongishu lahars. On the other hand, the average content of calcium in soil was 7% in the Ongadongishu lahars, 6.5% in Ngarenanyuki lahar, 5.5% in Tengeru lahar and 4% within the main core and mantling ash groups. Sodium content showed similar trend whereby it was 9% in the Ngarenanyuki, 8% within the Ongadongishu, 6% in Tengeru lahars and 3.7% within the main cone and mantling ash groups.

Magnesium content in soil was not only low but also almost constant across the sediments. The other element that had values almost constant across the sediments is phosphorus, which ranged from 1% to 2%. The highest value for titanium was 3% at AKH 9 and that potassium was 5% in NGARE 5. Nevertheless the elements did not indicate consistent trend across the sediments.

3.3. Correlation Studies

Correlation studies between fluoride in water and selected major element in soil show that element which can impact fluoride variability held strong relationship between them and fluoride in water, Figures 4 and 5. The strongest relationship established was between the average aluminium content in soil and fluoride in water which was a negative one with correlation coefficient, r^2 being equal to 0.847.

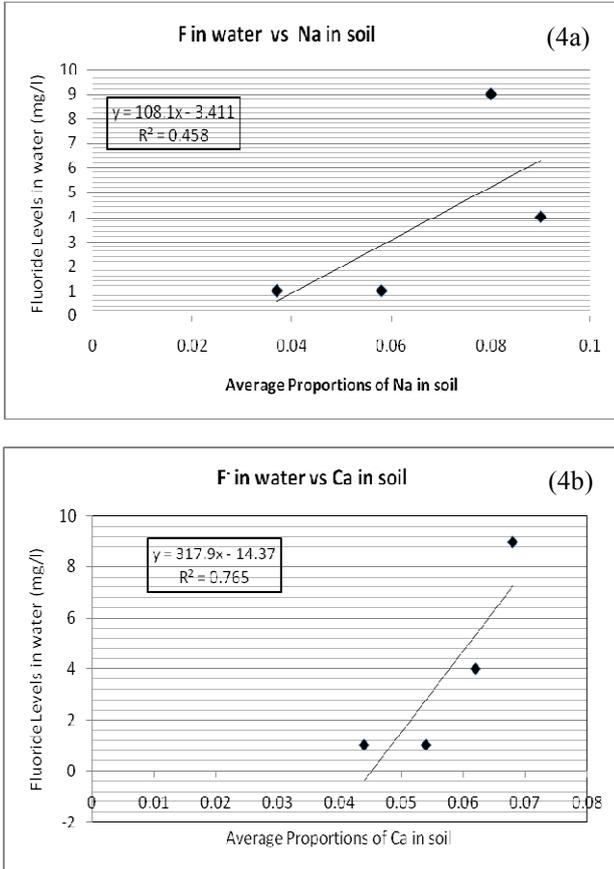


Figure 4. The correlation between fluoride in water against sodium (4a) and calcium (4b) in soil.

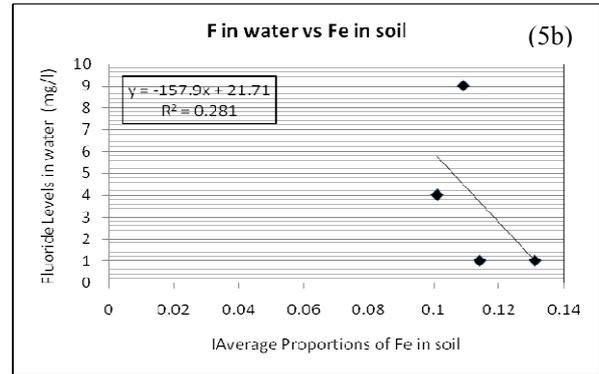
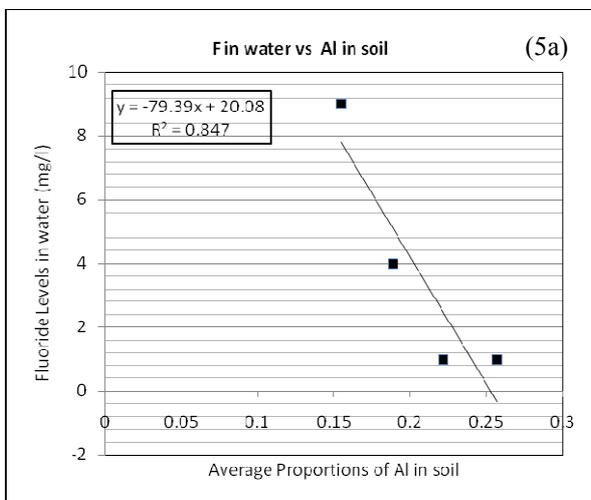


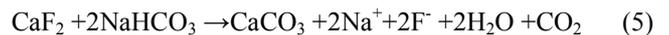
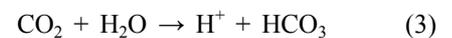
Figure 5. The correlation between fluoride in water against aluminium (5a) and iron (5b) contents in soil.

It has been pointed out that high aluminium contents are found within the main cone and mantling ash groups. The main cone group of Mount Meru has been reported to contain volcanic breccias and tuffs as well as some phonolitic and nephelinitic lava [19]. The correlation study between fluoride in water and iron in soil did not yield a strong relationship between although a weak negative correlation was indicated; with a correlation coefficient r^2 being equal to 0.281. Both aluminium and iron have the capability of complexing fluoride ion at low pH values [13]. Two possibilities are hence indicated, either high alumina lavas do not occur along with fluoride or fluoride is strongly complexed by the Al and Fe resulting into low fluoride availability in ground water.

Conversely, both sodium and calcium contents in soil showed positive correlations with fluoride concentration in water whereby the r^2 were 0.765 and 0.458 for calcium and sodium respectively. The positive correlation is in line with the observation that volcanic ashes may contain calcium compounds such as CaF_2 and $\text{Ca}_5(\text{PO}_4)_3\text{F}$ [24] along with other fluoride bearing compounds.

The occurrence of both Ca and Na in relatively high proportions in areas with high pH and fluoride concentration in water, suggest the formation of NaHCO_3 , Equations, 3-4. High levels of NaHCO_3 in water increases water pH thereby interfering with the normal precipitation of CaF_2 .

The presence of excessive sodium bicarbonates in groundwater increases the dissociation activity of fluoride [25] as expressed in Equation 5.



Ground water around in the eastern Meru has been reported to be alkaline in nature with high levels of sodium bicarbonate. As a result, high bicarbonate content has been associated with high fluoride in water. Therefore, this study has shown that sodium in lahar exist not only in water but also in soil. And that the areas with high sodium content in soil do contain high fluoride in their ground waters. And more important is that soil from such areas do contain minimal amount of aluminium.

Table 1. Distribution of fluoride in spring around Mt. Meru

Spring ID	UTM Zone	Easting	Northing	F-(mg/l)	pH	S*
Akh1a	37M	253092	9625729	1	6.25	ma
Akh1b	37M	253052	9625729	1	6.18	ma
Akh 2	37M	252705	9627008	0.9	5.95	ma
Akh 3	37M	252662	9627042	0.8	6.2	ma
Akh 4	37M	252754	9627346	1	6.13	ma
Akh 5	37M	253035	9627301	2	6.59	ma
Akh 6	37M	252542	962803	1	6.42	ma
Akh 7	37M	252382	9629128	1	6.79	mc
Akh8	37M	251844	9628100	0.7	6.42	mc
Akh9	37M	252598	9630735	0.8	6.37	mc
Akh10	37M	252595	9630735	0.9	6.72	mc
Akh11	37M	253058	9629590	2	6.62	mc
Ngare 1	37M	260474	9645864	5	7.07	N
Ngare2	37M	260394	9645947	5	7.33	N
Ngare 3	37M	261404	9648520	7	7.23	N
Ngare 4	37M	261849	9651643	3	7.37	N
Ngare 5	37M	261946	9651041	2	6.93	N
Ngare 6	37M	261958	9651001	4	7.15	N
Ngare 7	37M	259057	9651296	2	7.79	N
Akh 12	37M	256782	9623828	1	6.45	T
Akh 13	37M	255705	9625804	1	6.7	T
Akh 14	37M	256253	9625996	1	6.7	T
Usa1	37M	268514	9629442	2	7.63	T
Cha5	37M	264514	9634965	14	7.75	ON
Cha2	37M	267736	9629116	10	7.4	ON
Cha3	37M	268514	9631809	9	7.31	ON
Cha1	37M	265614	9629034	7	7.11	ON
Old2	37M	242333	9650006	8	6.8	n/a

S* sediment groups: ON= Ongadongishu, T= Tengere, N= Ngarenanyuki, mc= main cone and ma = mantling ash.

4. Conclusion

Table 2. Proportional ratios of major element in soil

Lithology	ID	Easting	Northing	Na	Mg	Al	K	Ca	Ti	Fe	Si	P
Ngarenanyuki	Ngare5	261946	9651024	0.11	0.01	0.18	0.05	0.07	0.01	0.10	0.47	0.01
-do-	Ngare6	261958	9651001	0.07	0.01	0.19	0.03	0.06	0.02	0.11	0.49	0.01
Ongadongishu	Cha3	268514	9631809	0.03	0.01	0.18	0.02	0.07	0.02	0.12	0.53	0.02
-do-	Cha4*	267291	9631837	0.13	0.01	0.13	0.03	0.07	0.02	0.10	0.49	0.01
Tengere	Usa 1	268514	9629442	0.09	0.01	0.20	0.03	0.07	0.02	0.12	0.45	0.01
-do-	Akh 12	256782	9623828	0.06	0.00	0.28	0.02	0.03	0.01	0.09	0.49	0.01
-do-	Akh13	255705	9625804	0.07	0.01	0.19	0.02	0.08	0.02	0.13	0.47	0.02
-do-	Akh14	256253	9625996	0.02	0.00	0.22	0.03	0.04	0.02	0.11	0.55	0.01
Main & ash	Akh 4	252754	9627346	0.00	0.00	0.30	0.01	0.04	0.02	0.12	0.51	0.01
-do-	Akh 2	252705	9627008	0.09	0.00	0.21	0.03	0.04	0.01	0.09	0.51	0.01
-do-	Akh 9	252598	9630735	0.02	0.01	0.27	0.02	0.04	0.03	0.18	0.42	0.02

Cha4* seasonal spring

Fluoride in ground waters around mount Meru is attributed to volcanoes which took place in the area about million years ago. The volcanic eruption happened in a series of eruptions leading to deposition of sediments with various compositions. This study has shown that fluoride availability in groundwater in the southern, eastern and northern eastern parts of Mount Meru do differ in the levels. The spring with high fluoride

levels in water occur within the Ongadongishu lahar where the median fluoride is 9mg/l. Water sources within the Ngarenanyuki lahar also record high fluoride concentration whereby the median fluoride levels in such areas is 4mg/l. On the contrary, sources within the Tengere lahar have low fluoride concentration in water, the maximum concentration being 2mg/l. The sources with the least fluoride levels follow

within the mantling ash and the main cone group. The median concentration in such springs is 1mg/l. Therefore, most of the springs within such sediment groups have fluoride levels in water within the recommended fluoride levels by WHO and local guideline. Soil analysis indicated that areas with low levels of fluoride in water contained much aluminium in soil than sodium. At the same time areas which had high fluoride in water, contained much sodium in soil than aluminium. The correlation studies indicated that aluminium in soil show negative correlation to fluoride in water with $r^2=0.847$. On the contrary, calcium content in soil show positive correlation to fluoride in water with $r^2=0.765$. Besides, the means of sodium content in soil had a fairly weak positive correlation to fluoride content in water, with $r^2=0.458$.

In general, calcium and sodium have been shown to exist together and at considerable high levels in lahars in contrary to the main cone and the mantling ash group. Fluoride also occur at considerable high levels in sources emerging from the lahar. On the other hand, aluminium is found mainly in the main core and mantling ash groups, the areas characterised by low fluoride in waters. Therefore, further study is needed to establish whether much fluoride is complexed by the minerals within the lahar or those within the main group and the mantling ash. All in all, soil composition can be informative enough to help prediction of fluoride variability, at least within the alkaline volcanic systems.

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