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The Origin of Fluoride in Groundwaters of the Paleozoic Sedimentary Formations of Ghana-A Preliminary Study in Gushiegu District

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Abstract: The occurrence of dental fluorosis and dental carries due to the occurrence of elevated fluorides in groundwaters had been identified and investigated in Gushiegu District of Northern Region of Ghana using an integrated approach involving water quality analysis and petrographical thin-section analysis of rock cuttings obtained from drilled samples from boreholes in the study area. Fluoride concentrations were found to range from 0.05 to 5.8 mg/L, with a mean concentration of 2.18 mg/L. Analysis of samples from 36 wells showed that 11.12% had fluoride concentrations below 0.05 mg/L, 44.44% had concentrations above 1.5 mg/L whilst 44.44% had concentration between 0.5-1.5 mg/L. Petrographical thin-section analyses revealed meta-mudstone, slate/meta mudstone, meta-mudstone/slate and meta-siltstone and carbonate quartz-schist as the rock types. Major rock minerals include quartz, iron oxide clay minerals, carbonates minerals, sericite and opaque whilst lithic fragments and fluorites make up minor minerals components. The range of modal composition of the major mineral components are opaque minerals (35-60%), quartz (20-60%), iron oxide (10-55%), sericite (3-35%), clay minerals (2-33%) and carbonate minerals (10-20%) with minor mineral components (lithic fragments and fluorite) having less than 1% in all the samples. Sericite appears to be the main mineral associated with the occurrence of elevated fluoride concentration in the groundwaters with carbonate minerals playing a minor role. Dissolution of fluoride into groundwater could be due to the interactions of groundwater with rocks containing sericite and probably carbonate minerals.

Keywords: Fluoride, groundwater, Ghana, sericite, voltaian sedimentary formation

INTRODUCTION

The Voltaian Paleozoic sedimentary formation underlies about 45% of entire land-area of Ghana and forms one of the two major hydro-geological provinces of Ghana. The other is formed by the Crystalline Basement Complex. This formation is sub-divided into three hydro-geological sub provinces by earlier geologists on the basis of lithology, field relationships and groundwater fortunes, namely; the lower Voltaian, middle Voltaian and the upper Voltaian (Junner and Hirst, 1946, Soviet Geological Survey Team, 1964), which are now referred to as The Bombouaka supergroup, Pandjari supergroup and Kodjari supergroup (Griffis *et al*., 2002). It underlies a portion of Ghana which is associated with the most poverty endemic rural settlements and with serious potable water supply challenges in terms of both quantity and quality. The Voltaian Basin is the least in terms of groundwater potential as far as groundwater potential of Ghana is concerned (Dapaah-Siakwan and Gyau-Boakye, 2000). Gushiegu District of Northern Region of Ghana is underlain by the Middle Voltaian formation, which forms part of the semi-arid region and amongst the poorest region of Ghana. Groundwater is the main source of potable drinking water and in some cases, as irrigation water to promote economic activities for almost all rural communities in this region. Groundwater is seriously depended upon due to its potential to be available throughout the whole year as compared to other sources such as dams and pipe-born water that are from surface sources and potentially not reliable throughout the whole year in terms quantity and quality. Groundwater is the safest in terms of quality; however, groundwater everywhere may not always be safe in terms of quality.

Fluoride concentrations outside the WHO recommended range of 0.5-1.5 mg/L in groundwaters in the Paleozoic sedimentary formations of northern Ghana in the Gushiegu-Karaga District had been reported (Dogoli, 2004). Earlier research in the district revealed the incidence of dental fluorosis with a prevalence rate in some communities as high as 61%, severe type of dental fluorosis and also the emergence of skeletal fluorosis

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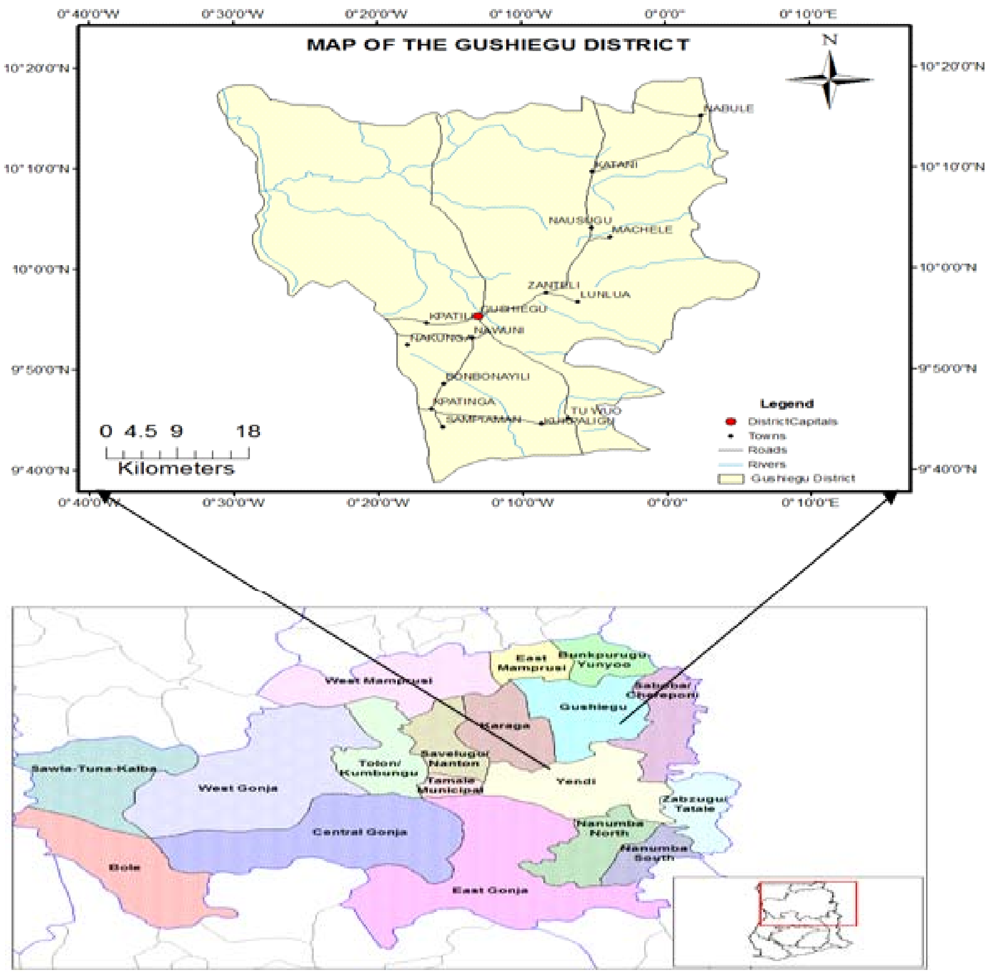

Fig. 1: Location of study area showing sample points soil, geology and hydrogeology

(Anongura *et al*., 2004). The source or origin fluoride in groundwaters of the Voltaian sedimentary formations of Ghana has however not been established. The purpose of this study is therefore to investigate the possible genesis or origin of high fluoride concentrations in groundwaters within the paleozoic sedimentary formations of Ghana. In this study, an integrated approach involving both waters and the aquifer materials (rock formations) in contact with the groundwaters were undertaken to establish the water/rock interactions. Laboratory tests involving hydrochemical analysis of groundwater and petrographical analysis of rock cuttings obtained from drilled samples from boreholes were carried out to help unravel the possible genesis of fluoride.

MATERIALS AND METHODS

Location: The study area (i.e., Gushiegu District) is located in the northeastern corridor of Northern Region of Ghana and lies between latitudes 9º40′0″ and 10°20′0″N and longitudes 0º40′0″W and 0º10′0″E. The District is 94 km from Tamale, the regional capital of Northern Region. It is bordered in the north by East-Mamprusi and Bunkpurugu/Yuyoo Districts' in the south by Yendi District, east by Saboba/Chereponi District and in the west by Karaga District (Fig. 1). The total land area of the district is 5,796 km^2. It is the fourth largest district in the Northern Region with about 469 communities, with the capital located in Gushiegu (www.ghanadistricts).

Climate and vegetation: The climate of the area is relatively dry and characterised by a single rainy season that normally begins in May and ends in October. The rainy season occurs between May to October follow by prolonged dry season. The main annual rainfall is between 100 and 120 cm whiles the dry season occurs between the months of November to April. The highest mean annual monthly temperature of about 33ºC occurs in March and

April and the lowest of about 26ºC in August. High relative humidity of 70 to 85% is recorded during the rainy season, the lowest is observed at about 20% during the peak of the dry season. (Dickson and Benneh, 1980). The vegetation type is guinea savannah, characterized by tall grasses interspersed with drought resistant trees such as the shear, Acacia, Neem, Mango and *dawadawa* trees.

Relief and drainage: The land is strewn with several streams, most of which are tributaries of the major river (the White Volta) in the northern region. The major rivers running through the district include the Nasia, which flows between Nambrugu and Bagli. The head waters of the Daka River are found in the district. Only the Nasia River is perennial. All the others dry up completely during the long dry season but flood the immediate surrounding land during the rainy season. The relief is fairly undulating with height ranging from about 140 m at the valley bottoms to about 180 m above sea level in the plateau surface (Kesse, 1985).

The soils are mainly savannah ochrosols, groundwater laterites formed over granite and Voltaian shales. Small areas of savannah ochrosols with some lithosols and brunosols are also very low. The laterites are similar in acidity and nutrient level to the ochrosols, but are poorer in physical properties, with substantial amounts of concretionary gravel layers near the top horizons and more suited for road and other constructional works than supporting plant root systems (Pelig-Ba, 2000).

The district lies entirely within the middle Voltaian basin-a sub-division of the paleozoic Voltaian sedimentary formation, specifically within the Kodjari formation of the Oti group which is highly heterogeneous in terms of lithology and dominated by quartzites, shales, mudstones, siltstones, conglomerates and limestones. This group overlies the Panabako formation of the Bombouaka Mega-sequence or the Lower Voltaian sub-division (Affaton *et al*., 1980; Wright *et al*., 1985; Hoffman, 1999). The northern tip of the district is underlain by Bombouaka Mega-sequence which consists of fine to coarse-grained and more or less feldspathic sandstones (Bertrand-Sarfati *et al*., 1990). The groundwater fortunes of this terrain have been extensively investigated (Wardrop and Associates, 1980; Acheampong, 1998; Agyekum, 2004). Aquifers in the study area are generally semi-confined and structurally controlled-developed by secondary porosity in the form of fractures and primary porosity has been destroyed through compaction and slight metamorphism (Junner and Hirst, 1946; Bannerman, 1975; Dapaah-Siakwan and Gyau-Boakye, 2000). According to Wardrop and Associates (1980), analysis of the available hydrogeological and lithological data from wells drilled in the study area indicated that fractured aquifer provides most of the well water.

Sampling and analysis: Between September and October 2010, available hydrogeochemical data from previous drilling records on boreholes were reviewed and analyzed to establish the general distribution of fluoride concentrations in groundwaters in the entire northern of Ghana. Elevated levels of fluoride concentrations (1.5 mg/L) were observed along the north-eastern flanges of the region, notably in the Gushiegu and Karaga districts. Gushiegu District had the most of the boreholes with elevated fluoride concentrations. Subsequently, groundwater sampling was therefore carried out taking into consideration the previous hydro-geological, geological and geochemical studies of the area between the months of October and November. Thirty-six (36) water samples from randomly selected borehole wells were collected and their geographical locations measured with Global Positioning System (GPS). Samples were collected after 5 min of pumping to purge stagnant waters, filtered through 0.45 mL cellulose nitrate filters and stored at 4ºC in carefully cleaned and sterilized 100 mL polyethylene bottles, transported in ice chest for fluoride analysis. Physical parameters namely; pH, electrical conductivity and temperature were measured in the field. The pH and electrical conductivity were measured using an YS16600-calibrated probe. All water samples were sent to Water Research Institute (WRI) laboratory at Tamale in the Northern Region of Ghana for analysis. Rock samples from each drilled borehole sampled were collected and sent to University of Ghana (Legon), Earth Science Geology Laboratory, Accra, Ghana. Thin sections of the rock samples was prepared and studied under transmitted light microscope for petrographic analysis.

RESULTS AND DISCUSSION

The results of water quality and petrographical thin section analyses of rock samples are presented in Table 1. Boreholes were generally of medium depths varying from 41-60 m averaging around 52 m. The acidity (pH) of groundwaters fell within the acceptable range of 6.5-8.5 (World Health Organisation, 2004); however, most of them (91.7%) were slightly basic whilst 8.3% were slightly acidic. pH values ranged from 6.8 to 7.6 with an average value of 7.17. temperature's ranged from 27-29.6ºC with an average of 28.25ºC. Conductivity values ranged from 630-1137 μS/cm with an average value 950 μS/cm. Fluorine concentration in groundwater ranges from 0.005 to 5.8 m/g. Out of 36 wells sampled within the study area, 4 wells representing 13.33% have fluorine concentration less than 0.5 m/g, 16 wells representing 44.44% have fluorine concentration greater than 1.5 mg/L while 16 wells representing 44.44% have fluorine concentration between 0.5 and 1.5 m/g (WHO acceptable range for drinking water). The fluoride concentrations show that some communities are potentially vulnerable to health hazards associated with fluoride concentrations outside the acceptable range of 0.5-1.5 mg/L for drinking water (McDonagh *et al*.,

Table 1: Summary of water quality and petrographical analyses

								Modal composition of major rock mineral (%)					
No	Community	GPS cordinates	Ph	Conducti vity (µS/m)	Temp (º C)	Fluoride conc(mg/L)	bh depth .(m)	sericite	clay minerals	iron oxide	carbonate minerals	opaque minerals	quartz
1	Gaa	-0.447361; 9.805194	7.02	997	28.30	0.05	42	<1	7	38	-	-	55
2	Bilsinga	-0.522306; 9.597056	7.60	786	28.80	0.70	52	-	20	30	15	-	35
3	Galiwei	-0.448639; 9.622306	7.30	875	29.10	1.00	41	-	2	50	-	-	48
4	Sambug	-0.006218; 9.9550906	7.10	908	28.40	5.80	50	30	10	24	-	-	36
5	Kasare	-0.021647; 9.980267	7.09	997	27.90	3.20	46	25	-	30	-	-	45
6	Limo 1	-0.593944; 9.611778	7.40	1045	28.70	1.70	48	12	5	-	-	55	28
7	Limo2	-0.593694; 9.6217083	7.12	983	29.30	0.26	58	<1	15	-	-	60	25
8	Kpanmange	-0.032111; 9.823444	7.32	872	28.20	0.67	50	<1	10	34	-	-	56
9	Tinguli	-0.010505; 9.956441	7.10	907	28.10	2.20	49	15	-	55	-	-	30
10	Gbanbu	-0.015375; 9.954391	6.80	1004	27.90	2.10	60	15	-	50	-	-	35
11	Chabiya 1	-0.0539720; 9.849806	7.20	985	27.70	1.00	52	<1	4	36	-	-	60
12	Chabiya 2	-0.0540280; 9.9849778	7.13	913	27.30	0.10	50	-	7	55	-	-	38
13	Kpugi	-0.0479472; 9.627167	7.30	945	28.00	2.03	48	3	-	-	-	50	46
14	Natinga	-0.0122671; 9.879110	6.89	1068	28.30	1.68	58	5	10	45	-	-	40
15	Lamanem 1	-0.0022780; 9.986000	7.50	675	27.90	0.56	60	<1	-	35	10	-	60
16	Lamanem 2	-0.0024720; 9.884639	7.30	1103	28.60	1.10	56	<1	33	12	-	-	55
17	kanshiegu	-0.014251; 9.954345	7.10	1067	28.20	2.40	48	7	-	20	18	-	55
18	Yawungu	-0.187000; 9.876389	7.20	893	28.10	0.80	56	<1	10	32	-	-	58
19	Gushiegu 1	-0.214250; 9.914667	6.90	788	27.90	1.10	52	-	5	55	-	-	40
20	Gushiegu 2	-0.214250; 9.926583	7.30	854	28.70	0.09	52	-	4	60	-	-	36
21	Gushiegu 3	-0.006218; 9.925139	7.20	1011	28.90	1.80	56	15	5	-	-	60	20
22	Gushiegu 4	-0.222806; 9.928750	7.32	788	28.40	1.30	50	-	7	55	-	-	38
23	Gushiegu 5	-0.218389; 9.928750	7.12	988	29.50	1.45	52	<1	10	55	-	-	35
24	Gushiegu 6	-0.228083; 9.907417	7.10	866	27.90	1.16	56	-	5	58	-	-	37
25	Gushiegu 7	-0.222861; 9.926028	7.60	977	27.00	1.20	52	<1	10	60	-	-	30
26	Gingbani	-0.0025830; 9.883651	7.11	1042	27.90	4.36	52	35	25	10	-	-	30
27	Nakondugu	-0.217165; 9.904329	7.09	1105	28.80	1.90	52	7	-	20	18	-	55
28	Saguli 1	-0.404833; 9.9620528	7.16	914	28.40	1.34	48	<1	8	36	-	-	56
29	Saguli 2	-0.403806; 9.620528	7.20	1076	28.00	1.38	50	<1	10	30	-	-	60
30	Zulogu	-0.032111; 9.823444	7.03	978	28.10	1.97	54	20	20	24	-	-	36
31	Kasela 1	-4.058567; 33.173517	7.06	970	27.00	0.87	50	<1	-	32	18	-	50
32	Kasela 2	-4.054566; 33.1616	7.36	979	27.00	1.72	55	8	-	24	20	-	48
33	Lubaga	-3.99900; 33.173400	7.17	911	28.30	1.87	48	24	18	22	-	-	32
34	Kabale	4.014133; 33.193150	7.14	630	28.60	1.08	52	-	15	55	-	-	30
35	Itonjamandi	-4.018200; 33.166517	7.08	1085	28.20	2.39	52	12	-	20	18	-	50
36	Mwagundu	-4.026167; 33.173917	7.17	1137	29.60	2.54	50	20	-	25	10	-	45

2000; Mazet, 2002; Edmunds and Smedley, 2005; World health Organisation, 2004). Fluoride has gained global recognition in recent times as far as groundwater quality issues are concerned. Fluoride is known to have both beneficial and adverse effects on humans, depending on the total intake. Fluoride in acceptable concentrations has been found to promote the growth and strength of the human bone and the teeth. However, consumption of water containing fluoride concentrations outside the WHO recommended range of 0.5-1.5 mg/L could result in serious health implications, including dental caries, dental fluorosis and skeletal fluorosis and crippling fluorosis. According to Pontius (1991), waters containing fluoride with concentrations less than 0.5 mg/L can cause dental caries Waters containing fluoride concentrations above 1.5 mg/L but less than 4 mg/L causes dental fluorosis whilst waters containing fluoride above 4 mg/L but below 10 mg/L can cause skeletal fluorosis. Waters containing fluoride concentrations above 10 mg/L can result in crippling fluorosis. In Ghana, groundwaters with high fluoride concentrations above WHO limit of 1.5 mg/L and associated health implications of mottled teeth and dental fluorosis had been widely studied and found to be occurring in Bongo and Bolgatanga districts in north-eastern Ghana, which are underlain by granitoids associated with the Precambrian Birimian meta-volcanics and meta-sediments. The occurrence of elevated fluorides in groundwaters occurring in the granitoids of Northeastern Ghana has been found to be associated with microcline-rich granitoids (Apambire *et al*., 1997).

Petrographical thin-section analyses revealed the following rock types; meta-mudstone, slate/meta-mudstones, meta-mudstone/slate, meta-siltstones and carbonate quartz-schist. Analysis of petrographical thin sections of rock samples under the ore microscope revealed five mineral components, namely; quartz, iron oxide, sericite, clay minerals opaque mineral and carbonate minerals. Estimated modal compositions of the above minerals identified quartz, iron oxides and clay mineral as major minerals components while sericite, carbonate minerals and some opaque minerals constituted the minor mineral components. Quartz mineral is ubiquitous and most abundant with modal composition ranging between 20-60% and averaging around 42.5% followed by iron oxide, which occurred in 32 rock samples out of thirty-six with modal compositions ranging from 10-60% and averaging around 37%. Clay minerals occurred in 25 samples with modal compositions ranging between 2-33% and averaging around 11.5%. Sericite, Carbonate minerals and opaque minerals occurred in 16, 8 and 4 rock samples with modal compositions ranging from 3-35 and 10-20% and opaque minerals 50-60% and averaging around 16.20, 15 and 55.80%, respectively (Table 1).

Analysis of modal compositions from thin section and measured fluoride concentrations from water samples showed that all rock samples containing sericite mineral

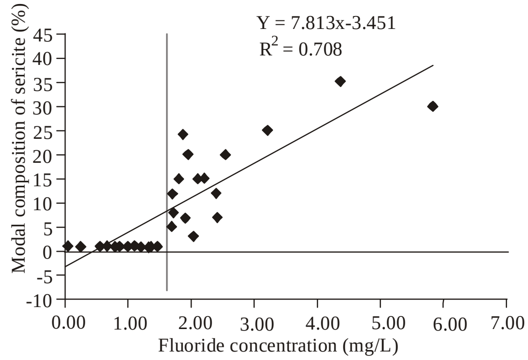

Fig. 2: Variation of fluoride concentration with modal composition of sericite

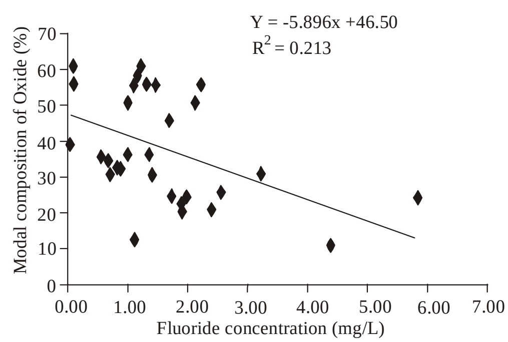

Fig. 3: Variation of fluoride concentrations with modal composition of iron oxide

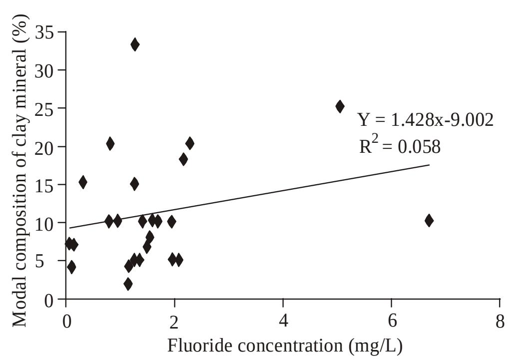

Fig. 4: Variation of fluoride concentration with modal composition of clay minerals

had elevated fluoride concentration above 1.5 mg/L. Furthermore, there is a strong correlation between fluoride concentration and modal composition of sericite (Fig. 2). Where modal composition of sericite is less than 1% (1) fluoride concentration were found to be less than 1.5 mg/L. This pre-supposes that the occurrence of elevated fluoride concentrations in groundwaters within the study area could be associated with sericite minerals occurring in the Voltaian sedimentary rock formations. This correlates with an earlier proposal by Affaton *et al*. (1990)

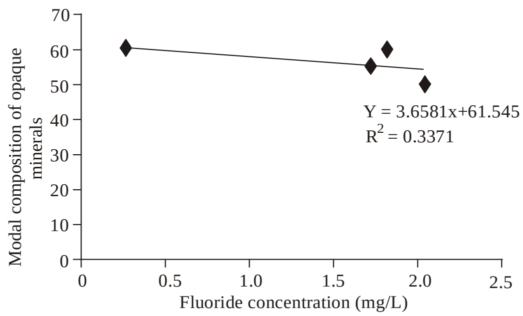

Fig. 5: Variation of fluoride concentration with modal composition of carbonate minerals

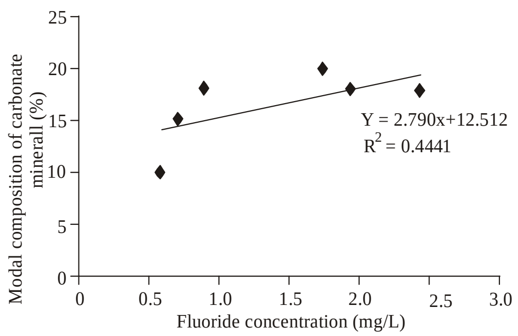

Fig. 6: Variation of fluoride concenttion with modal composition of opaque minerals

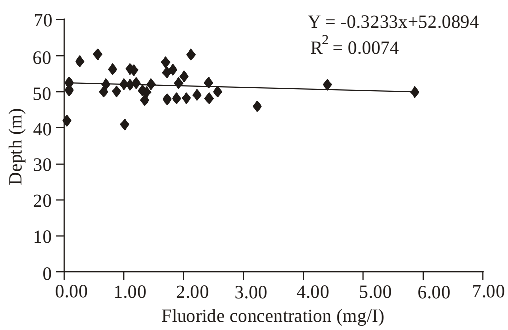

Fig. 7: Variation of fluoride concentration with borehole depth

that the Voltaian formation is of marine environment, which normally is characterised by volcanic eruption and Sericite is known to be formed from crystallisation of hydrothermal fluid during volcanic eruptions.

Carbonate minerals could be possible sources or associations of elevated fluoride concentrations in groundwaters in sedimentary formations where fluoride easily proxies for hydroxyl ions (Josephus *et al*., 1977). There seem to be some level of contribution of elevated fluoride concentration from carbonate minerals next to sericite. However, the correlation between fluoride concentrations and modal composition of carbonate minerals, even though positive, is reasonably weak (Fig. 3). Clay minerals had very little or no contribution to the occurrence of elevated fluoride concentrations in groundwaters in the study area (Fig. 4). This presupposes that clay mineral plays very little or no significant role in the accumulation of fluoride in groundwaters in the study area. There are inverse correlations between fluoride concentration and the modal compositions of iron oxide and opaque and minerals and borehole depth, even though the correlations is quite weak (Fig. 5, 6 and 7).

CONCLUSION

Elevated and low fluoride concentrations in groundwaters had been identified in Paleozoic Voltaian sedimentary formation Gushiegu District of Northern Region of Ghana. In this study, the maximum fluoride concentrations in groundwaters up to a maximum of 5.8ml/l, implying that some communities depending on groundwater as their main and some cases, the only source of potable drinking water for livelihood could potentially be at risk of contracting health effects associated with consuming waters containing fluoride concentrations lying either below or above the WHO acceptable range 0.5-1.5 mg/L. Such diseases include dental caries (F^- <0.5 mg/L), dental fluorosis (1.5 mg/L <F^- <4 mg/L) and skeletal fluorosis (4 mg/L <F^- <10 mg/L). Waters containing fluoride concentrations above 10 mg/L can result in crippling fluorosis, thus, apart from crippling fluorosis, communities in the study area could be affected.

The occurrence of elevated fluoride concentrations above the upper limit of World Health Organisation (WHO) acceptable range 0f 0.5-1.5 mg/L could mainly be associated with the presence of sericite minerals (major contributor) and carbonate minerals (minor contributor) in the Voltaian sedimentary rock formation. Clay minerals do not seem to play any significant role in the accumulation of elevated fluoride concentration in ground waters of the study area due to the very low correlation. This implies that there are no hydroxyl ions present in them to serve as proxies for the anionic exchange with fluoride ions. The correlation between depth of boreholes and the occurrence of elevated fluoride concentrations is very weak, suggesting that dissolution of fluoride into groundwater may be due to groundwater-rock interaction or groundwater-unconsolidated zone interaction or both.

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