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FLUORIDE CONTAMINATION OF GROUNDWATER AND HEALTH IMPLICATIONS IN ZANGO, KATSINA STATE, NIGERIA

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ABSTRACT

The consumption of drinking water is the principal route of exposure to fluoride for human beings and other living things. Different living organisms are more or less sensitive to fluoride. It can affect their growth, activity, or survival above certain concentrations. This study presents the impact of fluoride in groundwater in Zango Local Government Area of Katsina State, Nigeria. Standard sampling and analytical methods were employed in the study. Results revealed that the alkaline pH and high bicarbonate are responsible for release of release of fluoride-bearing minerals into groundwater. The arid climate of the region, the granitic rocks and the low freshwater exchange due to periodical drought conditions are responsible for the higher incidence of fluorides in the groundwater resource. The people dependent on groundwater resources in the area are prone to dental fluorosis and mild skeletal fluorosis. Fluoride is both beneficial in strong teeth and bone formation (<1.5mg/l) and also damage both the teeth and bone that it helped in their formation if the concentration exceeds 1.5mg/l. Preventive measures, including education programmes to raise awareness on risks/detrimental effects that fluoride in groundwater has on health, multidisciplinary work and joint actions between health and water managers, are necessary to supply safe water to the population affected.

Key words: *Fluoride, fluorosis, groundwater, health impacts, Zango, Katsina*

INTRODUCTION

Water quality is a consequence of the natural physical and chemical state of the water as well as any alterations that may have occurred as a consequence of human activity. Zango Local Government Area is fully characterized by heavy farming activities as well as extreme use of both organic and inorganic fertilizers. Poor sanitary practices are also rampant in the area. The usefulness of water for a particular purpose is determined by the water quality. If human activity alters the natural water quality so that it is no longer fit for a use for which it had previously been suited, the water is said to be contaminated or polluted. It should be noted that in many areas, water quality has already been altered by human activity but the water is still usable e.g water having a high concentration of certain minerals such as calcium which makes it become hard and therefore unsuitable for lather forming but still within suitable drinking limits, water having high concentration of certain minerals such as fluoride above the drinking limits which can still be used for other domestic purposes (washing, gardening, construction amongst others).

Fluorine is the most electronegative of all chemical elements and is therefore never found in nature in elemental form (Gupta, 1999; Gupta *et al*., 2013). Combined chemically in the form of fluorides (F^-), it ranks 17^{th} in the abundance of elements in the earth's crust representing about 0.06 – 0.09% of the (WHO, 2006). It occurs in a combined form in rocks and soil in a wide variety of minerals such as fluorspar (fluorite, CaF_2), cryolite, ($NaAlF_6$), apatite Ca_5 (PO_4) F) and topaz Al_2SiO $(FOH)_2$ (Buxton & Shernoff, 1999; Hamilton, 1992).

Fluorite (CaF_2) is the principal fluoride mineral, mostly present as an accessory mineral in granites. Dissolution of such minerals can constitute a major source of F in groundwater (Shaji *et al*., 2007). The concentration of fluoride in groundwater varies and depends on the level of impact caused geological formation, water and temperature. pH, solubility of Fluoride bearing minerals and the presence or absence of other precipitating or complexing ions do contribute to fluoride concentrations (Parkhurst and Appelo, 1999). Because of the large number of variables, the fluoride concentrations in groundwater can range from under 1mg/L to more than 35mg/L (WHO, 2006). High concentrations in groundwater also result from evapotranspiration, which may trigger calcite precipitation as a result of reduction in the activity of Ca^{2+} (Jacks *et al*., 2004). Several studies have pointed out an increase in dissolved Fluoride concentrations with increasing groundwater residence time (Edmunds & Smedley, 2005; Genxu & Guodong, 2001). Relatively high Fluoride concentrations have been found in some deeply circulating groundwaters (Kim & Jeong, 2005; Kundu *et al*., 2001). Abu-Rukah & Alsokhny, 2004; Ghorai & Pant, 2005 reported that the assimilation of Fluoride by the human body from drinking water with concentrations above 1.5mg/L might result in fluorosis.

The wide spread of fluoriferous groundwater seen in the study area is thought to be associated with the heterogeneous nature of the sediments mineralogy (Goni & Bura, 2009). Dissolution of aquifer matrix could also be a possible source of high fluoride in the middle and lower aquifer zones. The principal factors controlling the occurrence of fluoride in the area include the long residence time of groundwater with the aquifer matrix coupled with the depth and confining nature of the aquifer zones.

The main justification for this study therefore is the realization that groundwater quality may have become degraded due to some contamination from the source rock or human activities. This study therefore evaluates the fluoride content of well in different parts of Zango, Katsina State, Nigeria with emphasis on the health risks/implications as well as provides baseline data/information, the natural background concentration of fluoride in the groundwater system, the geospatial distribution in the form of maps and its relationship with fluorosis prevalence and the need for government and other stakeholders in the water and health sectors to take urgent practical steps to reduce the

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incidence of dental and skeletal fluorosis in the area.

MATERIALS AND METHODS

The Geology and Genesis of Fluoride in the Study Area

The lithologies include the Gundumi formation emanating in the Sokoto Basin, those of the Chad Formation in the Chad Basin as well as Granites and Rhyolites from the Basement Complex Rocks and Younger Granites suites (Fig.1). Basement Complex Rocks as well as the Younger Granites suites. The study area (Fig. 2) lies within the basement complex.

Fluorite, the main mineral that controls the geochemistry of fluoride in most environments is found in significant amount in granite and granite gneisses of the study area. Favourable condition exists for the dissolution of fluoride-bearing minerals present in the granite and gneissic rocks in the area. Fluoride is a typical lithophile element under terrestrial conditions and studies have revealed their association with granitic rocks. It is a major constituent insilicate rocks especially those of late magmatic stages typified in apatite, Fluorspar, Cryolite and Fluorapatite as well as villiaumite and syenites.

The petrographic studies of the area revealed that the rocks which are granites, rhyolites and sandstone contain galena-chalcopyrite-sphalerite mineralization (Chae *et al* 2007; Amadi, 2009). During weathering process, the fluorite is released and is leached downward via infiltration into the groundwater system. Mineralogical evaluation of granitic rocks suggests the presence of fluoride bearing minerals such as fluorite, fluor-apatite and apatite as well as nacaphite. Studies have shown that the sodium concentration increases with the solubility of fluoride bearing minerals due to similarity in their charges (Chae *et al.*, 2007). This implies that the high fluoride content observed in the groundwater system in Zango Local Government Area of Katsina, North-western Nigeria is purely geogenic due to weathering and dissolution of fluorite-rich minerals contained in the host-rock. The level of enrichment of any mineral (fluoride) in groundwater is a function of chemistry of the aquifer, well depth, hydrological conditions, residence time and geological structures (Amadi & Tukur, 2014).

Fluorides in the groundwater are derived from the weathering and subsequent leaching of fluoride-bearing minerals in rocks and soils. A substantial amount of this fluoride is retained in subsoil horizons, where it complexes with aluminium that is associated with phyllosilicates (Vaish and Vaish, 2000). Larsen and Widdowson (1971) observed that the solubility of fluoride in soils is favoured by pH <5 and pH>6 values. The predominant retention mechanism is that of fluoride exchange with the OH group of amorphous materials, such as al-hydroxides. Studies have shown that weathering of rocks and evaporation of groundwater are responsible for high fluoride concentration in groundwater of an area apart from anthropogenic activities including irrigation which accelerates weathering of rocks (Rao *et al*., 1993; Murthy *et al*., 2003; Amadi *et al*., 2012).

Sampling and Analytical Methods

A sampling pattern was adopted to cover a substantial portion of the entire Zango Local Government Area. At each sampling point, the longitude and latitude were taken with the aid of a handheld Global Positioning

System (GPS). Boreholes and hand-dug wells were the sources that were sampled for this study. The sampling covers the Gundumi Formation which extends from the Sokoto Basin and the Chad Formation which is part of the Chad Basin and Basement Complex Rocks including the Younger Granites suites. A total of 79 samples were collected for analysis while 31 samples were collected from boreholes installed with hand pump, 12 from those with solar powered and 9 from those with submersible pumps; 47 from hand-dug wells and 1 from a tube well. In terms of geology, 42 water samples were collected from the Gundumi Formation, 19 from the Chad Formation and 18 from the Basement Complex. The measurement of the following physical parameters i.e pH, temperature, conductivity and turbidity were carried out in-situ (on-site) using appropriate instruments in line with the specified standards (APHA, 1998). Glass and plastic containers were used to collect the samples at each location. Two drops of concentrated HNO_3 were added to the water samples in the plastic container (Schroll, 1975). The water samples in the glass container were for the determination of the anions. The samples were collected during the peak of the dry season. The determination of the physical parameters was carried out in the field, while the analyses of chemical and bacteriological parameters were done in the laboratory using standard methods. The samples were analyzed at the National Research Institute for Chemical Technology Laboratory (NARICT) Zaria, Nigeria using Atomic Absorption Spectrophotometry (AAS) Model 6400 Shimabzu Spectrophotometer and the Nigerian Geological Survey Agency Laboratories (NGSA), Kaduna and Aqua Tetra Sundry Services Katsina. Fluoride was determined using colorimetric method. The total and faecal Coliforms were determined using incubation method at the Katsina State Water Board Laboratory, Ajiwa, Katsina State, Nigeria.

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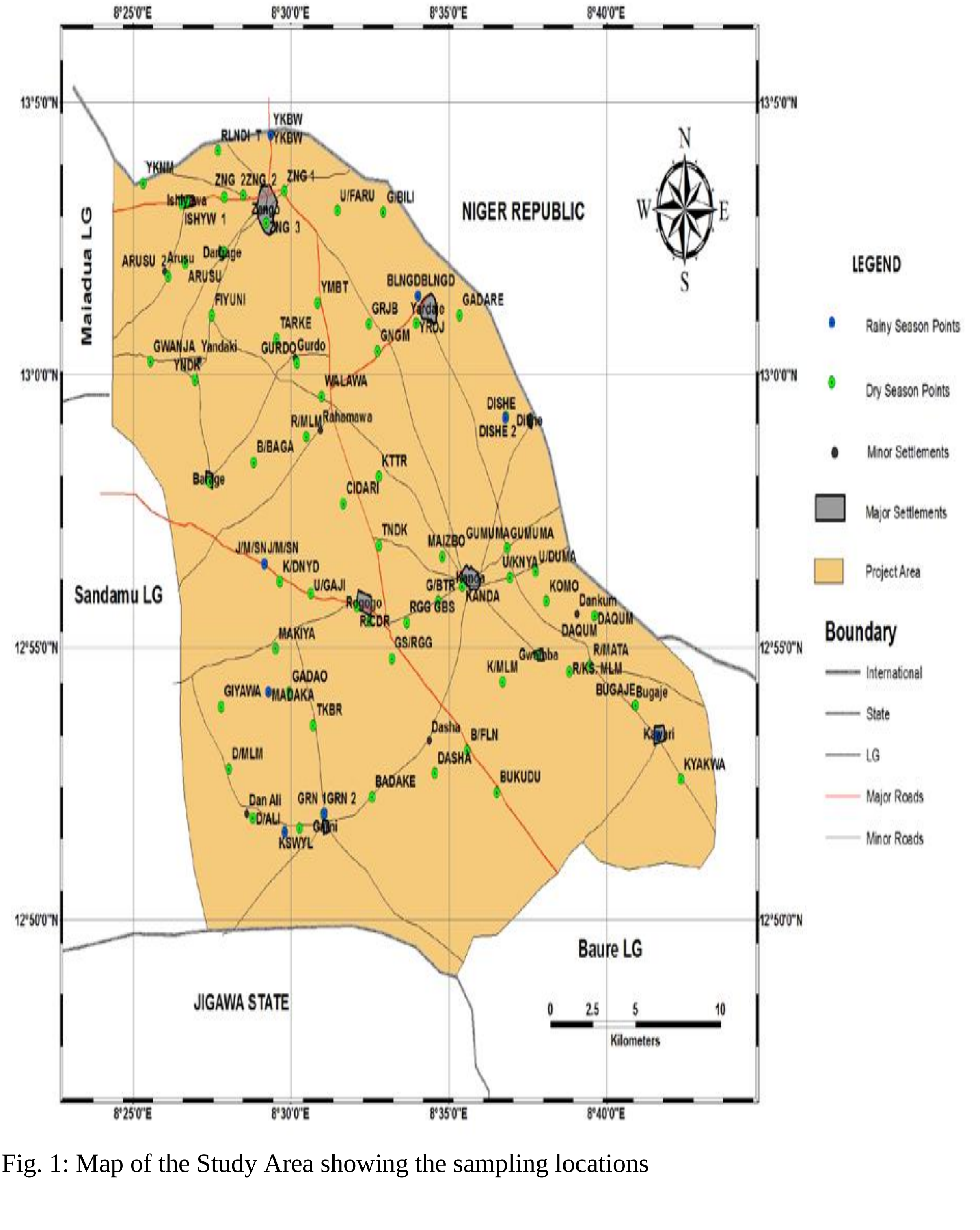

Fig. 1: Map of the Study Area showing the sampling locations

RESULTS

Table 1: The statistical summary of results obtained from the analysis

Parameter	Max	Min	Mean	Range	Variance	S.D	WHO (2006)
pH	7.85	5.15	6.489	2.7	0.37	0.611	6.5 – 7.5
EC (μS/cm)	3050	22.7	305.0	3027.3	22865	473.14	180
Temp (^{O}C)	34.5	28.1	32.32	6.4	1.47	1.21	27.00
TCC/100ml	94	0	19.27	24	2735.09	52.29	0
FCC/100ml	94	0	6.87	94	319.43	17.87	0
Salinity (mg/l)	35.67	0	11.84	35.67	68.87	8.36	200
CI (mg/1)	21.62	0	7.21	21.62	25.59	5.06	250
HCO_3(mg/1)	126	6	23.79	120	389.12	19.73	-
F(mg/1)	3.16	0.10	0.81	7.06	0.813	0.90	1.50
Colour(Hazen)	15	1	2.81	14	4.422	2.10	20
Ca (mg/1)	127.82	0.06	5.82	127.76	240.03	15.49	70
Mg (mg/1)	3.19	0	1.71	3.19	1.44	1.2	30
Na (mg/1)	31.18	0.07	6.79	31.11	48.76	5.86	200
K (mg/1)	28.22	0.19	8.35	28.03	39.38	6.28	200
Mn (mg/1)	0.48	0.01	0.09	0.47	0.012	0.11	0.2
Fe (mg/1)	0.3	0.01	0.122	0.01	0.01	0.08	0.30
Cu (mg/1)	0.08	0.01	0.041	0.07	0.01	0.022	1.00
Cr (mg/1)	0.1	0.01	0.03	0.09	0.05	0.02	0.05
Pb (mg/1)	0.05	0.01	0.02	0.04	0.01	0.01	0.01
Zn (mg/1)	0.2	0.01	0.02	0.19	0.02	0.04	3.00
SO_4(mg/1)	78.65	0.06	11.69	78.59	275.91	16.61	100
NO_3(mg/1)	19.12	0.06	6.28	18.52	21.14	4.59	50
PO_4(mg/1)	9.4	0.01	1.744	9.39	4.02	2.05	0.05

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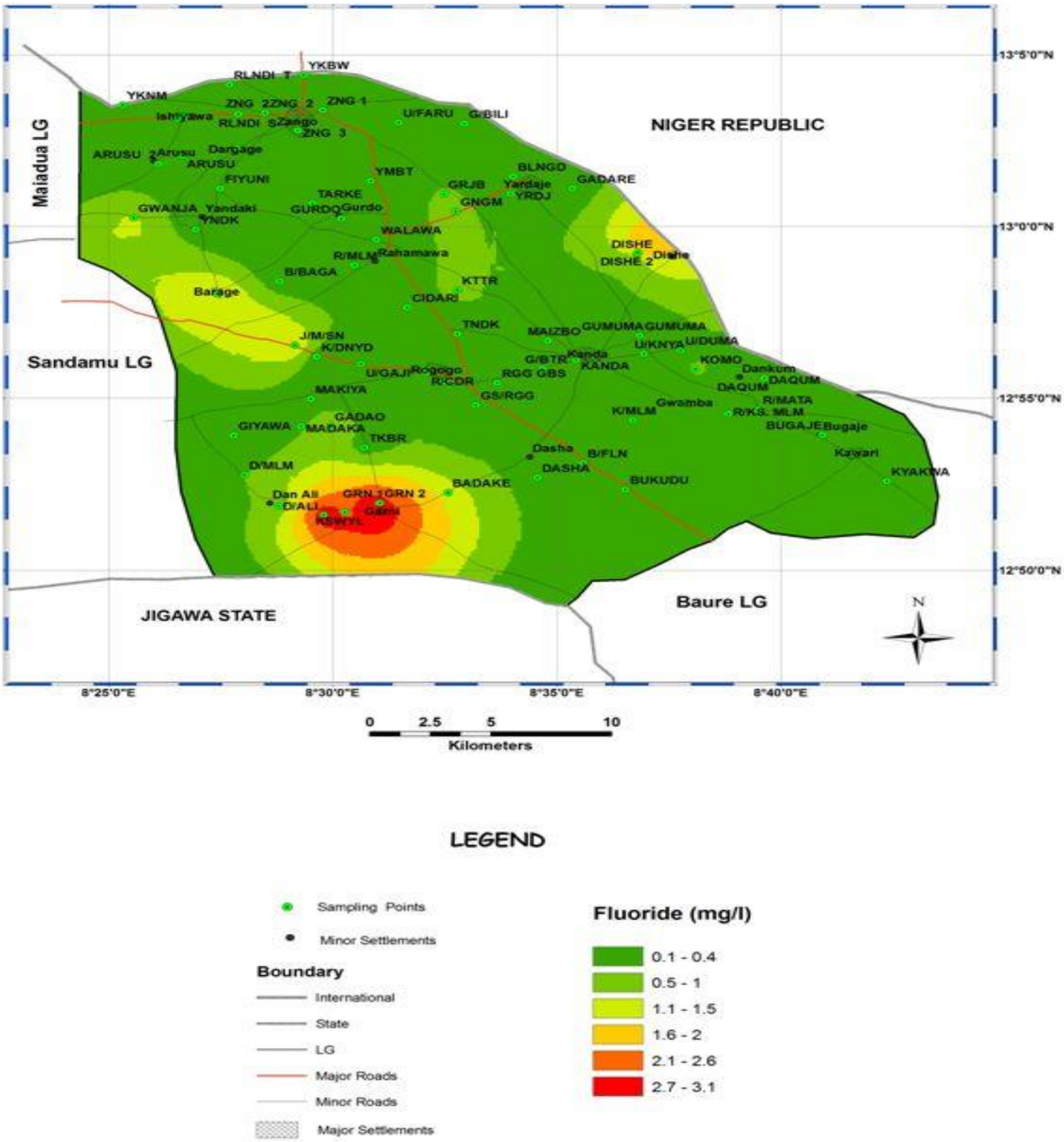

Figure 3: Map of Zango Showing the Concentration of Fluoride

Hydrochemical Facies and Implications

The concentration of eight (8) major ions (Na^+ ,K^+, Mg^{2+}, Ca^{2+}, Cl^-, CO_3^{2-}, HCO^-_3 and SO_4^{2-}) are represented on trilinear diagrams by grouping the (K^+ with Na^+) and the (CO_3^{2-} with HCO^-_3), thus reducing the number of parameters for plotting to six. The ionic strength of the groundwater is generally dominated by major cations (Sodium, Potassium, Calcium and Magnesium) and anions (Bicarbonate, Chloride, Carbonate and Sulphate). Hydrochemistry of the groundwater aquifer are summarized through the trilinear plots of the principal anions and cations. On the piper diagram, the relative percentages of the cations and anions are plotted in the lower triangles, and the resulting two points are extended into the central field to represent the total ion concentration.

The plot clearly revealed that the analyzed well-water samples from the study area had multi- hydrochemical facies with Ca-Mg-HCO_3-SO_4, Ca-HCO_3, Ca-Mg-HCO_3-Cl, Na-HCO_3 and Ca-Mg-K-HCO_3 being the most dominant water types. Piper's trilinear plot revealed that the dominant types are Earth-Alkaline (Ca or Mg). These dominant water types are indicative of the fact that the wells from which the water samples were collected tap water from the shallow regolith zone. This condition as well as short residence time of groundwater greatly enhances the effect of the subsurface geology on the groundwater chemistry.

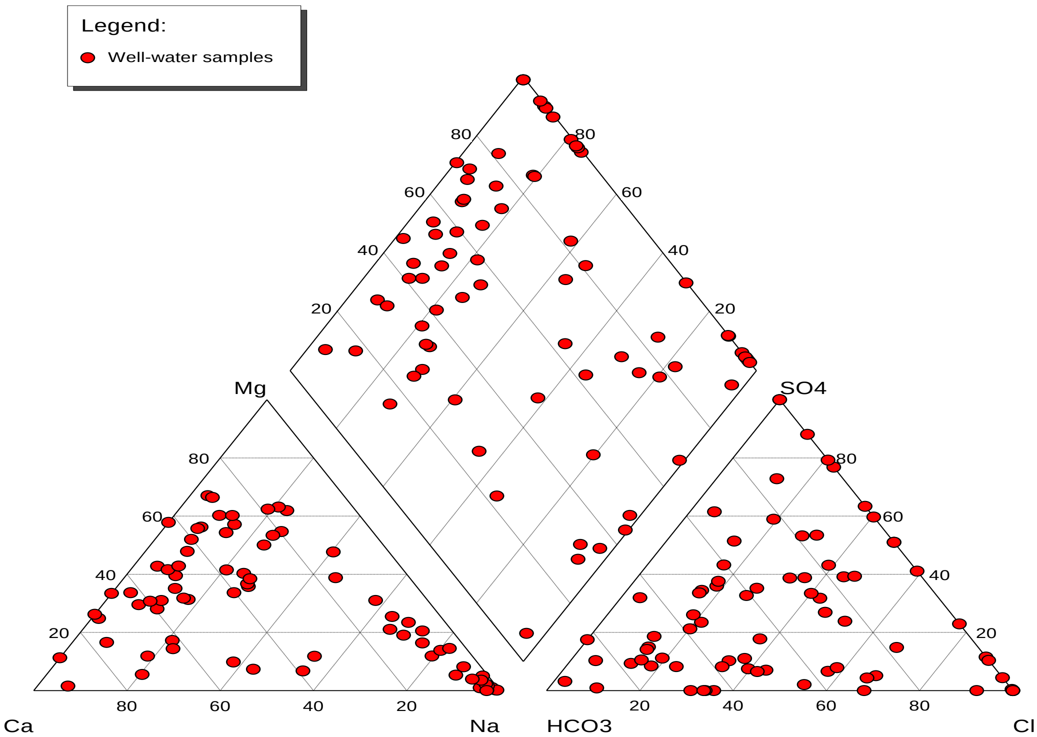

Figure 4: Piper diagram of groundwater in the area

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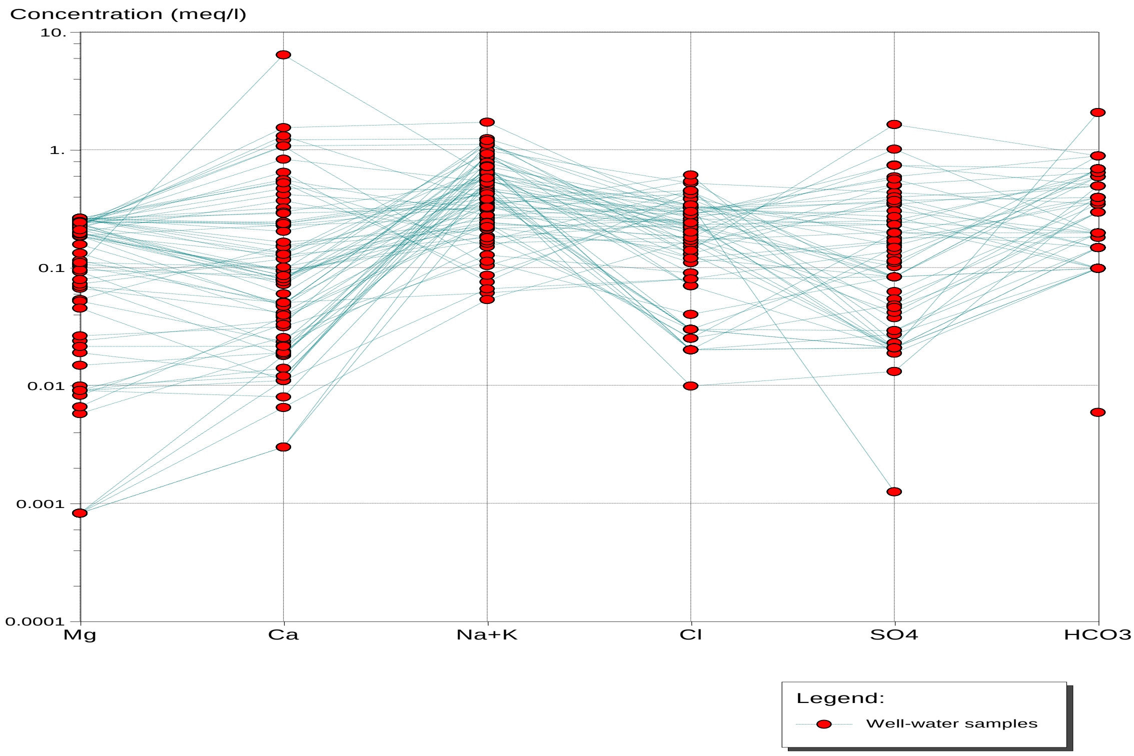

Figure 5 : Schoeller plot of well water samples in the area

Health Implications of Fluoride in Groundwater

Fluoride has beneficial and negative impact on human health (WHO, 2014). Small concentration of fluoride is essential for normal mineralization of bones and formation of dental enamel (Fung *et al.*, 1999; Shomar *et al.*, 2004). However, excessive fluoride intake above 1.5mg/l causes dental enamel to lose its lustre into either mild form of dental fluorosis, which is characterized by white, opaque areas on tooth surface, or into severe form of dental flourosis, which is characterized by yellowish brown to black stains on the teeth (Chaobasia and Sampura, 1996). Excessive fluoride intake may also result in slow, progressive crippling scourge known as skeletal fluorosis.

Fluorosis is a disease caused by deposition of fluorides in the hard and soft tissues of the body. It is usually characterized by discoloration of tooth and crippling disorders (UNICEF, 1999). Crippling skeletal fluorosis, which is associated with the higher levels of exposure, can result from osteosclerosis, ligamentous and tendinous calcification and extreme bone deformity. Dental fluorosis is the damage of tooth enamel, which is caused by the long-time consumption of water with high fluoride content during period of development of tooth (WHO, 2004). Hence, older children and adults are not at risk for dental fluorosis. Teeth impacted by fluorosis have visible discolouration from white spots to brown and black stains. Dental fluorosis has been classified in a number of ways. The most accepted classifications of dental fluorosis are based on the level of tooth damage hence dental fluorosis can be classified into: very mild, moderate and severe categories (Chaturvedi, *et al*., 1990).

High concentration of fluoride in groundwater causes a disease known fluorosis which affects mainly the tooth and bones of animals/man plate 1 & 2. This study revealed that the enrichment factor of the ions on the groundwater is possibly geogenic and related to the local geology of the area. The alkaline pH and high bicarbonate are responsible for release of fluoride-bearing minerals into groundwater. The arid climate of the region, the granitic rocks and the low freshwater exchange due to periodical drought conditions are the factors responsible for the higher incidence of fluorides in the groundwater resources. Apart from these prevailing natural conditions, years of neglect and lack of restoration programs on terrestrial and aquatic environments have led to accumulative impacts on groundwater, soils, plants, and animals including humans.

The people dependent on these groundwater resources are prone to dental fluorosis and mild skeletal fluorosis. Table 3 shows the concentration of fluoride and the corresponding health implications. Fluoride is both beneficial in strong tooth and bone formation (<1.5mg/l) and also damage both the tooth and bone that it helped in their formation if the concentration is > 1.5mg/l (Table 3). Fluoride when consumed in inadequate quantities (<0.5mg/l) causes health problems like dental caries, lack of formation of dental enamel and deficiency of mineralization of bones, especially among the children (Fluhler *et al*., 1982; FAN, 2001). Also fluoride when consumed in excess (>1.5mg.l), leads to several health complications such as skeletal and dental changes/metabolic processes are progressively affected negatively (plates 1 & 2).

The difficulties in diagnosing the early stages of fluorosis from the arthritic symptoms results to damage before detectable bone changes are evident (Wagner *et al.*, 1993). The early symptoms are usually misdiagnosed as rheumatoid or osteoarthritis due to similarity in symptoms (Susheela, 1999). The misdiagnosis of possible skeletal fluorosis, leads to severe disability and damage to the brain resulting in low intelligent quotient (Bassin *et al*., 2006).

Table 2: Degree of Dental Fluorosis (After Dean, 1934)

Index	Tooth Damage
i	Very mild fluorosis: This is small opaque paper white areas scattered irregularly over the tooth and not involving up to 23% of tooth surface
ii	Mild fluorosis: The white opaque surface of the enamel is affected up to 50% of tooth surface
Iii	Moderate fluorosis: This affect all the enamel of the tooth surface, and the surface shows wears, brown stains as distinguishing feature
Iv	Severe fluorosis: This affect all the enamel and hyperplasia, the general form of the tooth shows wide brown stains and corroded appearance

Table 4: Fluoride Content in Drinking Water and the Corresponding Health Impact (Chaturvedi *et al*., 1990)

Fluoride Content (mg/l)	Level of Impact on Human Health
<1	Safe limit, beneficial in tooth and bone formation
1< 3	Dental fluorosis
3 ≤4	Stiff and Brittle bones
>4	Deformation and crippling of bone, permanent and irreversible paralysis

(a) (b)

Plate 1: Dental Fluorosis in the southern part of the study area (Garni).

(a) (b)

Plate II: Dental Fluorosis in Jama'ar Malam Sani area Dental fluorosis observed in Dishe Area

DISCUSSION

Results of study revealed that the groundwater in the area is characterized by relatively low pH, high electrical conductivity, total coliform count, faecal coli count, bicarbonate, fluoride, calcium, manganese, iron, chromium, lead, zinc and sulphate. There is a corresponding wide range and high variance and deviation for those parameters with high values. This implies that the quality of groundwater in the area has substantial temporal and spatial variation.

The concentration of total coliform count (TCC) ranged between 0.0 - 24.0 cfu/100ml with a mean value of 19.27 cfu/100ml and 0.0 - 23.00 cfu/100ml with an average value of 5.25 cfu/100ml. Similarly, faecal coliform count (FCC) varied from 0.0-94.0 cfu/ml with a mean value of 6.87 cfu/ml. The maximum allowable limit is 0.0cfu/ml to 10.0cfu/100ml (NSDWQ, 2007; WHO, 2006) were exceeded at the following locations: Dishe, Dasha, Gidan Bili, Gingimi, Rahamawar Malamai, Garba Bature, Rogogo Cidari, Kwanar Danyada, Giyawa, Baba Daga, Yakubawa, Gwanja, Daqum, Kasuwayal, Bulungudu, Walawa, Arautaki and Zango, all of whose samples come from open dug wells with the exception of Giyawa and Walawa whose samples come from boreholes. The presence of TCC and FCC in water is a clear indication of groundwater contamination by human or animal faeces. Faecal contamination of groundwater is responsible for most water borne diseases such as cholera, typhoid, meningitis and diarrhea (Amadi, 2009; Egharevba *et al*., 2010; Amadi *et al*., 2013).

Poor sanitary situation of an area such as close proximity of unlined soakaway/pit-latrine can introduce TCC and FCC into the shallow aquifer via infiltration.

Fluoride content in groundwater of the area ranged between 0.10 to 3.16mg/l with a mean value of 0.94mg/l. The study revealed that about 75% of the groundwater samples in the area that are suitable for human consumption fall within the sedimentary rocks of the Gundumi and Chad Formations as well as the Basement Complex rocks while 25% of the samples with high fluoride concentration above the permissible limit of 1.5mg/l are from the Younger Granites Suites. Fluorite is a hydrothermal mineral in granite and due to its fast dissolution Kinetics and it is probably the source of fluoride in the groundwater in the area. The fluoride concentration map (Fig. 3) developed for the study was in agreement within the interpretation as the area dominated with sedimentary formations show low concentration of fluoride while region occupy by younger granites high fluoride concentration. This implies that fluoride-rich groundwater in the area emanates from the granite aquifers and the problem of fluorosis in the area is purely by natural processes.

This study has established the problem of fluorosis in the groundwater in the area. The high fluoride concentration of the area was attributed to weathering and dissolution of the fluoride rich mineral host rock. Communities living in the granite/rhyolite dominated region where cases of fluorosis have been observed should discontinue the use of groundwater from the area for drinking purposes. There is need for regular groundwater quality monitoring, mainly of fluoride content in the area. It is also very necessary to promote joint action between health ministries and water managers in order to make health an important issue in water administration in the area. Meanwhile, the Federal and State governments should urgently provide an alternative source of drinking water for the people living in the basement complex area of the Local Government Area.

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