

## Seasonal fluctuations in groundwater fluoride concentration and health risk assessment: Tolon District, Northern Ghana

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**Abstract:** Groundwater contamination with fluoride is a major global issue, contributing to 65% of endemic fluorosis cases worldwide. This study evaluates the seasonal variations in groundwater fluoride concentrations and their connection to health risks in the Tolon District, Ghana. A total of ninety-seven groundwater samples were examined over two seasons to assess fluoride ( $F^-$ ) concentrations. Levels of  $F^-$  varied from undetectable levels to  $1.30 \text{ mg}\cdot\text{dm}^{-3}$  and had an average of  $0.24 \text{ mg}\cdot\text{dm}^{-3}$  during the rainy season. In the dry season, it varied from undetectable levels to  $2.08 \text{ mg}\cdot\text{dm}^{-3}$  and had an average of  $0.36 \text{ mg}\cdot\text{dm}^{-3}$ . Significant spatial and temporal variations were observed, with lower fluoride levels in the northern part of the area and higher levels in the southern region during both seasons. Approximately 84% and 74% of samples fell into group I (optimal for dental health), and 16% and 21% into group II (moderate risk of dental fluorosis) during rainy and dry seasons, respectively. Additionally, 5% of the samples during the dry season fell into group III (high risk of dental fluorosis). Hazard quotient ( $HQ$ ) values for fluoride varied widely, with higher risks observed in children compared to adults during both seasons. This study highlights that children in the Tolon district face greater risks of fluorosis than adults, emphasising the need for targeted mitigation strategies. The research contributes significantly to addressing the pressing global issue of water quality and public health, offering insights that can guide both immediate interventions and long-term sustainability efforts in affected regions.

**Keywords:** fluoride contamination, groundwater, health risk, spatio-temporal variation, Tolon district

### INTRODUCTION

The impact of environmental and groundwater pollution on human well-being is a significant concern, especially in semiarid and arid regions worldwide (Li, 2016; Adimalla and Venkatayogi, 2018). Poor drinking water quality contributes to nearly 80% of illnesses worldwide. Fluoride concentrations alone are responsible for 65% of epidemic fluoride exposure cases worldwide (Felsenfeld and Roberts, 1991).

Approximately 200 mln individuals globally are estimated to ingest water containing F levels that exceed the allowable limits set by the WHO. This situation poses substantial health risks, particularly in Africa, where tens of millions are vulnerable (Edmunds and Smedley, 2012). While low concentrations of  $F^-$  in

potable water can strengthen tooth enamel and lower dental caries, prolonged contact with high levels of  $F^-$  might lead to several medical issues, including dental and skeletal fluorosis, neurological and renal diseases, and myopathy, none of which currently have a proven cure (Adimalla and Venkatayogi, 2017; Narsimha and Rajitha, 2018). Many people in Ghana, especially those living in rural regions, do not have access to safely managed water supplies. Aquifers often serve as their main source of domestic water, yet they often contain elevated levels of fluoride, increasing the incidence of dental fluorosis. Young children, particularly those under two years old, are especially vulnerable to fluoride's detrimental effects, retaining 80–90% of ingested  $F^-$  compared to 60% in adults. Approximately 15% of Ghana's territory, primarily in the northeast, faces an elevated risk of  $F^-$

contamination, affecting an estimated 920,000 individuals, constituting about 3% of the national population. Within these high-risk areas, an estimated 240,000 children aged 0–9 years are particularly susceptible. Zango *et al.* (2019) and Araya *et al.* (2022) indicate that  $F^-$  concentrations in groundwater ranging between 0.05 to 13.29  $mg \cdot dm^{-3}$ , with an average value of 3.26  $mg \cdot dm^{-3}$ , significantly exceeding guideline of WHO's acceptable limits of 1.5  $mg \cdot dm^{-3}$  in the northeast region of Ghana.

The composition of host rocks, climate, recharge water mixing, geochemical settings, aquifer properties, and hydrogeological dynamics are some of the factors that affect the natural occurrence of  $F^-$  in the aquifers (Apambire, Boyle and Michel, 1997; Ravindra and Garg, 2006; Subba Rao, Subrahmanyam and Babu Rao, 2013; Li, Wu and Qian, 2016; Zango *et al.*, 2019). Fluoride concentrations in groundwater can rise due to alteration and preferential dissolution of fluoride-bearing minerals during prolonged water-rock interactions across various rock types (Chae *et al.*, 2007; Li *et al.*, 2016; Sunkari *et al.*, 2019). Calvi *et al.* (2016) identified fluorite ( $CaF_2$ ) and fluoridated hydroxyapatite [ $Ca_5(PO_4)3F$ ] as the main sources of fluoride in sedimentary aquifers. These minerals are typically found alongside clayey rocks and are naturally present in micas. However, the contribution of these minerals to fluoride levels in groundwater can vary, and other sources may also play a significant role in rising fluoride concentrations. Minerals such as calcite can reduce the reactivity of calcium through chemical weathering processes in areas with high transpiration and evaporation rates. Additionally, recent studies have demonstrated that human activity contributes to rising groundwater fluoride concentrations. According to Kim, Kim and Kim (2011), these activities include the use of phosphate

group fertilisers, pesticide storage degradation, indiscriminate sewage dumping, irrigation with high-fluoride waters, and groundwater table depletion. The Tolon District is predominantly engaged in agricultural activities, cultivating maize, pepper, yam, rice, peanut, and cowpea. The district's land is highly suitable for maize and pepper, but less so for cowpea. Smallholder farmers in the district utilise mineral fertilisers, especially for crops like soybeans. The adoption of these fertilisers and other soil fertility management practices is influenced by factors such as farm size, access to extension services, and herd size (GSS, 2010; Nakasone, Ghimire, and Suvedi, 2021). The study attempts to assess the spatio-temporal variations of  $F^-$  levels in underground water in the Tolon District, Northern Region of Ghana. It employs the inverse distance weighted geospatial method to map the distribution of fluoride. Additionally, the study aims to evaluate the potential health hazards associated with fluoride exposure by calculating Hazard Quotients ( $HQs$ ) for adults and children, following the methodology established by USEPA.

## MATERIALS AND METHODS

### STUDY AREA

The research was carried out in the Tolon District, Northern Region of Ghana, spanning approximately 1,355  $km^2$  between latitude coordinates  $9^{\circ}15'$  and  $10^{\circ}02'N$ , and longitude coordinates  $0^{\circ}53'$  and  $1^{\circ}25' W$ . It is bordered by Sagnarigu District to the east, Gonja to the west, Kumbungu to the north, and Central Gonja to the south (Fig. 1). The district's population is 118,090,

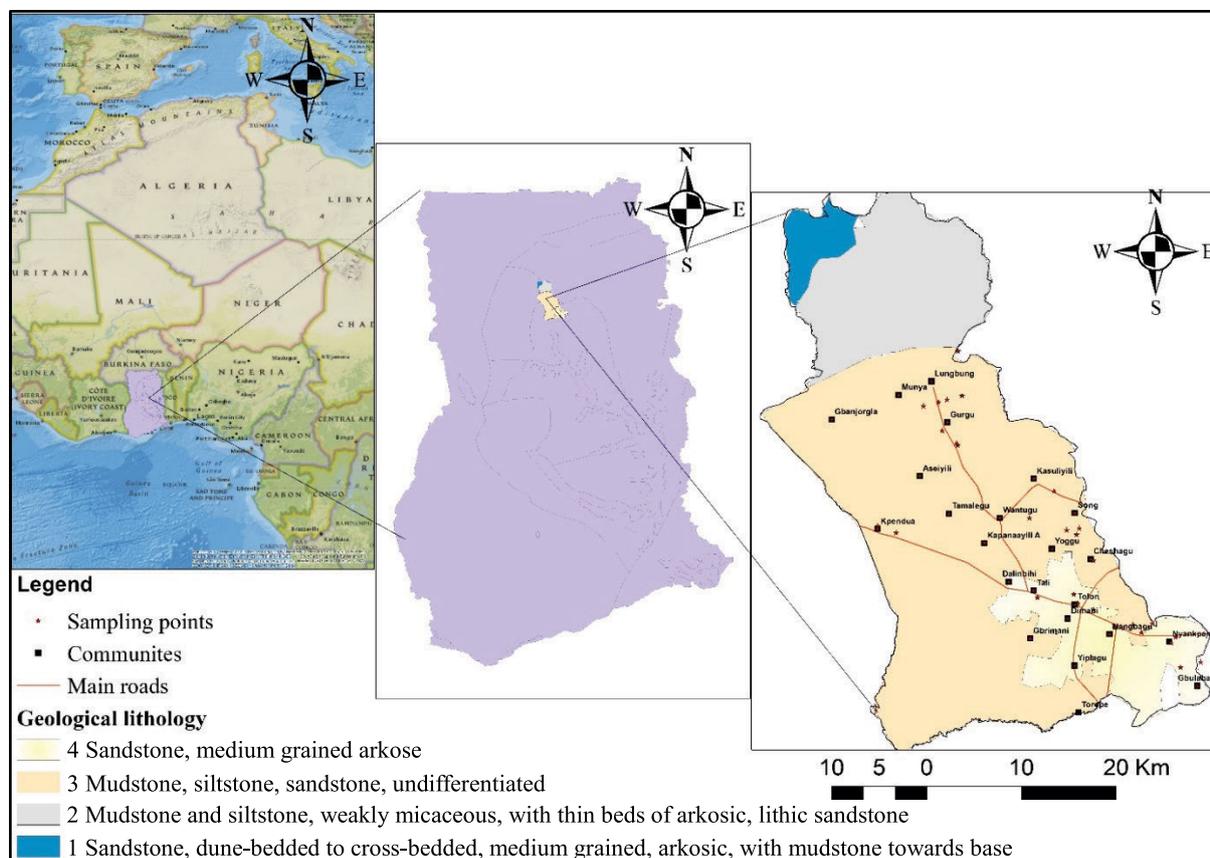


Fig. 1. Geological and study area map; source of geological map; own elaboration based on Jordan *et al.* (2009)

predominantly engaged in agriculture (StatsBank, 2021). Its topography is undulating, ranging between 94 and 187 m, characterised by several rivers that aid drainage and sporadic depressions (Salifu, 2013; Abdul-Ganiyu *et al.*, 2017).

Geologically, the area is situated within the Voltaian Supergroup, a complex assemblage of ancient sedimentary rocks dating from the Neoproterozoic to the early Paleozoic era. These rocks comprise silty mudstones, sandstones, limestones, conglomerates, and glacial deposits (SNC-Lavalin/INRS, 2011). The Voltaian Supergroup covers a significant portion of Ghana and extends into neighbouring countries such as Togo, Burkina Faso, and Niger (Trompette, 1994; Affaton, Sougy and Trompette, 1980). The geological formations in the Tolon District can be categorised into four main units (Jordan *et al.*, 2009) as shown in Figure 1.

1. Sandstone, dune-bedded to cross-bedded, medium grained, arkosic, with mudstone towards base – consists of diverse lithologies including argillaceous and strata of micaceous at the base transitioning into medium-grained sandstones. Evidence suggests deposition in braided river systems and Aeolian dunes.
2. Mudstone and siltstone, micaceous, with beds of arkosic, lithic sandstone – predominantly located in the northern part, this unit is characterised by green-grey mudstones, siltstones, and feldspathic, lithics-rich sandstones. It shows distinct compositional variations and is identified through gamma spectrometry.
3. Mudstone, siltstone, sandstone, undifferentiated – the youngest in the Volta Basin, this formation includes flysch-type beds in the north and molasse-type sandstones in the south. It exhibits variegated shallow-water deposits with occasional sub-aerial exposure, characterised by calcareous mudstones, siltstones, and conglomerates.
4. Sandstone, medium grained arkose – found prominently in Tamale, this formation represents fluvial red-bed deposits from the Pan-African orogeny. It features medium-grained arkosic sandstones with cross-bedding, indicative of south-southeast currents, alongside pink-weathering laminated sandstones.

## GROUNDWATER SAMPLING AND ANALYSIS PROCEDURES

Groundwater sampling and analysis were conducted over the rainy season (August–September 2023) and the dry season (March 2024). The samples (97) were collected from operational wells in the rainy (55 samples) and dry (42 samples) seasons. Boreholes typically vary in depth from 50 m to 100 m, with a success rate of approximately 50%. Groundwater recharge is mainly driven by precipitation and runoff, affecting the flow of groundwater from highlands to valleys and rivers. Seasonal fluctuations in groundwater levels are common, with lower levels observed during the dry season and higher levels in the wet season (SNC-Lavalin/INRS, 2011). However, the samples for this study were collected from depths ranging between 5 and 63 m. The sampling sites were mapped using GPS (maps.me), as shown in Figure 1. Before sampling, wells were purged for 5–10 min to stabilise physicochemical parameters. Pre-cleaned 1-dm<sup>3</sup> polyethylene bottles labelled with masking tape were used for sampling, stored at 4°C, and transported to the West African Center for Water, Irrigation and Sustainable Agriculture

(WAWISA) and the Water Research Institute (WRI) laboratories. The sampling protocol established by Weaver, Cave, and Talma (2007) was followed to ensure accuracy and consistency.

Eleven parameters were analysed in this study. Physicochemical parameters, including total dissolved solids (TDS), potential of hydrogen (pH), and electrical conductivity (EC), were measured using a handheld water quality meter LAQUA WQ-330 (HORIBA Advanced Techno, Co., Ltd., Kyoto, Japan) probe calibrated with standard solutions. Cations such as K<sup>+</sup>, Na<sup>+</sup>, and Ca<sup>2+</sup> were detected at the WACWISA laboratory using an FP 910-5 flame photometer (PG Instruments Ltd, Lutterworth, United Kingdom). Anions like Cl<sup>-</sup>, F<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and HCO<sub>3</sub><sup>-</sup> were analysed at the Water Research Institute (WRI) laboratory using a Janeway 6305 spectrophotometer (Janeway Laboratory equipment supplier, Felsted, United Kingdom) for sulphate (SO<sub>4</sub><sup>2-</sup>) and fluoride (F<sup>-</sup>); the alkalinity strong acid titration method for bicarbonate (HCO<sub>3</sub><sup>-</sup>); the argentometric titration method for chloride (Cl<sup>-</sup>); and the EDTA titration method for magnesium (Mg<sup>2+</sup>). During the dry season, analyses were conducted at the WACWISA laboratory using a DR6000 spectrophotometer (HACH, London, United Kingdom) for SO<sub>4</sub><sup>2-</sup> and fluoride (F<sup>-</sup>), and titration methods for Mg<sup>2+</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and HCO<sub>3</sub><sup>-</sup>.

The ionic balance error (IBE) was used to verify the analysis accuracy. The sum of anions and cations, expressed as meq·dm<sup>-3</sup>, must balance in potable water as it is electrically neutral. A ±5% charge balance error is generally acceptable, indicating a good balance of cations and anions in the parameter analysis. The test is based on the percentage difference (Eq. 1). All groundwater samples in the study fell within the IBE's ±5% tolerance (Friedman and Erdmann, 1982; Lipps, Braun-Howland, and Baxter, 2022).

$$IBE = \frac{\sum \text{Cations} - \sum \text{Anions}}{\sum \text{Cations} + \sum \text{Anions}} \times 100 \quad (1)$$

## GEOSTATISTICAL INTERPOLATION TECHNIQUE

The study applied the inverse distance weighting (IDW) geospatial method to create distribution maps based on fluoride concentrations derived from groundwater samples. Using ArcGIS software, spatial and chronic risk assessment maps of fluoride were generated through IDW interpolation. The IDW, a geostatistical method, estimates attribute values at unsampled locations by weighing values at sampled points inversely to their distance from the point of interest (Isaaks and Srivastava, 1989). The primary goal of geospatial analysis was to visualise concentration variations across different areas, revealing crucial insights for exploration purposes. Geostatistical interpolation models like IDW are vital in natural resource management and biological conservation efforts. The growing need for continuous spatial data on environmental factors underscores the significance of such tools (Li and Heap, 2008).

## HEALTH RISK ASSESSMENT OF FLUORIDE

Assessing health hazards entails determining the probability that exposure to polluting elements will result in negative health impacts over a certain time frame. This assessment quantifies the risk levels associated with non-carcinogenic health effects

(Bortey-Sam *et al.*, 2015; Tirkey *et al.*, 2017). The U.S. EPA Risk Assessment method was utilised to evaluate the health hazard posed by F<sup>-</sup> in groundwater. Chronic daily intake (CDI) estimates the quantity, rate, and period of exposure to pollutant metals (Tirkey *et al.*, 2017). To assess the non-carcinogenic risks of F<sup>-</sup>, Equations (2) and (3) were applied (Anim-Gyampo *et al.*, 2019; Yahaya *et al.*, 2020; Zakir *et al.*, 2020; Niknejad *et al.*, 2023):

$$CDI = \frac{F_c \cdot IR \cdot EF \cdot ED}{BW \cdot AT} \quad (2)$$

$$HQ = \frac{CDI}{RfD} \quad (3)$$

where:  $F_c$  = level of fluoride concentration (mg·dm<sup>-3</sup>),  $IR$  = drinking water consumption per capita (dm<sup>3</sup>·day<sup>-1</sup>),  $EF$  = frequency of exposure (day·year<sup>-1</sup>),  $ED$  = period of exposure (year),  $BW$  = weight of the total body (kg per person),  $AT$  = average

length of time (days) – the calculation of this parameter for non-carcinogenic risk is determined by multiplying 365 by the exposure duration according to Mesdaghinia, Nasseri and Hand (2016),  $HQ$  = hazards quotient, and  $CDI$  = long-term daily exposure (mg·kg<sup>-1</sup>·day<sup>-1</sup>),  $RfD$  = the dose of reference (their values and other assumptions related to F-exposure through the consumption of water are detailed in Table 1).

The results of  $HQ$  were compared with the assessment of the chronic-risk scales outlined by USEPA (1999a).

## RESULTS AND DISCUSSION

### GENERAL PHYSICO-CHEMICAL OF GROUNDWATER

A statistical overview of the physico-chemical characteristics of groundwater is presented in Table 2. According to international (WHO, 2022) and Indian Bureau (IS 10500: 2012) guidelines, the

**Table 1.** Human health risk factors and their values

Exposure factor	Unit	Value for		Reference
		adults	children	
$F_c$	mg·dm <sup>-3</sup>	concentration of F <sup>-</sup> in groundwater		current study
$IR$	dm <sup>3</sup> ·day <sup>-1</sup>	2.2	1.8	Anim-Gyampo <i>et al.</i> (2019)
$EF$	day·year <sup>-1</sup>	365	365	USEPA (2004)
$ED$	year	65.5 <sup>1)</sup>	12	WHO (2013)
$BW$	kg	60	10	WHO (2022)
$AT$ for non-carcinogens	day	23,907.5 <sup>1)</sup>	4,380	WHO (2013), Mesdaghinia, Nasseri and Hadi (2016)
$RfD$	mg·kg <sup>-1</sup> ·day <sup>-1</sup>	F <sup>-</sup> = 0.06		USEPA (2005), USEPA (2014)

<sup>1)</sup> Mean for adult males and females.

Explanations:  $F_c$  = level of fluoride concentration,  $IR$  = drinking water consumption per capita,  $EF$  = frequency of exposure,  $ED$  = period of exposure,  $BW$  = weight of the total body,  $AT$  = average length of time,  $RfD$  = dose of reference.

Source: own elaboration based on literature.

**Table 2.** Statistical overview of physico-chemical characteristics in groundwater

Parameter	Unit	Values for rainy season (n = 55)				Values for dry season (n = 42)			
		mean	SD	min.	max.	mean	SD	min.	max.
pH	-	7.39	0.64	5.91	9.36	7.56	0.33	6.74	8.32
EC	μS·cm <sup>-1</sup>	891.18	628.32	24.00	2,580.00	1,614.75	3,872.23	30.4	25,700.00
TDS	mg·dm <sup>-3</sup>	446.16	314.63	12.04	1,290.00	932.16	2,079.94	14.90	12,870.00
Ca <sup>2+</sup>		7.03	4.68	0.00	16.00	49.40	150.19	4.50	960.00
Na <sup>+</sup>		96.20	64.41	1.00	291.00	163.50	156.22	2.00	853.00
Mg <sup>2+</sup>		23.24	29.62	1.27	157.00	47.65	60.59	1.20	321.50
K <sup>+</sup>		12.41	17.46	0.10	67.00	3.78	2.45	1.00	12.90
HCO <sub>3</sub> <sup>-</sup>		242.95	197.92	12.20	805.00	403.80	267.85	12.80	1,003.70
Cl <sup>-</sup>		52.74	71.89	4.00	424.00	425.02	1,336.22	9.70	6,277.00
F <sup>-</sup>		0.24	0.38	0.00	1.30	0.36	0.46	0.00	2.080
SO <sub>4</sub> <sup>2-</sup>		93.98	106.92	8.70	587.00	26.11	36.50	0.00	136.00

Explanations:  $SD$  = standard deviation,  $pH$  = potential hydrogen,  $EC$  = electrical conductivity,  $TDS$  = total dissolved solids.

Source: own study.

pH of drinking water should range between 6.5 and 8.5. The mean pH of the well samples taken during the rainy season was 7.39 (from 5.91 to 9.36). The mean pH during the dry season was 7.56 (from 6.74 to 8.32). In the rainy season, *EC* ranged between 24 and 2580  $\mu\text{S}\cdot\text{cm}^{-1}$  (mean = 891.18), and in the dry season, between 30.4 and 25,700  $\mu\text{S}\cdot\text{cm}^{-1}$  (mean = 1,614.75). According to the World Health Organization (WHO), *EC* in drinking water should not exceed 1500  $\mu\text{S}\cdot\text{cm}^{-1}$  (WHO, 2011). Moderate to high salt levels were found in around 18% and 24% of groundwater samples that surpassed this threshold during the wet and dry seasons, respectively.

Similarly, high total dissolved solids (TDS) values imply potential contamination and health risks depending on specific pollutants present. For instance, the highest permissible level for calcium ( $\text{Ca}^{2+}$ ) in potable water has been set by the WHO at 100  $\text{mg}\cdot\text{dm}^{-3}$ . In the study area, concentrations ranged widely, from below detection to 16 (mean = 7.03)  $\text{mg}\cdot\text{dm}^{-3}$  during the rainy season, and 4.5 to 960.0 (mean = 49.4)  $\text{mg}\cdot\text{dm}^{-3}$  in the dry season (Tab. 2). All samples fell under the permissible range during the rainy season, but three samples (7%) exceeded the permissible level in the dry season. Magnesium ( $\text{Mg}^{2+}$ ) levels varied from 1.27 to 157.00  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 23.24  $\text{mg}\cdot\text{dm}^{-3}$ ) during the rainy season and from 1.20 to 321.50  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 47.65  $\text{mg}\cdot\text{dm}^{-3}$ ) during the dry season. Most samples exceeded the WHO level of 50  $\text{mg}\cdot\text{dm}^{-3}$  during both seasons, potentially impacting taste and scaling in water supply systems (Ramesh and Elango, 2011; IS 10500: 2012). Sodium ( $\text{Na}^+$ ) levels varied between 1 and 291  $\text{mg}\cdot\text{dm}^{-3}$  and from 2 to 853  $\text{mg}\cdot\text{dm}^{-3}$ , in the two seasons respectively. Potassium ( $\text{K}^+$ ) concentrations varied from 0.1 to 67.0  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 12.4  $\text{mg}\cdot\text{dm}^{-3}$ ) during the rainy season, and from 1.0 to 12.9  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 3.78  $\text{mg}\cdot\text{dm}^{-3}$ ) during the dry season (Tab. 2). The WHO guidelines recommend the permitted level of  $\text{K}^+$  in water designated for drinking at 12  $\text{mg}\cdot\text{dm}^{-3}$ . During the rainy season, 16 samples (29%) exceeded this level, and all samples except one exceeded it during the dry period. The level of bicarbonate ( $\text{HCO}_3^-$ ) was 12.20–805.00  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 242.95  $\text{mg}\cdot\text{dm}^{-3}$ ) and 12.80–1003.70  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 403.80  $\text{mg}\cdot\text{dm}^{-3}$ ) during the rainy and dry seasons, respectively. Chloride ( $\text{Cl}^-$ ) levels ranged 4.00–424.00  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 52.74  $\text{mg}\cdot\text{dm}^{-3}$ ) and 9.70–6277.00  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 425.02  $\text{mg}\cdot\text{dm}^{-3}$ ) in respective seasons (Tab. 2). The WHO standard for  $\text{Cl}^-$  in potable water is 250  $\text{mg}\cdot\text{dm}^{-3}$ , and 96% and 95% of samples not exceeded this threshold in the rainy and dry periods, respectively. Fluoride ( $\text{F}^-$ ) concentrations ranged from below detectable levels to 1.30  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 0.24  $\text{mg}\cdot\text{dm}^{-3}$ ) during the rainy period, and from below detectable levels to 2.08  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 0.36  $\text{mg}\cdot\text{dm}^{-3}$ ) during the dry season (Tab. 2). Sulphate ( $\text{SO}_4^{2-}$ ) concentrations ranged from 8.70 to 587.00  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 93.98  $\text{mg}\cdot\text{dm}^{-3}$ ) during the rainy period, and from below detectable levels to 136.00  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 26.11  $\text{mg}\cdot\text{dm}^{-3}$ ) during the dry period. All groundwater samples in the study region showed  $\text{SO}_4^{2-}$  levels under the WHO allowable level of 250  $\text{mg}\cdot\text{dm}^{-3}$ , except for three samples. There seems to be a potential relationship between the depth of the borehole and the fluoride concentrations in the groundwater. Specifically, it appears that boreholes with depths between 52 m and 61 m have higher fluoride concentrations. Generally, deeper groundwater tends to have higher mineral content due to longer contact with geological formations.

### SPATIO-TEMPORAL VARIATION OF FLUORIDE CONCENTRATION

To highlight the spatio-temporal variation and assess  $\text{F}^-$  levels in groundwater across the research region, fluoride levels were interpolated using the IDW geospatial method, complemented by the geometrical interval method for the rainy and dry seasons (Figs. 2, 3). Groundwater fluoride levels tend to be elevated, especially in regions where fluorite-bearing rocks undergo continuous water-rock interactions, exacerbated by arid climates, low precipitation, and high temperatures (Machender, Dhakate, and Narsmha Reddy, 2014).

Geological processes are the primary natural source of fluoride ( $\text{F}^-$ ) in groundwater, with typical concentrations ranging from 0.01 to 7.20  $\text{mg}\cdot\text{dm}^{-3}$ . In the rainy season, fluoride levels ranged from below detectable limits to 1.30  $\text{mg}\cdot\text{dm}^{-3}$ , averaging 0.24  $\text{mg}\cdot\text{dm}^{-3}$ . In the dry season, concentrations varied from below detectable levels to 2.08  $\text{mg}\cdot\text{dm}^{-3}$ , averaging 0.36  $\text{mg}\cdot\text{dm}^{-3}$  (Tab. 2). This study illustrates significant spatial and temporal variations in fluoride ( $\text{F}^-$ ) values, with lower values observed in the northern part of the region during both seasons, and higher values in the southern part, particularly in areas such as Woribogu Kuku, WACWISA, Gbulahagu, and Yiplagu (Fig. 3). Therefore, certain bicarbonate and sodium minerals may contribute to elevated fluoride concentrations in groundwater. When calcium-rich groundwater transitions to sodium-rich groundwater, the likelihood of fluoride dissolution increases (Ahmad *et al.*, 2022).

Fluoride levels rise notably in medium-grained arkosic sandstones (Figs. 1, 2). Additionally, box-whisker plots of fluoride concentration (Fig. 2) were applied to analyse seasonal variations, highlighting that the rainy season exhibits greater differences in fluoride levels between median and maximum concentrations. In contrast, observed differences in concentrations are generally elevated in the dry period. The increased presence of major ions in the dry period is likely due to enhanced leaching from soils and rocks, driven by reduced water availability and intensified evaporation. The dissolution of fluoride-content minerals, such as calcium fluoride ( $\text{CaF}_2$ ) is facilitated by elevated values of pH in groundwater (Chen *et al.*, 2017). The correlation matrix reveals significant associations between fluoride ( $\text{F}^-$ ) and several key ions in the examined samples. Fluoride shows moderate positive correlations with TDS ( $R^2 = 0.58$ ),  $\text{Na}^+$  ( $R^2 = 0.64$ ),  $\text{Cl}^-$  ( $R^2 = 0.50$ ), and  $\text{SO}_4^{2-}$  ( $R^2 = 0.59$ ). These findings indicate that increases in TDS,  $\text{Na}^+$ ,  $\text{Cl}^-$ , and  $\text{SO}_4^{2-}$  are accompanied by higher fluoride concentrations, while decreases in these ions correspond with lower

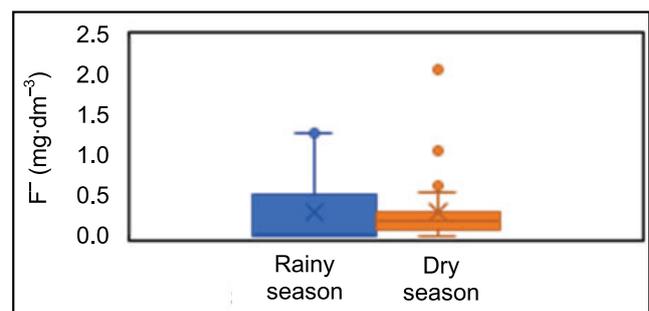
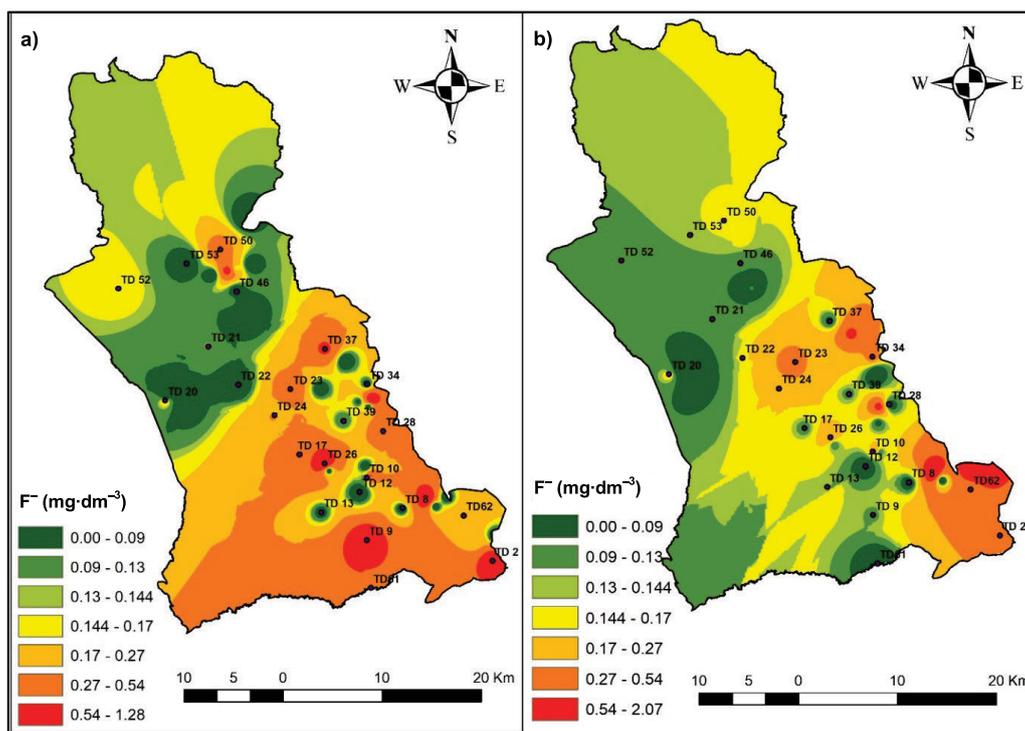


Fig. 2. Box-whisker graphs of fluoride concentration ( $\text{F}^-$ ); source: own study



**Fig. 3.** Spatial distribution of  $F^-$  in the Tolon district in: a) rainy season, b) dry season; TD1–TD53 = sampling points; source: own study

fluoride levels. These correlations underscore potential co-occurrences or interactions between fluoride and these ions, highlighting the interconnected nature of their presence and concentrations in the analysed samples.

#### HEALTH RISK ASSESSMENT OF $F^-$ INGESTION

Fluoride levels in natural sources of water typically range from 0.01 to 7.20  $\text{mg}\cdot\text{dm}^{-3}$ . Prolonged contact with levels of fluoride greater than 1.5  $\text{mg}\cdot\text{dm}^{-3}$  can result in fluorosis (IS 10500: 2012; WHO, 2022). In our research,  $F^-$  concentrations ranged from undetectable levels to 1.30  $\text{mg}\cdot\text{dm}^{-3}$  (average = 0.24  $\text{mg}\cdot\text{dm}^{-3}$ ) in the rainy season, and undetectable levels to 2.08  $\text{mg}\cdot\text{dm}^{-3}$  (average = 0.36  $\text{mg}\cdot\text{dm}^{-3}$ ) in the dry season (Tab. 2). These concentrations often fall under the recommended limits for drinking water, except for two samples (TD42-Woribogu Kuku; TD63-WACWISA wells) which exceeded the recommended thresholds. The assessment of health hazards associates with groundwater fluoride levels in the study area is

summarised in Table 3, where the hazard levels are divided into five groups.

Approximately 84% and 74% of the samples fell under group I, and 16% and 21% were in group II during the rainy and dry seasons, respectively; both groups are considered beneficial for promoting strong bones and teeth. Additionally, 5% of the samples collected during the dry season fell into group III, which may cause dental fluorosis (Dissanayake, 1991; WHO, 1996). The  $HQ$  values are commonly used to assess the potential health hazards posed by contaminants to humans by exposure to various media within the environment (Shams *et al.*, 2022). The  $HQ$  values for the noncarcinogenic risk of  $F^-$  consumption from groundwater in the study area are presented in Table S1, which is used to quantify potential human health risks. During the rainy season,  $HQ$  values for  $F^-$  varied from 0.00–0.79  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 0.15  $\text{mg}\cdot\text{dm}^{-3}$ ) for adults, and from 0.00–3.90  $\text{mg}\cdot\text{dm}^{-3}$  (mean = 0.73  $\text{mg}\cdot\text{dm}^{-3}$ ) for children. In the dry season, the  $HQ$  values ranged from 0.00 to 1.27  $\text{mg}\cdot\text{dm}^{-3}$ , with a mean of 0.28  $\text{mg}\cdot\text{dm}^{-3}$ , for adults, and from 0.00 to 6.24  $\text{mg}\cdot\text{dm}^{-3}$ , with a mean

**Table 3.** Health hazards of  $F^-$  ingestion (Dissanayake, 1991; WHO, 1996)

Group	$F^-$ ( $\text{mg}\cdot\text{dm}^{-3}$ )	Impact on human health	Number of samples in	
			rainy season	dry season
I	<0.5	favourable to dental cavities	46 (84%)	31 (74%)
II	0.5–1.5	encourages the growth of healthy teeth and bones	9 (16%)	9 (21%)
III	>1.5–4	dental fluorosis or teeth mottling	0	2 (5%)
IV	>4–10	fluorosis of the teeth and skeleton (pain in the neck and back)	0	0
V	>10	debilitating fluorosis	0	0

Source: own study.

of  $1.37 \text{ mg}\cdot\text{dm}^{-3}$ , for children. These *HQ* values were compared with the chronic hazard assessment scales defined by USEPA (1999a), as presented in Table 4.

According to Table 4, approximately 64%, 49%, 36%, and 21% of the wells for adults and children during the rainy and dry seasons, respectively, fall into the negligible to chronic risk category. Additionally, 36%, 29%, 62%, and 48% of the wells indicate a low risk. For children during the rainy season, 22% of the samples indicate a medium risk, while 2% and 26% of the samples for adults and children during the dry season, respectively, show a medium risk. Notably, 5% of the samples for children during the dry season indicate a high chronic risk. Elevated *HQ* values were observed in the dry season for children, suggesting that increased mineral dissolution during this period

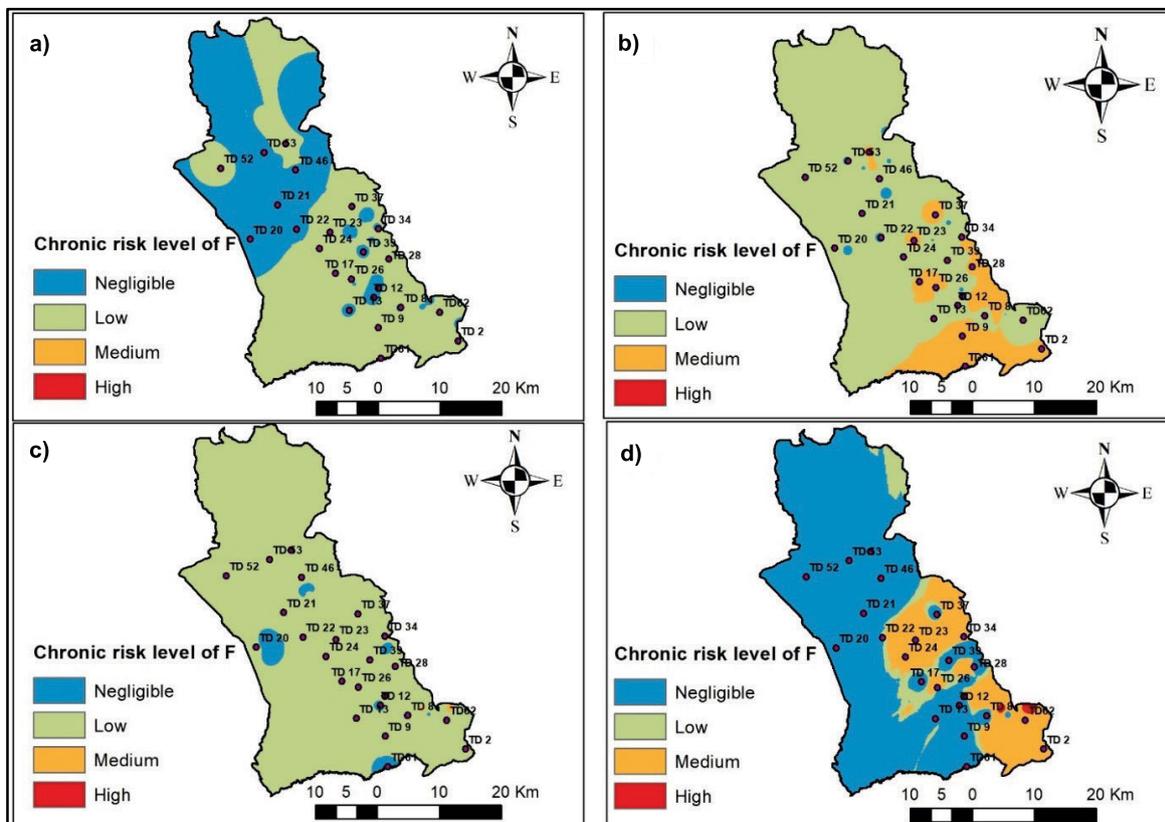
contributes to higher fluoride concentrations in the study area. The consumption of water with high fluoride levels presents health risks of fluorosis to children.

In both the wet and dry seasons, children exhibit a higher chronic risk assessment for fluoride ( $\text{F}^-$ ) exposure than adults, as shown in Figure 4. This suggests that children are more susceptible to fluorosis than adults. Their increase vulnerability stems from lower body weight and greater sensitivity to contaminants in drinking water (Wu *et al.*, 2020; Niknejad *et al.*, 2023). Chen *et al.* (2017) discovered a correlation between fluoride levels in water for drinking and body weight and size. Since residents in the study area mostly drink groundwater with high fluoride levels, they may be at risk for long-term fluoride-related health effects.

**Table 4.** Chronic risk assessment of fluoride in groundwater in the study area

Risk level	Hazard quotient ( <i>HQ</i> )	Chronic risk	Number of samples			
			rainy season		dry season	
			adults	children	adults	children
1	<0.1	negligible	35 (64%)	27 (49%)	15 (36%)	9 (21%)
2	(0.1; 1.0)	low	20 (36%)	16 (29%)	26 (62%)	20 (48%)
3	(1.0; 4.0)	medium	0	12 (22%)	1 (2%)	11 (26%)
4	$\geq 4.0$	high	0	0	0	2 (5%)

Explanations: risk level, *HQ* and chronic risk according to standards of USEPA (1999a). Source: own study.



**Fig. 4.** Chronic risk assessment maps of fluoride ( $\text{F}^-$ ) in groundwater in the study area for: a) adults during the rainy season, b) children during the rainy season, c) adults during the dry season, d) children during the dry season; TD1–TD53 = sampling points; source: own study

## CONCLUSIONS

This study investigated the spatio-temporal variations of fluoride concentrations in groundwater within the Tolon District, Northern Region of Ghana, employing the IDW geospatial method and hazard quotient (*HQ*) analysis according to USEPA guidelines. Significant spatial and temporal variability in fluoride levels was observed, with lower concentrations in the northern region and higher concentrations in the southern area, notably in areas characterised by medium-grained arkoses sandstones and pink-weathering laminated sandstones. Fluoride concentrations ranged from below detectable levels to  $1.30 \text{ mg-dm}^{-3}$  during the rainy season and peaked at  $2.08 \text{ mg-dm}^{-3}$  during the dry season. Health risk assessments based on *HQs* indicated varying degrees of risk, with a substantial portion of samples showing negligible to medium risk. Notably, 5% of the samples collected during the dry season indicated a high chronic risk for children. These findings underscore the significant fluorosis risks posed by elevated fluoride levels in drinking water, particularly among children. To mitigate these risks, regular monitoring of groundwater fluoride levels is essential. Public awareness programs should be intensified to educate communities about the risks associated with excessive fluoride intake and promote preventive measures. Furthermore, targeted interventions, such as water treatment or the provision of alternative water sources, should be implemented in areas where fluoride concentrations exceed safe limits. Future research should focus on expanding the understanding of factors influencing fluoride concentrations in groundwater and include longitudinal studies to monitor trends and the effectiveness of mitigation measures over time.

## SUPPLEMENTARY MATERIAL

Supplementary material to this article can be found online at [https://www.jwld.pl/files/Supplementary\\_material\\_65\\_Nogara.pdf](https://www.jwld.pl/files/Supplementary_material_65_Nogara.pdf).

## CONFLICT OF INTERESTS

All authors declare that they have no conflict of interests.

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