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Fluoride contamination in groundwater sources in Southwestern Nigeria: Assessment using multivariate statistical approach and human health risk

Chidozie PraiseGod Emenike[a,b,*], Imokhai Theophilus Tenebe[a], Peter Jarvis[b]

[a] Department of Civil Engineering, Covenant University, Ota, Ogun State, Nigeria
[b] Cranfield Water Science Institute, School of Water, Energy and Environment, Cranfield University, Bedfordshire MK43 0TE, United Kingdom

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ABSTRACT

The present study investigated the ionic and fluoride concentrations in tap water and its associated health risk to local dwellers of Ogun State (Abeokuta south), Nigeria. 63 samples were collected from twenty-one different locations. Results obtained revealed the mean concentration of fluoride (F^-) as 1.23 mg/L. Other water quality parameters such as total dissolved solids (TDS), electrical conductivity (EC), F^-, Fe^{2+}, and SO_4^{2-} surpassed the WHO guidance for drinking water. Strong positive correlation was observed between F^- and TDS; F^- and pH; TDS and EC; TDS and Mg^{2+}; TDS and SO_4^{2-}; TDS and HCO_3^-; EC and HCO_3^-; EC and SO_4^{2-}; Na^+ and Cl^-; SO_4^{2-} and Cl^-. In addition, Empirical Bayesian Kriging (EBK) model was employed to spatially distribute the concentration of the analyzed elements within the study region. The chronic daily dose (*CDD*) and hazard quotient (*HQ*) were also used to evaluate the health risk associated with F^-, considering dermal and ingestion as pathways. The results revealed that the associated *HQ* for infants between the age range of 6–12months within about 91% of the study region surpassed the accepted *HQ* limit. However, the *HQ* for age categories 11–16years; > 65years; 18–21years; 21years; 16–18years within 95.2%, 90.5%, 80.95% and 100% of the study location were less than 1. Conclusively, the *HQ* values obtained in this study should serve as a baseline information for water management authorities, policymakers and the society at large towards addressing these pollution issues.

1. Introduction

The presence of fluoride at elevated concentrations in drinking water has caused severe health effects in humans in some parts of the world (Bhatnagar et al., 2011; Shen and Schäfer, 2015). Fluorine exists in the environment through combination with other elements to form highly soluble fluoride compounds. The main source of fluorine in water is from natural deposition from geogenic sources in aquifers (EPA, 2010; Sun et al., 2013). The primary way by which humans ingest fluoride is through consumption of contaminated groundwater (Singh et al., 2013; Singh and Mukherjee, 2015; Subba Rao et al., 2013). The World Health Organization (WHO) recommended the concentration of fluoride that can cause minimal health risk to be 1.5 mg/l. However, more than 200 million people living across 20 developing and developed countries regularly consume water with elevated fluoride concentrations above the standard guidelines set by the WHO (Amini et al., 2008; Fawell et al., 2006; Shen and Schäfer, 2015). Some countries such as Tanzania in the East of Africa have a drinking water standard for fluoride of 4 mg/l, well above the WHO recommended value. This can cause a number of possible health problems, including dental and skeletal fluorosis. In the Rift Valley, East Africa, more than 80 million people display a range of symptoms consistent with dental fluorosis (Shen and Schäfer, 2015; Smedley et al., 2002). However, this should also be placed in the context of water scarcity, population growth and access to clean water in the region.

High concentrations of fluoride in humans can lead to various health problems such as nervous system damage (Kaoud and Kalifa, 2010), reduced fertility (Izquierdo-Vega et al., 2008), intellectual impairment in children (Ding et al., 2011; Shivaprakash et al., 2011), urinary tract disease (Jha et al., 2011), as well as dental and skeletal fluorosis in children and adults (Maguire, 2014). In turn, this can lead to significant lower back pains (Namkaew and Wiwatanadate, 2012). It has been proposed that regularly consuming water with fluoride concentrations of at least 0.9 mg/l is the cause of at least 37% of dental fluorosis cases (McGrady et al., 2012). Levy and Leclerc (2012) also correlated fluoride in drinking water with bone diseases (Osteosarcoma) in adolescents and children. Sun et al. (2013) discovered that cases of hypertension in adults could be linked to fluoride present in drinking water. They further emphasized that fluoride exposure could cause an increase in plasma Endothelin-1 (ET–1) levels. Liu et al. (2014)

* Corresponding author at: Department of Civil Engineering, Covenant University, Ota, Ogun State, Nigeria.
E-mail addresses: praisegod.emenike@covenantuniversity.edu.ng (C.P. Emenike), imokhai.tenebe@covenantuniversity.edu.ng (I.T. Tenebe), p.jarvis@cranfield.ac.uk (P. Jarvis).

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also identified a link between fluoride exposure from drinking water and carotid artery atherosclerosis in adults. In a recent study conducted by Irigoyen-Camacho et al. (2016), reports showed that increased morbidity and mortality rate could be correlated with nutritional deficiencies propagated by fluoride intake. It should be noted that at low concentrations, ingesting fluoride from drinking water can hinder dental caries and some health authorities deliberately add fluoride to drinking water to reduce the incidence of enamel decay (Freire et al., 2016).

In the urban regions of Nigeria, the wholescale provision of reliable access to drinking water has been made difficult due to increasing population and habitation spread within cities (Emenike et al., 2016; Odjegba et al., 2015, 2014), a similar case as seen in Tanzania. Many localities in southwestern Nigeria and Tanzania, in these regions, are supplied with surface water for their source of drinking water (Adekunle et al., 2013). However, in many cases, these surface waters are heavily contaminated and polluted, particularly with respect to the microbial quality of the water, and are subsequently poorly treated making this water unfit for human consumption (John-Dewole, 2012; Tenebe et al., 2017; Tenebe et al., 2016). For this reason, many people have accessed groundwater for their drinking water given its perceived higher quality (Adekunle et al., 2013). However, given the noted pollution of such groundwater systems with fluoride and the evidenced adverse effect on human health, a stronger link between fluoride concentration and its health impacts is needed to enable more thorough risk assessment and mitigation measures to be applied.

Recently, researchers have adopted the human health risk assessment (HHRA) model to determine the adverse effect of absorbing or ingesting chemicals at a range of concentrations (Yang et al., 2012; Zhai et al., 2017). Therefore, this study seeks to consider two common pathways (ingestion and dermal) by which human populations can be exposed to high fluoride concentrations. In the same vein, the study capitalizes on the risk assessment indicators endorsed by the US EPA (2011) to assess the extent of fluoride contamination in groundwater and evaluate in detail, the health risk associated with fluoride pollution in groundwater. In furtherance of this, reports of elevated fluoride concentration in drinking water and its potential health risk has been investigated in Tunisia (Guissouma et al., 2017), Iran (Battaleb-Looie et al., 2013; Dehbandi et al., 2018; Yousefi et al., 2018), Ghana (Craig et al., 2015; Salifu et al., 2012), China (Zhang et al., 2017) and United Arab Emirates (Walia et al., 2017). But, to the best of our knowledge, there are a dearth of literature in Nigeria with no previous investigation in the studied region in this regard. Therefore, this study focuses on the following objectives. First, to determine the spatial distribution of fluoride and other groundwater quality parameters in Abeokuta. Secondly, to analyze the interrelationship between fluoride and other water quality parameters. Thirdly, adopt geostatistical modeling in which semivariogram graphs will be used to describe and validate the extent of contamination via the interpolation of known concentration from sampled locations. Finally, assess the health risks associated with fluoride concentration in groundwater. To establish a more realistic base for judgement, ingestion and dermal pathways were investigated. Also, the receptors (individuals) were classified into seven age groups (6 – 12 months, 6 – 11 years, 11 – 16 years, 16 – 18 years, 18 – 21 years, ≥ 21 years and > 65 years). This will assist in adding relevant data to local rural water management, policy and decision makers to take adequate measures in safeguarding the lives of residents in endemic fluoride regions.

2. Materials and methods

2.1. Study area

Abeokuta, the capital of Ogun state, occupies 40.6 km^2 (latitude 7.17–7.25°N; longitudes 3.28–3.43°E). The population of Abeokuta is estimated to be 451,600 with an annual growth rate of 3.5% (National Population Commission, 2010). The area sits on a complex geological system, composed of a mix of rock from the Precambrian period. The complex spreads through the southwestern region of Nigeria, sharing a part with the Dahomey basin composed of sedimentary rock (Rahaman, 1976). The region is also linked to Lagos by river/canal (130 km) or by railway (77 km) and shares common borders with Ibadan, Shagamu, Ilaro, Ketuo, and Iseyin. The sample locations were targeted at regions with high population density.

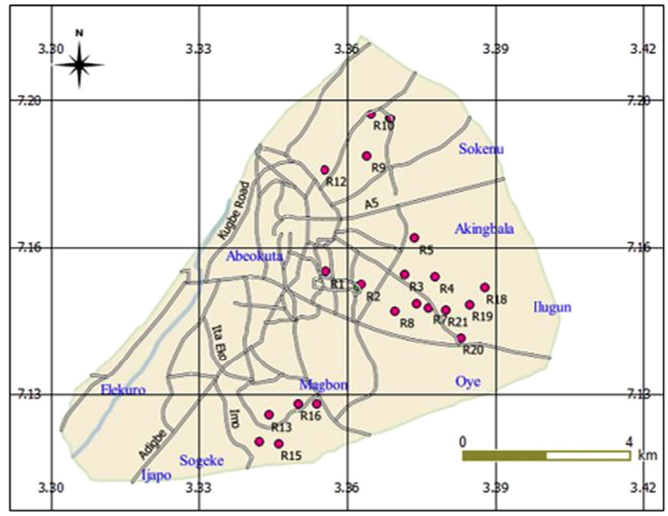

Fig. 1. Map of study area showing sampling locations.

2.2. Sample collection

The water samples analyzed in this study were obtained from taps that were connected directly to the underlying groundwater aquifer. In total, 63 water samples were collected from twenty-one locations (Fig. 1) in the study area (R1 – R21). The sampled taps were regularly used by householders mainly for consumption and domestic activities. At the point of collection, the taps were allowed to run for about 15 min before representative samples were collected to reflect the status of the aquifer and the water consumed by the householder. Sample bottles (made of polyethylene) were washed with distilled water (mixed with 20% nitric acid) prior to sampling. The sample bottles were further rinsed three times with distilled water to remove any trace of acid and then air-dried before transporting to the sampling site. At the sampling location, the bottles were rinsed three times with tap water prior to collection to ensure no acid interference. After sample collection, the containers were sealed with screw corks, labeled appropriately, placed in a container filled with ice and immediately transferred to a refrigerator regulated at 4 °C.

As soon as each sample was collected, unstable and sensitive water quality parameters such as total dissolved solids (TDS), electrical conductivity (EC), temperature (Temp.), alkalinity (Alka.) and pH were measured in situ with a calibrated multiparameter probe (HANNA – HI2030 Salinity/TDS/EC meter and HANNA – HI98130). Standard analytical procedures (APHA, 2005) were followed to measure the ionic concentration of major ions. Sodium ($Na+$), potassium (K^+), magnesium (Mg^{2+}), calcium (Ca^{2+}), iron (Fe^{2+}) and manganese (Mn) were measured using flame photometric method (Flame Atomic Absorption Spectrophotometry), dissolved silica (SiO_2) by molybdosilicate method with UV–Visible spectrometer and Nitrates (NO_3^-) concentration by UV–Visible spectrometer. Sulfate (SO_4^{2-}) concentration was determined by the turbidimetric method, bicarbonate (HCO_3^-), chloride (Cl^-), and carbonate (CO_3^{2-}) concentration were measured by volumetric method. A calibrated potentiometric ion-selection electrode (HANNA–HI5315) attached to a water-resistant portable ORP/pH/ISE meter (HANNA–HI98191) was used to measure the concentration of fluoride