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Fluoride in groundwater in the Bongo District, Ghana: an assessment, health impact and possible mitigation strategies

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ABSTRACT

Fluoride contamination in groundwater in the Bongo District of Ghana was investigated using samples from 323 boreholes covering three geologic zones; granite, greenstones, and igneous/metamorphic. The relationship between fluoride concentrations and the geologic zones, groundwater depth, pH, and conductivity were determined to assess the risk of fluorosis from the ingestion of groundwater in these zones and to potentially guide future borehole locations. Data clearly showed that the problem of groundwater fluoride contamination exists mainly in the area underlain by granite and the risk of fluorosis and other fluoride ingestion related diseases exist for about 39% of the population of the District living in the granite zone. No clear correlation was found between fluoride concentration and pH, conductivity or depth of the boreholes, which rules out alternative borehole locations as a solution to the problem. An option for fluoride removal is a hybrid pre-adsorption/ultrafiltration treatment system powered by solar panels in off-grid communities in the District. Under non-optimized field conditions, the performance was affected by the adsorption capacity of the unmodified, natural laterite and membrane type. Improvement on laterite capacity or use of other adsorbents, and membrane type selection and optimization of such a system would be required for the field application.

Keywords: Decentralized water treatment; Fluoride; Groundwater; Hybrid pre-adsorption/ultrafiltration

1. Introduction

1.1. Fluoride in groundwater

The problem of dissolved contaminants in general, and fluoride specifically in groundwater, is a widespread global challenge for potable water provision [1]. Fluoride contamination in groundwater has been a subject of research and discussion for many decades [2–9]. Removal of fluoride as a dissolved water contaminant is difficult and potentially expensive which leaves many communities unable to take effective remedial action. Often, communities and water providers assume that once a borehole with sufficient yield is installed, safe water is available. Water quality is a secondary concern over water quantity, and geogenic contaminants such as nitrate, fluoride, chloride, sulfate, and uranium are not naturally monitored. While a borehole typically provides microbiologically safer water than surface wells, chemical contamination is often not considered. In the absence of access to alternative water supplies, many communities continue to consume contaminated water and are often left with very severe health effects [10–14].

Fluoride in groundwater is generally known to come from two main sources, namely, anthropogenic activities and the geology of the area. The anthropogenic sources include the extensive use of fluoride-containing fertilizer on agricultural fields or industrial activities that involve the use of

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fluoride-containing materials [15–17]. In a review, Ozsvath [18] indicated that fluorine is preferentially enriched in highly evolved magmas and hydrothermal solutions, that include syenites, granitoid plutonic rocks, alkaline volcanic and hydrothermal deposits, as well as in sedimentary formations that contain fluoride-bearing minerals derived from the parent rock, fluoride-rich clays, or fluorapatite. High fluoride in groundwater is therefore associated with granitic and metamorphic rocks, biotite, albite, hornblende, fluorite and amphibole [8,19–25]. Clearly, fluoride is widespread and the global occurrence has been published by Amini et al. [26] while the geology appears to be a determining factor in fluoride in groundwater.

The mobilization of fluoride from the rocks into groundwater is known to depend on a number of factors including the rock type and the prevailing conditions in the contacting aqueous environment during the weathering process. According to Apambire et al. [4], the minerals that have the greatest effect on the hydrogeochemistry of fluoride are fluorite, apatite, micas, amphiboles, certain clays, and villiaumite. Among these, it is fluorite that is the main mineral controlling aqueous fluoride geochemistry. It has been suggested that the presence or otherwise of calcium ions influence the build-up of fluoride in groundwater [21,22,27–29]. Rukah and Alsokhny [6] and Guo et al. [30] have indicated that fluoride concentration in groundwater shows a positive correlation with pH. Fluoride release into the contacting aqueous medium was reported to be governed by ion exchange between the OH^- ions at the exchange sites with F^- ions as pH increases. Abdelgawad et al. [8] and Garcia et al. [23] have suggested that a longer residence time of the groundwater in the aquifer results in higher fluoride concentrations, which may be affected by the intensity of water abstraction through boreholes.

1.2. Health problems associated with fluoride in water and diet implications

Health problems due to the ingestion of fluoride from drinking water have become a topical issue among health practitioners and scientists. Much of the literature reports health implications that can be summarised as dental and skeletal fluorosis [1,31] (Takeda and Takizawa [32] and Ozsvath [18] cataloged the health risks to additionally include increased rates of bone fractures, decreased birth rates, increased rates of urolithiasis (or kidney stones), osteosarcoma, impaired thyroid functions, and lower intelligence in children. Chandrajith et al. [10] mentioned the occurrence of chronic kidney disease as one of the diseases that may be associated with a high intake of fluoride.

It has been suggested that higher risks of fluorosis exist in the absence of balanced diets, as may be the case for arid regions. While the consumption of fluoride-contaminated agro products can contribute to increased fluoride intake, diet can equally be a fluorosis mitigation measure. Indeed, Rango et al. [33] highlight the importance of diet in the management efforts to mitigate dental fluorosis. The use of nutrition intervention especially for children as a strategy for controlling fluorosis has also been suggested by Meenakshi and Maheshwari [34]; Andezhath and Ghosh [35] and Hussain et al. [36]. Nutrition as a very accessible mitigation strategy does not require expensive infrastructure investment. It is believed that with the appropriate nutrition, the incidence of fluorosis can be minimized. In addition to agricultural products containing high levels of vitamin C, some of the food items mentioned in the references include daily intake of milk, vegetables, and nuts with high calcium content.

1.3. Fluoride removal from water

Several mitigation measures have been suggested in the literature to reduce fluoride ingestion and the health effects thereof. Bhatnagar et al. [37] and Waghmare et al. [38] have presented reviews of methods for fluoride removal from drinking water. These include coagulation-precipitation, adsorption using various adsorbents, ion exchange, electrochemical and membrane separation technologies. Many of the methods prescribed are promising although most will face significant operational obstacles in the implementation in remote areas [39], some being exacerbated by the complexity of the method, problems with the reusability of the adsorbent, efficiency of the process, cost of implementation and maintenance of systems especially in the rural communities, and cultural concerns in the acceptability of materials used.

Fluoride removal systems requiring energy input would have to consider the use of renewable energy to power such systems given the absence of a reliable electricity grid. Shen et al. [14] and Schäfer et al. [40] have investigated fluoride removal membrane technology powered by renewable energy. Meenakshi and Maheshwari [34] have looked at the advantages and limitations of various technologies presented in the literature and advised that the selection of the treatment process should be site-specific to match the prevailing conditions.

1.4. Water supply in Ghana's Bongo District

Bongo is one of the nine Districts in the Upper East Region of Ghana. It lies between longitude 0.45°W and latitudes 10.50°N to 11.09 and has an area of 459.5 square kilometers, which is considered as semiarid. There is only one rainy season starting from May to mid-October, spanning about 70 d with rainfall ranging from 600 to 1,400 mm. The Ghana 2000 Population and Housing Census [41] put the population of the district at 77,885. A calculation by the District Planning Team indicated that the District has a growth rate of 2.8% [42]. However, the 2010 Population and Housing Census of Ghana (GSS, 2012) puts the population of the District at 84,545, an increase in population of 8.6% over 10 years. Available data for the district shows that there is no significant in-migration of population, the increase in population, therefore, arises from a high birth rate. This population increase, coupled with climate change, will result in further deterioration of the water supply. To ensure adequate water supply to the communities, a number of boreholes have been provided in addition to hand-dug wells. Household surveys by the District Water and Sanitation Team (DWST) with the support of WaterAid Ghana [43] (Anon, 2008) show that boreholes constitute the major source (over 80%) of water in the District. The quality has largely been affected by the geology/geochemistry of the aquifer.

Access to safe drinking water in the Bongo District has been a matter of concern for many years. The problem of relatively high fluoride concentrations (up to about 5 mg L^{-1}, compared to a WHO Guideline of 1.5 mg L^{-1} [1] in many boreholes in the district has a significant impact on the health of the people, especially young children who often exhibit severe dental fluorosis.

1.5. Fluoride in Ghana's Bongo District

The occurrence of fluoride in the Bongo District groundwater has been associated with the various geological formations in the two regions, namely the Birimian metavolcanics, metasediments and granitoids, and the Bongo granitic rocks [4]. Murray [44] indicated that the Bongo area consists mainly of Bongo granite and porphyritic microgranite; greenstone, lava amphibolite, tuff and minor occurrences of phyllite and schist; altered hornblende granodiorite, biotite granodiorite and adamellite; and, to a lesser extent, areas covered by foliated hornblende-biotite granodiorite and tonalite. Apambire et al. [4] noted that all rock formations in the study area contain different levels of fluorine, varying between trace and 2,000 mg/kg and that the fluoride concentration in the contacting water in the Birimian metavolcanics, metasediments, and granitoids were generally low, measuring less than 1 mg L^{-1}, whereas in the Bongo granitic rocks the values ranged from 0.61 to 4.6 mg L^{-1}. Abugri and Pelig-Ba [45] have found the presence of fluoride in soils in the upper regions including the Bongo area. Recent work by Abitty et al, [46] and Addae [47] have indicated that the Bole-Nangodi belt of which the Bongo granite is part contains among others K-feldspar with fluorite and apatite as among accessory minerals.

An expanded mapping of the fluoride concentration in groundwater in 278 boreholes in the Bongo District alone was done in 2008/2009 by Alfredo et al. [48] confirming high fluoride concentration in the Bongo granite. The studies have not established whether there was an association between fluoride concentration and borehole depth across the geologic formation, which may enable decisions on the depth of drills to minimize fluoride concentrations.

Government agencies responsible for the provision of potable water in rural areas as well as non-governmental organizations have been investigating the problem and are looking for solutions to provide good and safe drinking water. As at the time of the study, more than 340 boreholes have been installed in the communities, some of which have been capped because of high fluoride concentration.

1.6. Socio-economic implications of fluorosis in Ghana's Bongo District

The socio-economic implications of fluorosis in the Bongo District merit mention. A paper by Atipoka [49] discusses the accessibility of potable water in the Bongo District. She indicated that according to the household survey of 2008 [43], only about 7%–18% of people in the various area councils in the district could access water within 500 m or 7%–21% within 15 min walking distance. Also, only 7%–18% of the people have access to 35 L of potable water per day. This means that people, mostly women, and girls, spend a lot of time searching for water for daily use and will often resort to the collection of water from contaminated sources such as streams, untested wells and open hand-dug wells [50].

The available stock of boreholes provides no relief for some of the communities because of fluoride contamination. The consumption of fluoride-contaminated water has resulted in dental fluorosis in parts of the Bongo District. Individuals with dental fluorosis often exhibit signs of inferiority complex and are reluctant to reveal their severely stained teeth. Climate change has resulted in the drying up of safer sources of water [49] and both government and NGOs continue to provide more boreholes. Treatment of contaminated boreholes is an important mitigation strategy.

For sustainable management of water systems, adequate institutional arrangements must be made. Water governance issues are important in determining mitigating strategies for minimizing fluorosis. By institutional arrangement, the management of water resources in the districts of Ghana is a mandate of District Water and Sanitation Teams (DWSTs) through Water and Sanitation Committees (WATSAN) and Water and Sanitation Development Boards (WSDBs) [51,52]. The committees supervise community water delivery, including operation and maintenance, using the accepted principle of contributing to the capital cost of the facilities, their operation, and maintenance. Tariffs are set accordingly, most often above levels paid by urban consumers to ensure undisrupted water supply. Amongst other aspects, Rossiter et al. [53] discussed the often surprisingly high costs of water delivery and the ability or inability of communities to pay the agreed fees for system maintenance. According to Atipoka [49], the major source of income for the Bongo District is the central government subvention through the Common Fund, which is not used by the District Assemblies to support water delivery systems. In discussing strategies with district authorities on water delivery to communities, small decentralized water treatment systems were mentioned as the preferred solution and that the communities through the WSDBs were determined to fund their operations and maintain them.

1.7. Fluoride mitigation strategies in Ghana's Bongo District

Possible mitigation strategies for minimizing the effect of consumption of high fluoride-contaminated water in endemic areas include the use of household filtration units, rainwater harvesting, provision of on-site treatment systems, and off-site treatment and distribution within the communities affected [54–56].

The advantages and disadvantages of using household units have been widely discussed in the literature [57,58]. In the Bongo District, discussions revealed that household filters supplied by UNICEF were initially welcomed but abandoned after a short period of use because users complained of having to wait for a long time to have enough water from the units [59].

Rainwater harvesting is of limited impact because semiarid climatic conditions prevail in the Upper-East Region [60] within which Bongo is situated. Annual rainfall is between 800 and 1,100 mm [43] and hence rainwater can only be considered a useful water resource from about May until October. According to Atipoka [49], surface water resources dry up during the dry season, which has been worsened by climate change trends, poor watershed management and increasing demand from a growing population. The Vea Dam, built in 1960 for irrigation purposes, is heavily silted and unable to hold much water for both irrigation and domestic water usage.

Ultrafiltration (UF) and nanofiltration (NF) have been recognized as modern technologies for the removal of bacteria and viruses (UF) and fluoride (NF) from contaminated water [61–63]. The technique is suited not only for the removal of fluoride and other contaminants but also for water-borne bacteria and viruses. Even though the initial cost of installation is comparatively high and requires skilled personnel in operating and maintaining the system it offers the best chance to provide safe drinking water. Decentralized systems for remote areas incorporating solar-powered pumps are good solutions [64]. Such a decentralized system could be coupled in a hybrid system with fluoride pre-adsorption using local adsorbents to reduce the number of membrane modules and reduce the cost and maintenance cycles of the membranes.

The primary objective of this research was to expand coverage of sampled boreholes, investigate the relationship of fluoride concentration to a depth of borehole, pH, and conductivity across the geological formations. As such the identification of links between borehole parameters and fluoride occurrence would offer a solution to draw water from specific, uncontaminated areas. A secondary objective was to investigate population distribution around the boreholes to determine areas and the inhabitants at high risk for the prioritization of points of supply of good drinking water. A third objective was to evaluate through a preliminary field study, the feasibility to use of locally abundant laterite in a hybrid pre-adsorption/ultrafiltration set up for removal of fluoride.

2. Materials and methods

2.1. Data collection

Data on the location and distribution of boreholes within the Bongo District were obtained from the Bongo District Assembly [43]. A digitized geological map of the Bongo District was prepared at the Department of Geomatic Engineering, Kwame Nkrumah University of Science and Technology, Kumasi. The locations of the boreholes, as calculated from GPS coordinates and mapped onto the digitized geological map of the District is presented in Fig. 1. Data on the depth of boreholes was obtained from the Regional Community Water and Sanitation Agency (CWSA) in Bolgatanga [43]. Data on the fluoride concentrations, depth of each borehole, population of people the area using each borehole and the pH of the sample from the borehole were then mapped.

Population data around the boreholes was obtained through community mapping. In this process community members are guided to mark their community on the floor by indicating various houses, water points, and water and sanitation facilities in the community [65]. The houses were marked with the number of people living in the home and by indicating the number of people in each household.

2.2. Water sampling

A total of 323 water samples were collected in the Bongo District during the dry season, from January to March 2011. The boreholes are constantly in use by the communities and therefore would have similar water quality as that of the aquifer. After pumping for about 3 min to further purge the boreholes with hand pumps fitted to the boreholes, samples were collected in well acid-washed and rinsed 500 mL polyethylene bottles and preserved in a cool box in the field.

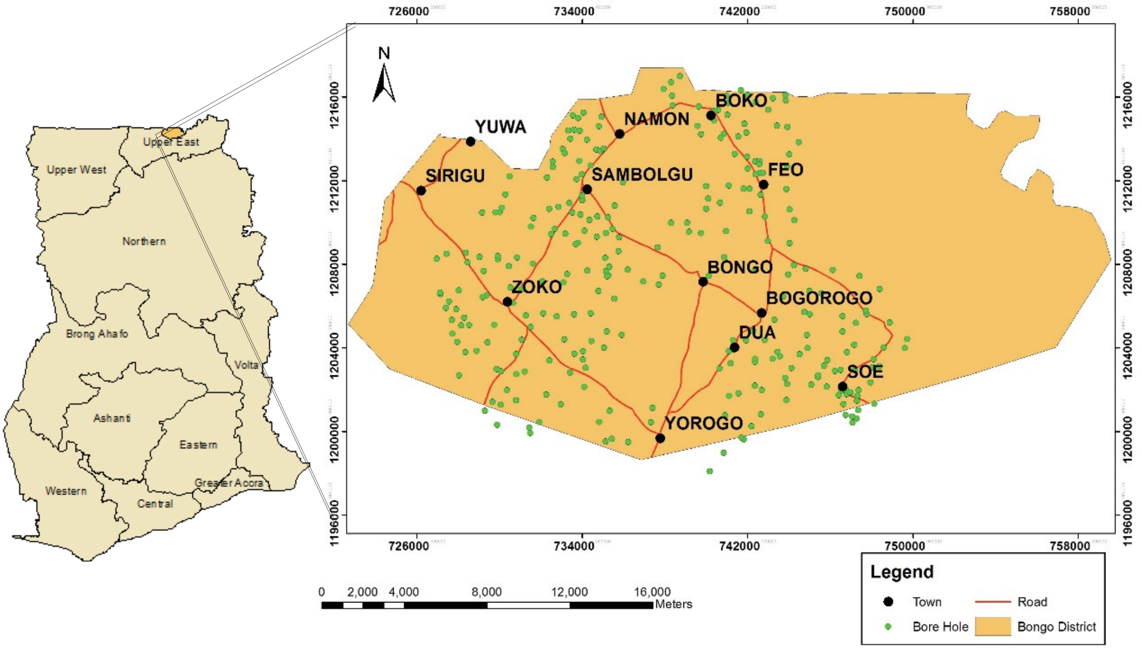

Fig. 1. Map of the Bongo District with towns and borehole locations (*Source*: Survey Department of Ghana).

The samples were then taken to the field laboratory, where 25 mL of each sample was filtered through a 0.45 µm syringe filter (Sartorius Minisart, non-sterile CA, UK) into the sample vials and subsequently stored in a fridge below 10°C before transporting to Edinburgh. On each day of sampling, the samples were analyzed for the pH and conductivity immediately.

2.3. Preliminary membrane filtration tests

Preliminary membrane filtration tests were conducted in the field using two membrane modules in a slurry containing laterite obtained from the locality as fluoride pre-absorbent. The work was carried out in the field laboratory in Bongo using natural waters.

2.3.1. Laterite collection, preparation and characterisation

Laterite was used as a pre-adsorption material in front of the membranes. Laterite was selected over other geomaterials as the pre-adsorbent material because of its availability locally virtually at low cost. Laterite samples were collected from an excavation site near Balungu, in the Bongo District, ground with a Retch orbital grinder, manually sieved with a 125 µm mesh size to obtain particles less than 125 µm. The chemical (by XRF) and mineralogical (by XRD) composition, as well as the surface area (BET), were determined.

2.3.2. Membrane and module characteristics

GE Zenon ZeeWeed hollow fiber module (ZW1) and Inge AG Dizzer d10 multibore module (D10K MB) were used for all experiments. The nominal pore size of the fiber membranes of ZW1 was 0.04 µm and the membrane surface area was 0.047 m^2. The membrane material was hydrophilic polyvinylidene fluoride (PVDF). The D10K MB modules had membranes with the nominal pore size of 0.02 µm and membrane surface area was 0.1 m^2. The membrane modules consisted of 13 fibers with 7 capillaries in each fiber and the membrane material was modified polyethersulfone (PESM). The submerged Zenon modules were operated in an "outside-in" configuration, while the Inge module was fixed in a vertical position and operated in a dead-end direct filtration mode. The flux changed from 70 to 130 $L\ m^{-2}\ h^{-1}$ for Zenon and 60 to 140 $L\ m^{-2}\ h^{-1}$ for Inge modules, respectively.

2.3.3. Membrane system design

Photos and schematics of the two membrane configurations set up in the Bongo field laboratory are illustrated in Fig. 2.

The Zenon module was a submerged configuration and permeate was extracted via vacuum while the Inge module was used in pressure filtration. A peristaltic geopump (AC/DC, CASE, EZ2 PH, Series II, manual variable-speed drive with 2 pump stations 1 (30–300 RPM) and 2 (60–600 RPM) was purchased from Geotech, USA. This was used with an EasyLoad II-S/S pump head, thick high performance (Geotech, USA) and Masterflex platinum-cured silicone tubing, L/S 15 of inner diameter 4.8 mm (Cole-Parmer Instrument Co., UK).

A pressure gauge (–1 to 1 bar) (Omega, UK) was connected in the permeate line to monitor the suction pressure for the Zenon system, while the Inge system was fitted with a feed side pressure gauge. An air compressor DC, ACO-003 (Hailea, China) provided air in the feed tank for both systems to keep the laterite in suspension during the experiments. In addition, air was supplied to Zenon modules from the top for membrane scouring.

Six 18 VDC solar panels with a size of 0.64 × 0.40 m (1.5 kg each) provided a maximum power of 30 W and were used to power the two pumps and the aerator. The maximum power required for each pump and the aerator were 70 and 25 W respectively. Six 12 V Cellite 12TSG20 18Ah GEL Batteries (Atlas Business & Energy Systems (ABES), Ghana) were connected to the solar panels (two solar panels for two batteries) to store power. Of these three pairs, two were used for the filtration systems, while the third was used to power the aerator. The location of the solar panels was changed manually depending on the direction of the sun during the day.

2.3.4. Membrane system operational protocol

The waters from boreholes with fluoride concentration less than 2.5 $mg\ L^{-1}$ is spiked with fluoride to add 3 $mg\ L^{-1}$ fluoride to obtain fluoride concentrations between 3.2 and 6.2 and be able to investigate fluoride removal in different water matrices. Each water was treated with Inge and Zenon modules. Seven consecutive water samples were taken during filtration.

The experiments were done is the recirculation mode. The laterite was kept in suspension with aeration provided by an aerator connected to the system. The airflow rate for laterite mixing was adjusted from 66 to 132 $mL\ s^{-1}$ for the Inge modules, while for the Zenon modules, the air flowrate for laterite mixing varied from 49 to 117, and membrane scouring from 67 to 107 $mL\ s^{-1}$. To investigate the influence of air flowrate on laterite mixing and thus the sorption, the same experiments were conducted with varying air flowrate conditions to account for variability during experiments.

The filtration was started when 15 g laterite was added into the feed solution and pH adjusted to 4.5 (volume 2.6–3 L). The pH was monitored throughout the experiment and readjusted if necessary. 50 mL samples were collected from the permeate line after 2, 10, 20, 30, 40, 60 and 90 min of filtration, which amounts to a total volume of 350 mL (about 10% of the feed solution). After taking the last permeate sample, the concentrated solution was mixed until a homogenous laterite solution was obtained and 50 mL of concentrated sample was taken.

Voltage, current, pH, pressure, temperature and the flow rate was recorded throughout the experiment. Flow rate data was obtained by measuring the permeate volume with a graduated cylinder for 30 s and 60 s for Zenon and Inge modules, respectively. Prior to the experiments, pure water flux measurements were done for half an hour in 10 min intervals.

2.4. Water sample analysis

The samples were analyzed using a fluoride ion-selective electrode (Metrohm, UK) immersed in well-mixed 5 mL

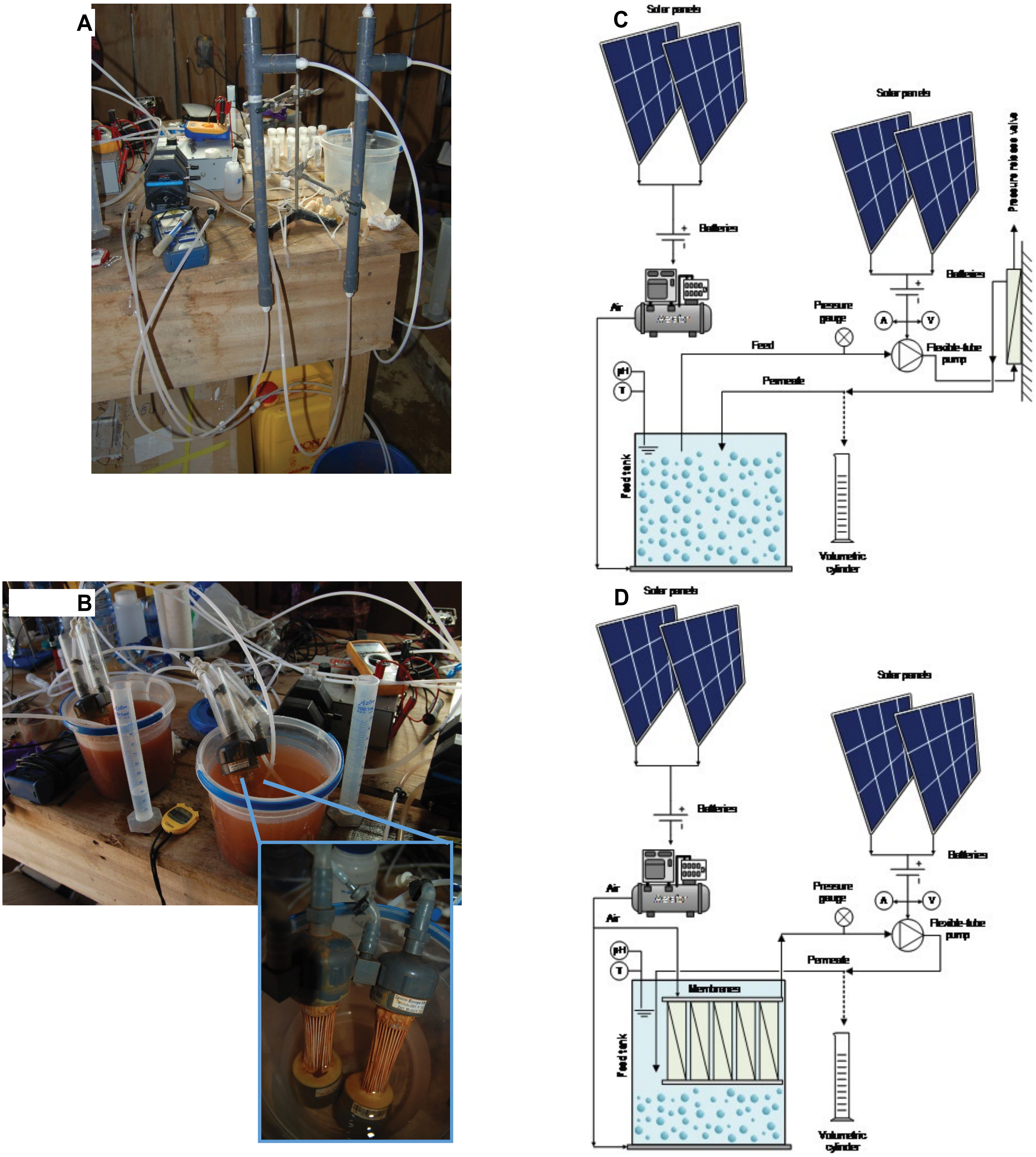

Fig. 2. Membrane set-up pictures and schematics of the Inge AG Dizzer d10 multibore module (D10K MB) (a,c) and the GE Zenon ZeeWeed hollow fiber module (ZW1) (b,d) as used in the Bongo field laboratory.

water samples and 5 mL of buffer solution, and the value recorded once the reading on pH/ISE meter 826 (Metrohm, UK) was stabilized. This method is based on the EPA (U.S. Environmental Protection Agency) Method 430.2 for fluoride determination and includes the addition of a total ionic strength adjustment buffer (TISAB) prepared from 1,2-cyclohexanedinitrilo-tetraacetic acid (CDTA) to control analytical interferences. The TISAB was prepared by adding 57.0 mL glacial acetic acid, 58.0 g sodium chloride and 4.00 g CDTA to 500 mL deionized water in a beaker, stirring and allowing to cool to room temperature.

The pH of the solution was then adjusted to between 5.0 and 5.5 using 5 M sodium hydroxide (NaOH). The solution was then transferred into a 1 L volumetric flask and filled to the mark with deionized water. This was transferred into a clean polyethylene bottle, then 5 mL of TISAB were added to 5 mL of water sample and fluoride content measured. A 100 mg L^{-1} sodium fluoride (NaF) stock solution was prepared by dissolving 221.0 mg of anhydrous NaF (purity 99+%, Fisher, UK) in deionized water and diluting to 1 L. The standards required for measurement were then prepared by appropriately diluting the stock solution.

3. Results and discussions

3.1. Fluoride occurrence and distribution in the Bongo District

The spatial distribution of borehole fluoride concentration mapped over the geological formations by the British Geological Survey (BGS) in Edinburgh is presented in Fig. 3. The data shows that groundwater within the granite geological formation contains higher levels of fluoride than in the other geological formations. Only 11% of the boreholes in the igneous/metamorphic geological formation have fluoride concentrations higher than 1 mg L^{-1}, compared to 94% in the granite and 36% in the greenstone formations. The geological map presented by Apambire et al. [4] showed the existence of a fault across the greenstone south-east of Bongo, linking the granite to the igneous/metamorphic formations, between Bongo and Yorogo. High concentrations of fluoride in boreholes located along this fault indicate that fluoride is leaching from the granite into the fault where the boreholes are located.

3.2. Fluoride concentration as a function of borehole depth

Fig. 4 presents the spatial distribution of boreholes with respect to borehole depth. Only 6 boreholes (about 2% of total sampled), located in the middle-eastern side of the granite, were identified to be deeper than 80 meters. 29 boreholes (about 9%) are of the depth between 60 and 80 m but scattered among the geological formations. The majority of the boreholes (89%) are between 40–60 m in depth.

Fluoride distribution according to borehole depth in the various geological formations is presented in Fig. 5. Results were examined for any correlation between fluoride concentration and borehole depth, which might be necessary to determine the depth of borehole for low fluoride concentration. Pearson correlation coefficient (r) was between 0.05 and 0.16 in all geologic zones, indicating very little correlation between borehole depth and recorded fluoride concentrations. Analysis of data indicated that 94% of the boreholes in the granite formation showed fluoride concentration above 1 mg L^{-1} irrespective of the borehole depth, indicating high-risk fluoride ingestion from boreholes by inhabitants living in this area. The risk is much lower in the igneous/metamorphic geologic zone where only about 19% of the boreholes had fluoride concentrations above 1 mg L^{-1} at depths between 40–60 m. Even though fewer boreholes were sunk in the greenstone formation zone risks exist for higher than 1 mg L^{-1} fluoride concentration for boreholes deeper than 40 m.

3.3. Fluoride concentration, pH and conductivity

Fig. 6 presents the distribution of pH in boreholes across the Bongo District and Fig. 7 shows the fluoride concentration as a function of groundwater pH. In the granite zone, the pH varied from 6.54 and 7.82, with a median of 7.03. In the igneous/metamorphic and greenstone formations, the pH was 6.22–8.36 and 6.7–7.9 respectively, with median values of 6.94 and 7.05 respectively

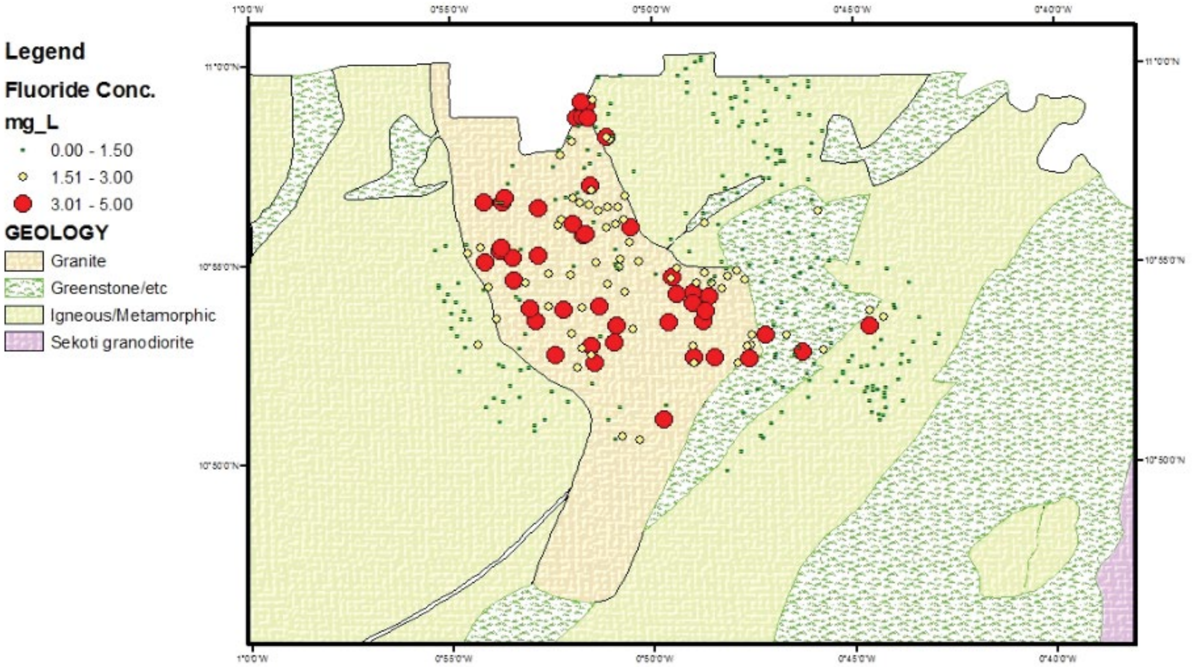

Fig. 3. Distribution of fluoride in the Bongo District for low (0–1.5 mg L^{-1}), medium (1.5–3 mg L^{-1}), and high (3–5 mg L^{-1}) concentration.

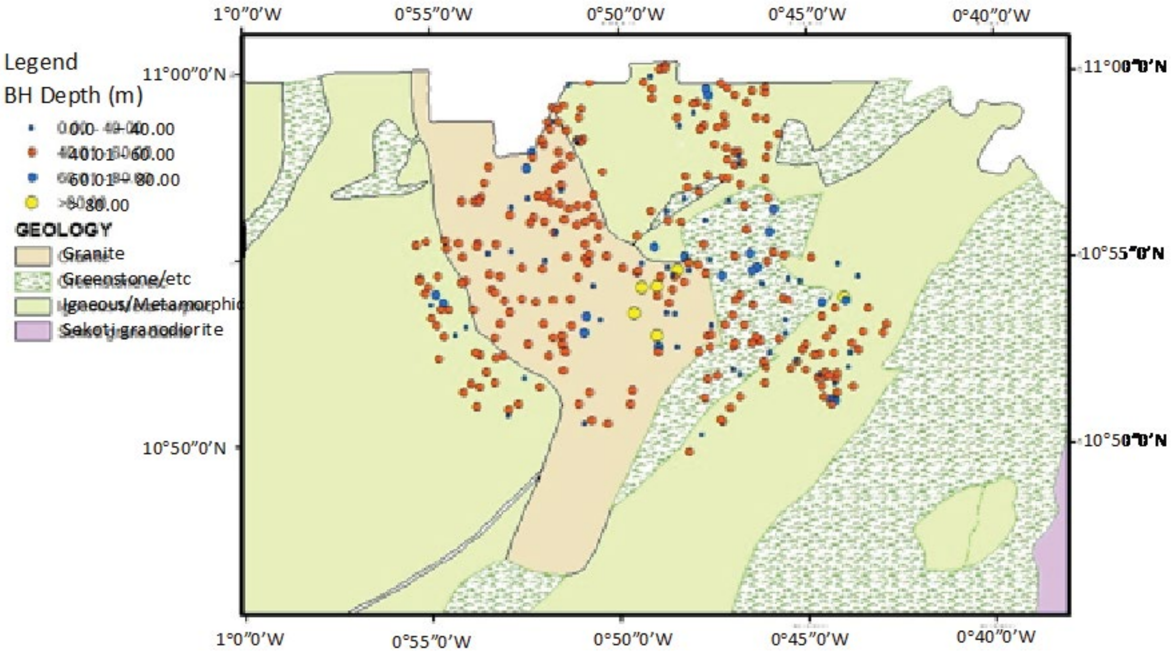

Fig. 4. Fluoride distribution with borehole depth in the Bongo District for shallow (0–40 m), medium (40–60 m), deep (60–80 m), and very deep (>80 m) depth ranges.

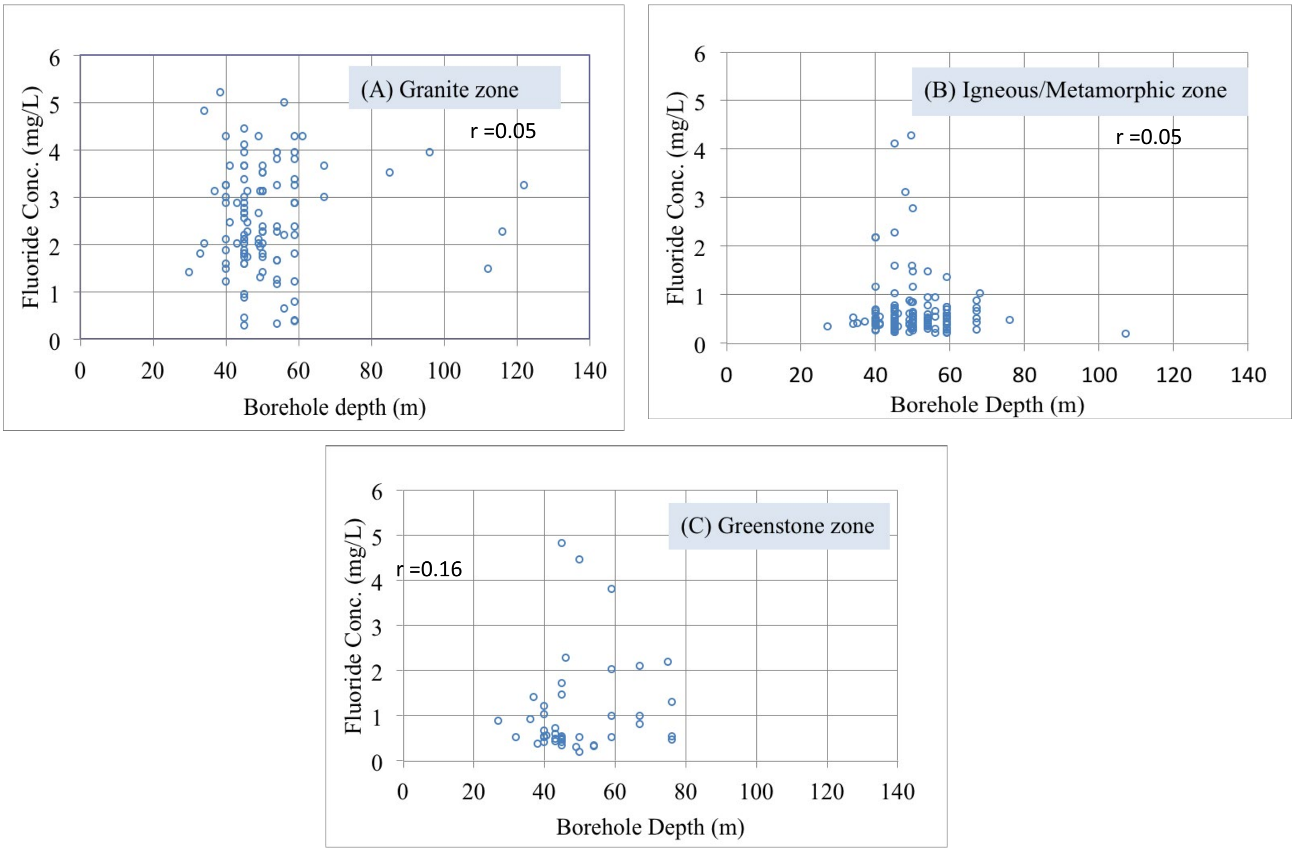

Fig. 5. Fluoride concentration as a function of borehole depth for three geologic zones (a) granite, (b) igneous/metaphoric zone, and (c) greenstone.

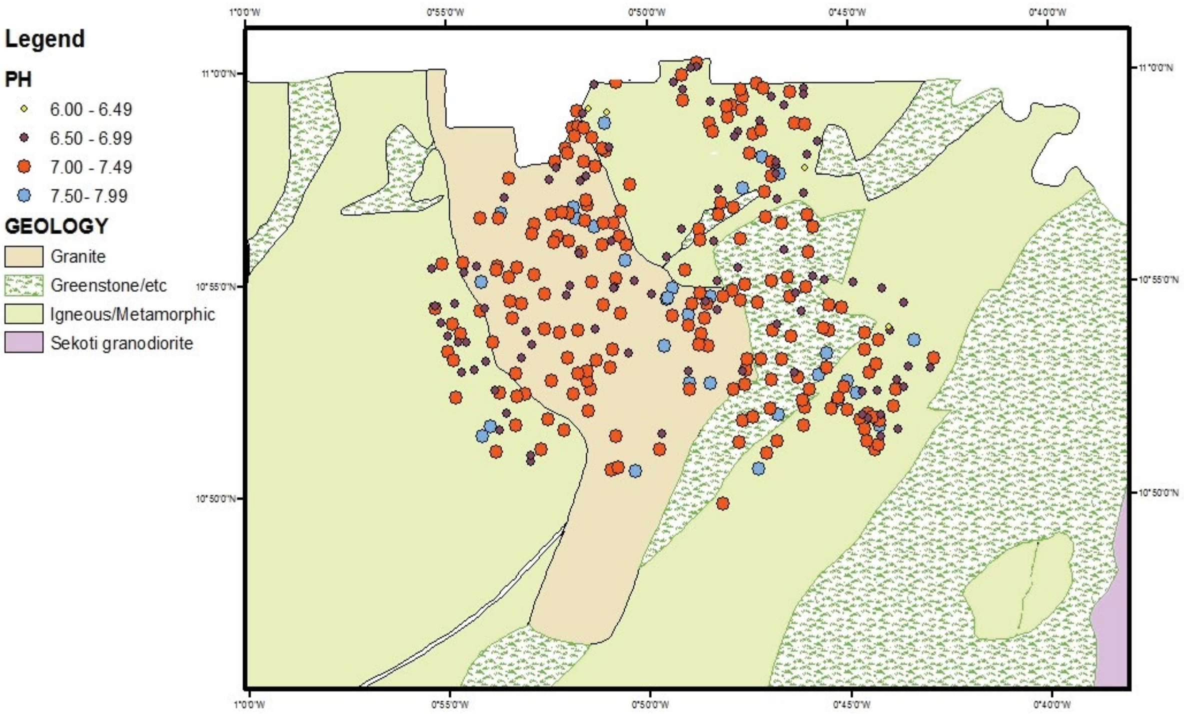

Fig. 6. Groundwater pH distribution in the Bongo District presented for the ranges of 6–6.5, 6.5–7, 7–7.5, and 7.5–8 concentration.

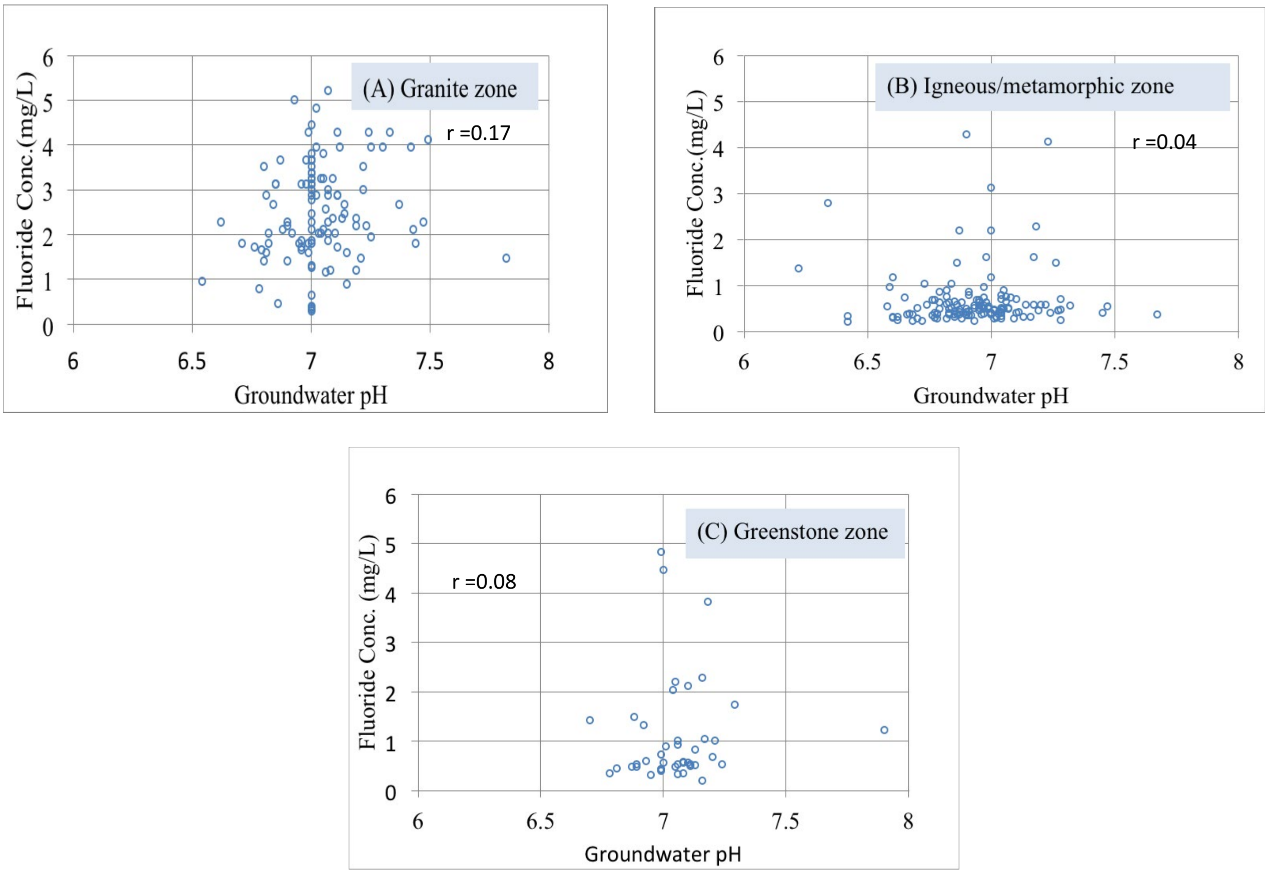

Fig. 7. Fluoride concentration as a function of groundwater pH for three geologic zones (a) granite, (b) igneous/metamorphic zone, and (c) greenstone.

Again, there was no correlation in our study between fluoride concentration and pH across all geologic formations, as suggested by Rukah and Alsokhny [6] and Guo et al. [30].

Similar results of no correlation were obtained for groundwater conductivity vs. borehole depth (Fig. 8). No correlation was observed between fluoride concentration and conductivity in the three geological formations (Fig. 9). The conductivity of Bongo borehole water is low and does not require treatment. While calcium was not measured in this study, previous sampling in Ghana [53] revealed calcium concentrations of between 8–42 mg L^{-1} in boreholes in the Bongo District.

3.4. Groundwater geochemistry

Abitty et al. [46] and Addae [47] indicated the presence of K-feldspar in the Bongo granite with fluorite and apatite as accessory minerals. Weathering reaction of K-feldspar in contact with carbonated water can be represented by:

$$2KAlSi_3O_8 + 2H^+ + 2HCO_3^- = Al_2Si_2O_5(OH)_4 + 2K^+ + HCO_3^- + 4SiO_{2(aq)} \quad (1)$$

This reaction shows that the weathering process may be influenced by the pH of contacting the aqueous medium. Arguing from the work of Handa [66] the presence of the bicarbonate ion favors the dissolution of fluoride from fluorine-bearing rocks. Houston [74] however suggested that the influence of pH is only significant at the initial stages of weathering and is minimized over the long term. Minerals in geologic formations are not uniformly distributed across the length and depth of the formations. Pelig-Ba [67] looked at the geochemistry of various samples from various locations in the Bongo area and indicated that the rock samples were of varying compositions. The weathering process involves hydrolysis, dissociation, and dissolution over time and depends on the structure of the rock minerals and the contacting solution. This may result in the absence of a correlation between fluoride concentration and borehole depth, pH and conductivity. A study conducted by Saxena and Ahmed, have suggested that there was no correlation between fluoride dissolution and the Ca, HCO_3, pH and specific conductivity of the contacting solution. Abugri and Pelig-Ba [45] investigations with the agricultural soils in the area also showed no correlation between soil pH, fluoride concentration and conductivity.

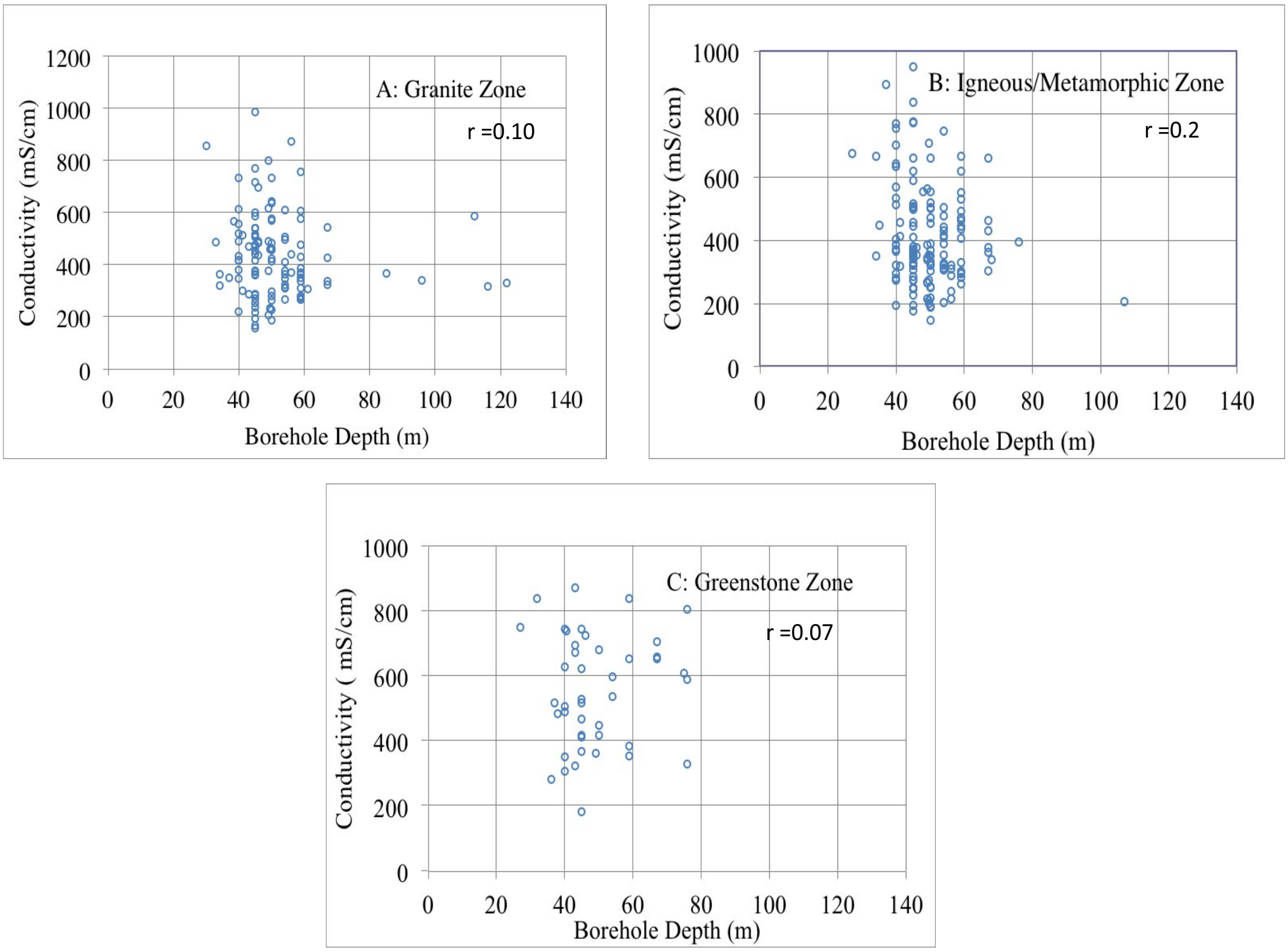

Fig. 8. Conductivity as a function of borehole depth for three geologic zones (a) granite, (b) igneous/metaphoric zone, and (c) greenstone.

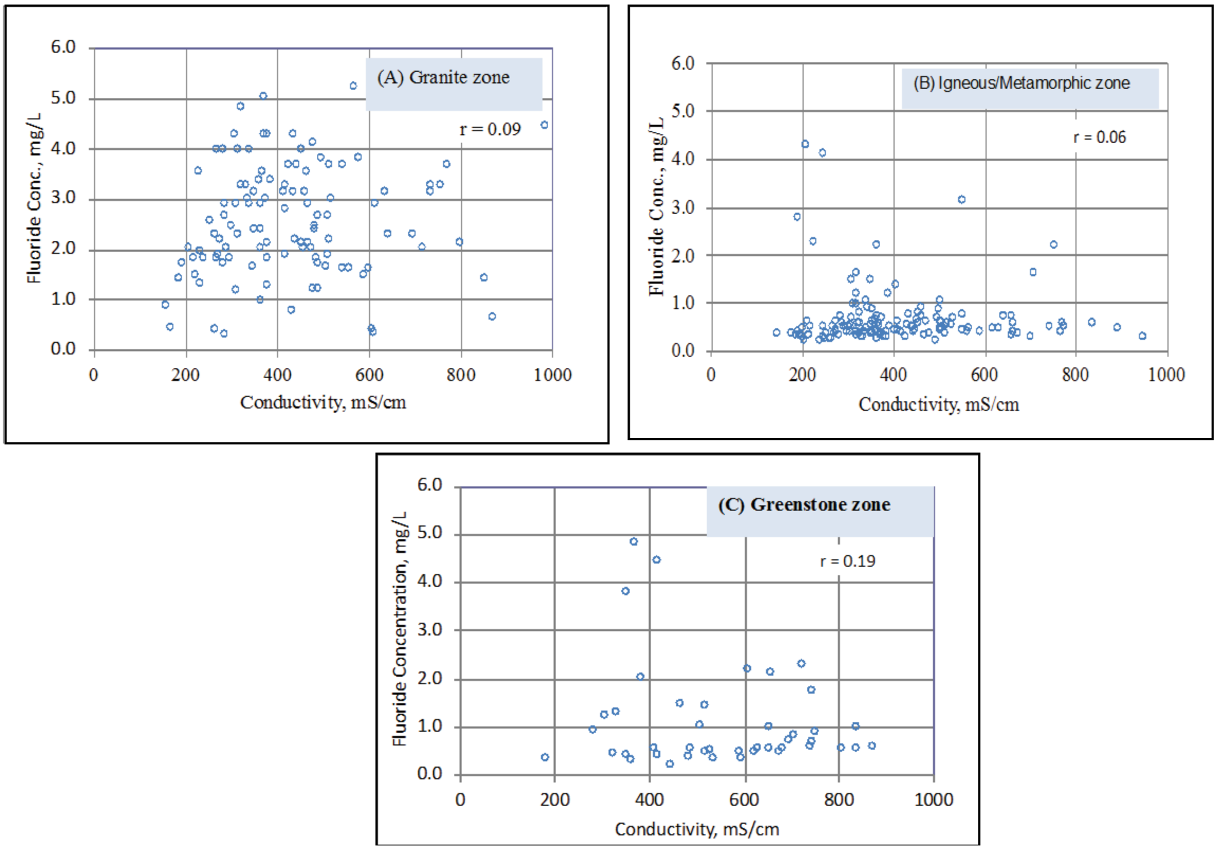

Fig. 9. Fluoride concentration vs. conductivity in the three geologic zones (a) granite, (b) igneous/metaphoric zone, and (c) greenstone.

3.5. Population distribution in the Bongo District and water demand

The population distribution in the district, especially in the proximity of the boreholes is an important factor in determining the groundwater usage and subsequent treatment capacity required. A plot of the spatial population distribution is presented in Fig. 10. The 2010 Population and Housing Census of Ghana [68] puts the population of the Bongo District at 84,545. Data collected during water sample collection shows that 39%, 47%, and 14% of the population live in the granite, igneous/metamorphic and greenstone geologic zones respectively. Thus, about 39% of the population is at risk of ingesting highly contaminated water.

The provision of fluoride-low water to address the occurrence of fluorosis in the Bongo District remains a particular problem especially for the inhabitants on the granite zone of the district. Household survey from 2008 [43] showed the per capita consumption to be between 3 and 7 L, much lower than the national basic norm of 20 L. Alfredo et al. [48], however, measured an average use of 30 L d^{-1} $person^{-1}$ by tracking all water from each borehole to the households.

3.6. Membrane filtration tests

3.6.1. Laterite characteristics

Table 1 presents both the chemical composition (by XRF) and the mineralogical content (by XRD) the laterite. The BET specific surface area was 30.36 m^2 g^{-1}, which is comparatively low for adsorbents.

The results show that the laterite sample contains about equal amounts of quartz and goethite plus hematite. Studies show that the minerals are normally intimately associated, which would determine the fluoride adsorption characteristics [69].

3.6.2. Membrane filtration

The feed and concentration for the seven consecutive samples are shown in Fig. 11a, whilst fluoride retention is presented in Fig. 11b. Of the >20 samples that were treated in this preliminary field study, a number of waters were selected in this paper, reflecting the overall trend of the data.

These results are obtained by using chemically unmodified local laterite added to waters and separated by a membrane filter that will simultaneously retain pathogenic contaminants. A number of typical local water were selected to investigate the scope of such simple technology for fluoride removal in a situation where fluoride concentrations are relatively low (3–6 mg L^{-1}) and a WHO guideline for drinking water of 1.5 mg L^{-1}.

Under the conditions of the preliminary field investigation, the WHO guidelines were not attained. Nevertheless, fluoride removal of 40%–50% was achieved, where Inge membranes demonstrated better performance. Zenon fluoride retention was about 30%, while that of Inge was between

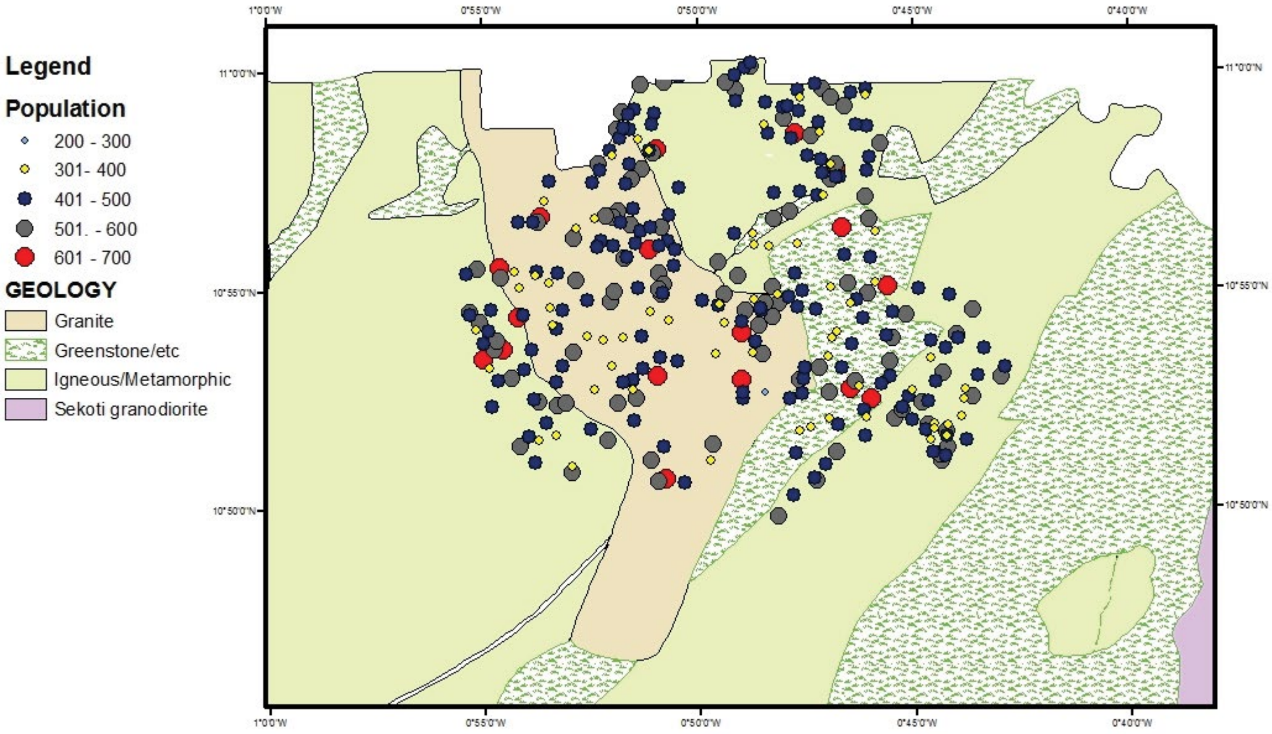

Fig. 10. Distribution of population per borehole in the Bongo District in categories of 200–300, 301–400, 401–500, 501–600, and 601–700.

Table 1
Chemical and mineralogical composition of the laterite used

Chemical composition by XRF	Mass (% oxide)	Mineralogical composition by XRD	Mass (% crystalline components)
SiO_2	38.82	Quartz	50.5
Al_2O_3	12.44	Goethite	41.1
Fe_2O_3	37.83	Hematite	8.4
MgO	0.16		
CaO	n.d.		
Na_2O	n.d.		
K_2O	0.221		
TiO_2	0.507		
MnO	0.655		
P_2O_5	0.117		
Loss on ignition	9.10		
Total	99.85		

40%–50%. From these preliminary investigations a number of conclusions can be drawn; firstly, the unmodified and natural laterite, has an ability to adsorb fluoride, but given the very low surface area this is not adequate to attain sufficient removal at a small dosage as applied in a membrane filtration system. Modified laterite [69] or other adsorbents [70,71] will be more suitable for such a process; secondly, the differences in performance can be attributed to the membrane properties. GE Zenon ZeeWeed hollow fiber module (ZW1) had a nominal pore size of 0.04 μm, while the Inge AG Dizzer d10 multibore module (D10K MB) 0.02 μm, hence about half. In consequence, the Inge module would retain more of the finer laterite fractions, that by nature are expected to have a higher surface area and hence a larger capacity for fluoride adsorption. For the future design of adsorption-UF systems, this is an important consideration unless the adsorbent is larger than the pore size.

Naturally, these preliminary studies served several purposes, from student experience to community-based demonstrations and can be modified into full scale or long-term

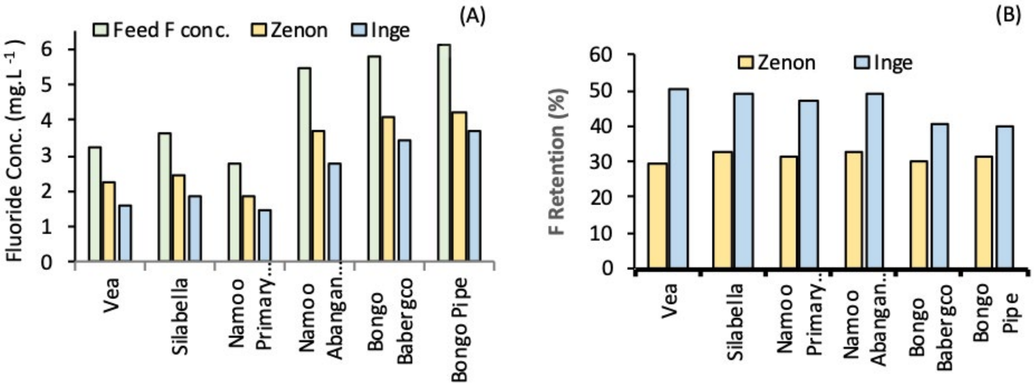

Fig. 11. Fluoride concentration before and after filtration (a) and fluoride retention for Zenon and Inge module configurations (b).

performance through further pilot studies and materials research. Further, it is critical for a successful long-term technology integration that the local stakeholders are buying into the technical solution. Long term operation and maintenance require problem-solving and resource management.

4. Conclusions

The problem of fluoride in the groundwater of the Bongo District of the Upper East Region of Ghana has been assessed using water samples from 323 boreholes. It can be concluded that:

- Over 39% of the boreholes in the Bongo District (and over 94% in the granite geological zone) have fluoride concentrations higher than 1.0 mg L^{-1},
- There is no correlation between the fluoride concentration and borehole depth, water pH or conductivity of the water and that the risk of fluorosis exists mainly for inhabitants living in the granite zone of the district.
- Under the field conditions applied, the performance of a hybrid pre-adsorption/membrane filtration treatment system used was affected by the capacity of the unmodified, natural laterite and membrane type.

5. Recommendation

To mitigate the occurrence of fluorosis from drinking water in the Bongo District the following recommendations are made:

- Install, test further and optimize decentralized on-site community hybrid, pre-adsorption plus ultrafiltration defluoridation systems using local material with enhanced adsorption capacity on selected boreholes. These would be managed by institutions already in place.
- Where power from the grid is unavailable or unreliable, solar water treatment systems recently tested in Tanzania (Shen et al. [14]) could be installed complemented with a reservoir for water storage and distribution.

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