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Utilisation of Agro-Wastes as Adsorbents for Fluoride and Co-Existing Ions Adsorption from Groundwater Sourced from Bongo, Ghana

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Abstract

Fluoride (F) contamination of groundwater through natural or anthropogenic activities presents health concerns to communities that depend on it as a source of drinking water. This study examined the levels of F^- and co-existing ions present in groundwater sourced from Bongo, a community in Ghana. The adsorption efficiencies and capacities using agro-waste (millet stocks and groundnut shells) for the removal of these ions from the groundwater were also assessed. The findings revealed that the mean concentration of F^- is 2.24 mg/L, which exceeded the permissible standard of WHO/GSA. The presence of co-existing ions in the groundwater with their mean concentration were 22.4, 45, 56.48, and 18.08 for Cl^-, SO_4^{2-}, PO_4^{3-} Mg^{2+}, K^+, respectively. The results showed that the adsorbents could effectively remove fluoride from the groundwater, with maximum adsorption capacities (and removal efficiencies) of 0.021 mg/g (37.95%), 0.016 mg/g (28.10%) and 0.015 mg/g (26.30%) for charred millet, groundnut husk and uncharred millet, respectively. Similarly, the adsorbents were able to remove the co-existing ions at varying adsorption efficiencies. In terms of behaviour, the fluoride adsorption was initially quick (2 h) followed by slow adsorption. The adsorption kinetic of the fluoride removal best fitted pseudo-second order model. Notably, commercially sourced anthracite exhibited similar behaviour but superior performance relative to the agro-waste adsorbents used for the study. The findings suggest that these abundant agro-waste adsorbents are promising and sustainable materials for fluoride and co-existing ions removal from contaminated groundwater.

Keywords

Defluoridation, Biomass, Adsorption Capacity, Fluoride Ion, Adsorption, Bongo

1. Introduction

Water is one of the most important resources for the sustainability of life on Earth, however, its quality, quantity, and accessibility are threatened by several factors, such as population growth, climate change, pollution, and overexploitation (Akhtar et al., 2021; du Plessis, 2023). Globally, millions of people depend on groundwater as a major source of fresh/potable water, especially in rural and arid areas, where surface water is scarce or unreliable (Connor & Miletto, 2022; Rohde et al., 2024). However, its vulnerability to contamination/pollution by biogenic and anthropogenic means affects its suitability for drinking and other purposes (Olatunde et al., 2022; Ghebremedhin & Gupta, 2023). Fluoride, a naturally occurring element in rocks and soils, has been identified as one of the major contaminants of groundwater across the globe (Mukherjee & Singh, 2018; Chaudhuri et al., 2024). Studies have shown that fluoride can dissolve into groundwater through weathering, leaching, and geochemical reactions, depending on the pH, temperature, redox potential, and mineral composition of the aquifer (Al Sabti et al., 2023). Additionally, it can also enter groundwater from human activities, such as fertilizer use, industrial effluent discharge, and mining (Abugri & Pelig-Ba, 2011). Notably, the presence of fluoride in groundwater offers two opposing effects on human health depending mainly on the level of concentration. At low concentrations, it is considered a beneficial element for human health, as it can prevent dental caries and strengthen bones. Contrarily, the high concentration or excessive intake of fluoride can cause adverse effects such as dental and skeletal fluorosis, brittle bones, cancer, infertility, and brain damage which are irreversible and debilitating (Everett, 2011; Prasad et al., 2025).

Currently, fluoride contamination/pollution of groundwater is noted as a global health problem, affecting millions of people in more than 60 countries, especially in Africa, Latin America, and Asia (de Carvalho Costa et al., 2024). Notably, the most affected regions are the arid and semi-arid zones, where groundwater (with fluoride concentrations that exceed 10 mg/L) is used as the main drinking water. Though World Health Organization (WHO) recommended guideline value for fluoride in drinking water is 1.5 mg/L (to be considered safe and optimal for human health) (Fawell, 2006; Alkan et al., 2008; WHO, 1997; WHO, 2011). However, this value may vary depending on the climate, diet, and water consumption of the population (Fawell, 2006; WHO, 2011; Craig et al., 2015). For instance, in hot and dry regions, where water intake is higher, a lower value of 1.0 mg/L may be more appropriate (Craig et al., 2015; Araya et al., 2022). Overall, defluoridation or the removal of fluoride from water (either surface or groundwater) to make it safer for millions of people at a lower cost is drawing a lot of attention due to its health implications (Mei et al., 2019). This has led to the development and application of various methods for fluoride removal from water. Broadly, these methods can be classified into three groups: chemical process, membrane process (nanofiltration, reverse osmosis, and electrodialysis), and adsorption techniques (Azari et al., 2015). It is worth noting that the adsorption process offers more satisfactory results for

fluoride removal from aqueous solution as compared with other techniques. This is because the adsorption process has higher operational efficiency, low operating cost, reusability, and reliability (Ali & Gupta, 2007; Fomina & Gadd, 2014).

Generally, adsorption is a surface phenomenon, where ions (called adsorbate) are attached to the surface of a solid material (called adsorbent), by physical or chemical forces (Rathi & Kumar, 2021; Han et al., 2021). The adsorption capacity, efficiency, and selectivity of an adsorbent depend on its surface properties, such as surface area, pore size, charge, functional groups, and affinity for the ions (Gusain & Bux, 2021). In the area of defluoridation/fluoride removal from water, several adsorbents such as activated carbon, alumina, clay, zeolite, bone char, metal oxides, hydroxides, sulphides, and polymers (synthetic and natural), have been examined over the years (Alkan et al., 2008; Ali et al., 2020). Unfortunately, most of these adsorbents have some inherent limitations such as low adsorption capacity, poor stability, inaccessibility, high cost, or difficulty in regeneration and disposal (Ali et al., 2020; Younas et al., 2021). Therefore, the need to find an alternative or develop novel and efficient adsorbents for fluoride removal from water, which can overcome these limitations and still meet the technical, economic, and environmental requirements is warranted.

Currently, the use of agro-waste to produce alternative adsorbent is gaining much attention since its potential promises to offer several advantages such as low cost, high availability, environmental friendliness, and value addition (Gupta et al., 2009; Aragaw & Bogale, 2021; Hamad & Idrus, 2022). Technically, agro-waste materials contain various organic compounds (hemicelluloses, cellulose, and lignin) that have the potential to bind or adsorb inorganic contaminants to the surface of the agro-waste adsorbent (Kanamarlapudi et al., 2018; Ahmad & Zaidi, 2020; Mathew et al., 2023). These materials can be converted into adsorbents through various thermal, physical, and chemical treatments to modify their surface properties and enhance their adsorption performance (Asim et al., 2020; Irshad et al., 2024). Additionally, agro-waste materials can be combined with other materials such as metal oxides, hydroxides, or sulphides, to form composite adsorbents, which can improve their adsorption capacity and selectivity for fluoride (Wei et al., 2022; Jaiswal et al., 2025).

In rural areas within Ghana (including the study area), abundant agro-wastes (e.g., rice husk, coconut, millet stock, and groundnut shells) which are considered an environmental nuisance are generated annually. Managing this form of waste is problematic to the community, hence exploring the potential of using them as adsorbents in fluoride removal from groundwater, which serves as fresh water for the rural communities in Ghana, is warranted. Particularly, this work examined the adsorption efficiency, capacity, and dynamics of using groundnut husks and millet stocks (charred and uncharred) in removing fluoride from groundwater sourced from Bongo within the Upper East Region of Ghana. The findings of this study will expand the scope of existing literature on the treatment of fluoride-rich groundwater from Bongo and provide valuable knowledge that could enhance the

quality of drinking water in the community.

2. Materials and Methods

2.1. Description of the Study Area

Bongo is one of the nine districts within the Upper East Region of Ghana. The study area lies within latitude 10˚54'28" north and longitude 0˚48'29" west. Bongo District has a total area of 459 square kilometers, and shares borders with Burkina Faso to the east and north, the Bolgatanga municipal district to the south, and the Kassena-Nankana district to the west (Abugri & Pelig-Ba, 2011; Ashong et al., 2024). Geologically, the district is situated in the Birimian Supergroup with the Bongo granitoids as its predominant rocks. The area is mostly covered by monzonite and granitoids that are rich in K-feldspar (Bongo granite) (Abitty et al., 2016; Ashong et al., 2024). The district is one of the driest in Ghana, and its topography is mainly flat and low-lying, with outcrops of granite rocks. In addition to protruding throughout the landscape, granite rocks are a source of materials used in the building industry (Ashong et al., 2024).

The area experiences a single-season precipitation pattern that begins in May/June and ends in October/November. Between June and September is when the most rainfall occurs with an annual mean of 935 mm (the yearly rainfall ranges from 600 to 1400 mm) (Ashong et al., 2024). Groundwater in the Bongo area is extracted from fragments of bedrock, typically at drilling depths of 40 to 60 meters. The aquifer flows from north to south and is limited by the amount of mica and clay it contains. It has high static water levels (2 - 16 m below ground level, m b.g.l.) (Ashong et al., 2024). The geography and thickness of the weathered mantle affect the aquifer's supply of water (Gómez-Escalonilla et al., 2022; Ashong et al., 2024).

2.2. Materials and Charring Process

Four different materials (**Table 1**); Millet stock (**Figure 1(A)**), groundnut shells (**Figure 1(B)**), charred millet husk (**Figure 1(C)**) and commercially sourced anthracite (**Figure 1(D)**), were used as adsorbents for the study. The millet stocks and groundnut shells were washed (to remove dirt) and sun-dried for 24 h before their usage. Moreover, a portion of the sun-dried millet stocks was charred whilst the groundnut shells were crushed a bit to prevent them from floating when in contact with the water sample.

Table 1. Raw materials and their sources.

Raw Material	Sources
Millet stocks	Wale wale
Groundnut shells	Wale wale
Charred millet stocks	Locally produced
Anthracite	Commercial

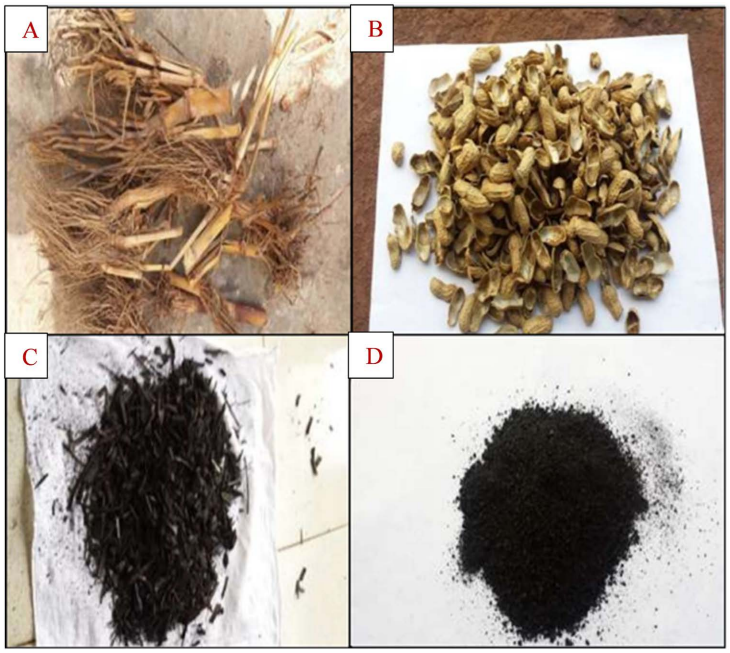

Figure 1. Biomass feedstocks used as raw materials: (A) Millet Stock; (B) Groundnut shells; (C) Charred millet; (D) Commercially sourced anthracite.

Charring Process

Figure 2 shows the equipment built locally for the carbonisation/charring of the millet stock. The carbonisation reactor was designed to have a low oxygen environment within, but able to withstand high temperatures. Hence, the bottom of the reactor was perforated to regulate the inflow of oxygen into the reactor since it is a closed system. For the charring process, the sundried millet stocks were fed into the reactor and covered. The reactor containing the biomass was fired on a traditional stone stove with firewood for about 10 min to avoid the production of ash. The charred biomass was then taken out of the reactor and spread on a pan to cool down.

Figure 2. Locally designed carbonisation/charring reactor.

2.3. Adsorption Procedure

For the adsorption study, 20 g of the adsorbents were contacted with 500 ml of the groundwater in plastic bottles and agitated on rollers for 24 h. Samples were drawn from the mixture periodically to examine the adsorption efficiency with time. These samples drawn were filtered and analysed using Atomic Absorption Spectrometer (AAS) Equation (1) and Equation (2) were employed for the calculation of the adsorption capacity (Qe) (mg/g) and the percentage of fluoride removal, respectively. The studies were conducted thrice to ensure that the analytical data obtained were repeatable.

Adsorption Capacity

The adsorption capacities of the agro-waste adsorbents used were determined by using Equation (1) (Okorocha et al., 2021; Al-Yosofy et al., 2024; Huang et al., 2023).

$$Qe = V\left(Co - Ce\right)/W \tag{1}$$

where: Qe in the equation is adsorption capacity in mg/g.

Co = initial concentration of fluoride ion in solution before adsorption.

Ce = final concentration of fluoride ion in solution after adsorption.

V = volume of solution in mL.

W = dry weight of biomass in g.

Adsorption Efficiency

The efficiency of fluoride ion removal by the biomass was determined for comparison. This was determined using Equation (2) (Al-Yosofy et al., 2024; Huang et al., 2023).

$$R = \left(Co - Ce/Co\right) \times 100 \tag{2}$$

where: R = efficiency of fluoride removal.

Study of Adsorption Dynamics

The adsorption kinetics or dynamics of the various biomasses were examined. Generally, the adsorption kinetics study explored the amount of adsorbate adsorbed as a function of time variation. This study enables one to determine the speed in the adsorption process. In this study, two commonly used models were employed; namely, pseudo-first-order (PFO) and pseudo-second-order (PSO), as shown in Equation (3) and Equation (4) (Lin & Wang, 2009; Amadi et al., 2024).

Pseudo-first-order (PFO) Rate Equation is expressed as:

$$\ln\left(Qe - Qt\right) = \ln Qe - k_1 t \tag{3}$$

Pseudo-Second-Order Rate Equation is expressed as:

$$dQt/dt = k_2\left(Qe - Qt\right)^2 \tag{4}$$

where Qe and Qt are the sorption capacities (mg/g) at equilibrium and at time t, respectively, and k is the rate constant of pseudo-second order sorption, g/mg. minutes). For the boundary conditions, $Qt = 0$ to Qt at $t = 0$ to t.

3. Results and Discussion

3.1. Concentrations of Fluoride and Co-Existing Ions

Table 2 shows the mean concentrations of fluoride and co-existing ions (Cl^-, SO_4^{2-}, PO_4^{3-}, Mg^{2+}, K^+) present in the groundwater from Bongo and their respective WHO/GSA permissible standards. The result indicates that the mean concentration of fluoride in the groundwater is 2.24 mg/L, which was higher than the GSA/WHO recommended thresholds of 1.0/1.5 mg/L. The higher mean concentration of F^- in the study could be attributed to two major sources, geologic and anthropogenic (Ashong et al., 2024; Sunkari, 2010). For the geologic source, it could be because of the weathering of fluoride-bearing minerals that characterise the rocks within the Bongo area, whilst the anthropogenic activities may be linked to industrial discharges and agricultural practices that release fluoride into groundwater (Sunkari, 2010). It is worth mentioning that the fluoride concentration recorded herein is within the ranges (0.75 - 4.4 mg/L) reported on the groundwater from the same enclave by Apambire et al. (1997), Smedley et al. (1995), Firempong et al. (2013); Annan et al. (2021). This also validates the account that the high fluoride content of the Bongo water accounts for the dental fluorosis incidence recorded among school children (Firempong et al., 2013).

The mean concentrations of the co-existing ions, Cl^-, SO_4^{2-}, PO_4^{3-}, and Mg^{2+}, and K^+ were 22.4, 45, 56.48, and 224.89 mg/L, respectively. Comparatively, the mean concentrations of PO_4^{3-} and K^+ were above the GSA standards whilst those of Cl^-, SO_4^{2-} and Mg^{2+} were below. The presence of these co-existing ions supports the earlier claim that the geological weathering of the rocks is one of the major sources of the ions including the fluoride. That is, the ions are believed to be byproducts of dissolved mineral contents (such as chlorine byproducts, alkaline substances, bicarbonate, and sulphur materials and dissolved salts) (Kashyap et al., 2021; Rao et al., 2021; Ashong et al., 2024).

Table 2. Mean Concentrations of fluoride and co-existing ions in the groundwater from Bongo.

Ions	Mean Concentrations (mg/L)	*GSA standards (mg/L)	*WHO standards (mg/L)
F^-	2.24	1.00	
Cl^-	22.40	200	
SO_4^{2-}	45.00	200	
PO_4^{3-}	56.48	0.50	
Mg^{2+}	18.08	50	
K^+	224.89	12	

*Ghana Standard Authority (GSA) (2017) GS 175: Drinking Water—Specification.

3.2. Adsorption of Fluoride Ions from the Groundwater

Figure 3 and Table 3 show the results for the adsorption capacities and efficien-

cies of the agro-waste adsorbents for removing fluoride ions from the groundwater after 24 h, respectively. Figure 3 shows that the amount of F^- removed by charred millet, groundnut shells, and uncharred millet were 37.95, 28.1 and 26.3%, respectively. The relatively higher removal percentage exhibited by the charred millet compared with uncharred millet and groundnut shells is not surprising since the charring/calcination enhances the adsorption capabilities/efficiencies of the resulting product (Shaikhiev et al., 2024).

In terms of adsorption capacity, the results (as shown in Table 3) indicate that charred millet husk exhibited the highest adsorption capacity of 0.021 mg/g followed by charred groundnut shells (0.016 mg/g) and uncharred millet (0.015 mg/g). This trend was expected because the calcination/charring of organic materials enhances their adsorption capacities because of increased active sites/surface area and/or microspores responsible for the adsorption activities (Shaikhiev et al., 2024).

Expectedly, commercially sourced anthracite showed superior adsorption capacity and removal efficiency relative to locally sourced and charred materials. The difference can be linked to the conditions under which each product was produced, thus, anthracite had nearly perfect conduction for its production.

Table 3. Adsorption capacity and efficiency.

Adsorbent type	Adsorption capacity (mg/g)
Charred millet stock	0.021
Uncharred groundnut shells	0.016
Uncharred millet stock	0.015
Anthracite	0.032

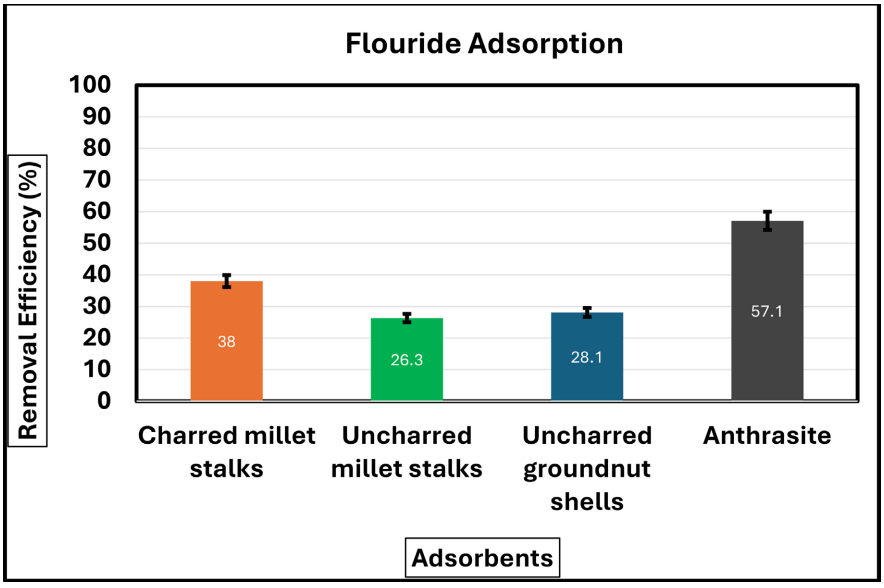

Figure 3. Fluoride adsorption efficiencies by the various adsorbents.

3.3. Adsorption of Co-Existing Ions

The removal efficiencies of the agro-waste adsorbents for co-existing ions from the groundwater were also examined after 24 h. Figure 4 and Figure 5 show the amount in percentage (%) of cations and anions adsorbed or removed from the groundwater by the adsorbents after 24 h, respectively. The cations adsorption efficiencies (Figure 3) exhibited by the various adsorbents indicate that the charred millet adsorbed the highest amount of Mg^{2+} (98.2%) and K^{+} (99.1%) ions, followed by groundnut husk (that adsorbed Mg^{2+} (97.2%) and K^{+} (80.0%) ions) and the uncharred millet (that adsorbed Mg^{2+} (60.0%) and K^{+} (0%) ions).

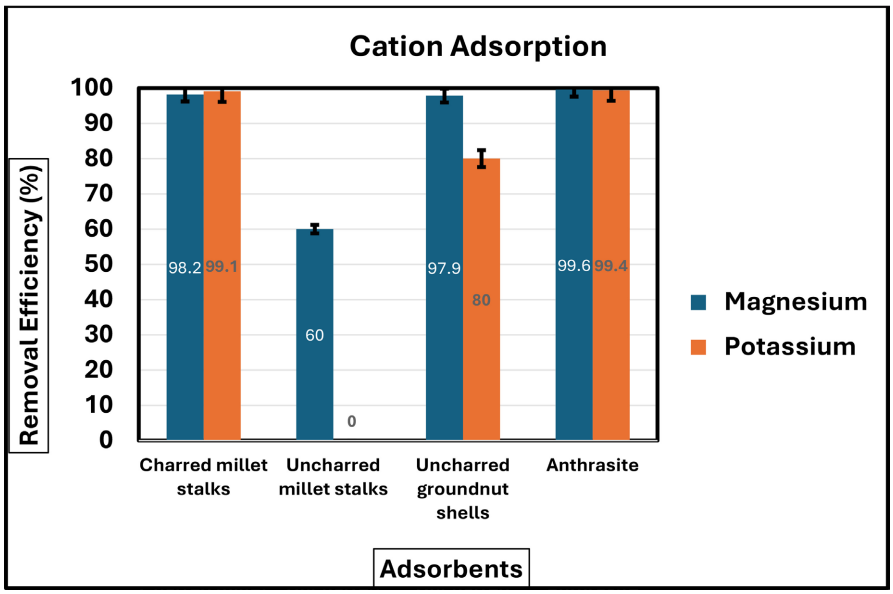

Figure 4. Cation adsorption efficiencies by the various adsorbents.

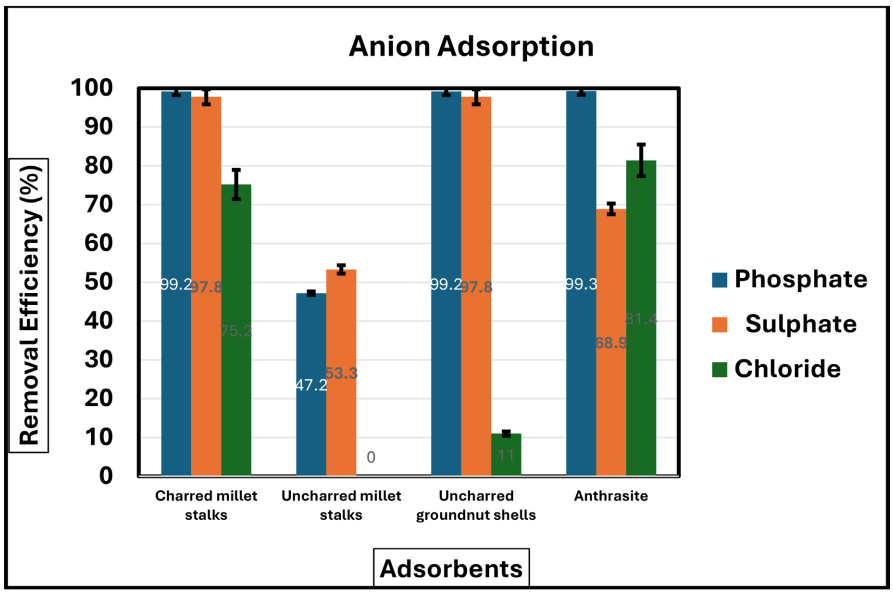

Figure 5. Anions adsorption efficiencies by the various adsorbents.

For anion adsorption, the results (Figure 4) showed that the amount (%) of SO_4^{2-}, PO_4^{3-}, and Cl^- ions adsorbed differed significantly with the various adsorbents. For SO_4^{2-} adsorption efficiency, charred millet adsorbed 97.8% followed by uncharred groundnut (97.8%) and uncharred millet (53.3%). The adsorption efficiency for Cl^- ions was 75.2% and 11.0% for charred and uncharred millets, respectively. Interestingly, there was no adsorption in the case of groundnut adsorbent but rather the Cl^- ion concentration was increased. Further study is required to unravel the reason underpinning this phenomenon. For PO_4^{3-} ions, both charred millet and groundnuts showed equal adsorption efficiency of 99.2% whilst uncharred millet exhibited 47.2% adsorption efficiency.

Generally, this result suggests two contrasting effects; (i) these agro-waste adsorbents could help the communities deal with their high conductivity issues associated with high levels of dissolved ions at lower cost and (ii) the presence and adsorption efficiency of co-existing ions is an indication that these ions can impact on fluoride removal hence the low adsorption efficiency for fluoride as shown Table 3.

3.4. Effect of Contact Time on Fluoride Removal or Adsorption

Due to the equilibrium-driven nature of the adsorption process, the period required to achieve equilibrium plays a critical role in the effective defluoridation process and the design of adsorption equipment (de Carvalho Costa et al., 2024). Therefore, the effect of the contact time on the defluoridation capability of the selected adsorbent (charred millet husks) was examined. The removal efficiency was measured at time intervals of 2, 4, 8, 12, and 24 h as shown in Figure 6. The results show that, in the first 2 h (120 min), fluoride adsorption was very quick (about 21.43% removal), suggesting rapid adsorption rates in the beginning and slower rates after that, as removal efficiency after 24 h was 37.95%. The observed rapid adsorption within the first 2 h could be attributed to the fact that the surface area or the adsorption sites were devoid of any solute molecules, resulting in substantial concentration differences between the bulk solution and the adsorbent surface. This concentration gradient facilitated the fast transfer of mass or the adsorption process onto the adsorbent surface (Al-Ghouti et al., 2004; Garcia-Reyes & Rangel-Mendez, 2010; Usman et al., 2020). In other words, the higher amount of F^- adsorbed within the first 2 h can be linked to the fact that at that moment there were a lot of free active sites which provided a greater surface area for adsorption. Additionally, as the adsorption of the fluoride progresses, the number of active surface areas decreases, hence a reduction in the rate of removal.

Generally, the overall removal percentage of F^- after 24 h seemed to be low in contrast to expectation. This is believed to be due to the presence of other cations/anions (especially Cl^-, SO_4^{2-}, and PO_4^{3-}) within the groundwater. Thus, these ions are believed to be acting as interfering ions by competing with F^- for active sites on the adsorbent (Amin et al., 2015). The presence of these competitive anions, thus, decreases the attraction between the F^- and active surfaces of biomass by posing a coulombic repulsive force which repels and decreases the

chances of the F^- interactions with the active sites of the biomass surface, hence the lower adsorption (Amin et al., 2015).

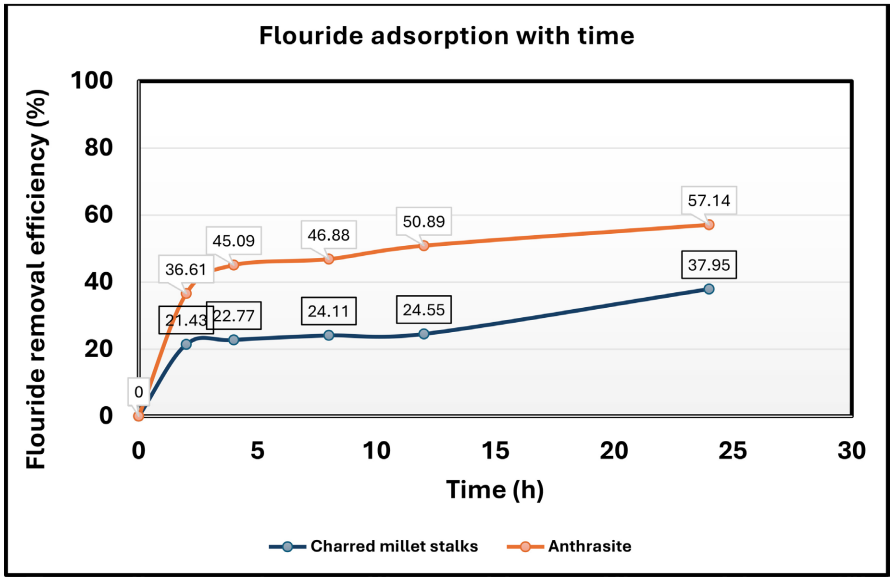

Figure 6. Charred millet stalks and anthracite adsorbents' fluoride removal efficiencies with time.

3.5. Adsorption Dynamics

The adsorption dynamics or kinetics of fluoride adsorption onto the charred millet adsorbent were investigated using two types of kinetic models for solid/liquid adsorption: pseudo-first order (PFO) and pseudo-second order (PSO) models. The fit of the two models for the kinetic data obtained from the experiment of fluoride adsorption is shown in **Figure 7(A)** and **Figure 7(B)**. **Table 3** summarizes the kinetic data for fluoride adsorption onto the adsorbent based on the two models. **Figure 7(A)** and **Figure 7(B)** show a linear relationship with an excellent correlation value. Thus, experimental data for the adsorbent could be fitted to a pseudo-first order kinetic model, but the pseudo-second order kinetic model ($R^2 > 0.9602$) provided the best fit. This indicates that fluoride adsorption over the charred millet adsorbent followed the pseudo-second order kinetic model throughout the entire adsorption process. This may mean that the rate-limiting phase is an absorption process involving electron sharing, valence forces, or exchange occurring between the adsorbate and the adsorbent (Srivastava et al., 2011; Mayakaduwa et al., 2016; Nayak et al., 2017). As a result, exchange on the adsorbent surface or existing electron sharing may aid in adsorption development (Neeti et al., 2024).

4. Conclusion

This study examined the adsorption or removal capacities, efficiencies, and dynamics of fluoride (F^-) and co-existing ions from groundwater (sourced from Bongo) using agro-waste (charred and uncharred millet and groundnut shells)

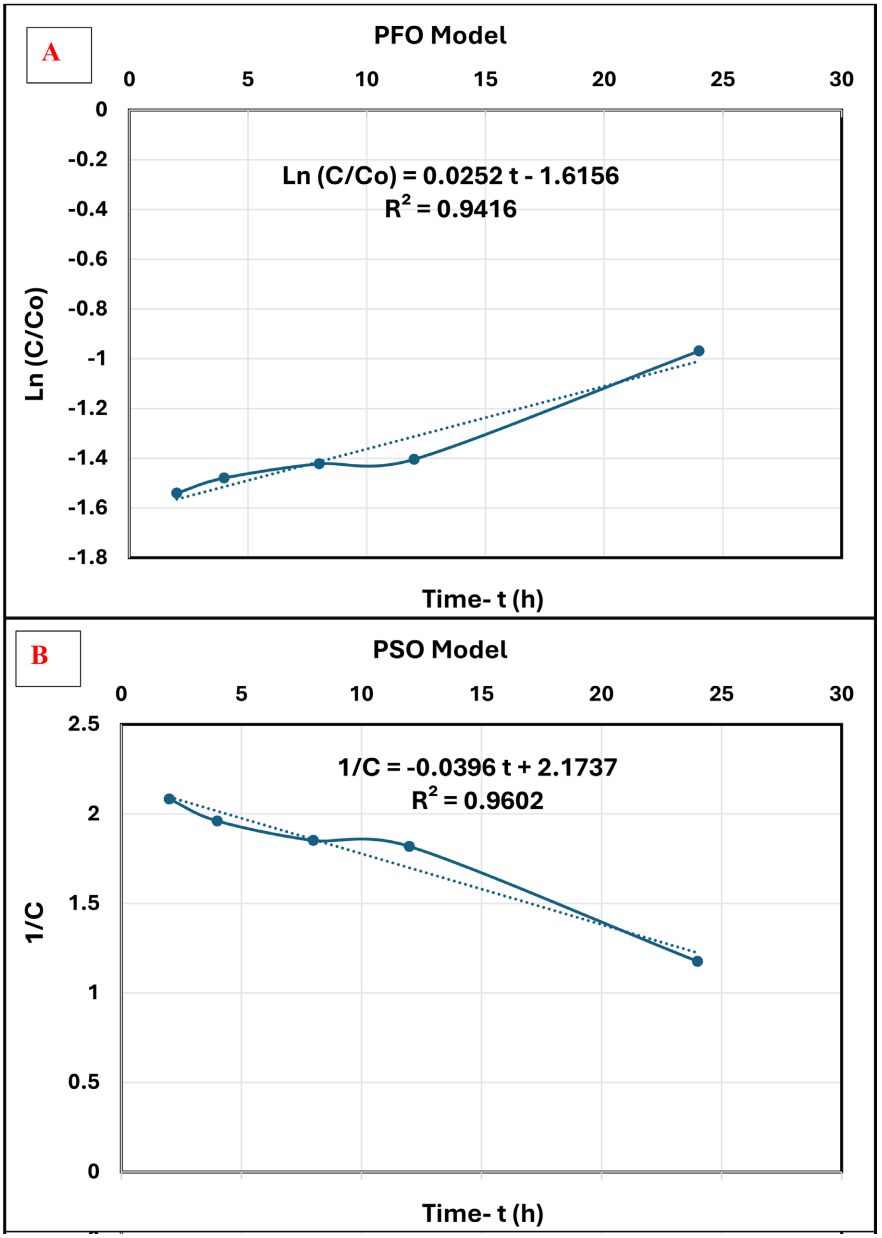

Figure 7. Plot of (A) Pseudo-first order and (B) Pseudo-second order on composite waste adsorbent.

adsorbents. The hydrochemical analysis revealed that the Bongo groundwater contained a high F^- concentration (2.24 mg/L), greater than the permissible WHO/GSA standard of 1.5/1.0 mg/L. The presence of co-existing ions: Cl^-, SO_4^{2-}, PO_4^{3-}, Mg^{2+}, and K^+ with respective mean concentrations of 22.4, 45, 56.48, and 224.89 mg/L, were recorded. The study showed that all the agro-waste adsorbents were able to remove fluoride and the co-existing ions at varying efficiencies and capacities with time. However, charred millet adsorbent demonstrated superior adsorption efficiency and capacity in the removal of fluoride and co-existing ions from the groundwater as compared with the uncharred groundnut shells and millet stalk. The results also showed that the contact time of the

adsorbent with the water influenced the adsorption rate, that is, the first 2 h of the adsorption was very active. Additionally, the finding revealed that the fluoride adsorption over the charred millet adsorbent followed the pseudo-second order kinetic model throughout the entire adsorption process. The adsorption behaviour and performance of the agro-waste adsorbents compared with the anthracite (commercially sourced adsorbent) revealed that the latter demonstrated similar behaviour but superior performance relative to the agro-waste adsorbents used for the study.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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