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Adsorption of Arsenic by Laterite, Sandstone and Shale in a Fixed-Bed Column

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Abstract

The removal of arsenic from water is essential for the protection of public health. To investigate the adsorption capabilities of laterite, sandstone, and shale for the removal of arsenic from aqueous solutions, column experiments were conducted. In this study, raw materials and heat-treated (calcined) materials were examined. The experiments assessed the influence of various parameters, including initial concentration, bed depth, and the effects of heat treatment. The findings revealed that the breakthrough curves were influenced by the initial concentration of arsenic, the depth of the bed, and the type of material used. For an initial arsenic concentration of 5 mg/L, columns containing 85 cm of calcined laterite, sandstone, and shale produced volumes of 7460 ml (1492 min), 3510 ml (702 min), and 4400 ml (880 min) of water with arsenic levels below 0.01 mg/L, respectively. These calcined materials demonstrate significant potential for the effective removal of arsenic from water.

Keywords

Geomaterials, Adsorption, Arsenic, Water

1. Introduction

Arsenic is commonly found in nature and is well known for its toxic properties ([1]-[3]). In certain parts of the world, arsenic contamination of groundwater poses a significant public health concern [4], since groundwater typically serves as the main source of drinking water for many populations. In groundwater, arsenic exists primarily in two oxidation states, + III and + V [5]. The primary

chemical forms of arsenic found in water are oxyanions or neutral inorganic compounds that contain soluble inorganic arsenic, which is highly toxic to living organisms, including humans and animals, when present in excessive amounts [5]. Prolonged ingestion can result in chronic arsenic poisoning, known as arsenicism. The range of arsenic's toxicity to human health encompasses issues from skin lesions to various cancers affecting the brain, liver, kidneys, and stomach and is influenced by the specific form of arsenic present. Specifically, arsenic in oxidation state III is significantly more toxic than arsenic in oxidation state V. Skin conditions resulting from chronic environmental exposure, such as hyperkeratosis in the palms of the hands and feet, can manifest with arsenic concentrations in water exceeding 0.01 mg/L [6].

In Africa, particularly in Burkina Faso, several aquifers with elevated levels of arsenic have been identified, believed to originate from eruptive and metamorphic rocks. This geological formation, known as Birimian, extends into neighboring countries such as Ghana, Mali, Guinea, Côte d'Ivoire, and Niger, indicating that the situation observed in northern Burkina Faso may not be an isolated event. In Côte d'Ivoire, research by Mangoua-Allali [7] has shown the presence of arsenic in well water in the Akouédo region, with concentrations exceeding 0.01 mg/L. This water, despite its contamination, is still used by the local population, which can pose health risks.

There are various methods to remove arsenic from drinking water, including conventional techniques such as precipitation using iron or aluminum salts, lime, manganese oxides, and oxidation processes. Additionally, membrane technologies such as reverse osmosis, nanofiltration, and electrodialysis are also employed. Biological methods, including biological oxidation, biomethylation, and biosorption, present further options ([8]-[10]). Although these arsenic treatment technologies have been successfully tested in contaminated water sources, many of them are prohibitively expensive and may require specialized personnel for implementation. It is essential to look for technologies that are innovative, straightforward, effective, cost-effective, require minimal maintenance, and utilize naturally available materials. Emerging adsorption technologies that employ natural supports such as clays, sand coated with iron and manganese oxides, and laterites are proving to be promising methods for treating arsenic-contaminated water ([11]-[13]).

Côte d'Ivoire has an abundance of materials, such as laterite, shale, and sandstone, which offer several advantages, including low acquisition costs, high availability, and the presence of oxides such as iron hydroxides and alumina ([14] [15]) that can help reduce arsenic levels. Previous studies investigated the adsorption of arsenic in water using these materials in a batch reactor setup [16]. The results indicated that arsenic concentrations can be effectively reduced by using laterite, sandstone, and shale in batch mode. However, more research is needed to explore this phenomenon in a continuous reactor (dynamic mode) to better understand the dynamic processes, as these factors may limit the practical

application of these materials for water treatment. The objective of this study is to examine the feasibility of using laterite, sandstone, and shale to remove arsenic adsorbed from drinking water. Research focuses on the effects of initial arsenic concentration, bed depth, and thermal treatment of the materials on arsenic adsorption through column experiments using laterite, sandstone, and shale powders.

2. Material and Methods

2.1. Preparation of Adsorbent Beds

Laterite (9˚35'N and 5˚23'W), sandstone (5˚21'07'N and 3˚56'30'W) and shale (6˚39'15'N and 4˚58'33'W) samples were collected in Côte d'Ivoire. Each type of material was crushed separately, reduced using a Retsch RS 100 crusher, washed, and then dried for 24 hours. The crushed materials were sieved using Saulas sieves (NF.X 11.501) (see Figure 1). Fractions with a particle size of 2 mm were retained to pack the column reactors, following the methodology established by Ezzati *et al.* [17]. Furthermore, 2 mm size fractions of each adsorbent were calcined at 400˚C for laterite and 300˚C for shale and sandstone, corresponding to the optimal calcination temperatures identified by Koua-Koffi *et al.* [18] [19] for column tests.

Geomaterials	Raw	Crushed	Crushed and calcined
Laterite			
Sandstone			
Shale			

Figure 1. Different materials used.

2.2. Sampling and Type of Water

The water samples were collected from wells in the village of M'Badon, situated at 5˚19' and 5˚22'N and 3˚55'N and 3˚56'W, near the former rubbish dump at Akouédo in Côte d'Ivoire. Before each sampling, the drums were rinsed three

times with the water intended for sampling. In situ measurements of pH and temperature were taken using a multi-parameter device. The collected water samples were then sent to the laboratory, where the total arsenic content was analyzed using an OPTIMA 2100 dual-view spectrometer (ICP-OES 2100 DV). To adjust the levels of arsenic in the water, a standard arsenic (III) solution was added to each sample can, adhering to the dilution protocols to achieve an arsenic concentration ranging from 1000 to 5000 µg/L for the column experiments.

2.3. Description of the Experimental Setup and Column Feed

The experimental setup included PVC columns with an internal diameter of 40 mm and a height of 95 cm, along with a feed barrel, a filter collection tank and a drainage system located at the lower base (Figure 2). Each column was filled from bottom to top with 5 cm of gravel (crushed granite) topped with a geotextile, followed by 30 to 85 cm of adsorbent material. The gravel acted as a drainage layer, while the adsorbents functioned as a filtration medium. The purpose of the geotextile was to prevent the loss of fine particles and to block the water outlets. Contaminated water was introduced into the columns at a flow rate of 5 ml/min. The experiment was deemed complete when the arsenic concentration (Ct) at the column outlet reached 90% of the initial concentration (C0).

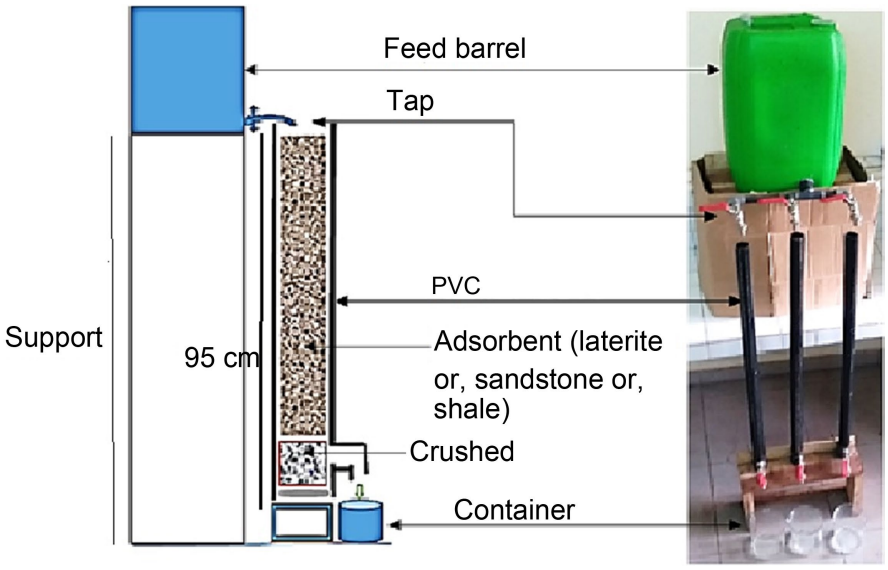


Figure 2. Presentation of the experimental setup used.

2.4. Parameters Influencing Column Adsorption

During raw material testing, we examined how the initial concentration, the height of the materials and the heat treatment of the materials affected the quality of arsenic removal from water. Regarding the effect of the initial concentration, we packed four columns with 30 cm of each type of raw adsorbent material, which were used to treat water with varying initial arsenic concentrations: 1 mg/L, 3 mg/L, 4 mg/L, and 5 mg/L. To investigate the impact of material height, we

identified the highest concentration that showed the least reduction during the first treatment trial and evaluated the influence of different material heights on the purification performance of the columns. This involved filling three columns for each material to heights of 30 cm, 40 cm and 85 cm, respectively. Additionally, heat-treated materials were used in the treatment tests. All experiments were carried out at a feed rate of 5 ml/min.

The total adsorbed As quantity or the maximum of column capacity (q_{total}), in mg, for a given feed concentration and flow rate ([20]), is provided in the following (Equation (1)):

$$q_{\text{total}} = QC_0 \frac{A}{1000} = \frac{F}{1000} \int_{t=0}^{t=t_{\text{total}}} \left(C_{\text{Ads}} \mathrm{d}t \right) \tag{1}$$

Here, t_{total} represents the overall flow duration in minutes, Q symbolizes the flow rate in ml/min, A denotes the total area beneath the breakthrough curve, C_0 is the initial concentration of the adsorbate and q_e (mg/g) is the column adsorption capacity. The theoretical total of As adsorbed by the column is determined by the Equation (2)

$$m_{\text{total}} = \left(Co * Q * t_{\text{total}} \right) / 1000 \tag{2}$$

With knowledge of the total mass of adsorbed ions and the overall sorbent capacity, one can compute the total percentage of As removed using Equation (3)

$$\%\text{Asremoved} = \left(q_{\text{total}} / m_{\text{total}} \right) * 100\% \tag{3}$$

2.5. Statistical Processing

Statistical analyzes were performed with R version 3.3.2, with a significance threshold established at $p < 0.05$. The univariate statistical tests used for data analysis included the Shapiro-Wilk test, Kruskal-Wallis test, t-test, Mann-Whitney test, and Wilcoxon test. The Shapiro-Wilk test was used to assess the normality of the data distribution. Comparisons of arsenic concentrations measured in the filtrates from different materials were performed using both the nonparametric Kruskal-Wallis test and the parametric ANOVA test. Additionally, arsenic concentration comparisons between the filtrates of raw and calcined materials were carried out using the nonparametric Mann-Whitney tests.

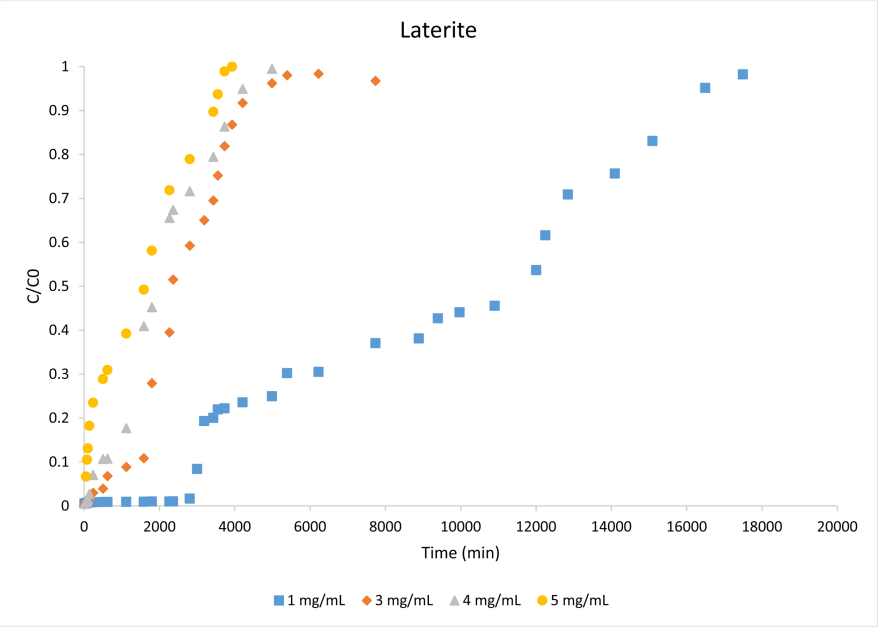
3. Results and Discussion

3.1. Effect of the Initial Concentration of Adsorbate

The effects of varying the inlet concentration of arsenic ions, set at 1, 3, 4, and 5 mg/L, while maintaining a consistent bed height of 40 cm and a flow rate of 5 mL/min, are illustrated by the breakthrough curves presented in **Figure 3**. It is evident that an increase in the inlet arsenic ion concentration results in a steeper slope of the breakthrough curve across all materials, leading to a reduced volume of treated liquid before the adsorbent bed reaches saturation. For laterite, the breakthrough curves for concentrations of 3, 4, and 5 mg/L exhibit similar

behavior, maintaining this trend up to a treated volume of less than 20,000 mL, which corresponds to 4000 minutes, before reaching saturation. On the contrary, for an initial concentration of 1 mg/L, laterite achieves saturation after a volume four times greater than that of the other concentrations tested. In the case of sandstone, the curve of 1 mg/L allows a larger volume of water to be treated compared to the other three concentrations. For shale, two distinct families of curves emerge. The first family pertains to concentrations of 3, 4, and 5 mg/L, with all curves reaching saturation after treating a volume of 15,000 mL (3000 minutes). The second family, represented by a concentration of 1 mg/L, has a gentle slope of less than 10%. Here, shale saturation occurs at 7735 minutes, corresponding to a treated volume of 30,000 mL. In particular, for the concentrations 3, 4, and 5 mg/L, the residual concentration did not decrease to 0.01 mg/L.

These findings indicate that a higher initial arsenic concentration leads to a greater driving force for mass transfer, causing the adsorbent to reach saturation more quickly. This results in a reduction in both the breakthrough time and the volume of water treated ([21]-[23]). On the contrary, a decrease in arsenic concentration to 1 mg/L produced a later breakthrough curve, with treated water volumes being 9000 mL, 2200 mL and 3000 mL for laterite, sandstone, and shale, respectively. These concentrations align with the WHO guideline value for drinking water at around 0.01 mg/L. This phenomenon may be attributed to the availability of active sites on the surface of the adsorbent in relation to the arsenic levels in the solution. In summary, the initial concentration of adsorbate ions has a significant impact on the breakthrough curve and overall column performance, highlighting the importance of carefully selecting the operating concentration to achieve optimal removal efficiency and breakthrough time [24].



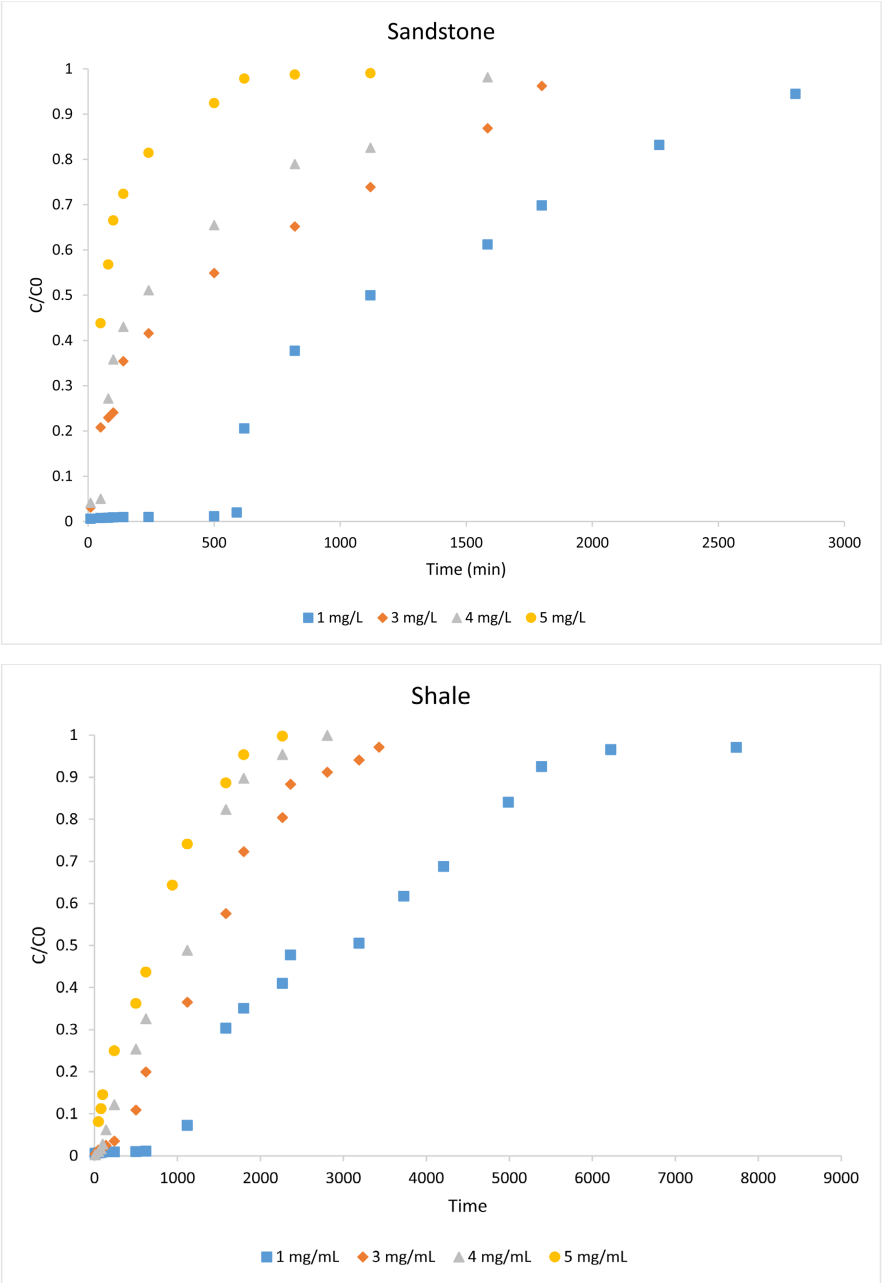


Figure 3. Effect of initial concentration on the breakthrough curve.

3.2. Effect of the Column Bed Height

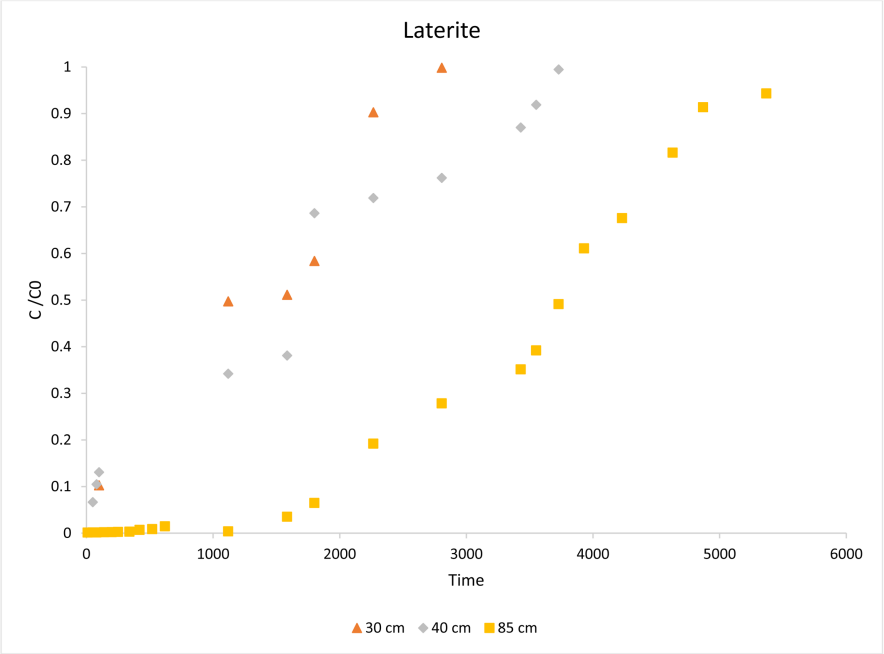
The influence of bed height on column performance is demonstrated by the saturation times or volumes depicted in the breakthrough curves (see **Figure 4**). For laterite, columns with material heights of 30 cm and 40 cm display similar trends, both reaching saturation after processing volumes below 20 L (approximately 4000 min). On the contrary, the column with a height of 85 cm attains saturation after 5369 minutes, corresponding to a treated volume exceeding 30,000 mL (see table).

The behavior of sandstone is similar, with clear initial slopes observed during

the treatment process. In particular, the 8cm column allows for a much larger volume of water treatment compared to the other two heights. In the case of shale, columns containing 30 cm and 40 cm of material reached saturation after treating a volume of less than 15,000 mL (3000 min). On the contrary, the column with 85 cm of material reached saturation after treating 20,000 mL (4000 min). However, the concentration of arsenic in the filtrates did not decline to 0.01 mg/L. During the treatment test, significant differences in arsenic concentrations were observed in the filtrates from the various crude laterite columns (Kruskal-Wallis test: $p < 0.05$). In particular, a significant difference was identified between heights of 30 cm and 85 cm, as well as between 40 cm and 85 cm (Mann-Whitney test: $p < 0.05$). The Kruskal-Wallis test ($p < 0.05$) also revealed differences in filtrates from columns packed with raw sandstone, specifically between columns of 30 cm and 85 cm (Mann-Whitney test: $p < 0.05$). ANOVA analysis showed a significant variation in residual arsenic concentrations between different reactors for filtrates treated with raw shale ($p < 0.05$), with differences observed between the 30 cm and 85 cm filtrates, as well as between the columns 40 cm and 85 cm ($p < 0.05$). In general, using bed heights of 30 cm, 40 cm, and 85 cm resulted in an increase in the breakthrough time and a higher percentage of arsenic removal as the volume of water treated increased (**Table 1**).

This phenomenon can be attributed to the increased surface area and the greater number of binding sites available for adsorption as the height of the bed increases [25].

The enhancement of the height of the bed for the removal of arsenic leads to a larger number of accessible sites for adsorption, as well as an extended contact time between the arsenic ions and the adsorbent material [26].



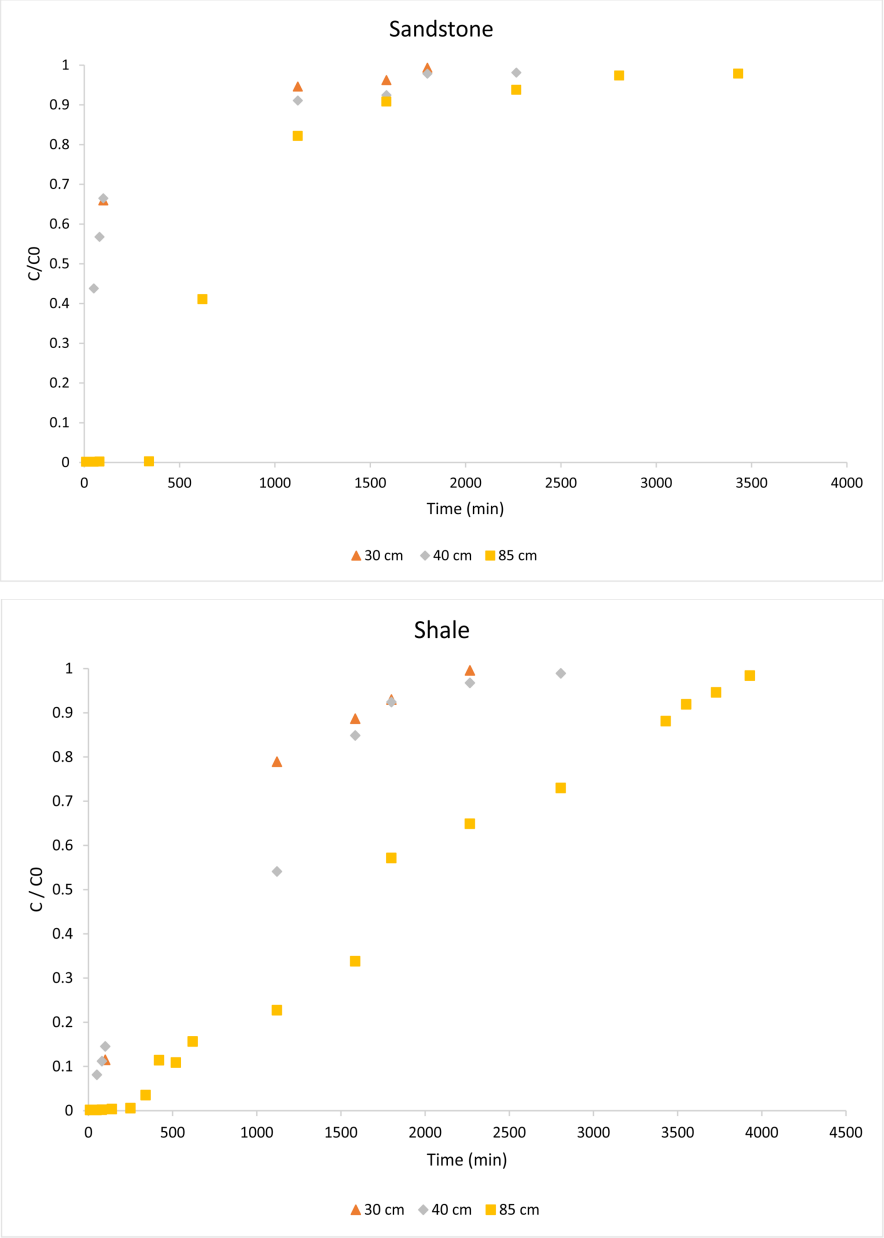


Figure 4. Effect of bed height on the breakthrough curve of water treated by laterite, sandstone and shale.

Similar findings were documented by Tan *et al*. [27], Han *et al*. [28], and Sadaf and Bhatti [29], who observed comparable results during the adsorption of basic dye using activated carbon derived from oil palm husks, the adsorption of methylene blue using phoenix tree leaf powder in a fixed bed column, and the in-column removal of indosol from groundnut husk, respectively. Roy *et al*. [30] and Maji *et al*. [31] also reported similar results when removing arsenic from a column using sugarcane charcoal and natural rock coated with iron oxide, respectively. Higher bed columns demonstrate improved performance, as noted in the literature [32]. Furthermore, the slower exhaustion of the adsorbent bed is advantageous for the

effective removal of arsenic ions from water. Therefore, greater bed heights are preferred, as they offer a longer duration of effective adsorption [33].

Table 1. Different parameters for fixed-bed adsorption under different operating conditions.

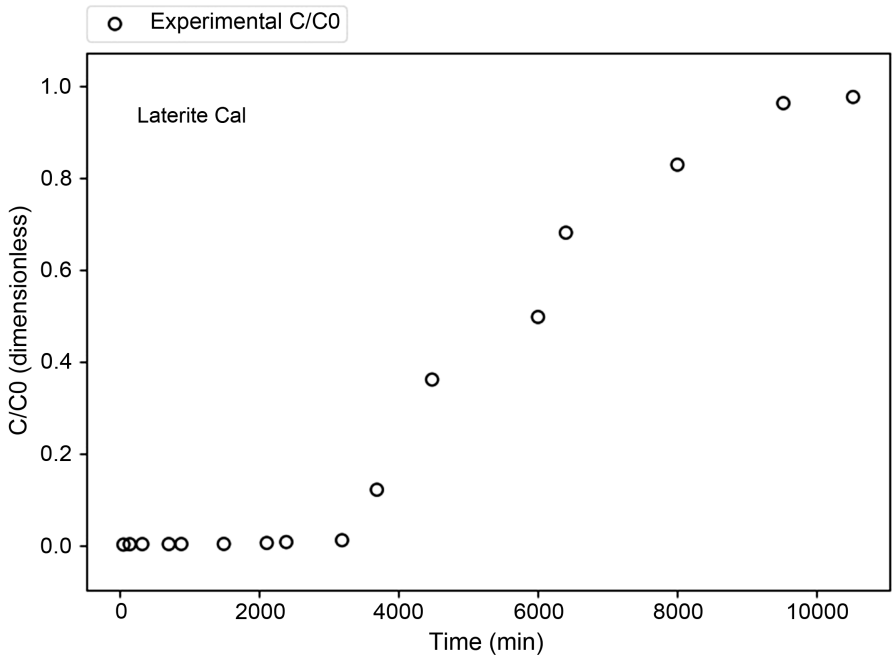
Experimental condition							V (mL)		
Materiaux	Initial concentration	Bed height (cm)	Flow rate (mL/min)	q_{total} (mg)	m_{total} (mg)	% Bed performance	V_b	V_T	q_e (mg/g)
Laterite	1 mg/L	40	5	49.13	263	56.17	14,030	87,460	0.12
	3 mg/L	40	5	40	116.037	34.53	2500	38,675	0.10
	4 mg/L	40	5	41.97	99.78	41.97	1000	24,945	0.10
	5 mg/L	30	5	19.38	56.65	34.21	194	14,030	0.06
	5 mg/L	40	5	19.5	56.65	34.3	247	18,645	0.5
	5 mg/L	85	5	88.78	134.235	55.24	8000	26,845	0.09
Calcined Laterite	5 mg/L	85	5	145.28	263	66.14	15,500	52,600	0.15
Sandstone	1 mg/L	40	5	6.81	70.15	48.52	2950	14,030	0.02
	3 mg/L	40	5	9.40	27	34.83	350	9000	0.02
	4 mg/L	40	5	9.09	31.7	28.67	250	7925	0.02
	5 mg/L	30	5	5.21	45	11.59	156	7000	0.02
	5 mg/L	40	5	7.22	56.65	12.74	203	11,330	0.02
	5 mg/L	85	5	22.43	85.75	26.16	2500	17,150	0.02
Calcined Sandstone	5 mg/L	85	5	99.85	200	49.93	8460	40,000	0.09
Shale	1 mg/L	40	5	15.49	38.679	40.06	4750	38,675	0.04
	3 mg/L	40	5	21.87	51.45	42.51	1200	17,150	0.06
	4 mg/L	40	5	20.97	56.12	37.37	650	14,030	0.06
	5 mg/L	30	5	19.38	56.65	34.21	181	11,330	0.07
	5 mg/L	40	5	19.5	56.65	34.45	185	14,030	0.07
	5 mg/L	85	5	48.36	98.235	49.23	2800	19,645	0.05
Calcined shale	5 mg/L	85	5	122.2	223	54.8	10,000	44,600	0.11

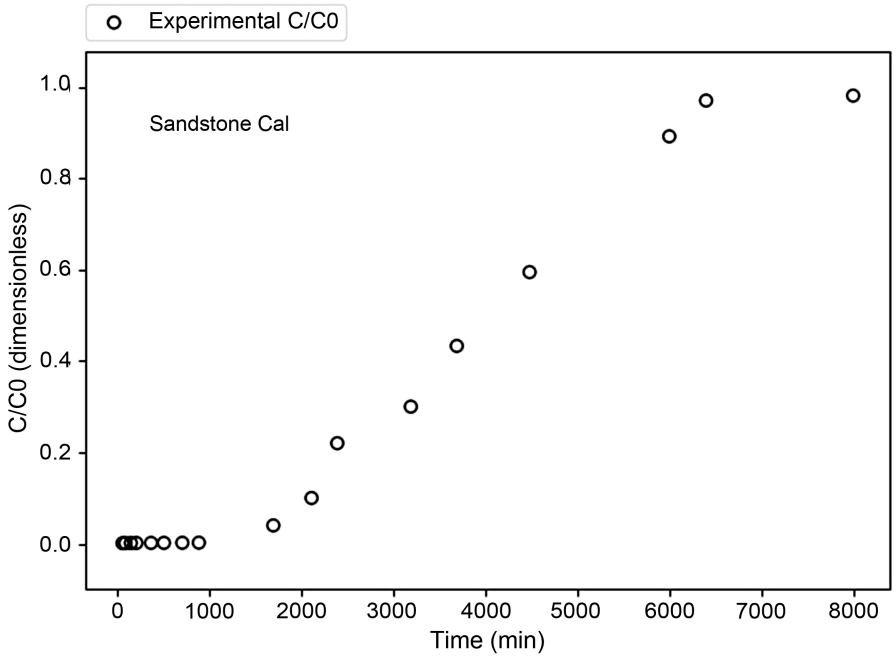
q_{total} = overall arsenic ion adsorption capacity (mg), m_{total} = overall mass of adsorbed arsenic (mg), V_b = volumetric output at breakthrough point (ml), V_T = total volumetric output at saturation (ml), q_e = adsorption capacity (mg/g).

3.3. Influence of Calcined Materials

The quantity of arsenic adsorbed and the volume of water treated with the calcined adsorbent within the columns are shown in Figure 5. For columns containing 85

cm of material, the arsenic concentration at the column outlet remained below 0.01 mg/L for treated water volumes of 7460 mL for laterite, 3510 mL for sandstone, and 4400 mL for shale. The calcined materials demonstrated a lower residual arsenic concentration compared to the raw materials. The effectiveness of the materials was classified as follows: sandstone < shale < laterite (see Table 1). Statistical analysis revealed a significant difference between columns filled with raw materials and those with calcined materials (Kruskal-Wallis test: $p < 0.05$). This difference was notably significant in the filtrates of the 85 cm columns containing raw materials compared to those containing calcined materials (Mann-Whitney test: $p < 0.05$).





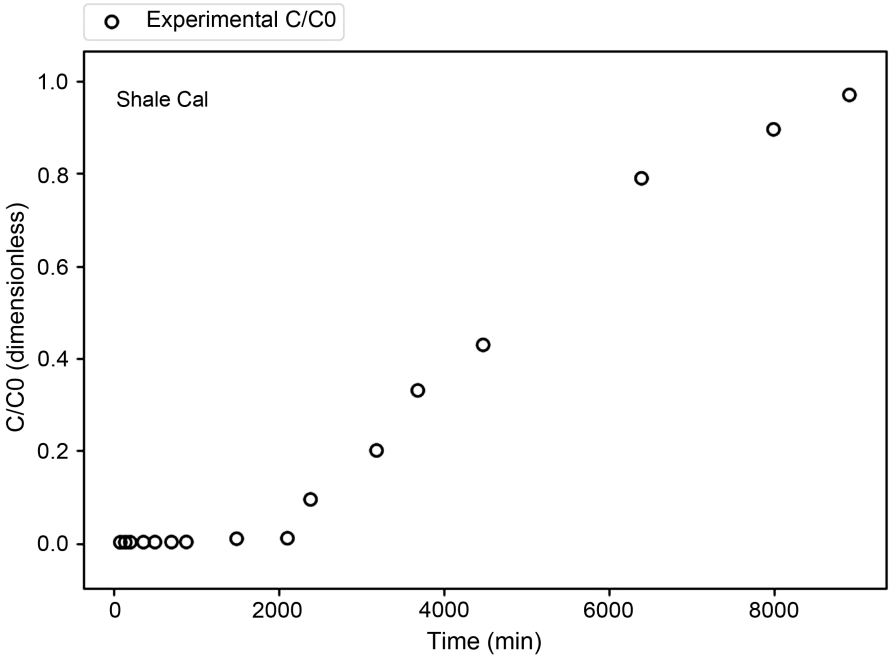


Figure 5. Influence of heat treatment of materials (laterite, sandstone and shale) on the volume of water containing treated arsenic.

The favorable results obtained with calcined materials can be attributed to the alteration, disappearance, or modification of certain minerals due to the effects of temperature [34], as well as the presence of minerals such as hematite, kaolinite, gibbsite, calcite, and goethite [18], [19]. Valix and Cheung [35] noted that calcination can influence both the chemical components and the physical structure of the adsorbent. This process effectively removes organic matter while altering the structure of the adsorbent, thereby enhancing its capacity for arsenic adsorption [36]. These adsorption values are lower compared to those achieved with granular ferric hydroxyl (GFH), which is a certified product for the removal of arsenic in water [37].

4. Conclusions

This study explored the adsorption of arsenic from well water in the Akouédo area using fixed bed columns filled with laterite, sandstone, and shale. Regarding the effect of the initial arsenic concentration on raw materials, the findings indicated that at a concentration of 1 mg/L, columns with a diameter of 40 mm and a bed height of 40 cm treated a total volume of 9000 ml (1800 min) with laterite, 2200 ml (440 min) with sandstone and 3000 ml (600 min) with shale, all achieving a residual arsenic concentration less than or equal to 0.01 mg/L. However, with higher concentrations of 3, 4, and 5 mg/L, the residual concentration did not drop to 0.01 mg/L.

For columns containing calcined materials, both the arsenic removal rate and the volumes of treated water increased significantly. At a concentration of arsenic of 5 mg/L and a bed height of 85 cm, the total volumes treated with water were 7460 ml with laterite, 3510 ml with sandstone, and 4400 ml with heat-treated

shale, all resulting in concentrations below or equal to 0.01 mg/L. The retention efficiency was classified as follows: laterite, shale, and sandstone.

The column treatment trials yielded significant results, revealing that the arsenic adsorption capacity of laterite surpasses that of sandstone and shale. In addition, both shale and sandstone are viable options for use in arsenic treatment. Future studies should focus on enhancing the adsorption capacity of these local materials, laterite, sandstone, and shale, for the remediation of arsenic-contaminated water. However, the results obtained are encouraging.

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Conflicts of Interest

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

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