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Adsorption of High-Fluoride Brackish Water from the Ndame Senegal Borehole on Coconut Shell Activated Carbon

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Abstract

The populations of the peanut basin are facing a groundwater quality problem due to high salinity and excess fluoride. The consumption of these waters constitutes a risk to public health through poisoning and cases of severe fluorosis. This study aims to propose a small-scale treatment accessible to the rural population. The methodology for defluoridation of hyperfluorinated water from the Ndame borehole by adsorption adopted consists of developing an activated carbon from coconut shells. Acidification of the medium with sulfuric acid, vinegar, and hibiscus is studied to improve treatment efficiency. The characterization of the activated carbon obtained gives iodine indices of 788.95 mg/g and 691.32 mg/g for methylene blue. Fluoride ions (F^-) are adsorbed at 64.50% (*i.e.* 4 mg/L to 1.17 mg/L) for water acidified with hibiscus, 70% (*i.e.* 4 mg/L to 1.42 mg/L) for water acidified with sulfuric acid, 70.75% (*i.e.* 4 mg/L to 1.20 mg/L) for raw water, and 72.75% (*i.e.* 4 mg/L to 1.09 mg/L) for water acidified with vinegar. Analysis of these results shows that in addition to the good adsorption capacity of activated carbon, the acidity of the medium and the presence of magnesium (Mg^{2+}) and calcium (Ca^{2+}) ions promote the elimination of fluorides. However, acidification with hibiscus is not an optimal method due to its high fluoride concentration. Thus, the physicochemical parameters of the environment impact the effectiveness of fluoride treatment. This study demonstrates that the activated carbon derived from coconut shells, in conjunction with a simple neutralization using locally available materials, can serve as a cost-effective and long-lasting solution for the elimination of fluoride in rural Senegal.

Keywords

Fluoride, Coconut Shells, Adsorption, Brackish Water, Ndame

1. Introduction

Access to drinking water is a recurring and major problem for the population, due to its availability and quality. On the one hand, this may be due to the nature of the environment, which influences the quality of water sources. This is the case in phosphate-producing countries, where confined water tables are polluted by fluorine ores (fluoroapatite), linked to phosphate ores. [1] [2]. On the other hand, this situation is intensifying with climate change, in particular drought (rainfall deficit), salinization of surface water and aquifers, as well as the increase in water needs [3] [4]. Agriculture and mining industries, the use of chemical fertilizers (phosphate fertilizers), and pesticides, also contribute to the pollution of these water sources [3]-[5].

In the Senegalese peanut basin, the quality of groundwater (deep Maastrichtian aquifer) is affected by excessively high levels of fluoride (1.5 to 7.5 mg/L) [6] [7] and salt (greater than 1.5 g/l) [8]. This is the central saline and fluoridated band (Kaolack, Fatick, Diourbel, and Thiès), where problems related to access to drinking water affect nearly a million people [9]. Indeed, long-term consumption of this water can lead to public health problems, such as poisoning and fluorosis [5] [7] [10].

Numerous studies are conducted in Senegal and other regions worldwide to address the issue of high fluoride levels in the population [11]-[14].

However, the different processes for treating hyperfluorinated water remain complex for reasons of cost, volume to be treated, and selectivity. These are chemical precipitation, electrodialysis, reverse osmosis, nanofiltration, ion exchange, and adsorption [15]. The latter is the most accessible to the rural population. It uses, in particular, activated carbon, whose useful capacity (quantity of fluorine eliminated) is 0.2 g/L of F^- per gram of material [15].

In the same vein as these studies, this study investigates the utilization of locally available, low-cost materials for fluorination. This study aims to propose an accessible, less expensive treatment suitable for domestic applications. To do this, the water from the Ndame borehole in the Diourbel region will be treated by adsorption with activated carbon made from coconut shells. Sulfuric acid and local products such as vinegar and white hibiscus are used to improve the treatment efficiency by acidifying the water, as several studies have demonstrated [16]-[18], reducing the competition of hydroxyl ions (OH^-) for adsorption sites [19].

Specifically, this involves taking samples for detailed analyses of the physicochemical parameters of the water. The results obtained provide an idea of the amount of fluoride in the water in order to deduce the proportions of treatment products.

2. Materials and Methods

2.1. Presentation of the Study Area

Ndame is located in the Diourbel region, in the northwest of the Mbacké department. Capital of the Ndame district, it is located between 14˚49′00″ north latitude and 15˚54′00″ west longitude in west-central Senegal (Figure 1).

Ndame has a Sudano-Sahelian climate, with an average rainfall of 400 mm to 600 mm per year and average temperatures varying between 30˚C maximum and 27˚C minimum [20].

The Maastrichtian sand layer is present there with a high salt and fluorine content, as reflected by a conductivity greater than 1500 µs/cm [21].

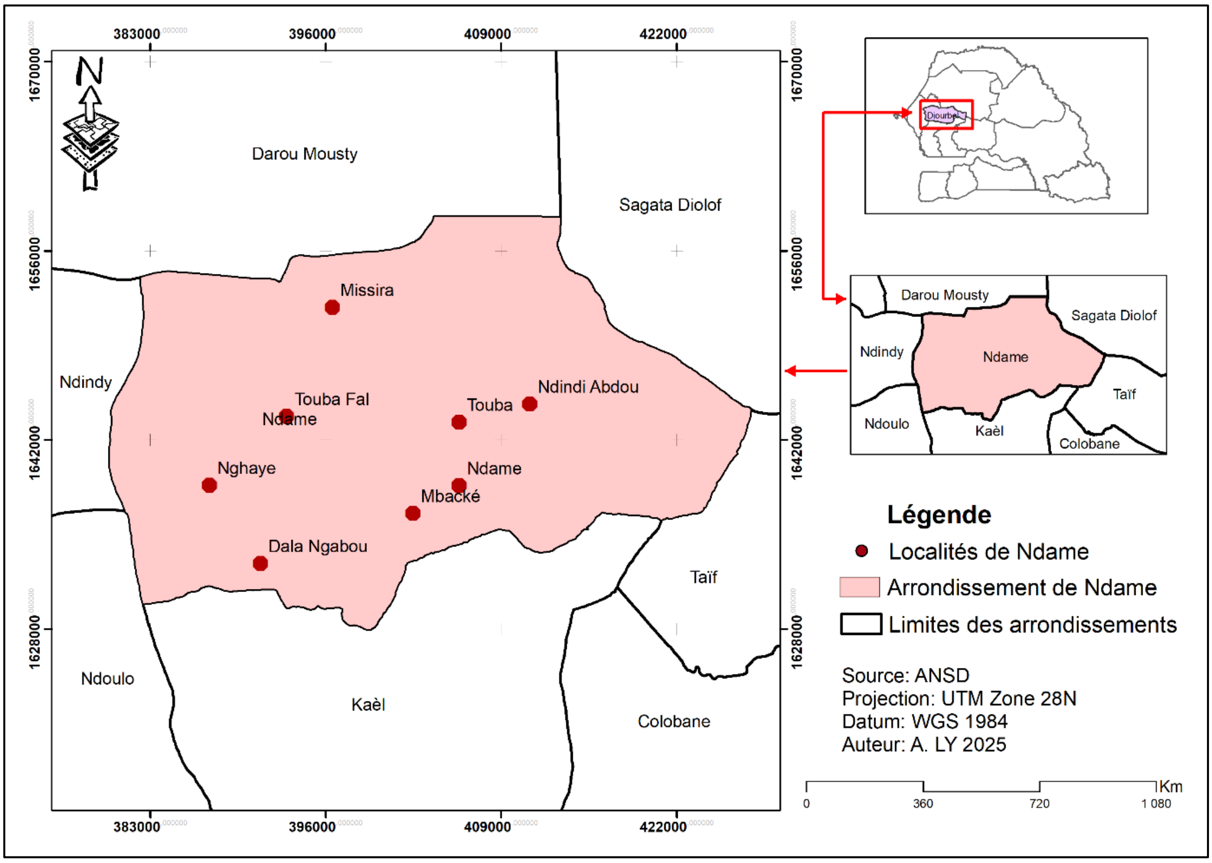

Figure 1. Location of the study area.

2.2. Sampling

In order to treat the hyperfluorinated drinking water of the commune of Ndame, it is important to carry out sampling. The latter must follow a strict protocol in order to preserve the characteristics of the sample. Based on this fact, the sampling was done using five-liter (5 L) plastic bottles. Once on site, the bottles were rinsed with the solution that must be sampled beforehand. Then, the in-situ parameters, namely pH, conductivity, temperature, and TDS, were determined with the METTLER TOLEDO multi-parameter device.

In the laboratory, different tests are carried out on the ions Ca^{2+}, Mg^{2+}, K^{+}, Cl^{-}, F^{-}, and SO_4^{2-}.

Coconut shells (**Figure 2**) were collected from coconut fruit vendors, and vinegar and hibiscus were obtained from the market.

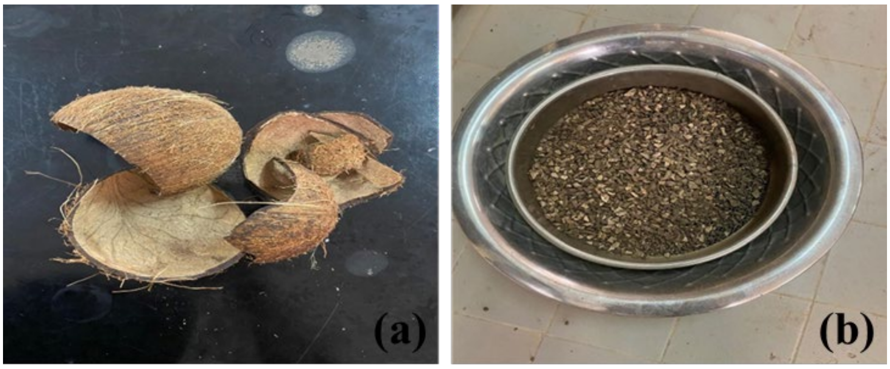

Figure 2. Coconut shells. (a) Raw coconut shells and (b) crushed coconut shells.

2.3. Preparation of Activated Carbon (AC): Carbonization and Chemical Activation

Activated carbon from coconut shells is prepared in the sanitary engineering laboratory of the Thiès Polytechnic School (EPT).

Coconut shells are broken into small pieces, washed with tap water and distilled water, and then oven-dried at 120˚C for 24 hours. They are then soaked in 85% phosphoric acid (H_3PO_4) for 24 hours before being oven-dried again at 120˚C for 24 hours.

Carbonization is carried out in a Nabertherm furnace at 600˚C for 45 minutes. The resulting product is washed with distilled water to remove impurities, after cooling it using a desiccator. This product is then dried for 24 hours at 105˚C in an oven. Finally, it is ground in a mortar and sieved to obtain two distinct particle sizes: a fraction less than 0.5 mm and another with particles with a diameter between 0.5 and 1 mm.

2.4. Adsorption Device (Column Filtration)

2.4.1. Filtration Equipment

Adsorption tests of high fluoride brackish water on coconut shell activated carbon were carried out using an adsorption device (**Figure 3**) single column that includes:

1) a raw water tank to be filtered;
2) an inlet valve;
3) A junction sleeve without a filter;
4) An activated carbon column;
5) A junction sleeve with a filter;
6) a shut-off valve;
7) A filtered water tank.

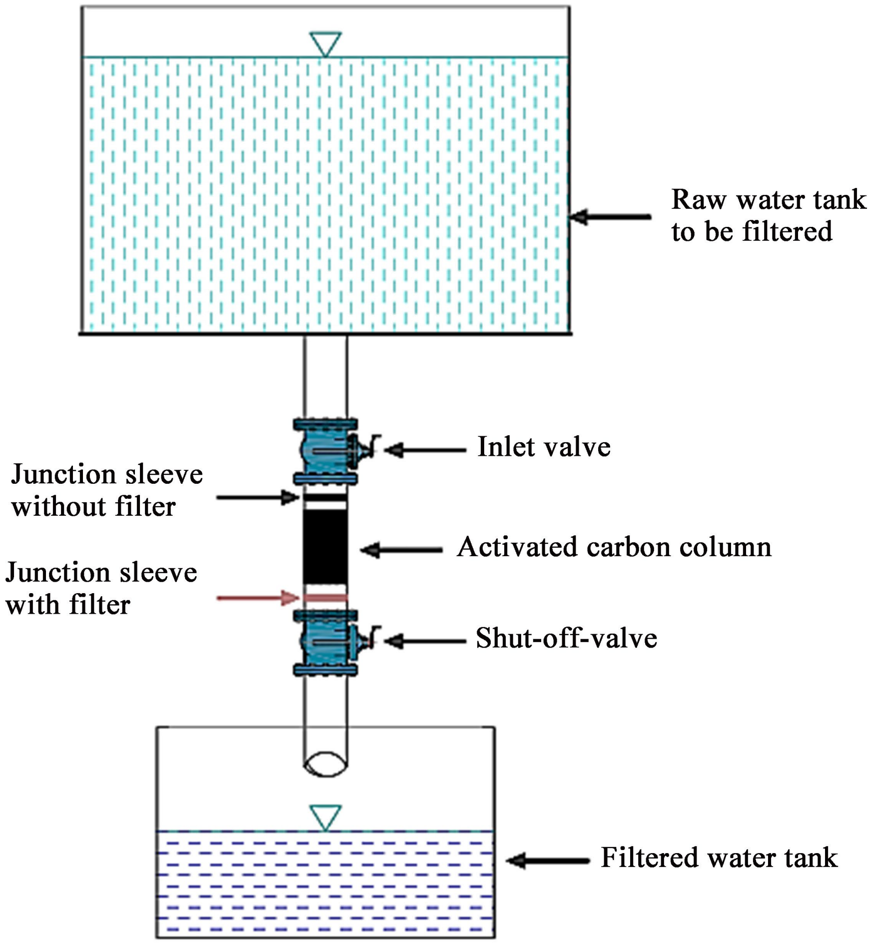

Figure 3. Adsorption device.

2.4.2. Filtration Procedure

Acidification with different acids (sulfuric acid, vinegar, and white hibiscus) available and accessible to the local population is carried out in order to improve the treatment results. Thus, four samples are obtained: raw water, raw water acidified with sulfuric acid, raw water acidified with vinegar, and raw water acidified with white hibiscus.

To obtain raw water acidified with sulfuric acid, sulfuric acid was added dropwise to 1 liter of raw water. The pH was measured after each addition, with the aim of reaching the optimal value of 4 [13].

For this study, white spirit vinegar titrated to 8% was used to acidify raw water. Tests were conducted to determine the amount needed to lower the pH to a favorable range. A volume of 0.1 mL of the acid solution was gradually added to 250 mL of raw water, until water with a low pH, neutral taste, and color was obtained. Thus, a volume of 2 mL of white vinegar is used for 1 L of water.

White hibiscus, due to its high acid content, was chosen for lowering pH. After testing solutions of different concentrations, the one composed of 400 mL of distilled water + 50 g of white hibiscus met the criteria already mentioned with a volume of 4 mL. Therefore, 16 mL was added to 1 liter of raw water to obtain our final sample of raw water acidified with hibiscus.

The batch filtration technique on an absorbent column was used on our different samples after characterization.

A volume of 1 L of water to be treated was introduced into the tank of the device and, using the shut-off valve, a filtration flow rate of 1.042 mL/s was set. This resulted in a contact time of thirty-two (32) minutes for a filter mass of 23 g.

After filtration, the treated water is collected before being analyzed.

The AQUA LYTIC Al 800 spectrophotometer was employed to quantify fluoride in water in accordance with the SPADNS colorimetric method. The detection limit is 0.05 mg/L. The calibration and adjustments are performed in accordance with Standard Methods [22].

The results of the analyses of these filtrates will be presented in the Results and Discussion section.

3. Results and Discussions

3.1. Study of the Physicochemical Parameters of Raw Water

The raw water analyzed is groundwater, from the Ndame borehole, which has a flow rate of 94 m^3/h and a depth of 140 m. The physical parameters (**Table 1**) show that the water is alkaline, with a pH of 7.636 and a temperature of 28.4˚C. Regarding the conductivity, it is high (4.62 ms/cm) and can be explained by the high salt content in the aquifers of the site studied. The TDS obtained (2310 mg/L), being higher than 1000 mg/L, allows us to deduce that the water is unfit for consumption [23].

Table 1. Physicochemical characteristics of raw water.

Physical parameters	pH	T (˚C)	C (µs/cm)	TDS (mg/L)							
value	7,636	28.4	4620	2310							
WHO standards	6.5 - 8.5	-	1200	1000							
Chemical parameters	Mg^{2+}	K^+	Ca^{2+}	Cl^-	SO_4^{2-}	F^-	Cl_2	Mn	Zn	Cr	Fe
Value (mg/L)	45.58	13	24.46	1189.15	160.8	4	0.39	0.02	0.03	0.19	0.1
WHO standards	50	12	100	250	250	1.5	5	0.1	3-5	0.05	0.3

Regarding the chemical parameters (**Table 1**), the Ca^{2+} and Mg^2 + ions have concentrations of 24.46 mg/L, 45.58 mg/L, 160.8 mg/L, respectively. Thus, we note that they

SO_4^{2-} comply with the standards accepted by WHO 2017 (Ca^{2+}: 100 mg/L, Mg^{2+}: 50 mg/L and SO_4^{2-} : 250 mg/L) and do not constitute a danger for human consumption [23].

Furthermore, the contents of K + ions (13 mg/L), Cl^- (1189.15 mg/L) and F^- (4 mg/L) exceed the recommended doses, which are 12 mg/L for K^+, 250 mg/L for Cl^- and 1.5 mg/L for F^- [23].

3.2. Characterization of Coconut Shells

The moisture content of the coconut shell samples was measured according to the AFNOR XP CEN/TS 14774-3 standard, which consists of heating the sample to 105˚C until all the water has evaporated. The dry matter content is calculated from the water content or by relating the dry matter to the total wet matter. The determination of the MV rates was carried out according to the XP CEN/TS 15148 standard. The determination of the ash content of a material is determined by the AFNOR XP CEN/TS 14775 standard. The fixed carbon content is obtained by the difference between the volatile matter content and the ash content.

The results of the characterization of coconut shells are recorded in **Table 2**.

Table 2. Characteristics of coconut shells.

Material	Humidity (%)	Dry matter (%)	Volatile matter (%)	Ash (%)	Fixed carbon (%)
Coconut shells	3,259	96,741	38,993	0.523	60,484

Analysis of these results shows that they contain a moisture content of 3.259% and an ash content of 0.523%. In fact, a moisture content of less than 8% and an ash content of less than 5% are considered low. Therefore, these results define a good adsorption capacity of coconut shells and are in line with those obtained by [19].

In addition, the high volatile matter (38.993%) and fixed carbon (60.484%) contents indicate that the hulls are rich in organic matter. This confirms the good quality of the absorbent material [19].

3.3. Characterization of Activated Carbon

To determine the adsorption capacity of activated carbon, we determined the iodine number and the methylene blue number.

The iodine value, expressed in (mg/g), is used to measure the adsorption capacity of small molecules by carbon. It was determined according to the AWWA B 600-78 standard [19].

The methylene blue index (mg/g) is used to assess the ability of carbon to adsorb medium and large organic molecules through its mesopores [19]. The 1986 European Chemical Industry Centre (CEFIC) method is used to determine the methylene blue index (mg/g) [19].

Table 3 represents the results of the characterization of activated carbon.

Table 3. Iodine and methylene blue indices of activated carbon.

Material	Iodine value (mg/g)	Methylene blue index (mg/g)
Activated carbon	788.95	691.32

The iodine and methylene blue values obtained are high, with respective values of 788.95 mg/g and 691.32 mg/g. In fact, the adsorption capacity of methylene blue greatly exceeds that of Merck commercial activated carbon (200 mg/g) [24]. In addition, activated carbon is capable of adsorbing small molecules if its iodine value is between 600 and 1100 mg [25] [26]. Good adsorption of iodine reveals the existence of micropores in activated carbon [27]. Similarly, strong adsorption of methylene blue indicates the existence of mesopores. This demonstrates the effectiveness of preparing activated carbon with a product with a high adsorption capacity for small and large molecules.

Therefore, activated carbon with H_3PO_4 and carbonized at 600˚C for 45 minutes is suitable for treating hyperfluorinated waters of Ndame.

3.4. Results of Filtrate Ion Analyses

Acidification of raw water with sulfuric acid, vinegar, and hibiscus yields final solutions with pH values below 4, as shown in **Table 4**.

Table 4. pH of acidified water.

Parameters	Water + filtered sulfuric acid	Water + filtered vinegar	Filtered water + hibiscus
pH	3,027	3,295	3,521

Analysis of the results (**Table 3** and **Table 4**) reveals that adsorption on coconut shell-based activated carbon had a variable impact on the concentrations of the different elements present in the filtrates of the treated water. Overall, activated carbon gives good yields for the adsorption of F^- ions for all the treated waters, with recovery rates between 64.50% and 72.75%.

These results indicate that coconut shells are a good material for the treatment of hyperfluorinated water. Thus, all treated waters meet the WHO standard limits for fluorides [23].

However, we find that activated carbon adsorbs more F^- ions in acidic environments, except in waters acidified with hibiscus. This is because the decrease in pH increases fluoride adsorption by reducing competition between OH^- ions and F^- ions. In addition, protonation of the adsorbing surface gives activated carbon a positive charge when the pH is below its zero-charge point pH (pZCP). This promotes the electrostatic binding of anions such as fluoride. [16] [18] [19].

Similarly, the presence of calcium and magnesium increases the adsorption of fluorides with the formation of fluorine (CaF_2) [17] [18].

It is important to note that hibiscus contains a high fluoride content. This could explain the low treatment efficiency rate compared to other acids, by releasing fluorides from hibiscus [17].

Furthermore, our results obtained, compared to those achieved with the adsorption on zircon of hyperfluorinated waters of Diouroup (67.5%), show that the activated carbon of coconut shells offers better performance [2]. It is also more competitive than activated carbon from eggshells, with an adsorption rate of 51.4% [18]. On the other hand, activated carbon from peanut shells has an adsorption efficiency of 82.3% [28], which is higher than our results.

However, it is necessary to take into account the contact time, the pH of the medium, the initial concentration of fluoride ions, as well as the quantity of adsorbent.

The adsorption of Mg^{2+} and Ca^{2+} ions is efficient, except for hibiscus acidified water, where their concentrations increased considerably (−118.14% for Mg^{2+} and −200.90% for Ca^{2+}). This increase can be explained by the high presence of Mg^{2+} ions (> 300 mg per 100 g of dry matter) and Ca^{2+} ions (> 1,500 mg per 100 g of dry matter) in hibiscus [29].

As for K^+ ions, a slight decrease for raw water (7.69%) and a notable increase for water acidified with sulfuric acid (−100%) are noted.

On the other hand, the concentration of Cl^- ions decreased significantly in the filtrates except for the water acidified with hibiscus (−38.84%).

S ions O_4^{2-} increased predominantly, reaching −90.30% for sulfuric acid acidified water, except for hibiscus acidified water (53.36%). Chlorine concentration remained constant for raw water (0.39 mg/L) and decreased for acidified water.

As for heavy metals (Cr, Zn, Mn) and Fe, their concentrations increased significantly in the filtrates except for Cr in the vinegar-acidified water, with a value of 10.53%. **Table 5** and **Table 6** are shown below.

Table 5. Ion concentrations of filtered water.

Chemical parameters (mg/L)	Raw water	Filtered raw water	Water + filtered sulfuric acid	Water + filtered vinegar	Filtered water + hibiscus
Mg^{2+}	45.58	20.9	12.91	12.08	99.43
K^+	13	12	26.00	14.00	19.00
Ca^{2+}	24.46	11.5	13.80	6.13	73.60
Cl^-	1189.15	1137	1175.50	1167.80	1651.00
SO_4^{2-}	160.8	169	306.00	207.00	75.00
F^-	4	1.17	1.20	1.09	1.42
Cl_2	0.39	0.39	0.31	0.28	0.32
Mn	0.02	0.09	0.24	0.24	-
Zn	0.03	0.03	0.04	0.04	0.03
Cr	0.19	0.2	0.22	0.17	0.22
Fe	0.1	0.1	0.10	0.21	0.25

Table 6. Recovery rate.

Chemical parameters in (mg/L)	Filtered raw water In %	Water + filtered sulfuric acid in %	Water + filtered vinegar in %	Water + filtered hibiscus in %
Mg^{2+}	54.15	71.68	73.50	−118.14
K^{+}	7.69	−100.00	−7.69	−46.15
Ca^{2+}	52.98	43.58	74.94	−200.90
Cl^{-}	4.39	1.15	1.80	−38.84
SO_4^{2-}	−5.10	−90.30	−28.73	53.36
F^{-}	70.75	70.00	72.75	64.50
Cl_2	0.00	20.51	28.21	17.95
Mn	−350.00	−1100.00	−1100.00	–
Zn	0.00	−33.33	−33.33	0.00
Cr	−5.26	−15.79	10.53	−15.79
Fe	0.00	0.00	−110.00	−150.00

4. Conclusions

The populations of the salt and fluoride belt of Senegal face a problem of access to drinking water, linked to the high salt and fluoride levels (greater than 1.5 mg/L) in groundwater. Indeed, the consumption of this water has harmful consequences for their health, including poisoning and cases of fluorosis. Thus, the treatment of this water is essential to preserve public health.

In this sense, the defluoridation of hyperfluorinated water from the Ndame borehole is performed by adsorption on activated carbon. The carbon is produced from coconut shells, activated with H_3PO_4 and carbonized at 600˚C for 45 minutes. This choice is justified by the efficiency and accessibility of the adsorbent. Indeed, this material offers a good adsorption capacity for small and large molecules, as confirmed by its high iodine index of 788.95 mg/g and methylene blue of 691.32 mg/g.

Knowing that the adsorption of F^- ions is more efficient in an acidic environment, the raw water is acidified with sulfuric acid, vinegar, and hibiscus. As a result, we obtain four (04) separate samples to be treated.

The adsorption of F^- ions with coconut shell activated carbon resulted in yields of 64.50%, 70%, 70.75%, and 72.75%, respectively, for hibiscus acidified water, sulfuric acid acidified water, raw water, and vinegar acidified water. It is worth mentioning that the results obtained meet WHO standards for drinking water.

These results reveal that the physicochemical characteristics of the environment influence the efficiency of fluoride treatment. Indeed, the acidity of the environment and the presence of fluorine (formed by calcium and magnesium) increase the rate of fluoride removal. On the other hand, acidification with hibiscus is not an optimal method because it contains a high level of fluoride.

Ultimately, defluoridation of hyperfluorinated water is efficient with coconut shell activated carbon, which has been shown to retain over 64% of fluoride. Acidification of the medium is a way to increase treatment efficiency. In this context, vinegar proves to be more effective, without health risks.

Looking ahead, studies on activated carbon regeneration, cost assessment, and field applicability should be conducted with a view to deploying this defluoridation solution among the local population in an effective and sustainable manner.

Conflicts of Interest

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

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