



# Removal of Fluoride Ions from Drinking Water by Activated Alumina and Activated Charcoal

Habiba Lebrahimi<sup>1\*</sup>, Mohammed Fekhaoui<sup>2</sup>, Abdelkabir Bellaouchou<sup>3</sup>.

<sup>1</sup>Department of Zoology and Animal Ecology, Laboratory of Zoology, Scientific Institute, Mohamed V University, Avenue Ibn Battouta, Agdal, 10090, Rabat, Morocco

<sup>2</sup>Department of Toxicology, National Institute of Hygiene, 27, Avenue Ibn Battouta, 769 Agdal, Rabat, Morocco

<sup>3</sup>Department of Chemistry, Faculty of Sciences Rabat, Mohamed V University, 4 Avenue Ibn Battouta, 1014 Agdal, Rabat, Morocco

## Abstract

The pollution of drinking waters by ions fluorides caused by industrial and natural activities, causes major problems for human health, what imposes to think seriously of the treatment of these waters. Experiments are carried out in the laboratory to remove fluoride ions in water, based on the adsorption process by activated alumina and activated carbon with a change in different parameters influencing the retention rate such as pH, amount of adsorbent and contact time. Good defluoridation requires a contact time between 30 min and 60 min with a retention rate of 53.51% and 67% for activated alumina, 50.27% and 56.35% for activated carbon. The retention rate for 3g adsorbent is 56% for activated alumina and 18.91% for activated carbon, this value increases with increasing adsorbent quantity for acid pH. According to these experiments the retention rate of fluoride ions by activated alumina is higher than that of activated carbon, which makes activated alumina treatment the most responsive for the removal of fluoride ions in water. In this work they try to find an appropriate method to eliminate the excess of ions fluorides in underground water after a study made on these ions in waters of the region of Khouribga in Morocco as phosphaté region the presence of ions of which results points out fluoride with a broad broadcasting of the illness of fluorosis to the population. According to tries made in the laboratory one there found that alumina speeded up and activated charcoal is very efficient for this action.

**Keywords:** Fluoride, Activated alumina, Activated carbon, Water treatment, Defluoridation, Adsorption

## 1 Introduction

Fluoride (F<sup>-</sup>) contamination in groundwater has been recognized as one of the serious problems worldwide. Fluoride is classified as one of the contaminants of water for human consumption by the World Health Organization (WHO), in addition to arsenic and nitrate, which cause large-scale health problems [1]. The fluorine as most part of the chemical elements (iron, manganese ...) effects of which their excess in Drinking Water causes failure for water and environment [2]. The quality of waters of drink is an obligation for everybody what obliges a strict treatment [3]. The classical techniques used for the treatment of waters of drinks are several categories among which in most cases the adsorption with activated charcoal, alumina speeded up and loam, the techniques of haste, physicochemical techniques (separation on membrane, oxidation / discount) and other biological techniques (biosorption, phytoextraction) [4]. The pollutions of underground waters of the region of Khouribga are a strict problem, because it causes an endemic illness to the population, what makes think seriously of the treatment of waters intended for human consumption. Everything results acquired from the physicochemical analyses of underground water of the region of khouribga are has norm except the content of fluorine which was superior has norm in several wells of the region. [5].

The removal of fluoride ions from groundwater is done by several techniques which differ from each other depending on the removal percentage (efficiency) and treatment cost. According to several studies [6], among these techniques, one

is based on the adsorption technique on activated carbon and alumina, which are the most responsive, effective, very simple and do not require much maintenance. Both elements are the most used industrially because of their excellent decontamination properties against the majority of pollutants. Considerable efforts are made to ensure a better protection of the environment of water resources. Scientific research is increasingly oriented towards the selection of the most effective means of depollution. Several methods of treatment of water containing fluorine, mainly adsorption by activated alumina and activated carbon, have given their effectiveness in removing fluoride ions dissolved in water [7]. This process is well suited to the elimination of fluoride ions with a need to control the ions but the pH of the treated water need to be control. The adsorption capacity depends on the specific surface of the material, the nature of the adsorbent, and the contact time. For our study one there chosen the treatment by the adsorption by alumina speeded up and activated charcoal seen that their fastness and their effectiveness according to results found without forgotten availability of this two adsorbants in Morocco as developing country [8]. The objective of fluoride removal meant the treatment of contaminated water in order to bring down fluoride concentration to acceptable limits. The defluoridation techniques are generally classified into 2 classes, specifically membrane and surface assimilation techniques. No study was made in this sense in Morocco contrary has the ladder international or they find many people work on this problem,

**Corresponding author:** Habiba Lebrahimi, Department of Zoology and Animal Ecology, Laboratory of Zoology, Scientific Institute, Mohamed V University, Avenue Ibn Battouta, Agdal, 10090, Rabat, Morocco, +212762567494, [zaidhabibalebrahimi@gmail.com](mailto:zaidhabibalebrahimi@gmail.com).

the treatment of waters of which becomes an obligation in our seen country has consumption there.

## 2 Materials and methods

### 2.1 Generality

The adsorption of a substance by a solid is the more or less irreversibly binding of the solute molecules in contact with the surface of this solid without any chemical reaction between them. This is a rapid phenomenon that results in a balance between the substance adsorbed and the substance remaining in solution. Balance depends on the concentration of the solute and the surface of the adsorbent body. There are two types of adsorption:

Physical adsorption: reversible, which does not modify the chemical identity of the adsorbed molecules.

Chemical adsorption: irreversible, characterized by chemical bonds between the adsorbate and specific adsorption sites. This process is well adapted to the specific elimination of fluorides but requires controlling the pH of the medium. The adsorption capacity depends on the specific surface of the material, the contact time, the pH and the initial fluoride concentration. [9].

Water defluoridation studies using several adsorbents such as aluminium hydroxide, activated carbon, and apatite and activated alumina have been carried out with encouraging results. [10]. For our study we used both activated alumina and activated charcoal because of their efficiency in the treatment as well as their cost and availability.

Activated alumina ( $\text{Al}_2\text{O}_3$  or aluminium oxide): To make two adsorbents assets an elimination of the molecules of water was made by calcination has a temperature of  $500^\circ$ . the technology used for the elimination of ions fluorides dissolved in waters of drink consists has percale some water to charge in fluorine on a bed of alumina speeded up allowing the retention of these ions, the most answering of which he is and assures a specific elimination of ions fluoride has strong concentration.[11]. The presence of fluoride ions with high levels in groundwater intended for human consumption requires serious efforts to treat them. In fact there are several treatment processes. Principle of adsorption: The principle of adsorption treatment is to fix the pollutants on an absorbent material. It is a separation process i.e. a simple transfer of mass from the liquid phase to the solid phase by fixing on their surface certain molecules, macromolecules and ions or certain gases.

### 2.2 Analytical method:

For our work, experiments were made in the laboratory for the elimination of fluoride ions in the ground water by adsorption processes through two products: activated carbon and activated alumina ( $\text{Al}_2\text{O}_3$  aluminium oxide). A solution prepared by dissolving 150 mg of sodium fluoride NaF. Dried in the oven for 3 hours, cooled in a desiccator in one liter of distilled water, and kept in a plastic bottle. All experiments were performed at ambient temperature of  $27.1^\circ\text{C} \pm 2^\circ\text{C}$ . Batch sorption tests were performed to determine the effect of control of related different parameters such as pH, contact time and adsorbent quantity. (Figure 1).

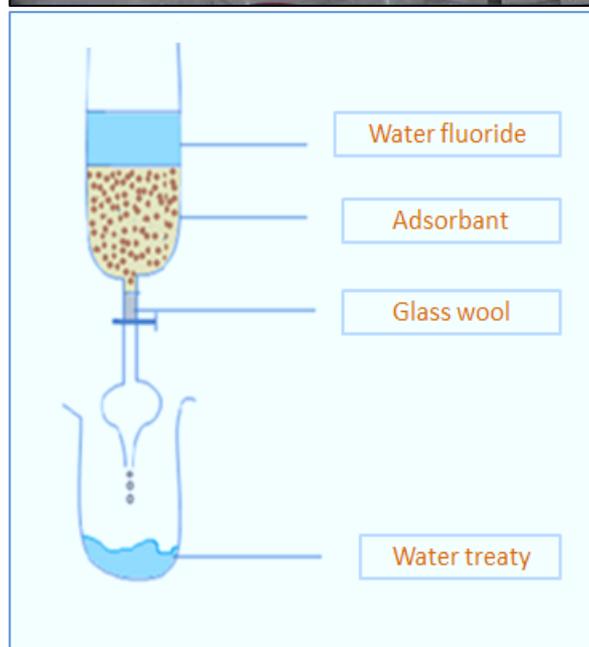


Figure 1: Elimination of fluorine by activated alumina / activated charcoal

The water samples taken were treated with activated carbon and activated alumina ( $\text{Al}_2\text{O}_3$  aluminium oxide) after they are activated at  $500^\circ\text{C}$ . The experimental protocol is to pass the sample through two columns each one containing one of the adsorbents, by three batches depending on the pH, the quantity of the adsorbent, and the contact time.

1st batch: to know the effect of the pH, the quantity of the adsorbent 3g is fixed, the contact time is the time of the filtration of a fixed volume of 50 ml of the prepared solution, and the pH of 5 and 8 is changed, the pH is adjusted by HCl (0.5 M) or NaOH (0.5M).

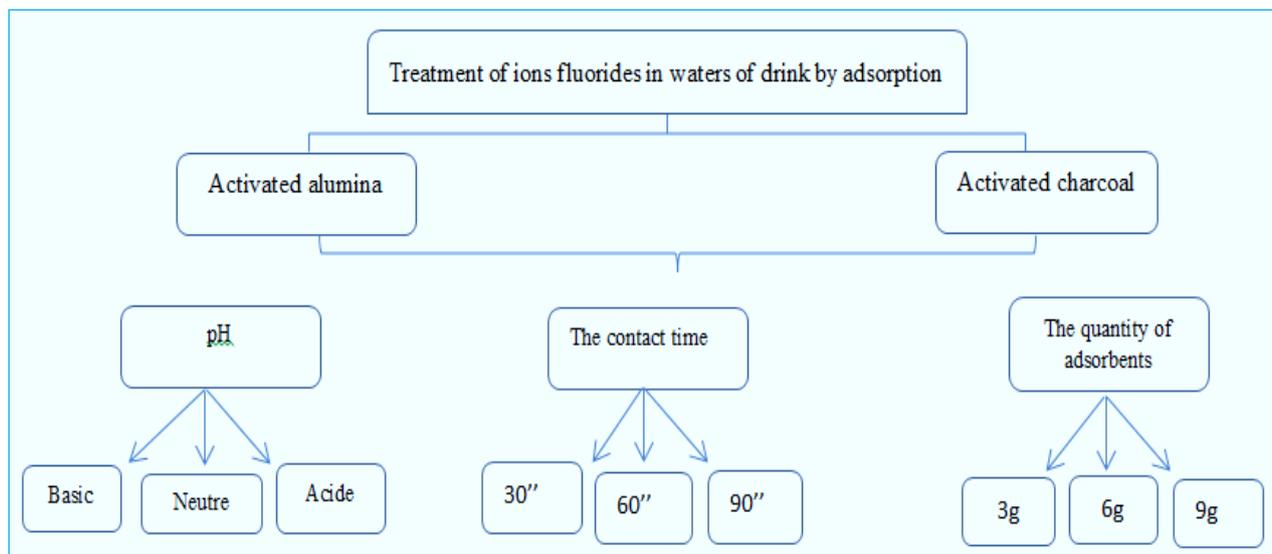


Figure 2: Flow chart of the working methodology

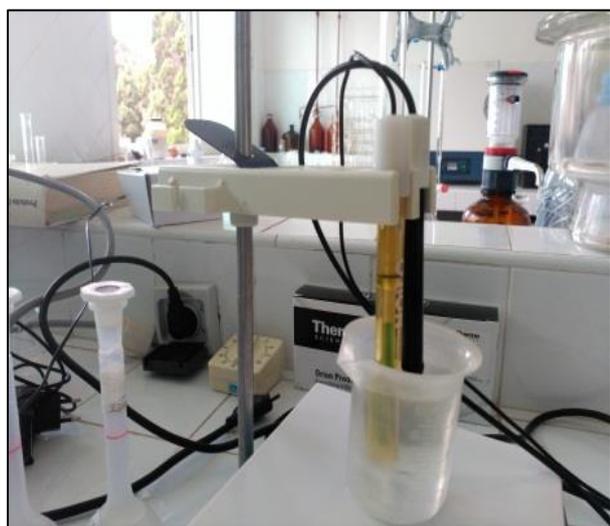


Figure 3: Measure of F- by the electrode specific

2rd batch: to study the contact time effect, the experiments were performed by varying the contact time by 30, 60 and 120 and passing a 50 ml volume of the prepared solution over two 3g columns of activated carbon and activated alumina for a pH of 6.1.

3rd batch: this is to study the effect of the quantity of adsorbents on the rate of elimination of fluoride ions in water, whose experiments are done with different quantities of 3g, 6g and 9g, using the same volume of 50 ml of the initial solution for a fixed pH 6.1 (Figure 2). The determination of fluoride ions is done before and after treatment with the specific electrode and the rate of elimination of fluoride ions is calculated by the following equation: (Figure 3).

$$R : (C_i - C_e / C_i) \times 100$$

where, R is the removal efficiency of fluorinated compounds (%),  $C_i$  is the initial concentration (mg/l) and  $C_e$  is the equilibrium concentration (mg/l) [6]. The order of activated alumina adsorptive force for anion in water is followed: OH-

>PO4<sup>3-</sup>->F<sup>-</sup>->SO3<sup>2-</sup>->Fe(CN)6<sup>4-</sup>->CrO4<sup>2-</sup>->SO4<sup>2-</sup>->Fe(CN)5<sup>3-</sup>->Cr2O7<sup>2-</sup>->I<sup>-</sup>->Br<sup>-</sup>->Cl<sup>-</sup>->NO3<sup>-</sup>->MnO4<sup>-</sup>->ClO4<sup>-</sup>-> S<sup>2-</sup>. It is apparent that activated alumina adsorptive force for phosphate anion is stronger than other anions, such as SO4<sup>2-</sup> and Cl<sup>-</sup>, so activated alumina adsorb phosphate anion in predominance. [12].

### 3 Results and discussion

#### 3.1 Contact time

Contact time adsorption tests for activated alumina and activated carbon show that the removal rate increases with increasing time until a given point in time when a constant value approaches that indicates the equilibrium point between treated water and adsorbent. According to this experience, the fluoride ion removal treatment procedure goes through three phases: The 1st was before 30 min of contact where the retention rate is relatively fast, 53.51% for activated alumina and 50.27% for activated charcoal, this speed due to the affinity of the adsorbent surface and the force of entrainment and diffusion in the porous medium. The 2<sup>nd</sup> is between 30 min and 60 min which consist of the migration of fluoride ions to the internal pores of the adsorbent, leading to high levels of fluoride retention rate, recording 67% for activated alumina and 56.35% for activated carbon. The 3<sup>rd</sup> phase when we exceed 60 min the retention rate is almost constant, reaching 69.46% for activated alumina and 58.78% for activated carbon (Figure 4 and Figure. 5). According to these results they point out that the percentage of the elimination of ions fluoride on alumina speeded up and activated charcoal augments fast with the increase of the time of contact especially for the first 60 minutes, of which beyond they note that the percentage of elimination gone towards stability for two adsorbents seen the occupation of the harbours of fixing of ions, therefore they can fix the time of contact has 60 minutes until 90 minutes, what points out that the time of contact has an effect on the elimination of ions fluorides. The results found are in agreement with those of BEN NASR et al 2014. Found for this work concerning the elimination of fluoride ions by the different treatment processes, also with the results found by fan et al, which fixes the time necessary for a good retention of these ions of less than 60 min.

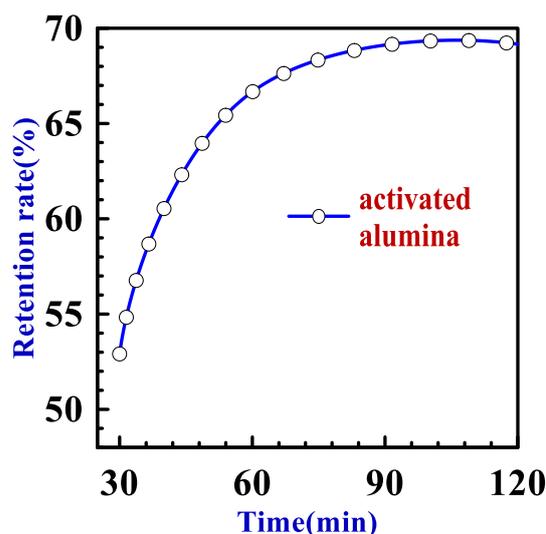


Figure 4: Retention rate % (activated alumina)

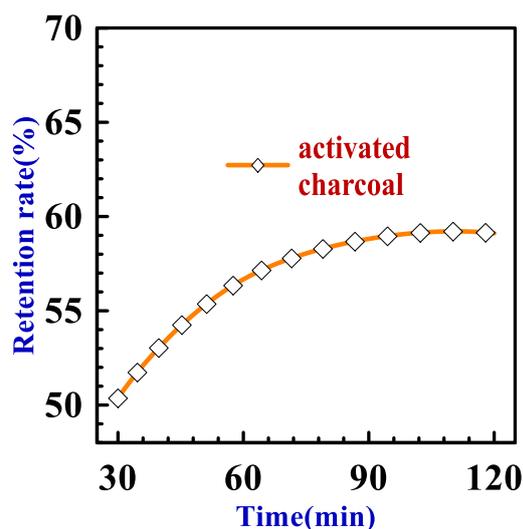


Figure 5: Retention rate % (activated charcoal)

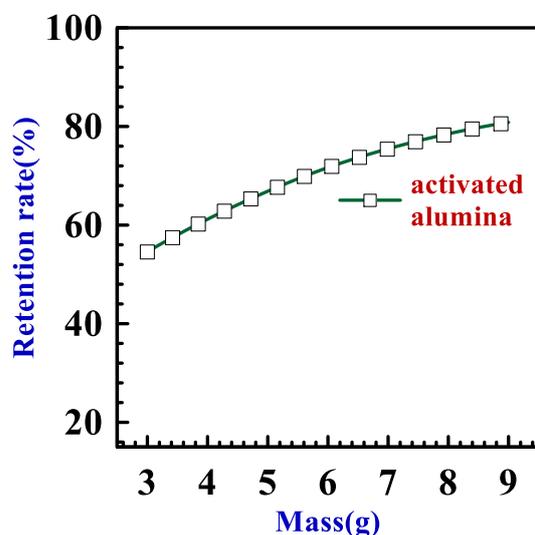


Figure 6: Retention rate % (activated alumina)

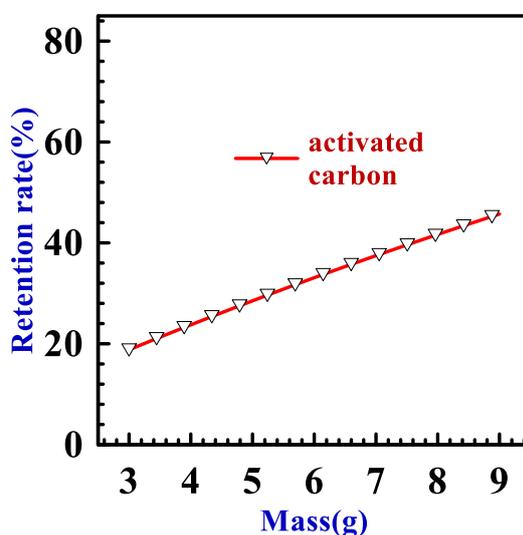


Figure 7: Retention rate % (activated carbon)

### 3.2 Effect of quantity of adsorbent

Tests of fluoride ions adsorption in water were carried out by varying the quantity of adsorbent by 3g, 6g and 9g which influences the retention rate according to the graphs below (Figure. 6 and Figure. 7). The results show that the retention rate increases with raising the amounts of adsorbent from 56% to 73.37% for activated alumina and from 18% to 32.83% for activated carbon in the case of 6g of the adsorbent. Whereas, when the amount of adsorbent is increased from 3g to 9g, we obtained a retention rate of 82.7% and 45.67% respectively for activated alumina and activated carbon (Figure. 6 and Figure. 7). These found results watch which the percentage of elimination of ions fluorides augments with the increase of the quantity of adsorbent at the same time activated charcoal is for alumina speeded up or, what points out that for a strong elimination of these ions needs an important quantity of adsorbents. This indicates that the rate of elimination of fluoride ions increases with increasing adsorption surface. This is in agreement with the results obtained by BEN NASR et al 2014, who found a retention rates of 96% and 89% in the case of adsorbent's amounts of less than or equal to 15g/l-1.

### 3.3 pH effect

The pH of the prepared water is 6.1, and in order to know the effect of pH on the retention rate of fluoride ions, the pH is adjusted by adding HCL 0.1 so that it becomes acid (pH 5) and NaOH so that it becomes basic (pH 11).

The results found after the determination of fluoride ions in these waters for both cases, show that the retention rate in the acid medium (pH=5) is 75.67% for activated alumina and 48.35% for activated carbon. However in the basic medium (pH=8) we obtained a retention rate of 47.3% for activated alumina and 41.35% for activated carbon (Figure. 8 and Figure. 9). Hence it appears that the pH of the medium considerably influences the rate of fluoride retention in water. These results are almost compatible with those found by Anis Ben nasr 2014 [6]. These results watch that the percentage of elimination of ions fluoride for a sour water is superior that a basic water for alumina speeded up and the opposite for activated charcoal what points out that the percentage of fixing varies according to nature da pH, they conclude therefore that the pH influences fluorides in general on the elimination of ions according to the chosen adsorbent.

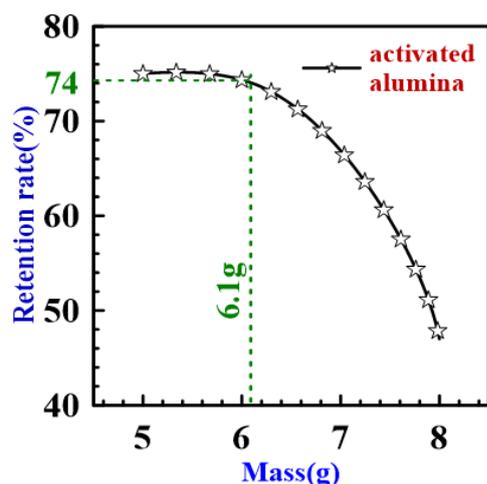


Figure 8: Retention rate % (activated alumina)

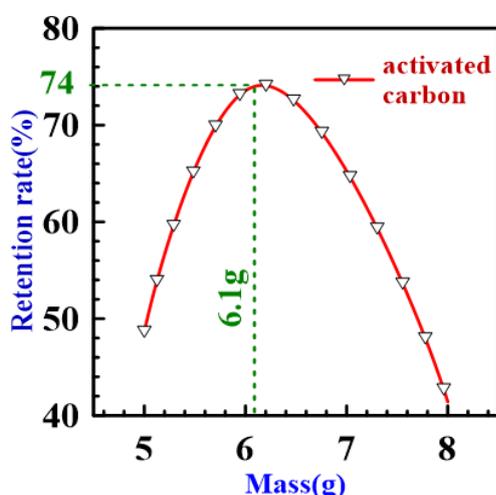


Figure 9: Retention rate % (activated carbon)

Other results found by Srimurali et al 2008. [7] and Bersillon et al 2006. [12, 13, 15] Suggest that the elimination of ions fluoride is quick if we leave 10 min of contact between alumina speeded up and the contaminated water. In the case of 60 min, the rate of fluorine retention becomes almost constant, with a pH between 2 and 6.5, reaching the maximum of retention. However when the pH exceeds 6.5, the rate of fluorine retention starts to decrease [10; 9]. In addition Karthikeyan and al. [9] found that the elimination of ions fluorides is influenced by three parameters, especially the pH, time of contact and the quantity of used adsorbent. In fact it is needed at least 20min of contact, 3g of the adsorbent for a pH of 7 to eliminate a given quantity of fluorine dissolved in water. [16]. Several works made to treat waters rich in fluorine such as that of Mr Menaksti. and al who results that alumina speeded up and activated charcoal among adsorbents the most efficient for this elimination, according to three mailmen pH, time of contact and the quantity of the used adsorbent. [9] also the study of Mwangi. and al who says that according to even mailmen among whom optimum of pH and time of contacts was found is 4,0 with a time of 20min and the concentration of retention is of 60mg / l [17].

## 4 Conclusions

As much as our country 'Morocco ' among phosphates countries, and most known by the illness of especially dental

fluorosis caused in general by the consumption of waters rich in fluorine, with which elimination of these ions become an obligation, alumina speeded up and activated charcoal is the most appropriate seen their effectiveness, their weak-willed person expenses and their availability in our country especially the region of khouribga. The quality of waters in the Khouribga region is characterized by high levels of fluoride causing, among others, dental fluorosis to the population of this region, what requires tries to eliminate this excess of these fluoride ions. The treatment of contaminated groundwater by fluoride is made by several techniques, the adsorption, one among them, is efficient and cost effective. In our case we have chosen the treatment by two chemical elements, namely activated charcoal and alumina speeded up to treat waters, which are most adapted and economic. The obtained results show that the best rate of retention has been recorded in the case of activated alumina comparatively to activated carbon. Moreover we found that the elimination of ions fluorides is influenced by different parameters, especially the pH, time of contact and the quantity of used adsorbent. Finally the acquired results show that the adsorption on speeded up alumina and activated charcoal can constitute an efficient technique for the elimination of the fluorine.

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## Ethical issue

They confirm that our publication is original.

## Competing interests

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

## Authors' contribution

All authors of this study have a complete contribution for data collection, data analyses and manuscript writing.

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## Authors Profiles



Habiba LEBRAHIMI, PhD in the scientific institute of Rabat, Morocco. Master sciences and techniques " management of food quality " in the faculty of sciences and techniques of Benimellal, Morocco. Licence sciences and techniques " protection of environment" in the faculty of sciences and techniques of Settat, Morocco. E-mail: [zaidhabibalebrahimi@gmail.com](mailto:zaidhabibalebrahimi@gmail.com).



Mohammed Fekhaoui, Manager of the Institute Scientifique, Mohamed university, Rabat. doctorate of EtatDomaine of environmental étudesScience in mohamed v university beating and university Claude Bernard Lyons. E-mail: [fekhaoui@israbat.ac.ma](mailto:fekhaoui@israbat.ac.ma).



Mohammed Fekhaoui, Manager of the Institute Scientifique, Mohamed university, Rabat. doctorate of EtatDomaine of environmental étudesScience in mohamed v university beating and university Claude Bernard Lyons. E-mail: [fekhaoui@israbat.ac.ma](mailto:fekhaoui@israbat.ac.ma).