Prediction of the location of fluoride pollution in Northern Algerian Sahara and the adsorption mechanism on the organic matrix (OM)

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Abstract:

This study aims to determine and manipulate the concentration of fluoride ions in groundwater (the northern Algerian Sahara) through the application of Geographic Information Systems (GIS). Water treatment using organic matrix (OM); the various factors affecting the removal efficiency (granular diameter diversity, temperature, contact time, -PH- adsorption dose) were changed. The physical and chemical analysis of the water showed that 62.19%. Of the well samples exceeded the limits of the World Health Organization (WHO). Standards in fluoride ion concentrations, where the high concentrations were mostly in The northeastern part, where the fluoride ions were treated through the process of physical adsorption between the highest concentration value of 2.78 mg / L and the matrix prepared from organic materials of animal origin (bovine bones), with very satisfactory results of 98%. The fluoride ion concentration Map could be used to identify and reduce health risks and mitigate groundwater pollution in the area, and on the other hand, water managers can use it to improve their water quality decisions by developing efficient and economical treatment mechanisms such as organic matter matrix used in this research.

Keywords: Ground water, Fluoride, Organic matrix, Adsorption

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Introduction

The use of modern technology in management of water resources and the protection of environmental pollution is one of the most important ways to make decisions and avoid risks that may to threaten public health. Water is a vital element for the development of life, as fresh groundwater is considered one of the most valuable sources of water on our planet. The global demand for high-quality drinking water of great importance is increasing, as it is directly related to our health (Villanueva, 2023; Pant, 2023). And our environment, where the issue of health and the environment has become one of the most important issues international modernity, and what the world has witnessed since the twentieth century of the industrial revolution and its results (Tianet et al., 2016) in advancing the wheel of development in various fields, which resulted in a rapid increase in demographic growth and thus increased demand for all sources of life. The increasing demand for fresh water is increasing day by day, especially with the climatic changes that the world is witnessing from drought. These climatic changes are one of the most important issues and topics of our time; In arid, semi-arid and dry sub-humid regions which are collectively defined as "drylands", available water resources are often too sensitive to climate variability compared to humid regions (AghaKouchak et al., 2013). In addition, generally, the more arid the climate becomes, the more long and severe droughts will occur (Li, 2021; Zhang, 2023) Africa is particularly vulnerable to global warming effects because of the important extent of arid/semi-arid regions and its overdependence on rain-fed agriculture, in fact during the last decades the continent witnessed numerous climate anomalies responsible of various food shortages and diseases; from the late 1960s to the early 1990 s persistent and severe droughts were witnessed in the Sudano-Sahelian region south of the Sahara. In the 1980s and 1990s many parts in the Horn of Africa, the Eastern Equatorial Africa and the South-Eastern South Africa experienced major hydrological anomalies ranging from severe droughts to extreme flooding (Kadomura, 2005). In more recent research (Mouhamed, 2013) revealed that extreme rainfall events have become more frequent in the West African Sahel during the 1991–2010 decade, compared to the1961–1990 period. Therefore, in order to meet the increasing needs for quality drinking water, it is necessary to go to the underground water reserves, the only way to fill this deficit. Groundwater acquires its physicochemical characteristics from the geological nature of the container rocks, and in this way determines the concentrations of the predominant mineral salts (K+ Na+, Fe2+, Mg2+, Ca2+, Cl-, SO42-, F-) and thus limits its use for drinking until it is treated according to the permissible standards. It has (OMS), European Common Market (CE) or national standards.

The Algerian desert, which covers two-thirds of the country's arid desert areas, has been the subject of many university studies, scientific articles and reports, and some studies have focused on geological exploration (Saibi, 2009), and hydro geologist (Radi, 2023). Despite the very arid nature of the climate, the regions of the northeastern Algerian-Tunisian desert contain the two largest systems of aquifers in the ground, the Continental Intercalaire aquifers. , which is a very deep layer in the Northeast Basin, containing hot water and often rich in minerals, and the "Complexe Terminal" aquifers, which are less deep but characterized by salinity. These different aquifers contain very important water supplies, which are currently widely exploited for domestic and industrial uses and for the irrigation of palm groves. Various studies have also focused on a broader assessment of aquifers, proposing different scenarios for their exploitation in the medium and long term (UNESCO., 1972; NESSON, 1978; OSS, 2003).

Despite the chemical quality of groundwater that exceeds the limit allowed by the World Health Organization (WHO). Many problems arise in its exploitation. As the people of these areas in southeastern Algeria suffer from high fluoride in the water, which has caused the spread of dental fluorosis (Messaitfa, A, 2008; Tabouche A. , 2004; Choubisa, 2022). Dental disease due to prolonged exposure to fluoride (Acharya, 2005; Arbab Chirani R, 2005; Chavaissieux P, Meuinier PJ, 1995) content in the exploited groundwater in those areas. (Aroua, 1981; Chavaissieux P, 1995) and the population's diet (eating tea and dates characteristic of the region) also plays an important role in increasing the incidence of this disease (Chavaissieux P, 1995; Baouia, K, 2017; Belmabdi A, 2011; Messaitfa, A, 2008; Choubisa, 2022.). As scientific contribution to this paper, the performance of the application of GIS was evaluated Ouargla, Touggourt and Ouad Souf state northern Sahara of Algeria. To predict the potential of fluoride in groundwater exceeding 1.5 mg/L and using GIS. Physicochemical analysis of groundwater in the region, to determine the spatial distribution of fluoride ions. It allows prediction and monitoring of fluoride concentrations to identify health risks and mitigate groundwater contamination with fluoride. On the other hand, the high concentration of fluoride ions was treated with the locally available organic matrix of animal origin (bovine bone), by physical adsorption. Water managers can use it to improve their water quality decisions. Fro the study area by developing effective and economical treatment mechanisms.

MATERIALS AND METHODS

1 Study area

The eastern region of the northern Sahara (Ouad Souf, Touggourt, and Ouargla) (Figure 1), which represents the study area as it is located in the north-east of the Algerian Sahara. Located between the northern latitudes 32 degrees and 34 "and the eastern longitudes 4 degrees and 8 degrees, the large basin of Ouargla covers an area of 99 thousand hectares, its length is 45 km towards the southwest and northeast, its width is 2-5 km and its circumference is at an average altitude 164 A.D. It occupies the lower end of Wadi Mih, which drains the northern slope of the Tidemet plateau and ends at Sebkhat Sefion, 20 km north of Ouargla. In the north, at an altitude of about 70 m, northeast of the desert, between Erg El Sharkia and the Aures Mountains, with an area of 1,334 square kilometers, as for Ouad Souf, in the far northeast of the study area, it is bordered to the south by Touggourt, to the southwest by Touggourt, Ouargla, and to the north by Biskra Tebessa and Khenchela to the east of the Tunisian Republic, and to the west of the state of Djelfa, occupying an area of 44,586 square kilometers.

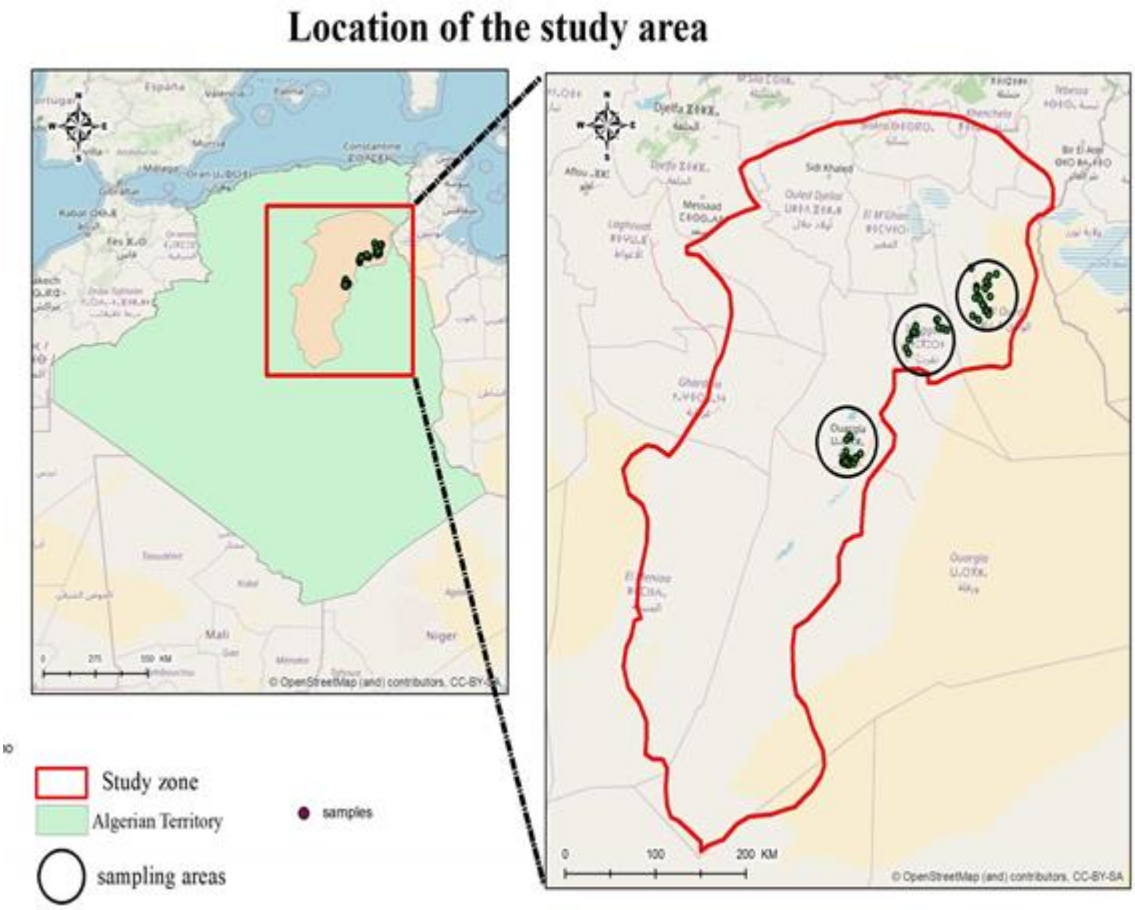

Figure 1. Location map of the study area (north-southeast) Algeria.

Eighty two (82) water samples (Figure 1) from two aquifers (Mio-Pliocene), were taken collected in 1.5 liter plastic bottles, and intended for physico-chemical analyzes during the period from March to June 2021. They were transported to the laboratory the same day. The samples were placed in a thermostat (Rodier, 1996). Our sampling strategy was to identify the different aquifers throughout the valley by accessing the maximum available water points exploited by the population of the region in the distribution on the territory of the municipality of Ouargla, Touggourt and Ouad Souf. . The layout of Mi-Pliocene boreholes for agricultural purposes, in most cases, while samples were taken from the boreholes intended for AEP located in the three cities.

2. Data analysis

Three parameters: hydrogen potential (PH), electrical conductivity (EC) and total dissolved solids (TDS) were measured in situ, immediately after sample collection. Portable "Multiparameter" measurement Type HI 9829 from the HANNA range with location of measurements by GPS function and ease of use.

Analytical measurements of the content of fluoride ions in groundwater. Uses the DR 6000 type spectrophotometer, with molecular absorption at 580 nm. And measurement steps; Using a pipette 10 ml of distilled water in an assay vial (the blank), having 02 ml of the "SPNDS" reagent. We put the sample vial in the cell and we press play. After 60 seconds, the device warns, according to a distinctive self-stimulation; the result will mark in [F-] mg/l.

Data preparation and GIS analysis geospatial database was created after extracting laboratory results obtained during the physical and chemical analysis process.

Available databases cover the study area of exploited Mio-Pliocene reservoirs. The collected data are processed with the technique of inverse interpolation of distance weighted (IDW) and spatial distribution maps are generated for the sampling communities. For waters in the Ouargla, Touggourt and Ouad Souf regions to avoid errors of the IDW method and make the spatial analysis applicable (Magesh et al. 2013; Kawo and Shankar 2018 Balamurugan et al. 2020b; Sarfo and Shankar 2020).

Weights were set for different parameters and the distance was calculated at each site, taking into account the closest selected sites. Figure 2

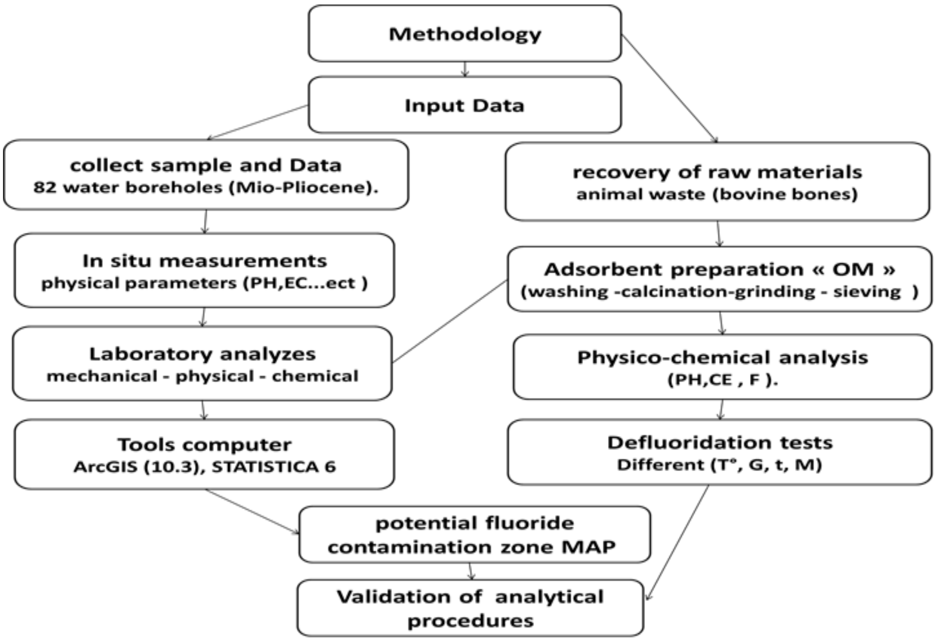

Figure 2. Stepwise procedure used to determine the concentration and removal of fluoride ions in groundwater

3. Prepare the OM and Defluoridation tests

The organic matter (beef bones) was washed with distilled water several times to remove dirt particles and water-soluble materials. The washed materials were then completely dried in an oven at 105°C, 200°C and 400 °C for 24 hours. They were crushed using a rotary jaw crusher. The latter was ground using a ball mill and then sieved using two sieves to retain only particles with a particle size between 50µm and 1 mm respectively. Without the addition of any chemicals (Figure 2).

The initial concentration of fluoride ions solution was 2.79 mg/l for all experi- ments. For fluoride ions removal kinetic experiments, the batch method was used because of its simplicity. About 1 g to 6 g of organic matrix was contacted with 40 mL of fluoride ions at PH = 7.49 solution in a sealed flask agitated vigorously by a magnetic stirrer controlled shaker at a speed of 150 rpm. The adsorption capacity was calculated by using the following equation:

$$qe = \frac{(C0 - Ce) * v}{m}$$

Where qe (mg/g) is the adsorption capacity, C0 and Ce (mg/l) are the initial and equilibrium concentrations of F- , and any time, respectively, V (L) is the volume of the solution and m (g) is the weight of used biosorben For the calculation of fluoride ions rate adsorption (R %), the following expression was used Al (Sagheer et al. 2009; Balarak et al. 2016):

$$R(\%) = \frac{(C0 - Ce) * 100}{C0}$$

RESULTS AND DISCUSSION

4. Physico-chemical characterisation of ground water:

Table 1 presents the descriptive statistics of the physico-chemical parameters of the study area. For each parameter measured, the minimum and maximum, the median value, the standard deviation and the coefficient of variation are indicated. CV in %) obtained from the expression CV=100 σ/moy makes it possible to show the degree of homogeneity of the data obtained. (Rodbard, D 1974) If CV < 2%, the measurements are very homogeneous; If 2% < CV< 30%, the measurements are homogeneous; and If CV > 30%, the measurements are heterogeneous (Reed, JF; Lynn, F; Meade, BD 2002).

Variable		CE	PH	TDS	F-
Touggourt	Mean	5362.9	7.3085	2874.2	2.156
	Median	5679	7.295	2823.5	2.275
	SD (σ)	1680.02396	0.24512671	933.928804	0.43987319
	Min	1216	6.83	1642	0.92
	Max	7658	8.1	6098	2.78
	Coefficient of variation in %	31.3	3.4	32.5	20.40
	Homogénéité des données	heterogeneous	Homogeneous	heterogeneous	Homogeneous

ourgla	Mean	3422.0625	7.34625	3370.4375	1.2715625
	Median	3382.5	7.325	2632	1.21
	SD (σ)	856.179056	0.20760928	3430.76253	0.39157341
	Min	2100	7.03	1275	0.58
	Max	5560	7.97	21460	2.58
	Coefficient of variation in %	25.0193869	2.82605791	101.789828	30.8
	Homogénéité des données	homogeneous	homogeneous	heterogeneous	heterogeneous

Table1.The principal statistical characteristics of the collected

4-1. Hydrogen potential (PH):

The pH of Mio-Pliocene groundwater remains homogeneous and close to neutral to slightly alkaline. It expresses the concentration of H^+ ions in the water. Thus, it determines the acidity or alkalinity of water with a report of the study area. PH values range from 6.83 to 8.10 with an average of 7.324. It is between 7 and 8.5 according to WHO and between 6.5 and 8.5 in relation to CEE The most common values are within the range of the drinking capacity criterion, that is, more 98% of the boreholes in the area are suitable for human consumption. Inland waters are bicarbonate alkaline, with a pH often below 8.3. This was indicated by some previous studies (Edmunds et al, 2003; Bouchahm et al, 2008). Most groundwater in areas that contain high concentrations of fluoride have a high pH and this is not indicated by the results obtained (Table 1). Which can be traced back to the geological age of this water, which over time has allowed it to adjust its acidity, since we see that the maximum reaches 8.10 and the ear is around 6.83 within the recommended limits by the WHO.

4-2. Electrical conductivity (EC):

The electrical conductivity (EC) in the Mio-Pliocene waters of the Touggourt region varies between 1216 and 7658 (µS/cm), with an average value of 5362.90 (µS/cm). These values reveal variable and excessive mineralization across the northeast to southeast direction of the study area. This mineralization increases from South to North, according to the presumed directions of flow (Figure 3). In the Ouargla valley, we find the (EC) values obtained vary between 2372 and 5425 µS/cm (Table 1) with an average value of 3450.34 µS/cm and a standard deviation (800.65 µS/cm). It thus expresses, a total mineralization exceeding drinking water standards, 1000 mg/l (WHO, 1994).

It shows changes in (EC) to mineralization, it is higher in the region, and this exceeds the recommendations of WHO. Therefore explains the high [F-] on the one hand and on the other hand, Figure 3 and 4. Which represents the [F-] in terms of (EC), shows an increase in high [F-

], the higher the (EC), and this is due to several factors, including the geological nature of the area in terms of soil and rock porosity, according (Mazet,P ,2002).

We find fluoride-rich water in sedimentary basins, volcanic regions, and rock-crystalline rocks. The hydrodynamics of the aquifer system also plays an important role in increasing fluoride concentrations. The contact time between water and rock (Chernet et al., 2001; Ponti et al., 2006), and the salts that pass through The water passes through it before it reaches the groundwater level. In addition, the chemical composition of groundwater and its ability to dissolve fluoride are all factors that contribute to the increase in fluoride concentration, and this is confirmed by (O.M.S 1972; Raop, 1988).

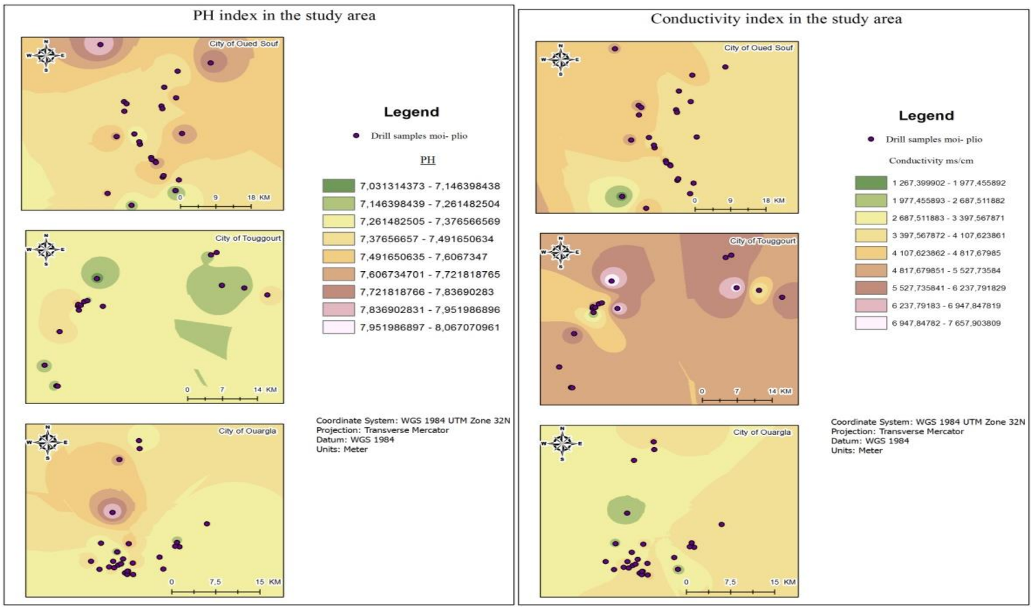

Figure 3: Spatial distribution maps showing the concentration of PH and Electrical conductivity.

4-3. Fluoride ion variation [F-]:

[F-] concentrations vary from 2.78 mg/L to 0.92 mg/L. The value of the standard deviation (0.58 mg/L) reflects a variation in concentrations with an average of 1.9 mg/L. Almost all the water points in the Touggourt region have fluoride levels that frequently exceed WHO standards. Where their concentration is slightly below the standard range such variations may be due to sampling conditions. The most frequent concentrations are between 1.8 and 2 mg/L and between 2 and 2.5 mg/L and represent approximately 95.17% of the samples.

No value of [F-] exceeds the potability standard defined for supply water in the Ouargla region (Table 1).Consequently, these waters do not present any potential risks to human health. Concentrations vary from 0.58 mg/L to 1.74 mg/L. These data are not very scattered because the standard deviation observed (0.27 mg/L) is low compared to the Touggourt region with an average of 1.206 mg/L. The most frequent concentrations between 0.98 and 1.49 mg/L reflect the affinity in the concentrations with an average of 1.042 mg/L. This groundwater is considered scientifically drinkable, but due to the nature of the hot zone and the way of life and foods (tea, dates, pulses, etc.), rich in fluoride, (Messaïtfa, 2008; Belmabdi and Messaïtfa, 2014; Baouï, 2017).These concentrations are considered high.

In relation to its distribution in the study area. The Map (Figure 4) shows the Interpolation Method (IDW). Of the distribution of fluoride ions according to the concentration. Where we find a clear difference from the southeast towards the northeast direction of the study area. to increase the value from 0.58 to 2.78 mg/L. so the high percentages are concentrated east of the Touggourt region. As we rise to the surface, which means that the geological, chemical and physical nature of the area and the high mineralization that has passed through It has a maximum electrical conductivity of 7568 µS.cm-1. Which means the presence of soluble minerals in water, especially calcium and bicarbonate ions, which have a clear effect on increasing or decreasing fluoride concentration, and this is what the above studies have shown (Chandra et al.. 1981) . Acidic pH can influence dissolution of salts from the aquifer rocks and increase the metal and TDS load in the ground water (Akhtar, 2014) pH, bicarbonate and calcium concentration also plays a crucial role for the dissolution of fluoride from rock minerals to ground water (Saxena, 2001) .

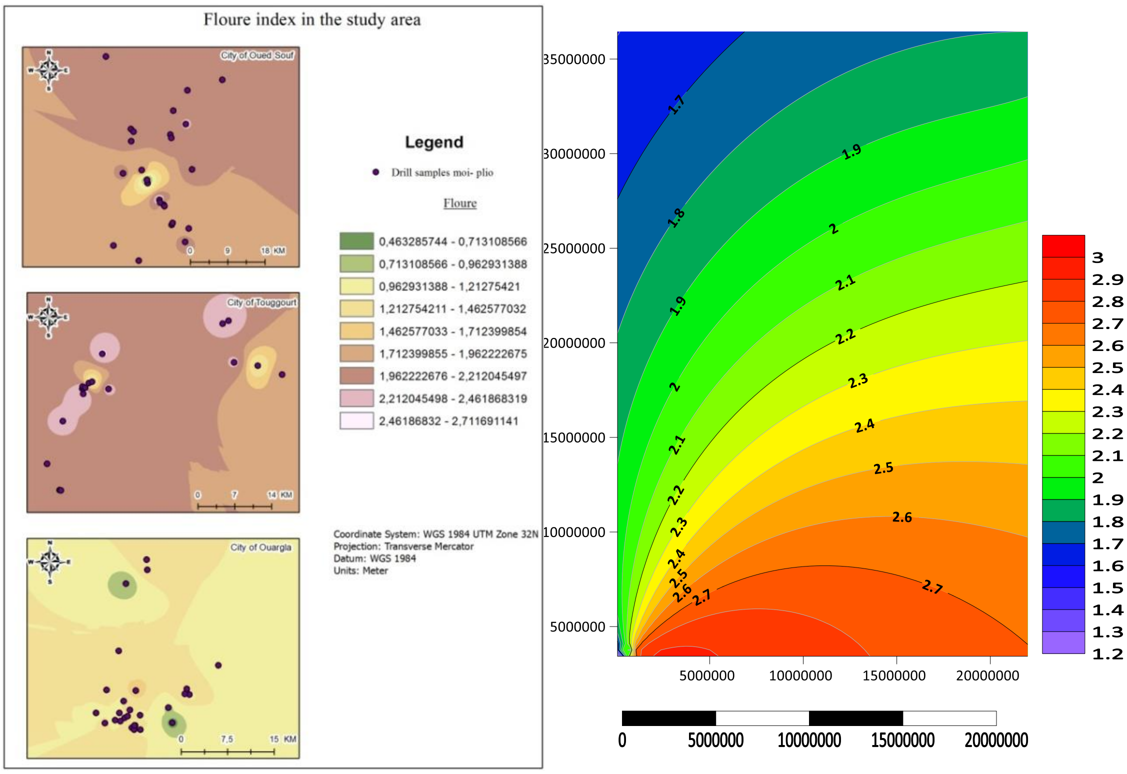

Figure 4 .Fluoride concentrations map of the study area.

5. Effect of operating conditions to remove fluoride:

5.1 Effect of contact time:

One of the factors affecting the adsorption is the contact time. In order to study the effect of the contact time between water and the (OM), prepared at different temperatures (105,200 and 400 C°) with different grain sizes (50,200,500μm and 1mm) on the adsorption of fluorine ions and deduce the optimal time by increasing the contact time from 5 to 150 min. According to (Figure 5, 6 and 7) in contrast, (Figure 8) represents the adsorption yield of fluoride ions on the organic matrix prepared at a different temperature.

The amount of polarization increases with the increase in the reaction time, most of the recorded results are consistent with the studies, (BEN NASER A, 2011; ISMAIL Z A.H., 2015; ISMAIL Z A.H., 2013). Where the fluoride ions decrease with the increase in the reaction time and from here, we adopt the appropriate reaction time in this study 60 min. That the concentrations of fluoride ions were recorded within the standards of the WHO, and at the same time the adsorbed amount remains constant until it reaches the stage of stability and saturation, which proves that this substance has the ability to remove fluoride ions and has the ability to permanently remove it from the water.

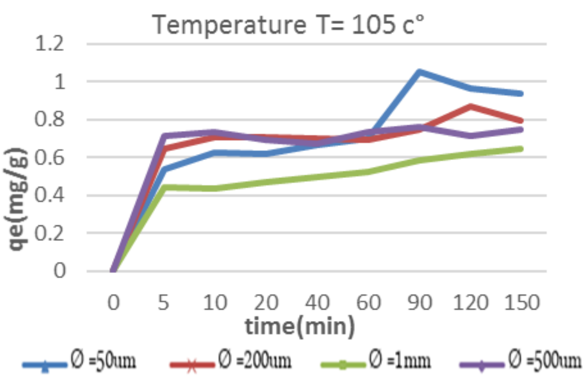

Figure 5 Effect of granular diameter diversity of OM on adsorption at a at a temperature 105 C⁰

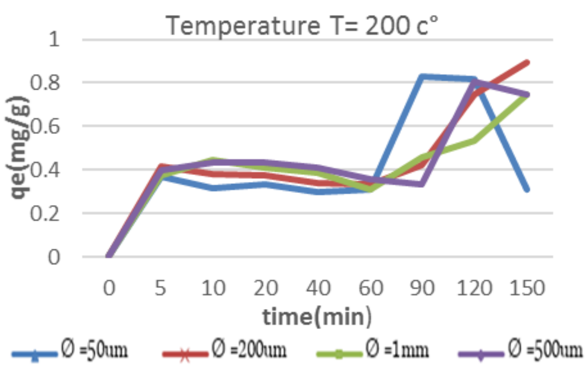

Figure 6 Effect of granular diameter diversity of OM on adsorption at a at a temperature200 C⁰

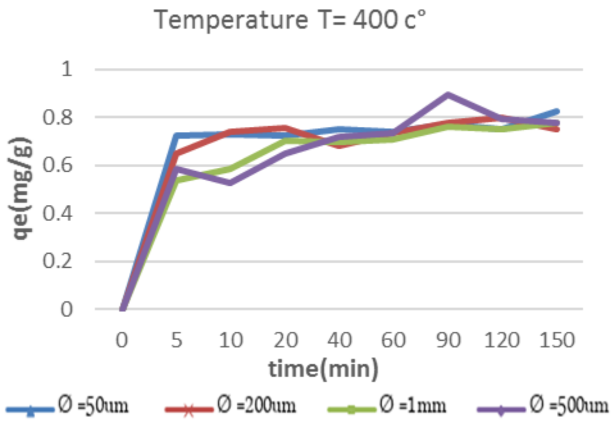

Figure7 Effect of granular diameter diversity of OM on adsorption at a temperature 400 C0

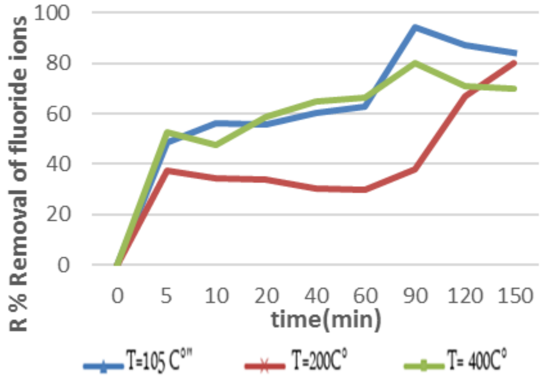

Figure 8 Effect of contact time on F- adsorption by OM (concentration: 2.79 mg/ Solution pH: 7.46 Agitation speed: 150 rpm, Dose 1 g/ 40 mL).

5.2- Effect of granular diameter diversity

5.3-Effect of the temperature

The difference in the granular diameter did not have a clear effect on this process. Although the increase in the specific area of the matrix is related to the granular diameter. Where the less the granular diameter, the greater the specific area and instead the number of active sites increases. Thus increases the adsorption efficiency, but in this case we consider it unimportant So that the diameters converge to each other, and this means a convergence in the specific area of the adsorbent. The convergence of the specific behavior of the matrix made our experiments possible to reach lower levels of fluoride ions, less than the limit recommended by the WHO. The polarization of fluoride in the reaction medium, where the average granular diameters Ø = 50 μm. Can be taken, chosen for the OM in this study (Ahmed D, Amar, M., & Sofiane, S, 2022).

By comparing the results recorded in Figure (5, 6, 7, and 8) during the adsorption process on the OM at a different temperature, we notice a clear increase in the amount of polarized fluoride and a good yield with the length of the contact time until it reaches or approaches 100% (Figure 8) with the increase of time interaction. When comparing the results in the first 5 min, we find the best polarization at a drying of 105C°. 90 min was sufficient to reach a quantity of polarization with a yield of R = 94.60 % at a grain diameter of 50μm. While at 200C° and 400C° the reaction took more than 90 min to reach an amount Polarization with a yield in the range of R = 70 %.

With regard to the temperature, we can rely on the organic matrix at 105 C° after the sterilization process as a first stage in the treatment to reduce fluoride ions in the water of the region, taking into account the organic matter resulting from the use of cow bones.

The phenomenon of adsorption on the organic matrix of cow bones and its ability to remove fluoride ions from the studied water is explained by the phenomenon of substitution between the fluoride ion in the water and the hydroxide ion (OH-) of the bones. The alternating adsorption mechanism between fluorine and one or both hydroxyl groups explained according to the interactions proposed by (Chidambaram S, 2003)

$$Ca_{10}(PO_4)_6(OH)_2 + F^- \longleftrightarrow Ca_{10}(PO_4)_6FOH + (OH^-).$$

5.4-Effect of Hydrogen potential

The results show a follow-up of the pH index, which is a very important factor in the adsorption phenomenon. Figure 9 shows the change in pH, which shows two periods. The first period notes that the interaction between the OM of cow bones with the treated water (the beginning of the reaction) increases the pH from 7.49 to a maximum value in limits of 8.34, when the OM dried at 0105C°. High PH values indicate the liberation of hydroxide ions in the medium (OH-) contained in the used bone hydroxyapatite, which confirms the phenomenon of substitution between F^- and OH^-.

As for the second period, we notice a decrease and then stability in the PH values. As we explain this decrease from 8.34 to 7.08. By liberating hydrogen ions in the medium H^+, and based on the principle of fluorine affinity for apatite (3 (Ca3 ($P0_4$) 2, CaC03)). Where carbonate ions can be replaced with fluoride ions; it is believed that under our conditions the adsorption result observed can be explained by the substitution between F^- ions and $3PO_4^-$ ions, according to the following reaction (Mazonie P, 1984; Srimulari M, 1998; Agarval M, 2003).

$$Ca_{10}(PO_4)_6(OH)_2 + 3F^- \longrightarrow Ca_{10}(PO_4)_5F_3OH + PO_4^{3-}$$

The two ions, released and hydrogen most likely resulting from the ionization of water, yield phosphoric acid, which is responsible for this lower PH.

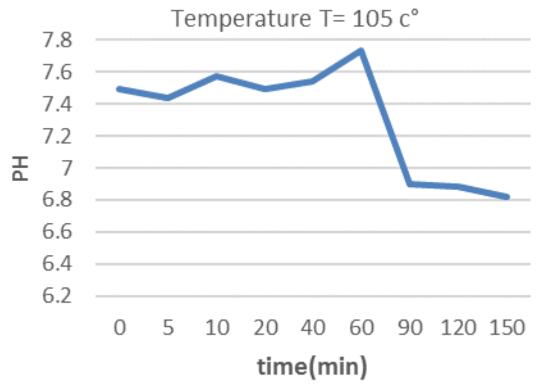

Figure 9 The PH evolution of the reaction of OM and treated water

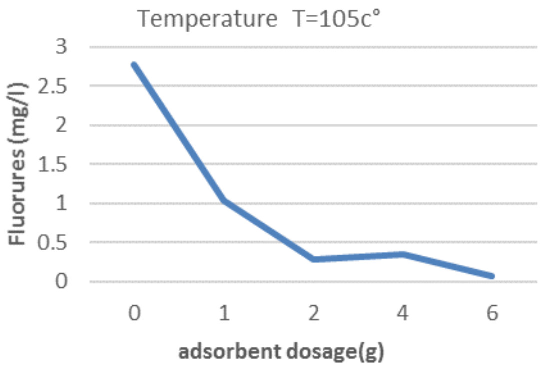

Figure 10 Effect of adsorbent dosage on F- adsorption by OM (concentration: 2.79 mg/L, Solution pH: 7.46 Agitation speed: 150 rpm, Temperature T=105c°, granular diameter 50 µm)

5.5-Effect of adsorbent dosage

Adsorbent dosage is one of the important parameter studied while conducting batch adsorption studies.The effect of adsorbent dosage on removal of F^- ion concentration was studied by varying dosage from 1 to 6 g as shown in Figure 10.

It was found that adsorption increased with increase in mass of adsorbent (OM) That is, increasing the active sites and thus increasing the percentage of fluoride ions removed, (Hanumantharao, Kishore, and Ravindhranath, 2012). Maximum removal efficiency of 100% for F^- (from initial concentration of 2.79 mg/l) was observed when 6g of (OM) was used.This result confirms what (Piekos R, 1998; Moufti A, 2004; Diawara CK, 2004; Samb F., 2004). Has found in reducing fluoridation.

CONCLUSION

In North Africa, the inhabitants of southern Algeria, particularly the south-east (northern Sahara), depend on groundwater, the only source of life in the region. This water has a very high mineralization and has caused many problems. The dissolved salts in the body, which results in kidney exposure to kidney failure and could be considered as a future threat to humans, animals and plants. Water quality monitoring shows that the electrical conductivity in the waters of the region varies between 1216 and 7658 (µS/cm). On the other hand, the pH of the Mio-Pliocene remains homogeneous and close to neutral to slightly alkaline .98% of drilling suitable for human consumption.

Currently, groundwater in the southeast is characterized by increasing concentrations of fluoride ions represents high concentrations that reach 2.78 mg/l. Exceeds WHO standards. We relied in this study to a large extent on the value of fluoride in groundwater, where 70% of the well samples reached a concentration higher than the permissible limit in the World Health Organization. Which has an impact on teeth and bones, is difficult to reduce its levels in the diet. For this, individuals and locals should follow a diet and avoid foods that contain high levels of fluoride, such as tea, dates and fish. It is recommended to use toothpaste without fluoride or use miswak (chewing stick) What the Prophet Muhammad (peace be upon him) recommended centuries ago to improve oral health and prevent cavities.

On the other hand, the use of processes Defluoridation of drinking water. By the low concentration of this substance in the water which remains the best solution to obtain a level of fluoride in the human body according to the recommendation of the WOH. The results obtained show that the adsorption of fluoride ions on calcined bones is very satisfactory. Lower than the WHO recommendation (1.5 mg/L). Despite the difference in calcination temperatures and particle size. The Bovine bones have been shown to be very effective in adsorbing fluoride ions. We can improve these results and apply them in practice to reduce fluoride pollution, recover organic waste in water treatment, and preserve the environment and human health in the context of sustainable development.

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