

Defluoridation of Groundwater with the Help of *Azadirachta indica* leaves as Bioadsorbent in Korba, Chhattisgarh, India

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ABSTRACT

In this paper, we have used a removal technique of fluoride from groundwater in Korba district, Chhattisgarh, using thermally activated neem (*Azadirachta indica*) leaves as adsorbents. For this purpose, we collected the groundwater sample in January–March 2021. The Ion-Selective Electrode (ISE) technique was used to assess the fluoride concentration in groundwater samples. Neem leaves were efficient at removing fluoride in this study. Fluoride has a split personality in the human system, having a damaging impact when fluoride concentration is more than 1.5 mg/L, causing dental and skeletal fluorosis, and a positive effect when concentration is less than 1.0 mg/L, causing caries preclusion, and health promotion. This small project provides the outcomes of a study on neem leaf powder for water defluoridation. The analysis here discusses the applicability of inexpensive leaf adsorbents for successfully remediating fluoride contaminated water: contact time, pH, and adsorbent concentration all influence fluoride ion sorption effectiveness. The effects of treated leaf powder on pH, adsorbent dose, and contact time with aqueous solutions containing 2.28–10.04 mg/L fluoride ions were investigated. Fluoride adsorption is most substantial at pH 2. Fluoride removal diminishes dramatically when the pH exceeds 2. At adsorbent doses of 10 g/L, the necessary time for fluoride ion adsorption equilibrium is 120 minutes, and the highest removal efficiency attained was 85%, during that amount of adsorbent was 12 g/L. This research also discusses fluoride's adsorption isotherm and kinetics by activated neem leaf powder.

KEY WORDS: ADSORPTION, AZADIRACHTA INDICA, BIOADSORBENT, DEFLUORIDATION, REMOVAL EFFICIENCY.

INTRODUCTION

Fluoride ions are negatively charged and are found in various minerals and may be in water and soil (Tolkou et al. 2021). Fluoride in ordinary water has developed into a severe anxiety in many parts of the world for humankind, and it should be within 0.6 – 1.5 mg/l (Patil et al. 2015; Prasad et al. 2021). India is one of the countries with many people affected by fluorosis due to drinking groundwater. Dental and skeletal fluorosis, neurotoxicity, endocrine impacts, and attention deficit hyperactivity disorder are among the side consequences of excessive fluoride exposure (Akafu et al. 2019; Kashyap and Ghosh 2021).

Because the difference between the intended and unwanted fluoride dose is so tiny, it's critical to monitor and consider the quality of drinking water and, when required, eliminate

excess fluoride from water to safeguard human health (Premathilaka and Liyanagedera 2019). The National Research Council of the United States evaluated EPA fluoride standards in (2006), They concluded that fluoride has direct and indirect effects on the brain, that excessive fluoride levels in water is also neurotoxic, for which a lot of analysis is required (Philippe Grandjean 2019). Although groundwater provides protected potable water for billions of people worldwide, there are rare instances where pollution levels are too high, and groundwater cannot be utilized for potable purposes without treatment. Fluoride poisoning of drinking water is a significant environmental hazard that affects much of the world's populace (Gronwall and Danert 2020; Kashyap and Ghosh 2021).

Fluoride concentrations in the water are so high in nations like India, Ghana, China, Pakistan, Bangladesh, and Tanzania (Yadav et al. 2019; Nde-Tchoupe 2019) that hundreds of thousands of people suffer from fluoride-related diseases. Fluoride concentrations in the water are so high that thousands of individuals are affected by fluoride-related

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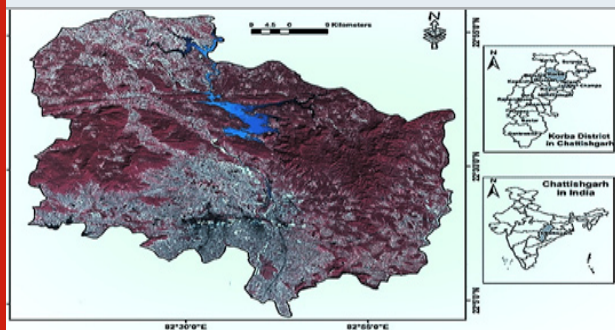
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conditions. Effective fluoride exclusion in these areas is a complex and fascinating undertaking due to chemical hurdles preventing fluoride from being successfully sorbed by most traditional adsorbents utilized in the drinking water industry (Rasool et al. 2018; Yadav et al. 2019). In addition, a lack of required infrastructure and technological know-how plays a crucial role. On the other hand, China and India are putting up significant effort to resolve the issue, and the situation is slowly improving (Wang et al. 2019; Kisku and Sahu 2019; Kashyap and Ghosh 2021).

Consequently, fluoride exclusion from drinking water has attracted a lot of interest in recent years, with many methods being explored in labs or used in the field (Yadav et al. 2018; Tolkou et al. 2019). Bio-remediation is the most common technology used to eradicate fluoride from drinking water. Adsorption methods are becoming more popular due to their ease of use and extensive selection of adsorbents. With varying degrees of success, research has been conducted on various types of low-cost adsorbents (Kanaujia et al. 2018; Kumar 2019; Yadav and Bhattacharya 2020). Plant-based bioremediation to enhance water quality has become a hot topic of research. For defluoridation, several plant materials such as coconut shells, tamarind seeds, neem leaves, and neem bark have been utilized as adsorbents. In the treatment of water, activated carbon is frequently utilized. For the cleaning of polluted water, bioremediation is widely regarded as a cost-effective and environmentally beneficial solution. This research aims to scrutinize the usage of neem tree leaf powder for groundwater Defluoridation (Mihavo et al. 2021).

Figure 1: Location of Korba district in Chhattisgarh (www.korbadistmapimage.com)



MATERIAL AND METHODS

The Korba district in Chhattisgarh's coal-based thermal power center from 22°02'50" to 23°01'20" N and from 82°07'20" to 83°07'50" E. The study area in different areas is full of natural possessions (Singha et al. 2019). Coal is the largest resource in the field of research. Talchir, Karaharbhari, Barakar, and Kamthi formations are characteristic of the Gondwana rock supergroup. The construction of the Talchir consists mainly of shale, sandstone, and a boulder bed. The formation of Karaharbhari is made up of subgraywacke, sandstone, and gallstone (Singha and Pasupuleti 2020). The appearance of Barakar includes arcose sandstone, shale, and carbon seam. Kamthi formation consists of coarse ferruginous carbonated sandstone with extremely fine coal

seams, whereas the CGC consists of undifferentiated granite and metamorphic litho units (Singha et al. 2019; Singha and Pasupuleti 2020).

Table 1. Fluoride Concentration in selected groundwater samples of Korba district in mg/L

SN	Sample spot and Sample code	Fluoride Concentration in groundwater samples in mg/L
1	Tanera (S1)	2.28
2	Fulsar (S2)	4.52
3	Aamatikra (S3)	6.0
4	Aamatikra basti (S4)	7.46
5	Pondikala (S5)	8.18
6	Pondikala basti (S6)	10.04

Table 2. The significance of the initial adsorbate concentration, Operating order for the consequence of initial adsorbate concentration, Adsorbent dose - 10 g/L, Temperature was adjusted 27°C ± 0.5°C, Time of contact - 120 Minutes, Volume of sample - 100 ml, and pH 2.

SN	C _i (mg/L)	C _e (mg/L)	Elimination % of F-
1	2.28	0.04	98
2	4.52	0.36	92
3	6.0	0.90	85
4	7.46	1.49	80
5	8.18	1.75	80
6	10.04	2.20	78

For the sampling of Groundwater and materials, borewells and hand pumps gathered field samples in various places around the Korba district. At least 1000 ml of each sample was acquired in cleaned plastic cans or bottles and kept at room temperature in the lab for analysis. Six field samples were obtained. For this study, groundwater samples were collected from the highly fluoridated zone of the Korba district. This study was conducted from January to March 2021. This device has the ISE and meter (Thermo Scientific, Orion Star A214), TISAB-III, and a fluoride standard of 100 mg/L. In the fluoride analysis, plastic goods were employed.

For the preparation of adsorbent, the leaves of the neem tree (*Azadirachta indica*) were exploited as a low-cost natural adsorbent. These were found in Naya Raipur, India, behind the Kalinga University Campus. The neem leaves were dried, crushed, and thoroughly washed with double distilled water. They were dried for 24 hours in a hot air oven at 60–90 °C, following which the dried material was crushed in a jaw crusher and screened at ASTM 50 m mesh. For the alkali treatment, a leaf biomass fine particle sample (40 gm) and 400 ml of 0.5 N NaOH was mixed in a 1000

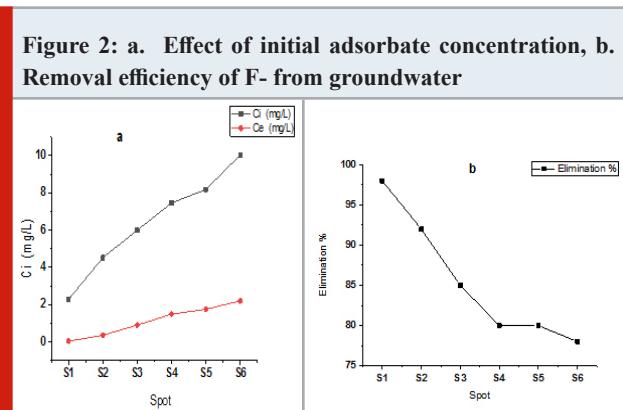
ml round bottom conical flask. The liquid was progressively heated on the burner for 20-25 minutes after it began to boil. The treated biomass was rinsed with double distilled water until the color was gone entirely, and very clear water was generated.

RESULTS AND DISCUSSION

For this investigation, high fluoridated six groundwater samples were selected from Korba district regions for fluoride analysis of samples used the ISE method. The fluoride content in this region varied from 2.28 to 10.04 mg/L shown table 1.

The successful use of the adsorption process necessitates developed low-cost, non-toxic, conveniently accessible, and locally available materials. Bioadsorbents meet these requirements. If the ideal circumstances were known, a better design and modelling process would be ushered in. From a kinetic standpoint, various essential factors, such as pH, contact duration, starting adsorbate concentration, and the adsorbent dose, were studied. Experiments like batch adsorption investigations were carried out (Birhanu et al. 2020). Experiments were conducted by agitating 100 ml of obtained groundwater samples containing F⁻ at a speed of 200 strokes per minute while shaking 10 g/L of adsorbent. To maintain samples containing F⁻ at the proper pH, 0.5 N HNO₃ was added. At room temperature (27°C ± 0.5°C), all of the measurements were taken (Birhanu et al. 2020).

Effect of Initial Adsorbate Concentration: During the optimal time of contact, the rate of a purely adsorptive reaction varies in lockstep with the concentration of adsorbate. As the initial fluoride ion concentration grows, the capacity of the adsorbent materials is rapidly exhausted. The adsorption capability of treated biosorbents was studied methodically by increasing the starting concentration of F⁻ from 2.28 to 10.04 mg/L.



The percent of elimination of fluoride ions varies through starting concentration at various initial pH levels. When the starting concentration of F⁻ was raised from 2.28 to 10.04 mg/L with a constant sorbent dosage of 10 g/l at a pH of 2, treated bioadsorbents were shown to be reasonably active in decreasing fluoride ions from 98 to 78% shown in table 2 and figure 2 a and b (Philip et al. 2021).

Adsorption Isotherm model: To fit the investigational adsorption equilibrium data (Yuan et al. 2020; Philip et al. 2021) of fluoride on a bioadsorbent (neem leaves), the isotherm models of Langmuir, Freundlich, and Temkin were applied (Ali and Ismail 2021). These models are mathematically expressed as follows:

Langmuir isotherm:

$$\frac{C_e}{q_e} = \frac{1}{q_m} C_e + \frac{1}{K_a q_m}$$

Where C_e is the equilibrium concentration of adsorbate (mg/L), q_e denotes the quantity of adsorbed per gram of the adsorbent at equilibrium (mg/g), K_a represents the Langmuir isotherm constant (L/mg) and q_m is the highest adsorption capacity (mg/g) of the adsorbent. As shown in Table 3 and figure 3 a.

Freundlich isotherm:

$$\log q_e = \log K_f + \frac{1}{n} \log C_e$$

Where K_f is the Freundlich isotherm constant $\text{mg}^{1-(1/n)} \text{L}^{1/n} \text{g}^{-1}$, n indicates the adsorption intensity, (Priyantha and Kotabewatta 2019; Yuan et al. 2020). As shown in Table 3 and figure 3 b.

Temkin isotherm:

$$q_e = \frac{RT}{b} \ln K_T + \frac{RT}{b} \ln C_e$$

Where b is Temkin constant which is related to heat of sorption (J mol^{-1}), (Altun et al. 2021) and K_T is Temkin isotherm constant (Lg^{-1}). As shown in Table 3 and figure 3 c.

Effect of contact time: The elimination of F⁻ is shown to increase with increasing contact time duration to some extent. Increased contact time does not improve absorption due to fluoride ions on the accessible adsorption sites on the adsorbent material. According to early studies, fluoride ions appear to absorb quickly on the adsorbent material at their ideal pH values. Within the first hour of interaction with fluoride ions at a concentration of 10.04 mg/L and an adsorbent dose of 10.04 mg/L for treated bioadsorbents, 62% of the adsorption occurs. After this rapid adsorption, a more gradual approach to equilibrium occurs, with saturation happening in 1.5 to 3 hours. This contact time was used as the equilibrium period for further optimization of other parameters (Ali and Ismail 2021).

Adsorption kinetic studies: Adsorption kinetics is a critical parameter that indicates adsorption efficiency. Figure 4 depicts the fluoride adsorption rate on the surface of adsorbents as a function of time (Siva kumar et al. 2019; Akinyeye et al. 2020). The pseudo-first-order rate (Lagergren 1898), equation is represented by follows:

$$\log (q_e - q_t) = \log q_e - \frac{K_1 t}{2.303}$$

Where q_t and q_e are the total of fluoride adsorbed (mg/g) at contact time t and at equilibrium respectively and k_1 is the pseudo-first-order rate constant (min^{-1}). Pseudo-first-order rate constant k_1 and the equilibrium adsorption capacity q_e were determined from the slope and intercept of the plots of $\log(q_e - q_t)$ against time are shown in figure 4a and table 5 along with the correlation coefficient (R^2) (Ali and Ismail 2021).

Figure 3: a. Langmuir isotherm studies plot not fitted for activated neem (*Azadirachta indica*) leaves as bioadsorbents for used defluoridation of groundwater. b. Freundlich isotherm studies plot fitted for activated neem (*Azadirachta indica*) leaves as bioadsorbents for used defluoridation of groundwater. c. Temkin isotherm studies plot not fitted for activated neem (*Azadirachta indica*) leaves as bioadsorbents for used defluoridation of groundwater (Ali and Ismail 2021).

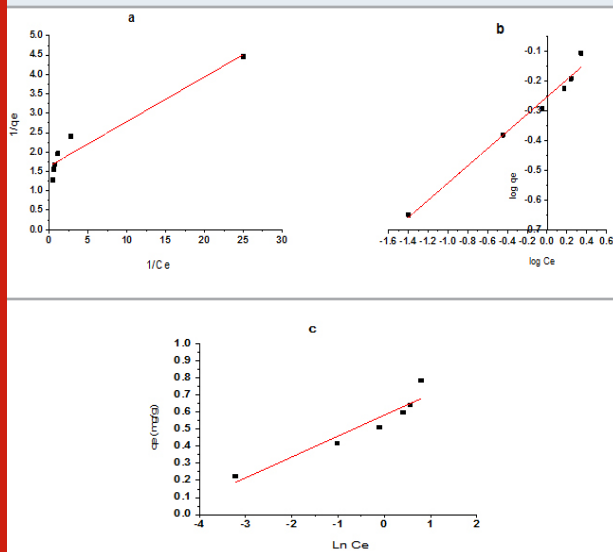


Table 3. In the case of bioadsorbent, a comparison of isotherm parameters from different isotherm models.

Langmuir isotherm		Freundlich isotherm		Temkin isotherm	
Q_m (mg/g)	0.611139	K_f ($\text{mg}^{-1} \cdot (\ln) \cdot \ln \text{g}^{-1}$)	0.55856	B (J mol^{-1})	0.1222
K_s (L/mg)	14.22985	$1/n$	0.28973	K_T (Lg^{-1})	117.0022
R^2	0.9196	R^2	0.97551	R^2	0.87558

The rate-determining step is the pseudo-second-order kinetic model, which may be represented as:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$$

Where k_2 denotes the adsorption rate, q_t is the total amount of fluoride adsorbed at any given time, and q_e denotes the equilibrium adsorption capacity. The slope and intercept of the plot of t/q_t against t (Wolowicz and Wawrzkiwicz, 2021) displayed in figure 4 b, as well as the kinetic data in table 5.

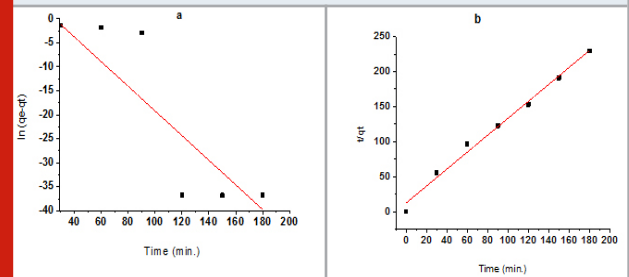
Table 4. Effect of contact time, Operating order for the consequence of contact time, Adsorbent dose - 10 g/L, Temperature was adjusted $27^\circ\text{C} \pm 0.5^\circ\text{C}$, $C_i = 10.04 \text{ mg/L}$, Volume of sample - 100 ml, and pH 2.

Time (min)	C_i (mg/L)	C_e (mg/L)	Elimination % of F-
30	10.04	4.61	54
60	10.04	3.81	62
90	10.04	2.71	73
120	10.04	2.20	78
150	10.04	2.20	78
180	10.04	2.20	78

Table 5. Virtual study of kinetic parameters in case of used bioadsorbent for defluoridation of groundwater.

Pseudo-first-order		Pseudo-second-order	
q_e (mg/g)	653.43953	q_e (mg/g)	0.82540
K_1 (min^{-1})	-0.00142	K_2 (g/mg.min.)	0.056069
R^2	0.73822	R^2	0.98862

Figure 4: a. Pseudo-first-order kinetic studies for activated neem (*Azadirachta indica*) leaves as bioadsorbents for used defluoridation of groundwater b. Pseudo-second-order kinetic studies fitted for activated neem (*Azadirachta indica*) leaves as bioadsorbents for used defluoridation of groundwater.



Effect of pH: The adsorption process is influenced by the pH of the aqueous solution (Mia and Jha, 2020). The effect of H^+ concentration was studied at pH values 2, 4, 6, and 8. This was attained by mixing 0.5N HNO_3 for 120 minutes with 100 ml of a standard fluoride solution containing 10.04 mg/L fluoride and a dose of 10 g/L treated bioadsorbent.

With increasing the pH of the resolution, we noticed a reduction in the amount of fluoride ion removal. At pH 2, this was found to be 78%, while at pH 4, it was found to be 75%. As a result, more research was done at these pH levels. In treated adsorbents, the entitlement of adsorption varies almost linearly between 2.0 and 8.0, achieving a maximum clearance at pH 2.0 after 120 minutes of contact time (Ali and Ismail 2021).

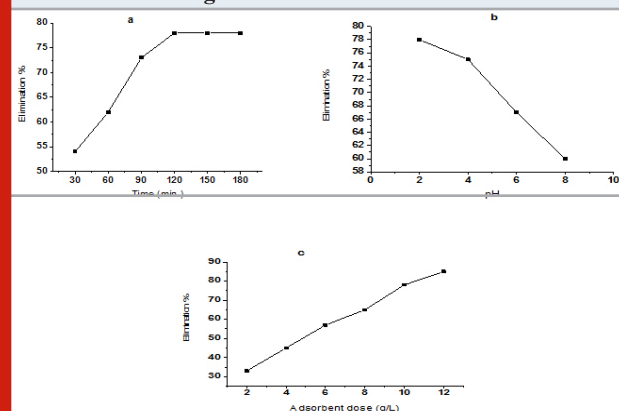
Table 6. Table for effect of pH, Operating order for the consequence of pH, Adsorbent dose - 10 g/L, Temperature was adjusted $27^{\circ}\text{C} \pm 0.5^{\circ}\text{C}$, Time of contact - 120 Minutes, and Volume of sample - 100 ml,

pH %	Ci (mg/L)	Ce (mg/L)	Elimination of F-
2	10.04	2.20	78
4	10.04	2.51	75
6	10.04	3.31	67
8	10.04	4.01	60

Table 7. Table for effect of adsorbent dosage, Operating order for consequence of adsorbent measure, Effect of contact time, Operating order for the consequence of contact time, Temperature was adjusted $27^{\circ}\text{C} \pm 0.5^{\circ}\text{C}$, Ci = 10.04 mg/L, Volume of sample - 100 ml, and pH 2.

Adsorbent dose (g/L)	Ci (mg/L)	Ce (mg/L)	Elimination % of F-
2	10.04	6.62	33
4	10.04	5.52	45
6	10.04	4.31	57
8	10.04	3.51	65
10	10.04	2.20	78
12	10.04	1.50	85

Figure 5: a. The effect of contact time relations on F-subtraction, b. The effect of pH on F- subtraction, c. Effect of adsorbent dosage on the subtraction of F-.



Higher hydrogen ion concentrations at lower pH values may have neutralized negative charges on the surface of the treated bioadsorbents in this example. This makes it simpler for negatively charged fluoride ions to diffuse onto the increased active surface of the modified bioadsorbent.

Effect of adsorbent dosage: The elimination of fluoride ions appears to augment when the quantity of adsorbent is increased. The starting fluoride ion concentration in all of these experiments was set at 10.04 mg/L. The adsorbent dosage in an aqueous solution was adjusted between 2 and 12 g/l at their optimum pH levels. Table 7 and figure 5c. demonstrate that treated bioadsorbent was efficient for 33 – 85% removal of fluoride ions, with maximum exclusion of 85 % recorded at 12 g/L, at a room temperature of $27^{\circ}\text{C} \pm 0.5^{\circ}\text{C}$ (Ali and Ismail 2021).

Contact duration, pH, and adsorbent concentration all affect the performance of neem leaf adsorbents in successfully remediating tainted fluoride water (Patel and Gupta, 2020; Pandey et al. 2020).

CONCLUSION

The findings of the present study used Azadirachta indica leaves as bioadsorbents to eliminate fluoride from groundwater samples from Korba district of Chhattisgarh. Fluoride in this region varied from 2.28 to 10.04 mg/L. As per the result analysis, we saw that contact duration, pH, and adsorbent concentration influence the effectiveness of low-cost leaf adsorbents in successfully remediating fluoride-polluted water. Maximum fluoride elimination was 78% at pH 2 and up to 85% at 12 g of the adsorbent dose. In the future, this technique of removing fluoride from highly fluoridated water will give positive results and be cheap and available everywhere.

Conflict of Interests: Authors declare no conflict of interests to disclose.

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Data Availability Statement: The database generated and /or analysed during the current study are not publicly available due to privacy, but are available from the corresponding author on reasonable request.

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