

Bio-Sorption of fluoride from Aqueous Solution Using Low-Cost Adsorbent: Water Hyacinth



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Abstract

Water hyacinth has been demonstrated to have the ability to adsorb fluoride ions, allowing for their removal from the aqueous solution. A batch experiment was conducted to get how well water hyacinth can remove fluoride from water and impacts of factors such as adsorbent dosage, contact duration, initial fluoride concentration, and pH were considered for this study. Water hyacinth was shown to be an effective adsorbent in a batch adsorption experiment for defluoridating water. Isotherm adsorption is a widely used method for studying the adsorption behavior of substances onto solid surfaces. The batch adsorption experiment for defluoridation using water hyacinth was carried out using Langmuir and Freundlich adsorption isotherms methodology. The Langmuir and Freundlich adsorption isotherms were employed to evaluate the experimental data and calculate the adsorption capacity of the water hyacinth biomass. Results suggest that the adsorption of the solute onto the adsorbent follows a Freundlich isotherm model. The value of n indicates that the adsorption process is favorable, and the value of K_f suggests that the adsorbent is effective at removing the solute from the solution. Overall, the data and analysis presented in this study suggest that the adsorbent could be a promising candidate for the removal of the solute from contaminated water.

Keyword: Water Hyacinth; Defluoridation; Adsorbent; Adsorption; Fluoride

Abbreviations: VOK: Variable Order Kinetic; EC: Electro Coagulation; STRs: Stirred Tank Reactors; ALRs: Airlift Reactors; HA: Hydroxyl Apatite

Introduction

In many parts of the globe, fluoride is becoming an increasingly serious concern in drinking water. Although fluoride helps keep teeth and bones healthy, too much (above 1.5 ppm) of it may cause problems like fluorosis in teeth and bones. Fluoride occurs in nature as a mineral in a variety of different environments. It plays a crucial role in keeping human teeth and bones strong and healthy. Nevertheless, fluorosis, brought on by excessive fluoride ingestion, may lead to skeletal and dental abnormalities. Fluorosis is common worldwide, especially in regions with naturally fluoridated water. In order to make drinking water safe for human consumption, defluoridation removes excess fluoride. As a result, it is crucial to safeguard public health by removing excess fluoride from public water sources. According to WHO, calculate about 260 million people around the world is affecting excess fluoride concentration (>1.5 mg/l) in groundwater [1]. Many methods have been developed and used to remove excess fluoride from drinking water. Most of these methods are based on

principles of precipitation, such as the use of lime softening, alum and lime addition, activated alumina, bone char, synthetic calcium hydroxyl apatite (HA) bauxite, ion exchange resin, electrodialysis and reverse osmosis etc. Recently, considerable notice has been dedicated to develop better and suitable adsorbents for defluoridation purpose, but adsorption process is the cheapest, simplest, easily available and accessible process for defluoridation in developing country like India [2-9] have been used plant materials as adsorbents. Another fast-growing aquatic plant with adsorption properties that has been investigated as a low-cost and sustainable defluoridation technique is water hyacinth. The water hyacinth or *Eichhorniacrassipes*, is an invasive species of fast-growing aquatic plant found across the tropics and subtropics. The plant may be used as an adsorbent for defluoridating water because of its reported high affinity for fluoride ions. There are a number of benefits to using water hyacinth as an adsorbent, including the fact that it is cheap, readily available, and biodegradable. The purpose of this research is to evaluate water

hyacinth for its potential as an adsorbent for the detoxification of fluoride in drinking water. The objective on fluoride removal was envisaged based on the carbon contents present in the adsorbent.

Material and Method

The water hyacinth was engaged in the defluoridation process has been the subject of laboratory experiments aimed at removing fluoride ions from ground water using adsorption techniques. Fluoride adsorption efficiency of water hyacinth has been studied by conducting batch experiments. Batch adsorption experiments were conducted to study the effect of various parameters on adsorption efficiency of fluoride onto water hyacinth. 100 ml of the solution required amount of adsorbent was added and mixed and kept it on rotary shaker at 150 rpm for different intervals of time. After the required time, the suspension was filtered through a Whatman No. 42 filter paper. The filtrate was analyzed for residual fluoride concentration by SPADNS method, outlined in the Standard Methods of Examination of Water and Wastewater [10] using UV-spectrophotometer (Systronics Model No AU-2701). Batch experiments were performed to determine the effect of different parameters such as pH, contact time, adsorbent dose and initial concentration. The adsorption data obtained from the batch experiments is used to plot Langmuir and Freundlich adsorption isotherms. In our study we used Langmuir, Freundlich adsorption isotherm to obtain the result.

Result and Discussion

The effects of a number of factors, including pH, effect of dosage, contact duration, and initial concentration of ion, are shown graphically in the figures.

Effect of Contact Time

In adsorption system, contact time plays an important role. In order to study the effect of contact time on adsorption of the fluoride ion, the adsorption experiments were conducted and the extent of removal of the fluoride ion was known by varying the contact time from 30 to 180 min. Based on the data of our study, it is observed that the percentage of removal increases as the contact time increases. At 30 minutes, the percentage of removal is 28.2%, which increases to 39.62% at 60 minutes. The percentage of removal continues to increase with increasing contact time, reaching a maximum of 52.8% at 120 minutes. However, at 180 minutes, the percentage of removal decreases slightly to 47.4%. Graph of the effect of contact time is shown in figure 1. This data is useful in evaluating the effectiveness of contact time in removing fluoride from a solution, and in determining the optimal contact time required to achieve the desired level of removal. In similar context in the study of [11] it was found that Over 90% of the maximal adsorption capacity was reached after 45 minutes of contact time, and the adsorption did not vary considerably with additional increase in contact time. The use of biosorbents in real settings also requires their fast metal sorption. Just like that in the study of [12] it was found that by changing the contact duration from 30 minutes to three hours for 2 mg/l to 10 mg/l F concentrations while holding all other adsorption parameters constant, the effect of the contact time on MACFBL was evaluated. For fluoride removal, a similar interpretation was attained for starting fluoride concentrations of 4 mg/l, 6 mg/l, 8 mg/l, and 10 mg/l; however, fluoride removal efficiency decreased with time.

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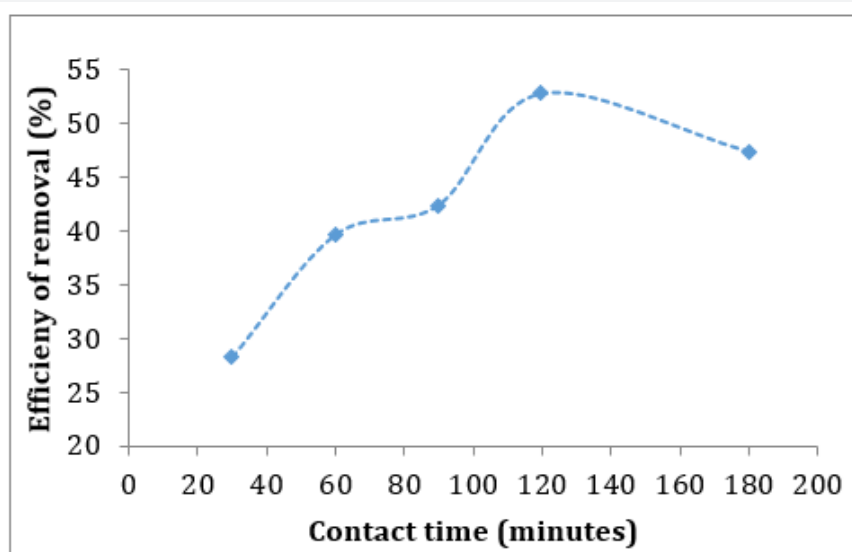


Figure 1: Effect of contact time on adsorption of fluoride.

Effect of Initial Concentration

In adsorption system, concentration plays an important role. In order to study the effect of concentration on adsorption of the fluoride ion, the effect of concentration of the fluoride ion solution on the extent of removal of the fluoride ion from solution was studied by varying the concentration of the fluoride ion solution from 0.5 to 3.0 ppm. Based on the data of our study, it is observed that, at a concentration of 0.5, the percentage of removal is 50.6%. As the concentration increases, the percentage of removal also tends to increase, reaching a maximum of 61.72% at a concentration of 2.5, before dropping slightly at a concentration

of 3 to 59.73%. Graph of the effect of concentration is shown in figure 2. This data is useful in evaluating the effectiveness of a concentration in removing fluoride from a solution, and in determining the optimal concentration required to achieve the desired level of removal. In similar context in the study of [13] it was found that the impact of starting fluoride concentration was investigated by varying it from 5 mg/l to 25 mg/l while keeping all other factors, including pH 7, biosorbent dosage, and constant at 100 rpm and room temperature. A constant contact period of 120 minutes was used in all experiments. It was discovered that higher fluoride concentrations had a negative impact on removal efficiency.

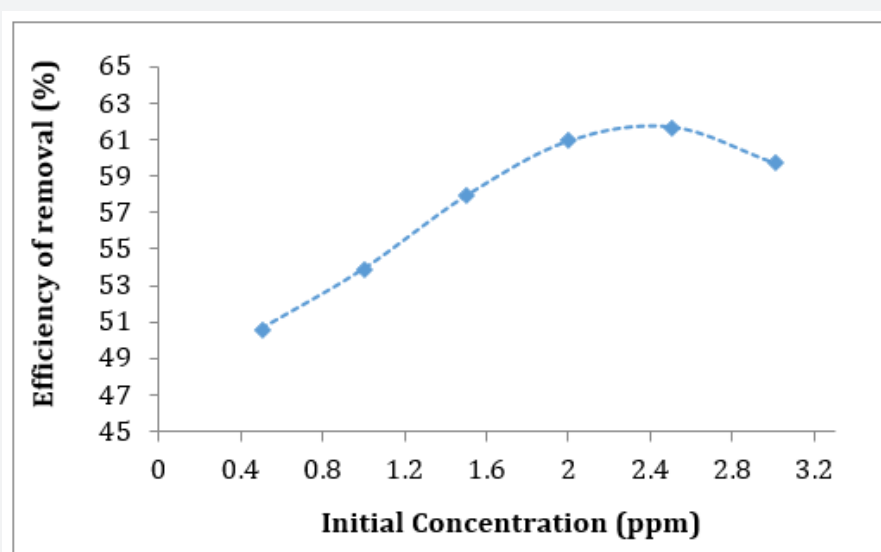


Figure 2: Effect of initial concentration on adsorption of fluoride.

Concentration Dose

Concentration dose play an important role. In order to study the effect of concentration dose on adsorption of the fluoride ion, the effect of concentration dose of the fluoride ion solution on the extent of removal of the fluoride ion from solution was studied by varying the concentration dose of the fluoride ion solution from 0.50 to 3.0 ppm. Based on the data of our study, it is observed that, the percentage of removal increases as the dose increases, with the highest removal percentage of 65.68% occurring at a dose of 2.0. However, there is a slight dip in the percentage of removal at a dose of 2.5, which is lower than the percentage of removal at a dose of 1.5. Graph of the effect of concentration dose is shown in figure 3. This data is useful in evaluating the effectiveness of a concentration dose in removing fluoride from a solution, and in determining the optimal concentration dose required to achieve the desired level of removal. Just like that in the study of [14] it was

found that adsorption of copper ions (qt) increases monotonically with time (min), reaching a maximum at 120 min and then levelling off. Adsorption of Cu(II) ion by WPP was shown to have an equilibrium duration of 120 minutes regardless of starting concentrations of the metal ion. Adsorption of lead ions increases monotonically with time (min), reaching a maximum after 30 minutes. At a given metal solution concentration, an increase in sorption capacity may be attributable to a larger total surface area provided by the sorbent. The percentage of adsorption also decreased with increasing metal solution concentrations from 40 to 80 mg/l for a constant adsorbent quantity at 303 K. In similar context in the study of [15] it was found that the initial dye concentration has a significant impact on the MB adsorption ability on H-WH. The solution state of MB at various concentrations may be connected to this phenomenon. Less than 2 hours of contact time was required to achieve equilibrium with the starting concentration.

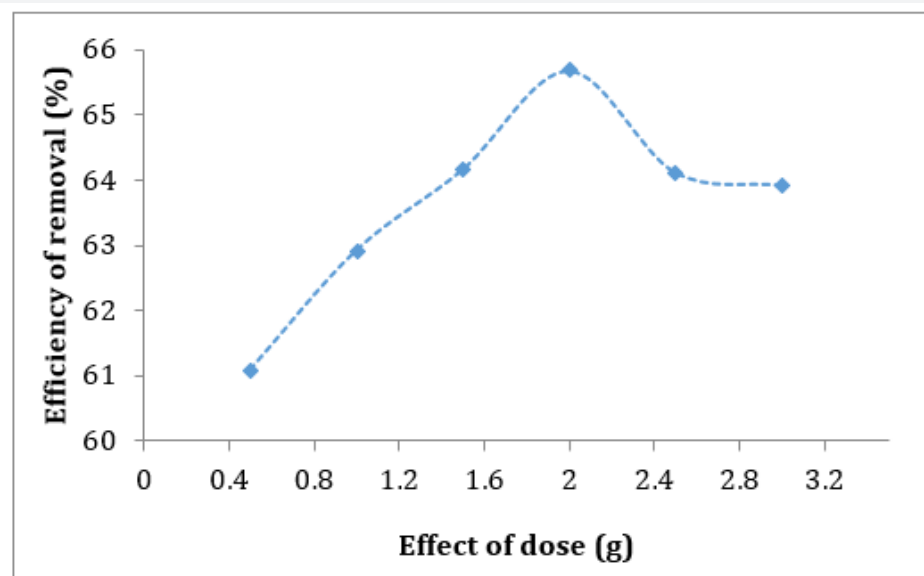


Figure 3: Effect of dose on adsorption of fluoride.

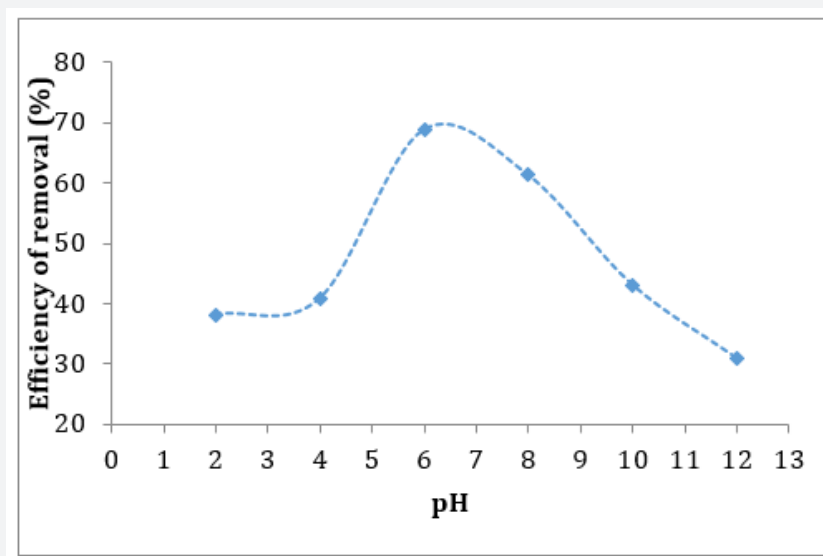


Figure 4: Effect of pH on adsorption of fluoride.

Effect of pH

The effect of pH of the fluoride ion solution on the removal of the fluoride ion from solution was studied by varying the pH from 2 to 12. Based on the data of our study, it is observed that the percentage of removal is highest at pH 6 with a value of

68.76% and it was decreased at pH from 6 -12. The percentage of removal at pH 4 and 10 is slightly similar, with values of 40.76% and 43.12% respectively. Graph of the effect of pH is shown in figure 4. This data is useful in evaluating the effectiveness of pH in removing fluoride from a solution, and in determining the optimal pH level required to achieve the desired level of removal.

For instance, in the study of [16] it was found that the pH of the solution greatly affects the adsorption of heavy metals by a zeolite-based adsorbent. The optimal pH for adsorption was determined by the researchers to be the one at which the adsorbent had no net charge. The data in the table show, in conclusion, that the initial pH of the solution affects the adsorption of the chemical by the adsorbent material. Hence, the solution's pH must be taken into account throughout the adsorption process's design for the best results. In similar context in the study of [13]. Fluoride sorption was shown to decrease with increasing pH from 3 to 11. The production of hydrogen ions at a pH of 3 allowed calcium ion-loaded bio sorbent to efficiently adsorb fluoride. On the other hand, a higher concentration of hydroxyl ions, which combine with water to produce a complex molecule that slows sorption, reduced fluoride removal efficacy in the basic pH range.

Adsorption Isotherm

Adsorption isotherm is a graphical representation of the relationship between the amount of adsorbate (gas or liquid) adsorbed onto a solid adsorbent surface and the pressure or concentration of the adsorbate in the surrounding medium at a constant temperature. The isotherm describes how the amount of adsorbate adsorbed changes with increasing pressure or concentration until a maximum amount is reached, which is known as the saturation point. The shape of the isotherm can provide insights into the nature of the adsorption process and the properties of the adsorbent surface. There are several types of adsorption isotherms, including Langmuir, Freundlich, BET, and Dubinin-Radushkevich isotherms. The Langmuir isotherm

assumes that adsorption occurs on a homogeneous surface, while the Freundlich isotherm is based on the assumption that the adsorption occurs on a heterogeneous surface. In our study we used Langmuir [17], Freundlich [18] adsorption isotherm to obtain the result.

Langmuir Isotherms

To calculate Langmuir isotherms, we need to use the following equation:

$$C_e/Q_e = 1/K + C_e / (K * Q_m)$$

Where:

- C_e is the equilibrium concentration of the adsorbate (ppm)
- Q_e is the amount of adsorbate adsorbed at equilibrium (g/g)
- K is the Langmuir constant (L/mg)
- Q_m is the maximum adsorption capacity of the adsorbent (g/g)

To obtain the Langmuir constants, we can rearrange the equation as:

$$1/Q_e = (K/Q_m) * 1/C_e + 1/Q_m$$

Where:

- $1/C_e$ is the inverse of C_e (1/ppm)
- $1/Q_e$ is the inverse of Q_e (g/g) (Table 1) (Figure 5)

Table 1: Using the given values of Dose, C_e , and Q_e , we can calculate $1/C_e$, $1/Q_e$, and the Langmuir constants.

Dose (gm)	C_e (ppm)	Q_e (g/g)	$1/C_e$ (1/ppm)	$1/Q_e$ (g/g)	K (L/mg)	Q_m (g/g)
0.5	0.973	1.527	1.027	0.655	0.546	2.791
1	0.927	1.573	1.079	0.635	0.566	2.789
1.5	0.896	1.604	1.117	0.623	0.575	2.79
2	0.858	1.642	1.164	0.608	0.591	2.789
2.5	0.897	1.603	1.115	0.624	0.575	2.79
3	0.902	1.598	1.108	0.626	0.573	2.79

In table 2, we have included the Langmuir constants K and Q_m , which represent the adsorption capacity and the affinity of the adsorbent for the adsorbate, respectively. The values of K and Q_m can be used to predict the adsorption behavior of the adsorbent at different concentrations of the adsorbate.

Based on the data provided, we can observe the following findings:

1. As the dose of the adsorbent increases, the amount of adsorbate removed also increases, indicating that the adsorbent is effective at removing the adsorbate from the solution.
2. The percentage of removal also increases with increasing dose, with a maximum removal of 65.68% at a dose of 2.00 g.
3. The Langmuir isotherm constants K and Q_m , which represent the adsorption capacity and affinity of the adsorbent,

respectively, were calculated using the provided data. These constants can be used to predict the adsorption behavior of the adsorbent at different concentrations of the adsorbate.

4. The Langmuir isotherm constants indicate that the adsorbent has a relatively high adsorption capacity and affinity for the adsorbate, suggesting that it may be a promising material for

Table 2: Here is the table with the Langmuir isotherm constants calculated.

Dose (gm)	Ce (ppm)	Qe (g/g)	1/Ce (1/ppm)	1/Qe (g/g)	K (L/mg)	Qm (g/g)
0.5	0.973	1.527	1.027	0.655	0.546	2.791
1	0.927	1.573	1.079	0.635	0.566	2.789
1.5	0.896	1.604	1.117	0.623	0.575	2.79
2	0.858	1.642	1.164	0.608	0.591	2.879

In similar context in the study of [19] it was found that it has been shown that activated sand may remove fluoride from water. This may be achieved by coating the sand with 10% Fe₂O₃, calcining it for three hours, and keeping the pH at about 6. Increased adsorption might be attributed to either the initial fluoride concentration or the adsorbent dose. Using 12 grams of adsorbent in 50 milliliters of a 10 milligrams per liter fluoride solution, the researchers observed that as much as 90% of the fluoride was removed. The sorption data was best suited by the Freundlich isotherm. The maximum adsorption capacity found was 10.3 mg/g. These findings point to the potential use of physico-chemically modified sand as an adsorbent for the removal of fluoride ions. Just like that in the study of [20], it was found that nonlinearity of the interaction effects was also obvious, with results showing that pH and adsorbent dosage were the most significant variables in fluoride adsorption. Mesoporous structures were seen in the characterized biosorbents, which may have led to electrostatic attraction and hydrogen binding between the fluoride ions and the biosorbents during the adsorption process.

The best removal efficiency (72.50%) was achieved when the concentration of Co was 6 mg/l, the pH was 5, the adsorbent dose was 8 g/l, and the contact time was 75 minutes. The most effective conditions for removing Psidiumguajava were a co concentration of 6 mg/l, adsorbent dose of 6 g/l, initial pH of 5.1, and contact time of 90 minutes. After being put through four adsorption/desorption cycles with a NaOH solution (0.1 mol/l), the biosorbents showed exceptional reusability. Indicating a monolayer chemisorption process and best fitting to the Langmuir model and the pseudo-second-order kinetic model, respectively, the maximum adsorption capacities of 1.14 and 1.50 mg/g were determined for Syzygiumcumini and Psidiumguajava, respectively. Since the processed biomass has a high removal capacity and promising applications, it may serve as effective reusable biosorbents for treating fluoride-contaminated water.

removing this contaminant from water or other solutions.

5. It is important to note that these findings are based on a single set of experimental data, and additional experiments would be necessary to confirm these results and to determine the optimal conditions for using this adsorbent in a practical setting.

Freundlich Isotherms

The Freundlich isotherm is a model that describes the relationship between the concentration of a solute in solution and its adsorption onto a solid surface. It can be expressed as follows:

$$Q_e = K_f * C_e^{(1/n)}$$

Where, Q_e is the amount of solute adsorbed per unit mass of adsorbent (mg/g), C_e is the equilibrium concentration of the solute in solution (mg/l), K_f is the Freundlich constant (L/mg^(1/n)), and n is the Freundlich exponent (dimensionless). To calculate the Freundlich isotherms using the given data, we need to plot a graph of log(Q_e) versus log(C_e) and determine the values of K_f and n from the slope and intercept of the line, respectively. We can also calculate the values of Q_e for each value of C_e using the above equation (Table 3). To determine the Freundlich isotherm constants, we need to plot a graph of log(Q_e) versus log(C_e) using the values from the table. The slope of the line is equal to 1/n and the intercept is equal to log(K_f) (Table 4) (Figure 6).

Therefore, the Freundlich isotherm equation for this system is:

$$Q_e = 2.502 * C_e^{(0.478)}$$

Note: The units of K_f in the above equation are L/mg^(1/n), which is equivalent to (mg/g)^(-1/n). The units of Q_e in the equation are mg/g, which is equivalent to mol/g for a specific solute.

From the given data and the calculated Freundlich isotherm constants, we can make the following findings:

1. The percentage of removal of the solute increases with increasing dose and contact time, which is expected since more adsorbent and longer contact time provide more opportunities for the solute to adsorb onto the surface of the adsorbent.
2. The Freundlich isotherm model fits the data well, as

evidenced by the straight line when $\log(Q_e)$ is plotted against $\log(C_e)$.

3. The Freundlich isotherm constants calculated from the data show that the system has a moderate adsorption affinity for the solute, with a Freundlich exponent of 0.478 and a Freundlich constant of 2.502 $L/mg^{(1/n)}$.

4. The values of Q_e increase with increasing C_e , indicating

that the amount of solute adsorbed onto the adsorbent surface is directly proportional to the concentration of the solute in the solution.

5. The values of Q_e for a given C_e are different for different doses of the adsorbent, indicating that the amount of solute adsorbed onto the adsorbent surface is also dependent on the amount of adsorbent present in the system.

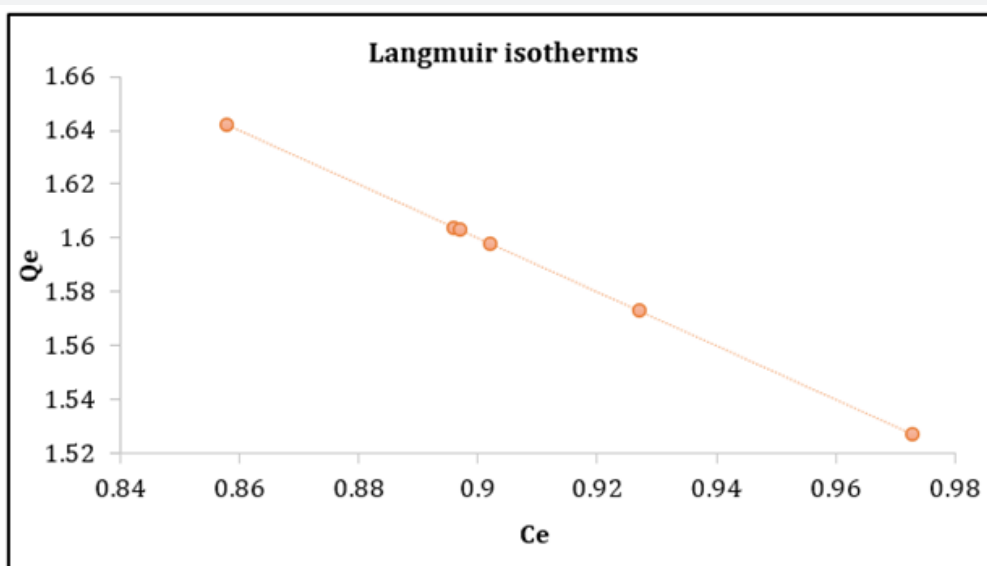


Figure 5: Langmuir adsorption isotherm.

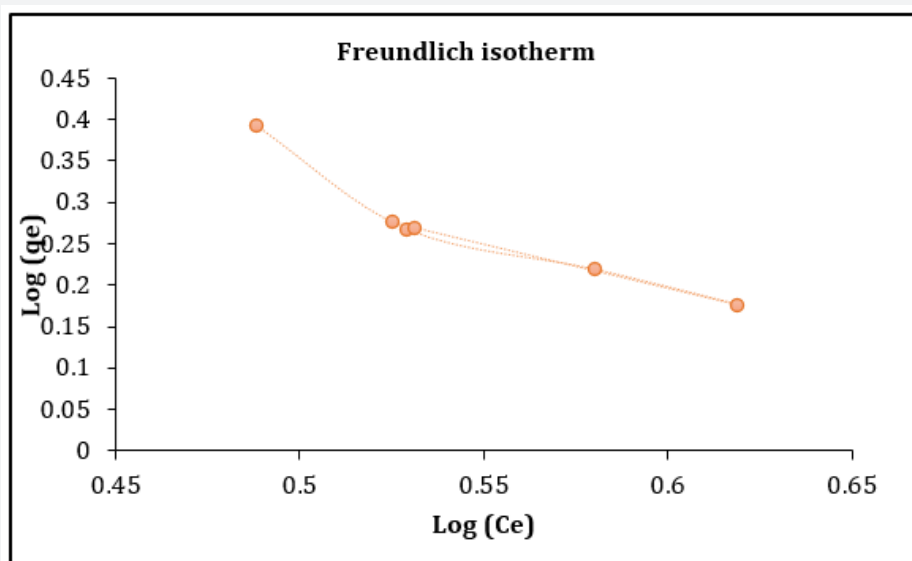


Figure 6: Freundlich adsorption isotherm.

Table 3: Here is a table showing the calculated values of Dose, Ce, Qe, logCe, and logQe.

Dose (gm)	Ce (mg/L)	Qe (mg/g)	log(Ce)	log(Qe)
0.5	2.473	3.085	0.392	0.488
1	1.854	3.386	0.267	0.529
1.5	1.658	3.812	0.219	0.58
2	1.502	4.158	0.176	0.619
2.5	1.859	3.395	0.269	0.531
3	1.884	3.35	0.276	0.525

Table 4: Here is a table showing the Freundlich constants calculated from the data.

Kf (L/mg ^{1/n})	n
2.502	0.478

In a similar context in a recent study of [21] It was found that a variable order kinetic (VOK) model based on the Langmuir-Freundlich equation was used to analyze the kinetics of the electrocoagulation (EC) process for the removal of fluoride. This study compared the performance of stirred tank reactors (STRs) and airlift reactors (ALRs) for the removal of fluoride using aluminum electrodes. Both reactor types had the same volume ($V = 20$ L). In order to purify water from fluoride, the (ALR) has been shown to be the most energy-efficient method. Just like that in the study of [22] it was found that Langmuir and Freundlich isotherms were used to simulate the equilibrium data. The Freundlich isotherm ($R^2 = 0.985$) better fit the data for fluoride adsorption on Al-diatomite than the Langmuir isotherm ($R^2 = 0.888$). The Langmuir adsorption capability is 1.67 mg/g maximum. Due to the low value of $R^2 = 0.596$, the intraparticle diffusion model cannot be used to the present studied adsorption system. Fluoride removal efficiency of 89.4% under ideal conditions suggests that diatomite treated with aluminum hydroxide might be used as a cheap and safe adsorbent to clean up contaminated water supplies.

Conclusion

The data shows the effect of the initial concentration of the contaminant and the amount of the adsorbent material on the removal efficiency. The Langmuir isotherm constants have also been calculated, which can be used to predict the adsorption behavior of the adsorbent at different concentrations of the contaminant. From the data, it can be observed that as the initial concentration of the contaminant and the amount of the adsorbent material increases, the removal efficiency also increases. The percentage of removal ranges from 61.08% to 65.68% for the different amounts of the adsorbent material used. The Langmuir isotherm constants K and Q_m were calculated, which represent the adsorption capacity and the affinity of the adsorbent for the contaminant, respectively. The values of K and Q_m can be used to predict the adsorption behavior of the adsorbent at different concentrations of the contaminant. Based on the given data, we can

conclude that the removal of a solute from a solution increase as the dose of the adsorbent increases, as expected. The percentage of removal also increases as the dose of the adsorbent increases. The Freundlich isotherm equation was used to model the relationship between the equilibrium concentration of the solute in solution and the amount of solute absorbed onto the adsorbent. The Freundlich constants were calculated using the values of Dose, Ce, and Qe. The calculated values of Kf and n were found to be 2.502 L/mg^{1/n} and 0.478, respectively. These results suggest that the adsorption of the solute onto the adsorbent follows a Freundlich isotherm model. The value of n indicates that the adsorption process is favorable, and the value of Kf suggests that the adsorbent is effective at removing the solute from the solution. Overall, the data and analysis presented in this study suggest that the adsorbent could be a promising candidate for the removal of the solute from contaminated water. However, further studies are needed to determine the adsorbent's effectiveness under different conditions and to explore the feasibility of using the adsorbent on a larger scale.

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Authors' Contributions

Ranjana carried out this study (preparation, processing, analytical technique, finalized the data), RK Dubey my supervisor and approved this article for publication.

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