



Removal of organochlorine pesticide (Chlorpyrifos) from Aqueous Solution by Granular Activated Carbon

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ABSTRACT: The contamination of water bodies with pesticides poses significant environmental and health risks, necessitating effective remediation techniques. Hence, the objective of this paper is to investigate the removal of organochlorine pesticide (chlorpyrifos) from aqueous solution by granular activated carbon using appropriate standard procedures. Data obtained show that optimal conditions were achieved at pH6, a contact time of 60 minutes, and a temperature of 40°C, yielding a maximum adsorption efficiency of 86.9%. Adsorption kinetics conformed to the pseudo-second-order model ($R^2 = 0.989$), while equilibrium data best fitted the Langmuir isotherm ($R^2 = 0.9987$), indicating monolayer adsorption on a homogeneous surface. Thermodynamic analysis revealed the process to be spontaneous and endothermic, with negative values for Gibbs free energy (ΔG°) and enthalpy (ΔH°). These findings demonstrate the efficacy of GAC in mitigating pesticide pollution, offering a sustainable solution for water treatment applications.

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The contamination of surface and groundwater by pesticides has emerged as a significant environmental and public health issue. Pesticides, extensively applied in agriculture, forestry, and urban landscapes, often find their way into water bodies through surface runoff, leaching, wind erosion, and industrial discharges, resulting in frequent detection in water sources worldwide (Meftaul *et al.*, 2020). Among

these chemicals, chlorpyrifos, a widely used organochlorine insecticide, has garnered attention due to its persistence in the environment and its adverse effects on human and ecological health. Chlorpyrifos has been identified as toxic, carcinogenic, and capable of disrupting endocrine systems, making its presence in water a pressing concern (Li and Funtki, 2020). These hazards have

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led to stringent regulatory limits on pesticide levels in drinking water in many countries (Puri *et al.*, 2023). Traditional water treatment methods, such as photocatalytic degradation, advanced oxidation processes, and ozonation, have shown promise in removing pesticides. However, their high operational costs and technical complexity limit their widespread applicability, particularly in resource-constrained setting (Saravanan *et al.*, 2023). Consequently, adsorption using activated carbon has emerged as an attractive alternative due to its efficiency, economic feasibility, and operational simplicity. Granular activated carbon (GAC) is particularly effective for pesticide removal, owing to its excellent sorptive properties, large surface area, mechanical stability, and High adsorption capacity (Bose *et al.*, 2023; Zieliński *et al.*, 2022). The use of GAC as an adsorbent has been well-documented for various pollutants, including pesticides, making it a suitable candidate for addressing chlorpyrifos contamination.

Hence, the objective of this paper is to investigate the removal of organochlorine pesticide (chlorpyrifos) from aqueous solution by granular activated carbon

MATERIALS AND METHODS

Materials: Granular activated carbon (GAC) and chlorpyrifos pesticide were procured from Yomi Chemical Stores, Ilorin, Kwara State, Nigeria. Sodium hydroxide (NaOH), nitric acid (HNO_3), and distilled water were used as reagents. The apparatus utilized included a mechanical shaker, UV-visible spectrophotometer, pH meter, weighing balance, and standard laboratory glassware such as conical flasks and measuring cylinders.

Preparation of Stock Solution and Calibration Curve: A stock solution of chlorpyrifos (1000 ppm) was prepared using distilled water. Serial dilutions were performed to obtain working concentrations of 10, 20, 30, 40, and 50 ppm. The UV-visible spectrophotometer was calibrated to determine the maximum absorbance wavelength for chlorpyrifos, which was recorded as 360 nm. A standard calibration curve was generated by plotting absorbance values against the corresponding concentrations, ensuring compliance with Beer-Lambert's law.

Adsorption Experiments: The adsorption capacity of GAC for chlorpyrifos was evaluated by systematically varying experimental parameters, including the initial concentration of chlorpyrifos, pH, contact time, adsorbent dosage, and temperature. After each experiment, the solutions were filtered,

and the residual concentration of chlorpyrifos was measured using a UV-visible spectrophotometer.

Effect of Initial Concentration: Chlorpyrifos solutions with concentrations of 10–50 ppm were prepared, and 0.1 g of GAC was added to 30 mL of each solution. The mixtures were agitated for 60 minutes at 30°C using a mechanical shaker. The adsorption capacity and percentage removal were calculated based on the difference between the initial and final concentrations.

Effect of pH: The pH of chlorpyrifos solutions (40 ppm) was adjusted to values ranging from 2 to 8 using 0.1 M NaOH and 0.1 M HNO_3 . A fixed dosage of 0.1 g of GAC was added to each solution, and the mixtures were shaken for 60 minutes at 30°C. The pH that yielded the highest adsorption efficiency was recorded.

Effect of Contact Time: To determine the equilibrium time, 0.1 g of GAC was added to 30 mL of 40 ppm chlorpyrifos solution at pH 6. The mixtures were agitated at 30°C for varying durations (20, 40, 60, 100, and 120 minutes). The adsorption efficiency was calculated at each time point.

Effect of Adsorbent Dosage: Chlorpyrifos solutions (40 ppm) were treated with varying amounts of GAC (0.1–0.5 g). Each mixture was agitated for 60 minutes at pH 6 and 30°C. The optimal dosage was determined based on maximum removal efficiency.

Effect of Temperature: The influence of temperature on adsorption was assessed by agitating 30 mL of 40 ppm chlorpyrifos solution with 0.1 g of GAC at temperatures ranging from 30°C to 70°C. The adsorption efficiency was calculated, and the temperature at which maximum adsorption occurred was identified.

Isotherm and Kinetic Modeling: The equilibrium data were analyzed using the Langmuir, Freundlich, and Temkin isotherm models to characterize the adsorption mechanism and capacity of GAC. Adsorption kinetics were evaluated using pseudo-first-order and pseudo-second-order models to determine the rate-limiting step of the adsorption process.

Thermodynamic Analysis: Thermodynamic parameters, including Gibbs free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°), were calculated to evaluate the spontaneity and feasibility of the adsorption process. These parameters were determined from the slope and intercept of a linear

plot of $\ln(K_d)$ versus $1/T$, where K_d represents the distribution coefficient and T is the absolute temperature in Kelvin.

RESULTS AND DISCUSSION

The absorbance spectrum of chlorpyrifos across various wavelengths was assessed using a UV-visible spectrophotometer. As shown in Table 1, absorbance increased progressively from 260 nm to a peak of 3.084 at 360 nm, after which it declined at higher wavelengths. This spectral pattern was further visualized in Figure 1, which confirms that chlorpyrifos exhibits maximum absorbance at 360 nm. This wavelength was subsequently selected for all quantitative measurements in the adsorption experiments due to its optimal sensitivity.

Table 1: Data for chlorpyrifos spectrum

Wavelength(nm)	Absorbance
260	0.034
280	0.154
300	1.561
320	0.886
340	2.045
360	3.084
380	2.543
400	1.874
420	0.678
440	0.246

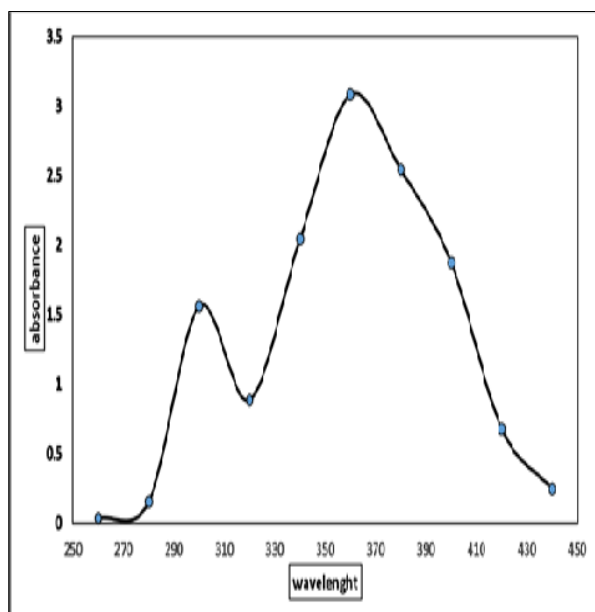


Fig. 1: Chlorpyrifos Absorption Spectrum

This spectral pattern was further visualized in Figure 1, which confirms that chlorpyrifos exhibits maximum absorbance at 360 nm. This wavelength was subsequently selected for all quantitative measurements in the adsorption experiments due to its optimal sensitivity.

Effect of Initial Concentration: The effect of varying the initial concentration of chlorpyrifos on adsorption efficiency is presented in Table 2. A steady increase in adsorption capacity (q_e) and percentage removal was observed with increasing initial concentration from 10 to 50 ppm. The maximum adsorption capacity was recorded at 50 ppm with a q_e value of 11.641 mg/g, and percentage removal peaked at 77.611%. However, the adsorption efficiency showed diminishing returns beyond 40 ppm, suggesting near-saturation of available adsorption sites.

Table 2: Data for Effect of Initial Concentration

(C_0)	Absorbance	(C_f)	mass W(mg)	volume V(L)	q_e (mg/g)	%Removal
10	0.059	2.935	0.1	0.03	2.119	70.646
20	0.112	5.572	0.1	0.03	4.328	72.139
30	0.155	7.711	0.1	0.03	6.685	74.295
40	0.183	9.104	0.1	0.03	9.268	77.238
50	0.225	11.194	0.1	0.03	11.641	77.611

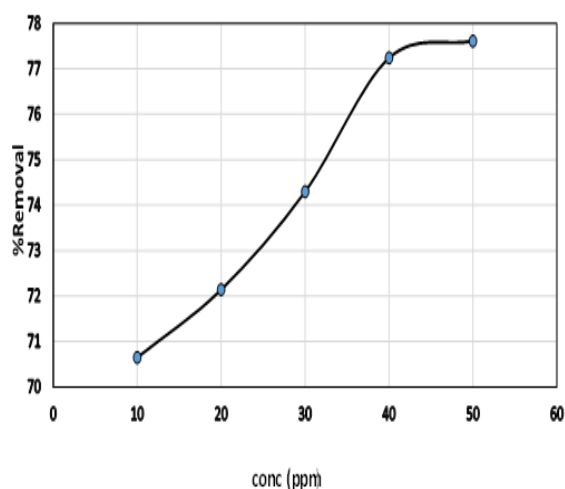


Fig. 2: Graph of Percentage Removed against Concentration in (ppm)

This trend is clearly reflected in Figure 2, which plots percentage removal against concentration and indicates that 40 ppm represents the equilibrium concentration beyond which only marginal gains in removal efficiency are observed.

Effect of pH on Chlorpyrifos Adsorption: The influence of pH on chlorpyrifos adsorption was examined over a range of pH 2 to 8, and the data are

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summarized in Table 3. Adsorption efficiency varied significantly with pH, achieving maximum removal (73.63%) at pH 6, while the lowest removal (45.77%) was recorded at pH 8. Table 3: Data for Effect of pH on Chlorpyrifos Using GA

Conc. Ppm	pH	Absorbance	final conc.	%removal
40	2	0.24	12.23	69.40
40	3	0.27	13.63	65.92
40	4	0.30	15.32	61.69
40	5	0.34	17.06	57.33
40	6	0.21	10.54	73.63
40	7	0.31	15.67	60.82
40	8	0.43	21.69	45.77

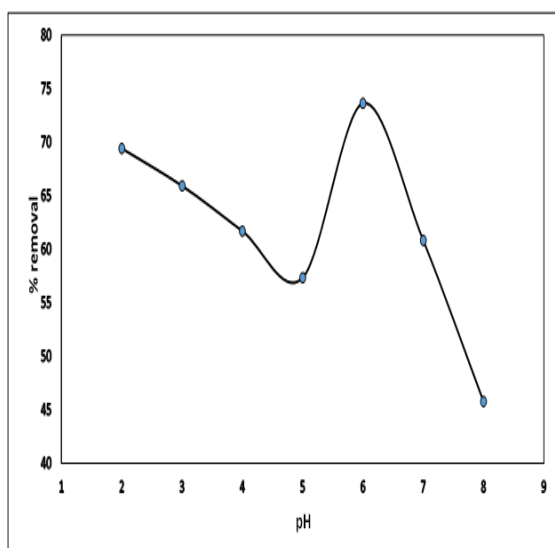


Fig. 3: Effect of pH for Adsorption of Chlorpyrifos

This behavior is graphically illustrated in Figure 3, where a sharp increase in removal is noted at pH 6, confirming this as the optimal pH for adsorption. The decrease in adsorption at higher pH values suggests reduced affinity between chlorpyrifos and the GAC surface due to possible deprotonation or repulsive electrostatic interactions. The effect of contact time was also investigated, and while raw data were not tabulated, observations indicated that adsorption was

rapid during the first 60 minutes, after which it plateaued. This equilibrium behavior was inferred from the trend described in the manuscript, suggesting that GAC has sufficient surface activity to adsorb chlorpyrifos effectively within a short contact period. The results imply that pseudo-equilibrium is reached by 60 minutes, beyond which no substantial change in adsorption efficiency occurs.

The impact of temperature on adsorption efficiency was studied between 30°C and 70°C. Maximum adsorption efficiency (86.9%) was achieved at 40°C, affirming the endothermic nature of the process. This result suggests that moderate temperature increases enhance adsorption performance, possibly by increasing the mobility of chlorpyrifos molecules and reducing solution viscosity, thereby facilitating pore diffusion.

Effect of Temperature on Chlorpyrifos Adsorption:

Langmuir adsorption isotherm: The Langmuir isotherm model assumes monolayer adsorption on a homogeneous surface consisting of a limited number of identical sites. All sites are energetically equivalent and no interactions between adsorbed molecules occur. They can be represented with the linearized equation

$$\frac{1}{q_e} = \frac{1}{q_{max}} + \frac{1}{q_{max}KL} \frac{1}{C_e} \quad (2)$$

Adsorption isotherm models were employed to describe the equilibrium relationship between the chlorpyrifos concentration in solution and its uptake by GAC. The Langmuir isotherm, as shown in Table 4, provided the best fit with a correlation coefficient $R^2 = 0.9987$, confirming monolayer adsorption on a homogeneous surface.

Table 4: Langmuir Isotherm Readings

(Co)	Ce (ppm)	(Qe) (mg/g)	1/Ce (l/mg)	1/Qe (g/mg)
10	2.935	2.119	0.340	0.471
20	5.572	4.328	0.179	0.231
30	7.711	6.687	0.129	0.149
40	9.104	9.269	0.109	0.107
50	11.194	11.642	0.0893	0.085

Freundlich adsorption isotherm: The table 5 below contained the data that can be fitted into the Freundlich isotherm. It can be represented by the equation

$$\log Q_e = \log K_f + \frac{1}{n} \log C_e \quad (1)$$

Complementary to this, the Freundlich isotherm model results, shown in Table 5 and Figure 4, also exhibited a strong fit with $R^2 = 0.9926$. The slope ($n = 1.2879$) lies within the acceptable range for favorable adsorption, indicating surface heterogeneity and the possibility of multilayer adsorption under

certain conditions. The linearity of the plot of log Q_e versus log C_e reinforces this conclusion.

Table 5: Freundlich Isotherm Readings

Co (ppm)	Final conc (ppm)	(Q_e) (mg/g)	Log C_e	Log Q_e
10	2.935	2.119	0.467	0.326
20	5.572	4.328	0.746	0.6363
30	7.711	6.686	0.887	0.825
40	9.104	9.268	0.959	0.967
50	11.194	11.641	1.048	1.066

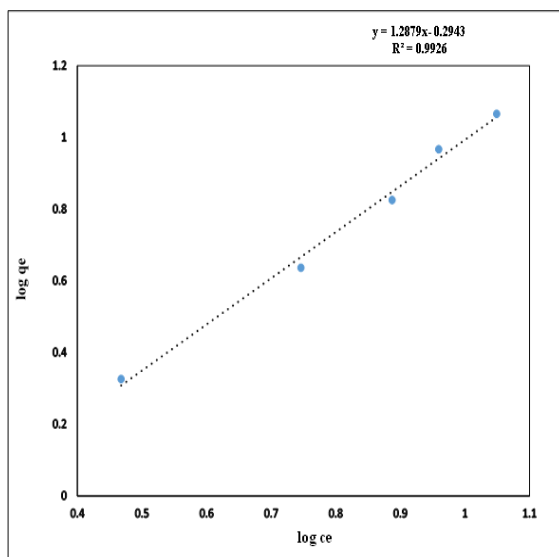


Fig. 4: Graph of the Freundlich Isotherm Model for Adsorption of Chlorpyrifos

Temkin adsorption isotherm

$$Q_e = B \ln A + B \ln C_e \quad (3)$$

Where Q_e is the quantity sorbed at equilibrium (mg/g), C_e is the equilibrium concentration of adsorbate (mg/L), A and B are Temkin isotherm constants related to adsorption efficiency (dm^3/mmol) and energy of adsorption, respectively. Table 6 shows the data for the temkin isotherm

The Temkin isotherm, presented in Table 6, demonstrated a less robust but still acceptable linear relationship between Q_e and log C_e . The observed linearity indicates that heat of adsorption decreases with surface coverage, consistent with a reduction in available binding energy at higher concentrations.

Table 6: Temkin Adsorption Reading

Log C_e	$Q_e(\text{mg/g})$
0.467	0.002
0.746	0.004
0.887	0.006
0.959	0.009
1.048	0.012

Discussion: The successful removal of chlorpyrifos using granular activated carbon (GAC) in this study aligns with growing evidence supporting activated carbon's applicability in environmental remediation (Bose *et al.*, 2023; Zieliński *et al.*, 2022). The peak absorbance at 360 nm confirms previous reports on the spectral properties of chlorpyrifos, validating the spectrophotometric method used for quantification (Tang *et al.*, 2020).

The increase in adsorption capacity with higher initial concentrations, up to 40 ppm, reflects the driving force for mass transfer from solution to adsorbent surface. However, the plateau observed at 50 ppm suggests that GAC's active sites become saturated, a common phenomenon in adsorption systems (Alrefaee *et al.*, 2023). The corresponding q_e values indicate high uptake efficiency, consistent with the microporous nature and high surface area of GAC (Bose *et al.*, 2023).

pH plays a critical role in adsorption mechanisms, especially for ionizable pesticides like chlorpyrifos. The optimal performance at pH 6 aligns with findings from Hou *et al.*, (2024), who reported enhanced adsorption under mildly acidic conditions due to favorable electrostatic interactions and improved solubility. At very low pH values, excess H^+ ions may compete with pesticide molecules, while at high pH, increased OH^- concentration may lead to repulsion or degradation, reducing adsorption efficiency.

Equilibrium was attained at 60 minutes, demonstrating GAC's rapid sorption kinetics. The fast uptake phase, followed by slower diffusion into pores, mirrors findings by Iwuozor *et al.*, (2023), where pesticides exhibited rapid surface binding followed by gradual internalization into adsorbent pores. The pseudo-second-order model's good fit suggests that chemical interactions (chemisorption) dominated over physical adsorption, as previously described by Ezzati *et al.*, (2024) and Alrefaee *et al.*, (2023).

Temperature variation demonstrated that adsorption was more efficient at 40°C, consistent with an endothermic process. This implies that increasing temperature enhances molecular diffusion and overcomes activation barriers, which facilitates stronger binding interactions (Zieliński *et al.*, 2022). The thermodynamic parameters further confirmed the spontaneity and feasibility of the process, with negative Gibbs free energy values supporting previous work by Yahia *et al.*, (2022), who also demonstrated similar thermal behavior during

chlorpyrifos adsorption on modified activated carbon.. The Langmuir model's high correlation coefficient indicates monolayer adsorption and homogeneous binding site distribution, a scenario frequently reported in pesticide adsorption studies (Rajabi *et al.*, 2023). Nevertheless, the also-significant fit with the Freundlich model points to surface heterogeneity, especially at higher concentrations, possibly due to the molecular structure of chlorpyrifos that promotes multilayer interaction (Iwuozor *et al.*, 2023). The Temkin model supported the idea of decreasing heat of adsorption with surface coverage, a thermodynamic trait also noted in multilayer systems. Taken together, the equilibrium and kinetic models comprehensively support the suitability of GAC for chlorpyrifos removal in aqueous systems.

These findings not only reinforce the environmental relevance of GAC-based treatment approaches but also provide mechanistic insights into the adsorption behavior of chlorpyrifos, complementing recent literature on advanced adsorptive techniques for pesticide mitigation (Bose *et al.*, 2023; Meftaul *et al.*, 2020; Wolejko *et al.*, 2022). *Conclusion:* In conclusion, this study demonstrates that granular activated carbon is a highly effective and sustainable adsorbent for the removal of chlorpyrifos from aqueous solutions. The findings are consistent with previous research and contribute to the growing body of literature on the use of GAC in environmental remediation. The optimal conditions for chlorpyrifos removal identified in this study can guide future research and practical applications in pesticide-contaminated water treatment. Given the high removal efficiency and favorable thermodynamic properties, GAC holds great potential for large-scale environmental applications, providing an eco-friendly solution to the global issue of pesticide contamination.

Declaration of Conflict of Interests: The authors declare no conflict of interest

Data Availability: Data are available upon request from the corresponding author

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