



Removal of lamivudine from synthetic solution using jamun seed (*Syzygium cumini*) biochar adsorbent

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ABSTRACT

Antiviral drugs such as lamivudine have been globally identified in the environment and marked as emerging pollutants of concern due to their bioactive extremity. Following therapeutic uses, approximately 70% of the oral dose of lamivudine is eliminated renally as the parent drug. Concerns have been raised for neighbouring aquatic bodies due to effluent produced from production plants containing high concentrations of antiviral drugs. Antiviral drugs, such as lamivudine, are extremely bioactive, prompting interest in their urgent removal from the environment. The purpose of the present study was to optimize the removal of lamivudine from the synthetic solution using jamun seed (JS) (*Syzygium cumini*) biochar. The influence of sorption parameters such as pH, lamivudine concentration, adsorbent dosage, contact time, and calcination temperatures on the removal of lamivudine was investigated and optimized using a response surface methodology (SRM) based on optimal design. The results indicated that, a quadratic model best fits data with a model regression coefficient R^2 , adjusted R^2 , and predicted R^2 of 0.9934, 0.9761 and 0.8340, respectively. The JS biochar calcined at 750 °C, at pH 8, initial lamivudine concentration of 10 ppm and contact time of 30 min indicated a maximum experimental removal efficiency of 84.9%. The residual standard error (RSE) value was 3.5% implying that the model was reliable. Isotherm data for the adsorption of lamivudine on JS biochar followed the Freundlich isotherm, with an R^2 value of 0.9977 while R^2 for the modified Langmuir model was 0.9852. These findings indicated that JS biochar is potentially useful for removal of lamivudine, and other organics from contaminated water and wastewater effluents. Therefore, this study presents an environmentally friendly remedy against lamivudine for a healthier ecology.

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1. Introduction

Viruses are among pathogenic agents causing several serious diseases such as respiratory syncytial virus infection, adenovirus infection, parainfluenza virus infection, severe acute respiratory syndrome (SARS), norovirus infection, rotavirus infection, infections, astrovirus infection, hepatitis C, and some adenovirus in humans [1–3], African horse sickness, African swine fever (ASF), bluetongue, BRSV, coronavirus, crimean Congo haemorrhagic fever, equine infectious anaemia, and equine viral arteritis in animals [4–7], and Johnson grass mosaic virus, celery mosaic virus, sweet

potato feathery mottle virus, subterranean clover stunt in plants [8], and some affects both [9–12]. Furthermore, viruses are responsible for more diseases than any other parasite [9,13,14]; they can cause effects such as blindness (SARS-CoV-2) [15], deafness (rubella virus) [16,17], paralysis (enterovirus (EV) virus) [18], congenital disabilities (talipes equinovarus (CTEV) virus) [16,19,20], and cancer (human papillomavirus (HPV)) [21], in some cases. Measles, mumps, smallpox, chicken pox, influenza, poliomyelitis, and yellow fever are some of the most well-known viral diseases [13,22–24]. Contrary to the effectiveness of using antibiotics in the treatment of bacterial diseases, antiviral drugs (ARVDs) used for the treatment of viral disease infection are less effective [9,13,14,25]. There are, however, several influenza drugs such as rapivab, relenza, tamiflu, xofluza [26,27], a couple of herpes virus drugs such as acyclovir, famciclovir and valacyclovir [28], and some new antiviral drugs such as lamivudine [22,29–31], for treating HIV and hepatitis C infections.

According to the WHO, approximately 28.7 million people across the world were expected to receive antiretroviral therapy such as lamivudine in 2021 [32], which is equivalent to more than 2700 tons of the drug. In turn, this might have significantly increased the active chemical (AC) load in the environment. Antiretroviral drugs, such as lamivudine, are used to treat HIV and other viral infections [33–35]. Although some antivirals are less effective, increased antiviral production and/or usage, release of contaminated effluents before or after partial treatment, and improper disposal of expired or out-of-use antiviral drugs have potentially contaminated terrestrial and aquatic ecosystems [36–49]. This may throw ecosystems off balance, leading to unsustainability. Antiviral drugs in the environment have gotten much attention because they avoid degradation in wastewater treatment plants (WWTPs) and end up in surface and groundwater sources [36–50], while creating harm.

Adeola and Forbes reported that efavirenz and nevirapine are the most persistent in effluents and are prevalent in surface water according to environmental concentrations [51]. Their study further observed highest concentration of efavirenz reported in Kenya was $12.4 \mu\text{g L}^{-1}$, concentrations as high as 119 and $140 \mu\text{g L}^{-1}$ have been reported in Zambia and South Africa [51], respectively. ARVD concentrations ranging from 670 to $34\,000 \text{ ng L}^{-1}$ (influent) and 540 to $34\,000 \text{ ng L}^{-1}$ (effluent) were determined in wastewater treatment plants in South Africa, compared to Europe, where reported concentrations range from less than the limit of detection (LOD) to 32 ng L^{-1} (influent) and less than the LOD to 22 ng L^{-1} (effluent) [51], including the report of exposure to aquatic ecosystems reported by previous researchers [46,52–62].

In addition, Kunene and Mahlambi reported that all the studied vegetables have the potential to uptake abacavir, nevirapine, and efavirenz from contaminated soil, be absorbed by the root, and translocate them to the aerial part of the plants. The total percentage of ARVD found in the individual plant was mainly attributed to abacavir, which contributed 53% in beetroot and 48% in spinach, while efavirenz (42%) was the main contributor in tomato. Abacavir was found at high concentrations to a maximum of $40.21 \mu\text{g/kg}$ in the spinach root, $18.43 \mu\text{g/kg}$ in the spinach stem, and $6.77 \mu\text{g/kg}$ in the spinach soil, while efavirenz was the highest concentrations, up to $35.44 \mu\text{g/kg}$ in tomato leaves and $8.86 \mu\text{g/kg}$ in tomato fruits. Spinach roots accumulated more ARVDs than beetroot and tomato. These results indicate that the presence of these drugs in effluents, soil, and other environmental compartments can harm the entire ecosystem through the food chain.

However, data on the associated risks following environmental exposure to nontargeted species are limited [52–54]. For example, the antiviral drug lamivudine is reported to be among the most toxic therapeutic classes in terms of toxicity to daphnids, fish and

algae [39,40,63]. Lamivudine is toxic to *Daphnia magna*, the exposure of 100 g/L of lamivudine to *Daphnia magna* in the freshwater resulted in a mortality rate of 85%, which can increase with prolonged exposure [33–35,62,64]. Although exposure to lower concentrations of lamivudine (10 g/L) was not toxic to *Daphnia magna* [64], yet a prolonged exposure to lower doses may lead to the development and dissemination of drug resistance that can threaten entire ecosystems [33–35,64]. Additionally, reports indicate a significant negative impact on root lengths and chromosomal aberration when *Allium cepa* was exposed into 100 g/L of lamivudine [64], while *Lactuca sativa* had a slight negative impact on both seed germination rate and hypocotyl length [64]. This indicates that lamivudine poses an ecological health risk to flora and fauna at different trophic levels at previously found environmental concentrations requiring intervention. The current environmental assessment and monitoring guideline does not consider emerging contaminants, and this calls for further research to generate data necessary for policy reforms for environmental sustainability.

Search for innovative ideas to improve and invent new techniques for remediation of these contaminants are required to ensure ecological safety [35]. Various techniques have shown promising results in the removal of organic contaminants such as lamivudine. However, the use of abundant environmental-friendly materials as an adsorbent, cost effectiveness, and no generation of toxic products with a low initial cost of implementation have attracted attention for remediation of lamivudine. Therefore, the current study investigated the removal of lamivudine from synthetic solution using jamun seed (*Syzygium cumini*) biochar adsorbent and optimized by response surface methodology.

2. Methodology

2.1. Samples

The current study used jamun seed (*Syzygium cumini*) biochar calcined at temperatures $400 \text{ }^\circ\text{C}$ (sample 1); $500 \text{ }^\circ\text{C}$ (sample 2); $600 \text{ }^\circ\text{C}$ (sample 3); and $750 \text{ }^\circ\text{C}$ (sample 4), and synthetic solution and lamivudine.

2.2. Materials

All equipment used for adsorption experiments, including shaker, analytical balance, and glassware's, were obtained from the College of Natural and Mathematical Sciences research laboratory of The University of Dodoma. Consumable analytical grade reagents including methanol, hydrochloric acid, and sodium hydroxide as well as distilled water and standard solution of lamivudine were locally secured.

2.3. Adsorbent preparation

Jamun seeds (*Syzygium cumini*) were collected, dried in the shade, pulverized, and sieved to obtain a uniform powdery material. The powder was then calcined at temperatures 400 , 500 , 600 , and $750 \text{ }^\circ\text{C}$ in order to obtain adsorbent material using a CARBOLITE tube furnace in the presence of nitrogen gas. Initial elemental characterization of the material was conducted using the CHNS analyser (Flash 2000), the FTIR was used to study the surface functional groups, and the characteristics of the surface area, volume, and pore size were obtained using a QUANTACROME 1000 L-Se series porosimeter.

2.4. Design of experiments and statistical analysis

Response surface methodology (RSM) is a method of empirical

modelling that determines the interaction of multiple operating and response variables. It outlines a systematic experimentation strategy for developing and optimizing an empirical model. The RSM is essentially a combination of mathematical and statistical approaches that can be used to model and analyze problems where the output is affected by input variables and their interactions [65–68]. An RSM based on the optimality criterion was used to optimize five independent and one response variables. The studied independent variables were the adsorbent dose that ranged between 50 and 1000 mg, calcination temperatures set at 400, 500, 600 and 750 °C, residence time set between 30 and 300 min, pH that ranged between 1 and 14, and lamivudine concentration at a range of 10 and 500 ppm against the lamivudine removal efficiency. Variables selection criteria were based on the data reported from similar studies [68–72].

Among the 55 experimental runs, 45 were model points, five were replicate points, and the last five were lack-of-fit points. The RSM involves five main steps including development of statistically designed experiments, generation of an empirical model, statistical analysis of the model, numerical optimization by using desirability function and finally the model confirmation. The experimental runs were randomized in order to diminish errors and effects of uncontrolled factors [73]. The observed responses were used to generate an empirical model conforming to the experimental variables. The obtained experimental results from the 55 runs were used to determine the regression coefficient of the quadratic model by using Design-Expert Version 13.0.5 software (Stat-Ease, Inc., Minneapolis, USA) [74]. The *R*-square values established the accuracy of the fitted model, and the significant model terms were evaluated by the *p*-value at a 95% confidence level.

Contour and the 3-D surface plots were developed to show the interaction of two independent variables while holding the third variable at the central value. The geometry of the surface plots provided information about the behavior of system on the variation of the processing variables within the design space. Optimization of adsorption conditions were numerically conducted using a desirability function of the Design-Expert software to maximize pollutant removal. Using the models created during analysis, the optimum-operating conditions that meet the goal of maximization of lamivudine removal were searched within the design space. Finally, the model validation was conducted by running three replicates' experiments of selected conditions.

2.5. Adsorption experiments

A batch adsorption experiment was conducted to evaluate the removal of lamivudine from a synthetic water solution. The stock solution containing 1000 mg/L lamivudine was prepared by dissolving 1 g lamivudine in deionized water. Then, working solutions were prepared from the stock solution through serial dilutions of 50 mL each. While maintaining the concentration of lamivudine, pH levels and varying doses of the J. S. biochar were used from different sets of experiments. Adsorption systems were continuously stirred at 220 rpm, and timely monitored. At the desired time intervals, the solutions of the adsorption systems were sampled. The sampled solutions' adsorbent was taken and centrifuged at 500 rpm for 15 min and filtered using a 0.45 µm syringe filter. The concentration of lamivudine in the grab sample was then determined spectrophotometrically at the wavelength of 270 nm following recommendations from previous studies [65,66,69–71,75–77]. The amount of lamivudine uptake at equilibrium (q_e) (mg/g) and removal efficiency (%) was finally calculated by adopting equations (1) and (2).

$$q_e = [(C_0 - C_t) V] / M \quad (1)$$

$$R = [(C_0 - C_t) / C_0] * 100 \quad (2)$$

where C_0 and C_t are the concentrations of lamivudine (mg/L) at the beginning and at time t of the process, respectively, V (L) is the volume of the solution and m (g) is the mass of the adsorbent. Table 1 presents experimental runs, experimental and predicted values of lamivudine removal efficiency.

2.6. Isotherm models

The validity of Langmuir isotherm for monolayer sorption is expressed in Equation (3) [78].

$$\frac{C_e}{Q_e} = \frac{1}{bQ_0} + \frac{C_e}{Q_0} \quad (3)$$

where C_e is the equilibrium lamivudine concentration (mg/L) and Q_e the amount adsorbed at equilibrium (mg/g). The Langmuir constants Q_0 (mg/g) represent the monolayer adsorption capacity and b (L/mg) relates the heat of adsorption [78–80]. The essential feature of the Langmuir adsorption expressed by R_L , a dimensionless constant referred to as the separation factor or equilibrium parameter for predicting whether an adsorption system is favorable or unfavorable [78–80]. R_L is calculated using Equation (4).

$$R_L = \frac{1}{1 + bC_0} \quad (4)$$

If the R_L values lie between 0 and 1, the adsorption is favorable. This Langmuir model can also be written in a non-linear form as presented by Equation (5)

$$Q_e = \frac{bQ_0C_e}{1 + bC_e} \quad (5)$$

The Freundlich isotherm is expressed in linear form as presented by Equation (6).

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (6)$$

where K_f indicates adsorption capacity and n is an empirical parameter related to the intensity of adsorption, which can vary with the heterogeneity of the adsorbent [78,80]. The greater the values of the $1/n$, the better the adsorption [78]. A higher fractional value of $1/n$ ($0 < 1/n < 1$) implies that the surface of adsorbent is heterogeneous.

3. Results and discussion

3.1. Adsorption of lamivudine onto JS biochar

Lamivudine removal efficiency observed from the 55 experimental runs ranged between 31.4% and 84% as indicated in Table 1. The maximum removal efficiency was observed at the 50th run, at a pH value of 8, initial lamivudine concentration of 10 ppm, adsorbent dosage 50 mg and contact time of 30 min, whereby the adsorbent was calcined at 750 °C. The obtained results were fitted to a generic quadratic equation, and models were selected using criteria presented in Table 2.

The significant of the model terms were evaluated and model reduction was made to improve the model by removing trivial terms similarly as reported by other researchers [81,82]. The final quadratic equation in terms of the coded variable presented in Equation (viii) was derived after the removal of $p > 0.1$ using the *p*-values backward elimination algorithm in the Design-Expert software.

Table 1
Experimental runs, experimental and predicted values of lamivudine removal efficiency.

Run	A: pH	B: Conc.	C: Ads. Dosage	D: Time	E: Temperature	Lamiv. % Removal	
	–	ppm	mg/l	Min	°C	Exp.	Pred.
1	0	10	50	212	250	77.9	75.02
2	0	10	1000	300	300	77.6	76.09
3	0	10	1000	30	500	74.9	73.02
4	0	500	50	165	0	40.7	41.97
5	14	500	1000	30	600	40.4	40.15
6	14	10	50	30	400	75.0	75.01
7	4	255	216	30	0	54.8	51.91
8	14	355	1000	30	750	40.9	40.86
9	14	10	1000	300	500	77.7	77.74
10	0	500	1000	30	400	31.4	31.47
11	7	245	507	158	400	46.9	46.72
12	14	500	1000	300	300	40.2	43.69
13	14	500	1000	30	600	40.4	40.15
14	5	10	50	300	0	67.8	68.56
15	14	10	50	300	300	79.9	77.66
16	3	248	1000	300	600	53.3	54.65
17	14	500	50	30	300	43.8	42.35
18	12	304	50	166	0	52.4	51.13
19	14	10	50	30	500	77.7	77.91
20	0	500	50	300	400	40.1	40.07
21	0	500	744	300	250	20.0	20.68
22	14	500	1000	118	250	35.8	32.96
23	0	10	1000	300	400	76.9	76.97
24	0	10	1000	30	0	76.6	76.55
25	14	500	1000	300	400	56.8	56.91
26	14	500	50	300	750	40.3	40.58
27	14	10	1000	300	250	80.4	83.04
28	14	10	1000	300	750	82.7	83.07
29	0	500	1000	30	300	41.7	39.4
30	14	10	1000	300	750	83.5	83.07
31	0	500	50	300	300	38.2	38.53
32	4	257	810	38	750	58.8	60.7
33	0	500	50	30	250	39.5	42.07
34	0	10	1000	30	250	79.0	78.57
35	13	110	50	30	600	54.7	55.76
36	0	152	50	300	750	59.8	60.3
37	14	10	311	30	250	79.0	79.66
38	0	10	649	30	600	77.2	76.48
39	0	125	582	207	500	39.7	42.22
40	0	500	50	300	400	40.0	40.07
41	0	500	50	300	500	37.6	36.89
42	14	255	1000	300	0	43.1	42.34
43	14	10	50	30	0	83.4	85.06
44	14	378	558	221	500	41.4	39.99
45	9	500	1000	30	0	49.5	50.83
46	0	500	50	300	600	50.3	50.22
47	0	10	50	30	300	69.9	73.36
48	0	500	50	300	600	50.6	50.22
49	14	500	1000	30	500	45.3	46.44
50	8	10	50	30	750	84.9	83.71
51	14	500	50	300	250	47.0	46.54
52	14	500	1000	300	400	57.0	56.91
53	14	10	1000	30	300	79.6	79.9
54	0	500	1000	184	750	57.6	56.23
55	14	10	538	300	600	79.9	79

Table 2
Fit summary.

Source	Sequential p-value	Lack of Fit p-value	Adjusted R ²	Predicted R ²	
Linear	<0.0001	<0.0001	0.7596	0.6991	Suggested
2FI	0.7736	<0.0001	0.7050	-3.6924	
Quadratic	<0.0001	<0.0001	0.9748	-0.5370	Suggested
Cubic	<0.0001		0.9998		Aliased

In this case A, B, C, D, E, AB, AD, AE, BE, CD, CE, DE, and C² combinations were significant in the formation of model input terms.

Therefore, equation (7) was useful in describing lamivudine

removal behavior, whereby negative and positive sign implied the antagonistic and synergistic effects of the factors, respectively.

Fig. 1 represents model diagnostic plots used to ascertain the validity of the suggested models. The plots of actual versus

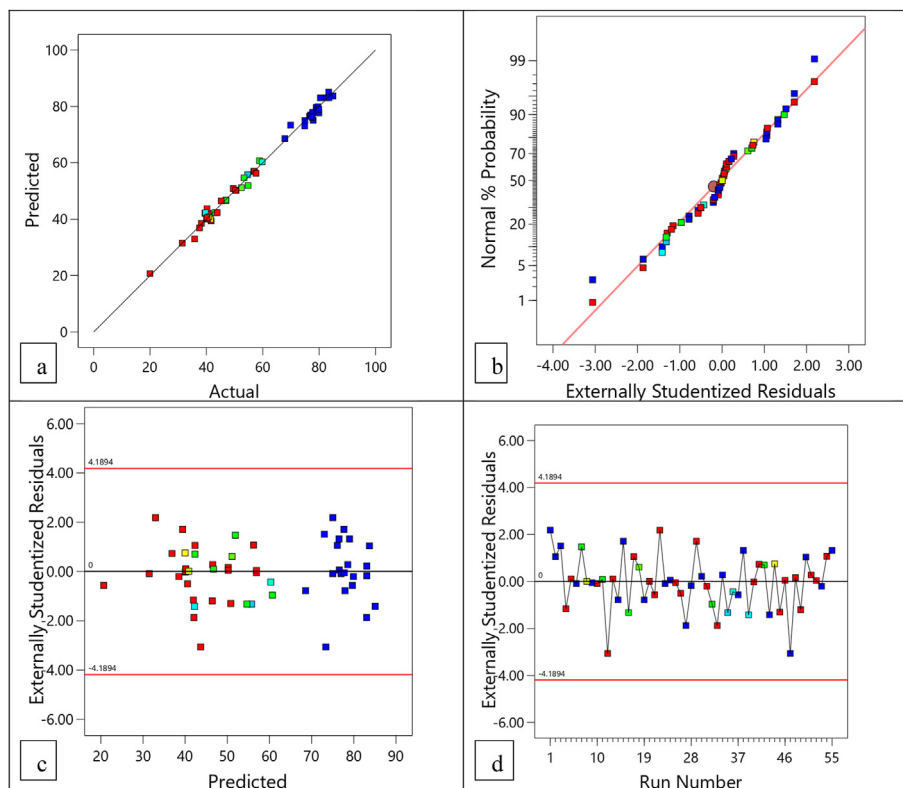


Fig. 1. Diagnostic plots ascertaining the validity of the data fitting in the selected models (a) actual vs predicted (b) normal probability (c) externally studentized residuals vs predicted, and (d) externally studentized residuals vs run number.

predicted values of lamivudine removal show that data were linearly fitted well and distributed along the line of best fit. It was an indication that the predicted values agree well with the experimental ones. The externally studentized residuals plot shows that the residuals are generally distributed within the limits, implying that the errors are normally distributed. Furthermore, the normal probability plot confirms that the model is normal.

equations 7–10, present removal of lamivudine from synthetic solution using JS biochar calcined at various temperatures.

% Lamiv removal at 400 °C

$$= 66.1754 + 2.16134A - 0.188753B + 0.0138865C + 0.0195324D + 0.00248677AD - 2.31644 \times 10^{-5}BC - 0.104781A^2 + 0.0002545B^2 + 2.09161 \times 10^{-5}C^2 \quad (7)$$

% Lamiv removal 500 °C

$$= 75.0022 - 1.88155A - 0.1748BA - 0.0196675C - 0.0426752D + 0.00248677AD - 2.31644 \times 10^{-5}BC - 0.104781A^2 + 0.000254545B^2 + 2.09161 \times 10^{-5}C^2 \quad (8)$$

% Lamiv removal 600 °C

$$= 78.5134 - 0.944567A - 0.182704B - 0.0137812C - 0.00212424 + 0.00248677AD - 2.31644 \times 10^{-5}BC - 0.104781A^2 + 0.000254545B^2 + 2.09161 \times 10^{-5}C^2 \quad (9)$$

% Lamiv removal 750 °C

$$= 94.0974 - 0.0370255A - 0.19102B - 0.00916401C - 0.0334989D + 0.00248677AD - 2.31644 \times 10^{-5}BC - 0.104781A^2 + 0.000254545B^2 + 2.09161 \times 10^{-5}C^2 \quad (10)$$

3.2. Analysis of variance

The results of ANOVA analysis for the reduced quadratic model are presented in Table 3. The model F-value was 57.47 implying that the model was significant. There was only 0.01% chance that an F-value this large could occur due to noise. Similarly, p-values less than 0.0500 as presented in Table 3, indicated that model terms are significant and can be used to explain the adsorption of lamivudine onto JS biochar.

The obtained 0.9934 value of the R-squared in the model was close to one implying that these data fitted well into the selected model. The predicted 0.8340 value of R² was in reasonable agreement with the adjusted R² value of 0.9761 whereby the difference being less than 0.2. The results of modal-fit statistics are presented in Table 4.

Adequacy precision measures the signal to noise ratio, with a ratio greater than 4 being desirable. The obtained ratio of 27.3995 as presented in Table 4, indicated an adequate signal. Therefore, this model is sufficiently useful for navigation of the design space. The suggested model gave a significant lack-of-fit with a p value less than 0.05, but other statistics parameters of model are significant and adequate precision are generally acceptable, which allows the model to be used for optimization purposes as previously reported by Dritsa and colleagues in 2009 [83].

Table 3
Results of ANOVA analysis for the reduced quadratic mode.

Source	SS	df	MS	F-value	p-value	Importance
Model	17016.22	39	436.31	57.47	<0.0001	significant
A-pH	215.99	1	215.99	28.45	<0.0001	
B-Concentration	13082.23	1	13082.23	1723.21	<0.0001	
C-Adsorbent dosage	4.00	1	4.00	0.5266	0.4792	
D-Contact time	21.04	1	21.04	2.77	0.1167	
E-Treatment temp	274.93	6	45.82	6.04	0.0022	
Interactions						
AD	116.20	1	116.20	15.31	0.0014	
AE	768.67	6	128.11	16.88	<0.0001	
BC	137.06	1	137.06	18.05	0.0007	
BE	242.22	6	40.37	5.32	0.0040	
CE	232.04	6	38.67	5.09	0.0049	
DE	465.38	6	77.56	10.22	0.0001	
A ²	58.48	1	58.48	7.70	0.0141	
B ²	889.57	1	889.57	117.17	<0.0001	
C ²	73.77	1	73.77	9.72	0.0071	
Residual	113.88	15	7.59			
Lack of Fit	113.51	10	11.35	156.16	<0.0001	significant
Pure Error	0.3634	5	0.0727			
Cor Total	17130.09	54				

SS- Sum of Squares, DF- Degree of freedom; MS- Mean Square.

Table 4
Modal fit statistics.

Std. Dev.	2.76	R ²	0.9934
Mean	57.66	Adjusted R²	0.9761
C.V. %	4.78	Predicted R²	0.8340
		Adeq Precision	27.3995

3.3. Optimization process and validation of the model

The validity of the predicted models was evaluated under three replicates confirmatory experiments randomized at pH value of 14, lamivudine concentration of 13 ppm, 999 mg of adsorbent dose, 249 min contact time, and treatment temperature of 400 °C. At these conditions, the predicted removal efficiency of lamivudine from the synthetic water solution was 99.06%, while the average experimental value was 90.38%. The Residual Standard Error (RSE) obtained using Equation (11) was 3.5%, indicating excellent agreement between experimental and predicted output values. Optimal conditions in Fig. 2 indicated that JS biochar may potentially be

useful for remediation of basic effluents, but further results presented on Figs. 6–9, shows that the adsorbent can work best on a wide range of pH that alter its surface charges leading to improved removal and may potentially be used to remediate emerging pollutants such as lamivudine.

$$\%RSE = \frac{\text{Exp.value} - \text{Pred.value}}{\text{Pred.value}} * 100 \tag{11}$$

Fig. 3 presents the modal confirmation conditions for lamivudine adsorption from synthetic solution onto JS biochar.

3.4. Effect of parameters on lamivudine removal efficiency

The effect of adsorption process variables such as initial lamivudine concentration, pH, adsorbent dosage, contact time, and JS biochar calcination temperatures on the removal efficiency of lamivudine are shown in Fig. 4.

Perturbation plots are used in examining the effects of individual input variables on the removal efficiency of lamivudine by the JS biochar. The plots allowed the comparison of all parameters

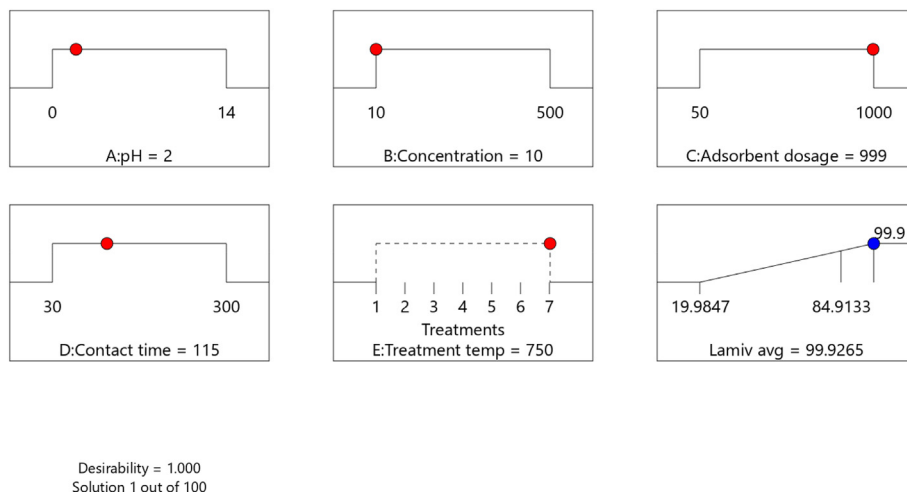


Fig. 2. Model optimization conditions.

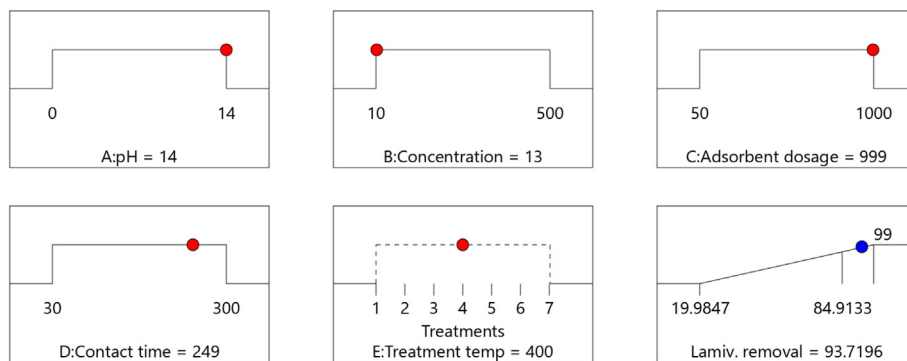


Fig. 3. Model confirmation conditions.

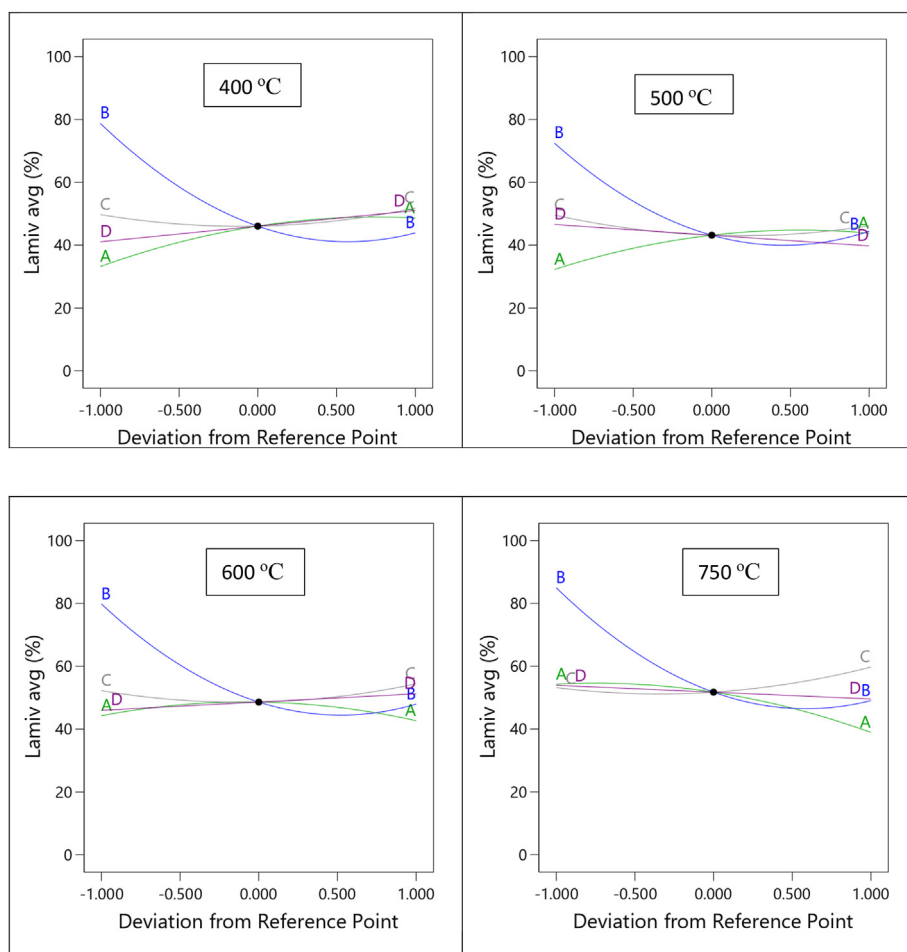


Fig. 4. Perturbation plots showing the sensitivity of various factors on the removal of lamivudine using adsorbents calcinated at 400, 500, 600, and 750 °C. A stands for pH, B for pollutant concentration, C for adsorbent ratio, and D for contact time.

that influenced the removal of lamivudine at a particular point in the design space by shifting one element while the rest are kept constant.

In general, the perturbation curve was drawn by selecting a reference point and modifying the operating range of each involved variable. Modification of reference point is then done at the center of each factor, and the results are plotted against the deviation from the reference point. A steeper or curved slope in the response reflected the amount of sensitivity to that parameter. A rather flat line indicated that the response is un-affected by changes in that

specific element [84]. The initial concentration of lamivudine in the solution had significant effect on the removal efficiency, followed by adsorbent dosage ratio, pH, and contact time.

3.5. Effect of variation of adsorbent calcination temperature

Generally, the removal efficiency of lamivudine onto JS biochar was observed to decrease as lamivudine concentration increased. It was an indication that biochar became saturated as the concentration of lamivudine increased. The effect of variation of adsorbent

calcination temperature on the removal of lamivudine onto JS biochar is presented by Fig. 5.

The removal of lamivudine onto JS biochar prepared at 400 °C were relatively like that of JS biochar prepared at 500 °C, due to these adsorbents having similar properties. The results of initial material characterization indicated surface area of 232.9 m²/g, pore radius of 27.02 Å, pore volume of 0.315 cc/g, and close carbon content of 77.25 and 79.38% respectively. The findings presented in Fig. 5 shows closeness at initial lamivudine concentration of 300–400 ppm. Further, for biochar prepared at 600 and 750 °C, are far apart, due to variation in adsorbent properties. The material prepared at 600 °C, surface area of 261.2 m²/g, pore radius of 24.33 Å, pore volume of 0.318 cc/g, and carbon content of 87.63% while for material prepared at 750, surface area of 220.8 m²/g, pore radius of 19.44 Å, pore volume of 0.215 cc/g, and carbon content of 76.61%. Further decrease of material properties at higher temperatures was due to pore clogging in the material microstructure.

Previous studies reported the potential of biosorbents and their modifications to effectively remove pollutants at lower concentrations [85–88]. Emerging contaminants are present and persist in the environmental compartments at lower levels creates harm and impair the sustainability of ecosystems [43,45–47]. The problem may be higher in countries with inefficient wastewater treatment facilities, as the likelihood of environmental release and limited infrastructure, presenting multiple challenges [89–92]. Report of the occurrence of lamivudine at 0.89, 242, and 130 ng/L in some in South Africa environments [93–95], whereas K'oreje [92], and Ngumba [90], presented significantly higher levels of 167 000 and 5428 ng/L, respectively, in Kenya. Report of the presence of antiviral drugs in Africa's aquatic environment are available, therefore the potential of harm to the ecosystems [91]. The antiviral drug lamivudine is reported to create ecological harm with concentration of 10–100 ppm [33–35,64]. In the current study, at this concentration, JS biochar was observed to efficiently remove, indicating its potential for remediation of organics, such as lamivudine. The removal of lamivudine using JS biochar was observed to increase as the calcination temperature increased from 400 to 750 °C, as presented in Fig. 5.

Previous research has reported a similar pattern [69,70,96–98],. Huang and Colleagues [97], prepared and tested rabbit manure biochar at 400 °C, 500 °C, 600 °C, and 700 °C and found that the

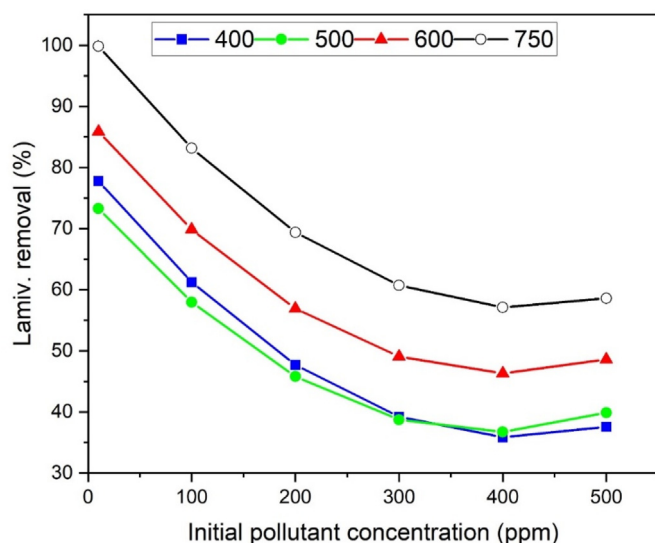


Fig. 5. Effect of adsorbent calcination temperature on lamivudine removal efficiency.

material had relatively similar effectiveness but the removal efficiency slightly increased as calcination temperatures increases, as observed in this study. The calcination temperature was reported to influence the physical chemical and adsorption capacity of a material as similarly observed in the present study. Again, Figs. 6–9, shows that the removal efficiency is largely affected by the initial concentration and adsorbent dose, whereas the pH and contact time had only a slight effect. The pH of solution has a significant impact on the adsorptive removal of a pollutant as it determines the form in which it exists. At lower lamivudine concentration the removal is more than at higher concentration. Similar results were reported by previous researchers [85–88].

3.6. Effect of interactions on the removal of lamivudine

The removal of lamivudine at various conditions are presented in Fig. 6(a–d) to 10 (a–d). The removal efficiency increased with the adsorbent dose, as the adsorbent dosage increases more surface area is available for adsorption and hence increased removal. The material removed about 80% of lamivudine at basic pH. Fig. 6 shows a contour and 3D plots presenting lamivudine adsorption onto JS biochar calcined at 400 °C (a) effect of concentration and pH, (b) adsorbent dosage and pH (c) contact time and pH, and (d) adsorbent dosage and initial lamivudine concentration. The results indicate that the removal of lamivudine onto JS biochar was increasing as pH of solution increases from 5 to 11 as presented in Fig. 6 (a). Furthermore, at an initial concentration of 10 ppm the removal was 80%, but the removal decreased to 70% when the initial concentration reached 80 ppm, indicating that JS biochar was more capable at lower concentrations, and at higher concentration the removal decreases. At pH of 7 JS biochar was capable of removing 70% of lamivudine, but the removal increases to 80% at pH of 11. The pH of solution have a great influence on the surface charges of the material leading to increased removal of contaminants as reported by previous scholars [99].

Fig. 6 (b) presents the contribution of adsorbent dosage on the removal of lamivudine. The lower adsorbent dosage was able to remove lamivudine from synthetic solution, but the removal increases as adsorbent dosage increases due to increase in the adsorptive surface area.

The results Fig. 6(c) also, indicates removal of lamivudine at lower contact times but increases as contact time increases, due to agitation of sample for longer time provides more area for adsorption. Fig. 6(d) indicates the removal of lamivudine is more at lower concentration and increases as adsorbent dosage increases due to the presence of more adsorptive surface areas.

Similarly, the material calcined at 500 °C managed to remove about 70% of lamivudine from synthetic solution. The removal was higher at lower lamivudine concentration and increases as adsorbent dosage increases.

The results indicated in Fig. 7 show that the removal of lamivudine from synthetic solution onto JS biochar reached maximum of 80% as presented in Fig. 7 (a). Furthermore, at initial concentration of 10 ppm of lamivudine the removal efficiency was 70%, which was obtained throughout the pH range. This was an indication that JS biochar can be used for remediation of lamivudine contaminated water and effluents at a variety of pH range that influenced its surface charges leading to the removal of lamivudine. Fig. 7 (b) presents the contribution of adsorbent dosage on the removal of lamivudine, whereby lower adsorbent dosage was able to remove lamivudine from synthetic solution, but the removal efficiency increased as adsorbent dosage increased. The results Fig. 7 (c) also, indicated removal of lamivudine at lower contact times but increased as contact time increases as longer agitation time provides more area for adsorption. Fig. 7 (d) indicates the

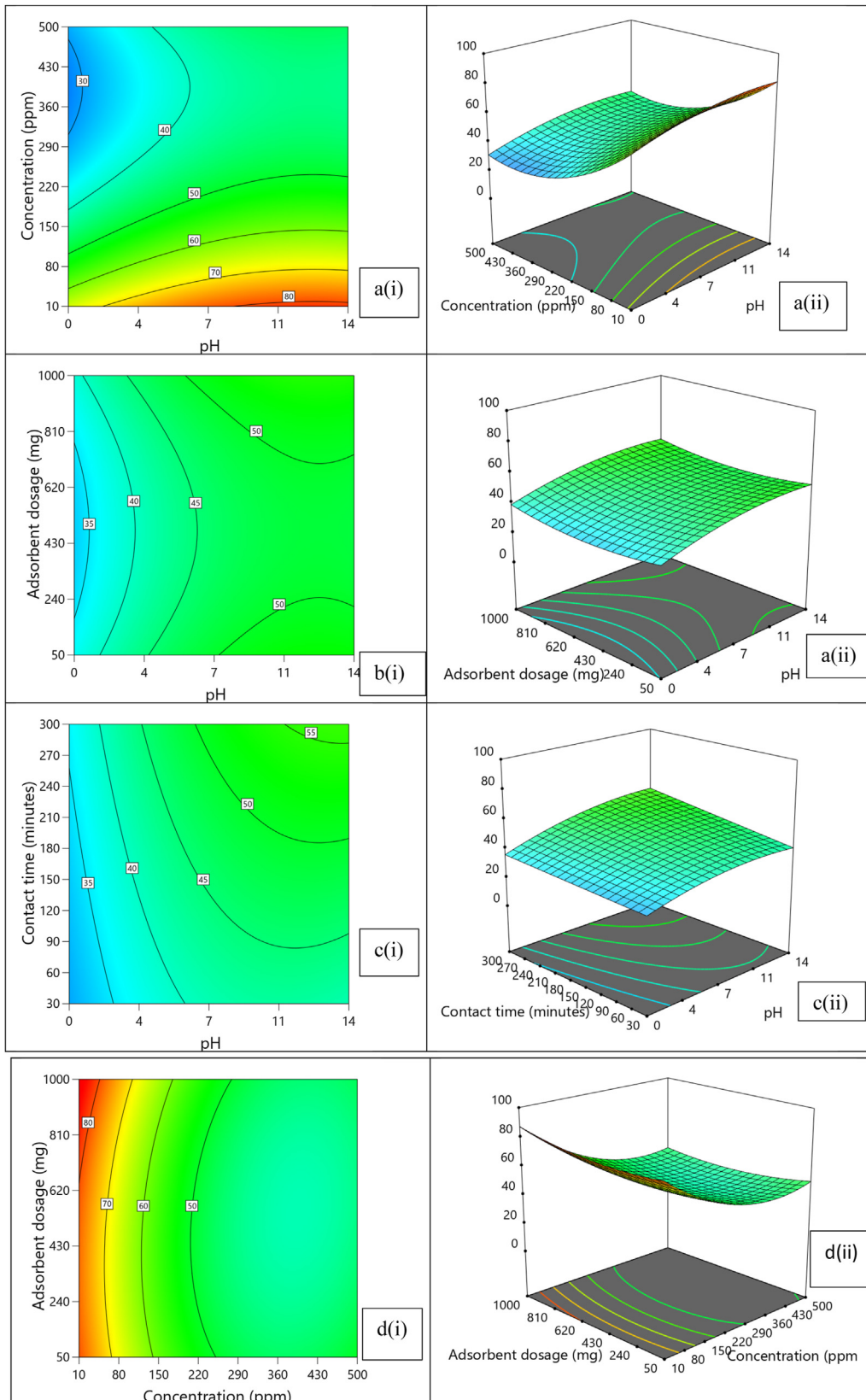


Fig. 6. Contour and 3D plots present the removal of lamivudine onto JS biochar calcined at 600 °C. Fig. 6a(i) and (ii) indicate the effect of concentration and pH, Fig. 6b(i) and (ii) indicate the effects of adsorbent dosage and pH, Fig. 6c (i) and (ii) indicate the effects of contact time and pH while Fig. 6d (i) and (ii) indicate the effects of adsorbent dosage and initial lamivudine concentration.

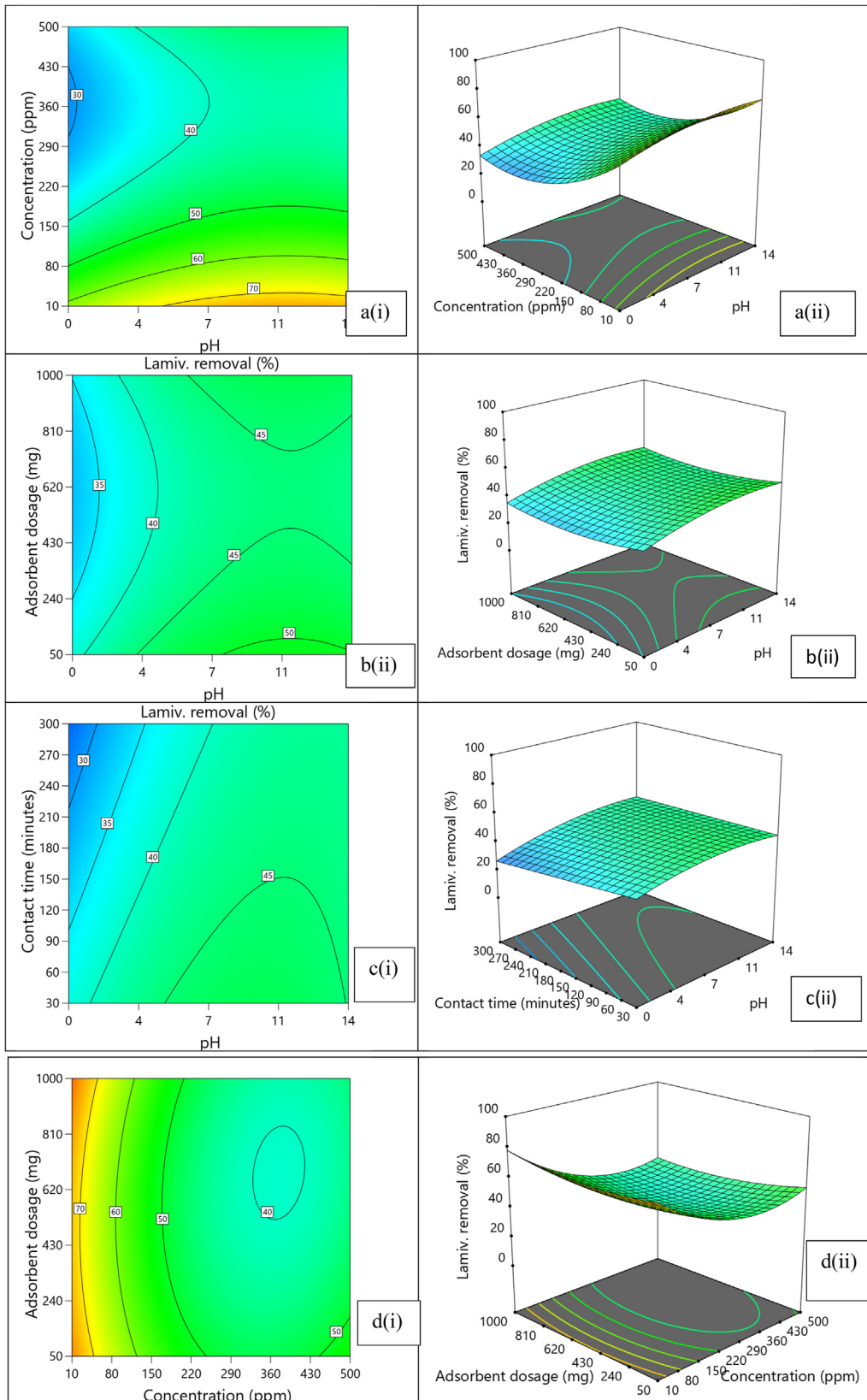


Fig. 7. Contour and 3D plots present the removal of lamivudine onto JS biochar calcined at 500 °C. Fig. 7a(i) and (ii) indicate the effect of concentration and pH, Fig. 7b(i) and (ii) indicate the effects of adsorbent dosage and pH, Fig. 7c (i) and (ii) indicate the effects of contact time and pH while Fig. 7d (i) and (ii) indicate the effects of adsorbent dosage and initial lamivudine concentration.

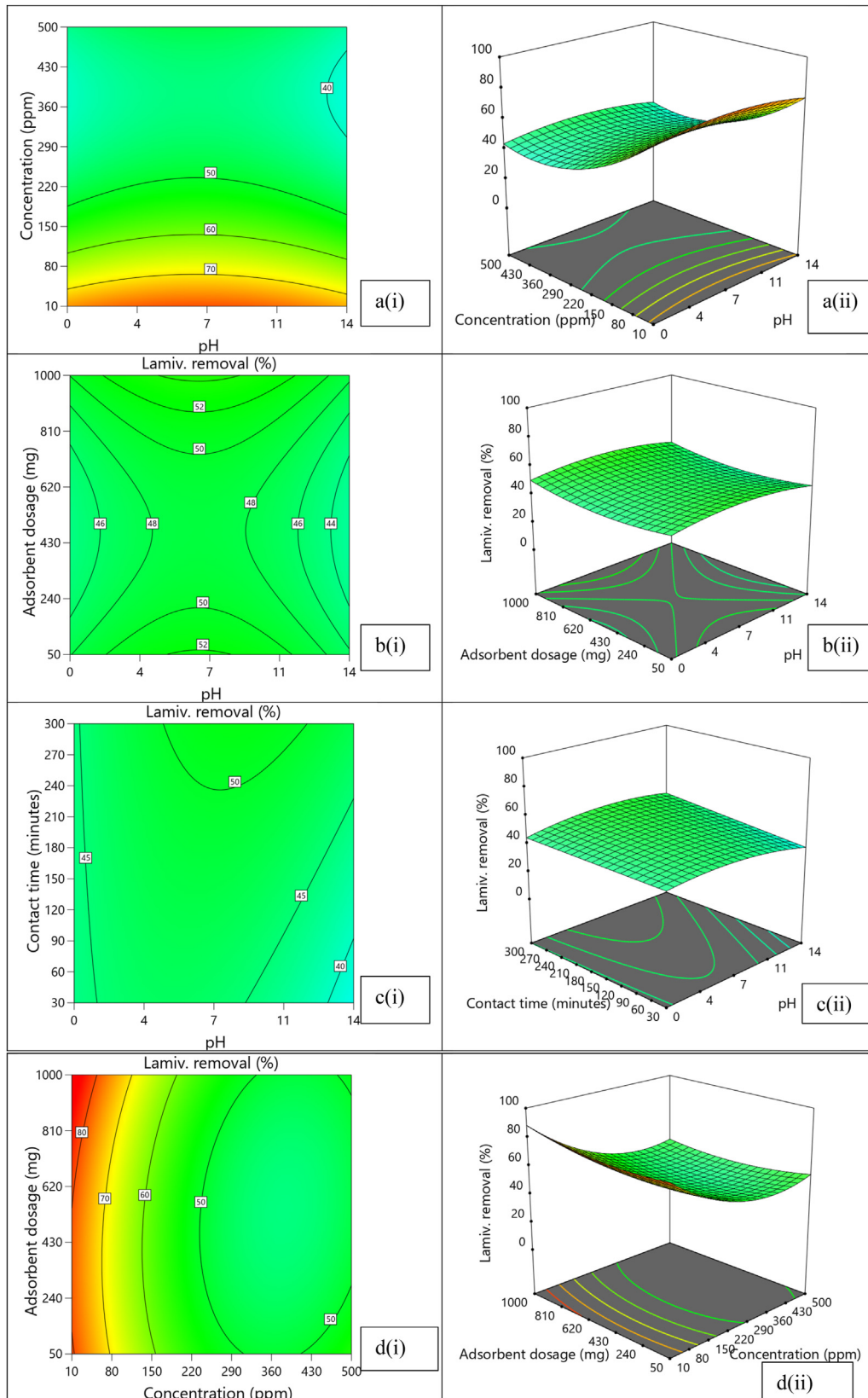


Fig. 8. Contour and 3D plots present the removal of lamivudine onto JS biochar calcined at 600 °C. Fig. 8a(i) and (ii) indicate the effect of concentration and pH, Fig. 8b(i) and (ii) indicate the effects of adsorbent dosage and pH, Fig. 8c (i) and (ii) indicate the effects of contact time and pH while Fig. 8d (i) and (ii) indicate the effects of adsorbent dosage and initial lamivudine concentration.

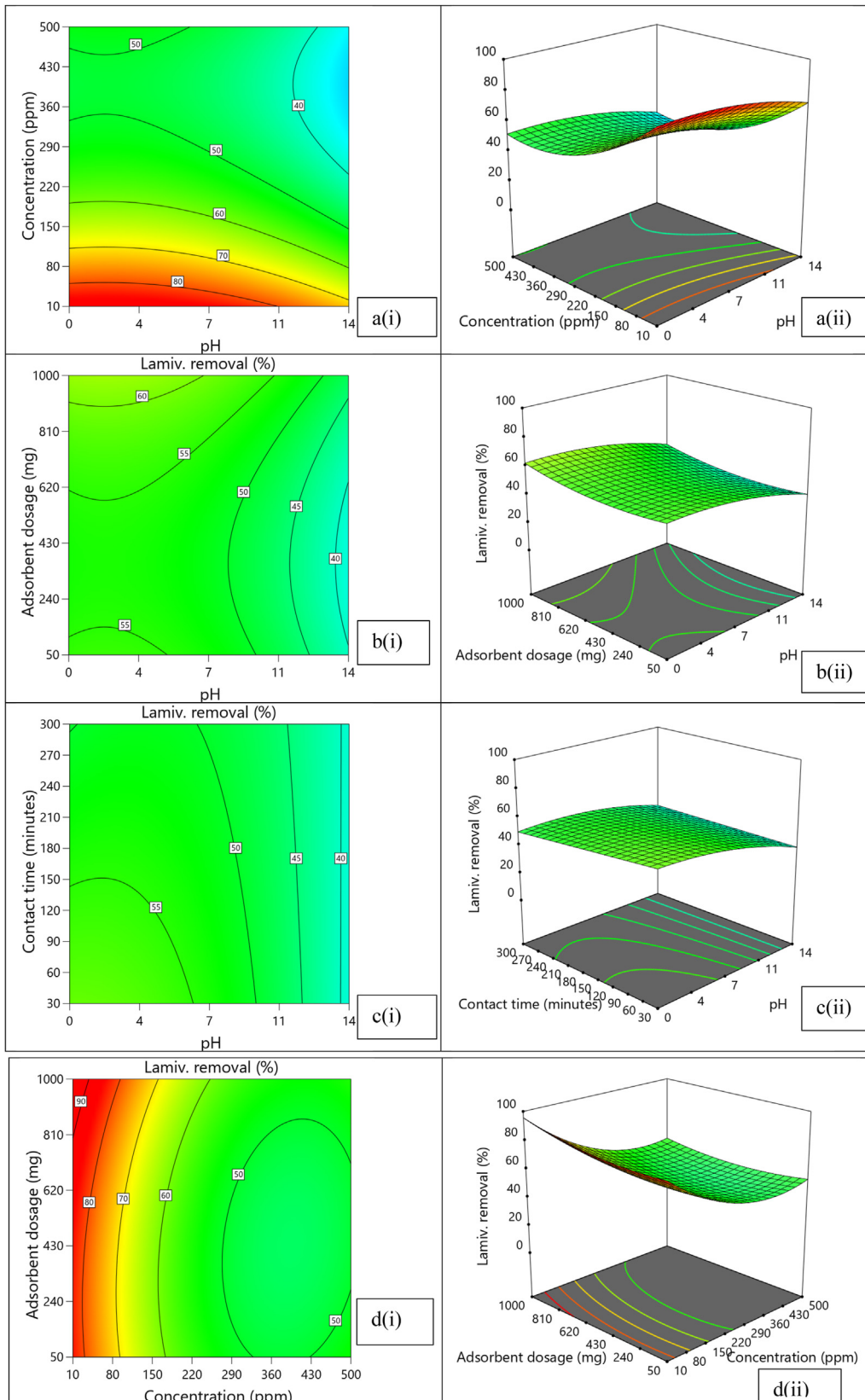


Fig. 9. Contour and 3D plots present the removal of lamivudine onto JS biochar calcined at 600 °C. Fig. 9a(i) and (ii) indicate the effect of concentration and pH, Fig. 9b(i) and (ii) indicate the effects of adsorbent dosage and pH, Fig. 9c (i) and (ii) indicate the effects of contact time and pH while Fig. 9d (i) and (ii) indicate the effects of adsorbent dosage and initial lamivudine concentration.

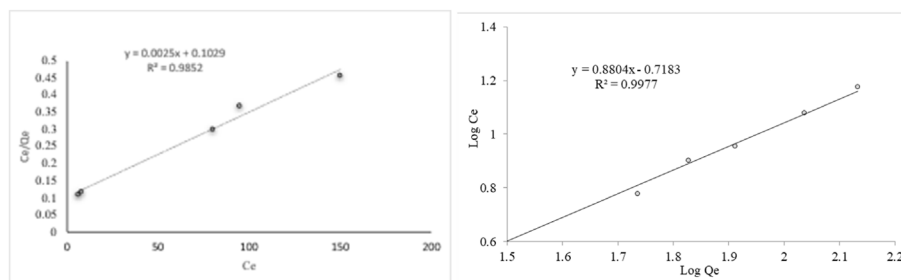


Fig. 10. Represents the isotherm models (a) modified Langmuir (b) Freundlich for the removal of lamivudine.

removal of lamivudine is more at lower concentration and increases as adsorbent dosage increases as surface area for adsorption increases.

The material calcined at 600 °C was capable of removing 70% of lamivudine from synthetic solution and the removal efficiency increases at higher adsorbent dosage to 80%, due to increase in surface area for adsorption. Fig. 8 Contour and 3D plots presenting lamivudine removal onto JS biochar calcined at 600 °C (a) effect of concentration and pH, (b) adsorbent dosage and pH (c) contact time and pH, and (d) adsorbent dosage and initial lamivudine concentration.

The results indicate that the removal of lamivudine onto JS biochar was 70% at a pH of solution increases from 5 to 11 as presented in Fig. 8. Furthermore, at initial concentration of 10 ppm JS biochar was capable of removing 70% of lamivudine from synthetic water solution. Fig. 8 (b) present the contribution of adsorbent dose on the removal of lamivudine, whereas lower adsorbent dosage was able to remove lamivudine from synthetic solution, with the removal increased as adsorbent dose increased. The results Fig. 8(c) also indicated removal of lamivudine at lower contact times but increases as contact time increases. Fig. 8(d) indicates the removal of lamivudine is more at lower concentration and increases as adsorbent dosage increases.

The material calcined at 750 °C could remove 80% of lamivudine from synthetic solution and increases to 90% at higher adsorbent concentration. Fig. 9 Contour and 3D plots presenting lamivudine removal onto JS biochar calcined at 750 °C (a (i and ii)) effect of concentration and pH, (b (i and ii)) adsorbent dosage and pH (c (i and ii)) contact time and pH, and (d (i and ii)) adsorbent dosage and initial lamivudine concentration.

The results indicated in Fig. 9 Show that the removal of lamivudine from synthetic solution onto JS biochar reached 80% from pH range of 5–11, and above which it decreased to 70% as presented in Fig. 9(a)(i) and (ii)). Furthermore, at initial concentration of 10 ppm, the removal efficiency was 90%, whereby as the concentration increased from 40 to 80 ppm the removal efficiency decreased to 70%, indicating that the JS biochar may potentially be used for remediation of lamivudine as presented by Fig. 9 (b) presents the contribution of adsorbent dosage on the removal of lamivudine, where lower adsorbent dosage was able to remove lamivudine from synthetic solution, with the removal increased as adsorbent dose increased.

The results presented in Fig. 9 (c (i and ii)) also indicate the removal of lamivudine at lower contact times but increased as contact time increased. Fig. 9 (d (i and ii)) indicates the removal of lamivudine is more at lower concentration and increased as adsorbent dosage increased, due to increase in surface area for adsorption.

As it can be observed from the current study, removal of lamivudine increased as the calcination temperatures increased due to increased surface area for adsorption. Previous researchers

reported similar trend [52,54,100–102], for adsorption of lamivudine and other organics onto biochar [103–106]. Kahilu and colleagues investigated the removal of HIV antiretroviral pollutant from wastewater using coal discards and sewage sludge derived-hydrochar [100]. Their study reported a significant removal of nevirapine by 97.19%, and lamivudine by 93.32% [100], which were relatively higher compared to the current study.

The contamination with antiviral drugs such as lamivudine have raised environmental concerns on the safety and sustainability of aquatics and entire ecology. These drugs are reported at relatively lower concentrations, but they are toxic to the ecology. They may potentially lead to the development of antimicrobial-resistant microbes, evolution of resistant genes and their dissemination, which threatens ecosystem health. Studies focusing on the removal of emerging contaminants such as antiviral are increasing to ensure safety and sustainability of ecology.

Tu and colleagues reported a significant removal of antiviral drug famciclovir, and that the adsorption process was a spontaneous process, whereas the hydrogen bonding, π - π interaction and C-H... π interaction enhanced the adsorption [107]. Similarly, the potential of biochar made from various biosorbent for the remediation of emerging contaminants such as lamivudine have been investigated, yet more data are needed to ensure public health awareness and safety [49,100,107–113].

The present study utilized reference surface methodology optimization for adsorption of Lamivudine onto JS biochar from synthetic solution, indicating potential future use, improvements, and adaptation for environmental remediation. The need for environmental remediation is prevalent for environmental safety and sustainability of ecosystems. Chen and Colleagues revealed that wastewater treatment and emerging contaminants received much attention in their bibliometric analysis conducted from 1998 to 2021 [61]. The study further proposed devotion into research and development of new green and environmentally friendly techniques for detecting and treating emerging contaminants in wastewater [61]. These techniques may potentially be utilized for remediation of the entire ecosystems once modified to meet the current demand.

The need for inclusion of Artificial Intelligence (AI) in technologies to solve problems was proposed by Zhang and colleagues [114]. Their study further reported that AI related technologies can effectively solve issues related to integrating renewable energy and power systems [114]. The significant use of AI related technologies can be applied to other settings such as environmental remediation. The AI related technologies faces some challenges, but apart from the challenges of AI related technologies and modals it can be included in various settings while continuously optimized [114].

3.7. Isotherm studies

Adsorption isotherms provide properties and equilibrium data

Table 5

The general representation of the graphs and calculations results.

Langmuir and Freundlich adsorption isotherm							
Temp(°C)	Langmuir Isotherm				Freundlich Isotherm		
	Q _e (mg/g)	Q _{max} (mg/g)	b(L/mg)	R ²	n	K _f	R ²
303	256.32	400	0.002429	0.9852	1.1	2.05	0.9977

that describe the quality of lamivudine adsorption onto JS biochar and are the main contributor to the optimization of adsorbent consumption. Establishing a proper relationship for the equilibrium curve and optimizing the adsorption system design for removing contaminants such as lamivudine [96,115,116]. The current study utilized modified Langmuir and Freundlich isotherm models presented in Fig. 10.

The isotherm data for removing ciprofloxacin onto JS biochar followed Freundlich isotherm. The R^2 value for the adsorption of lamivudine onto JS biochar for Freundlich isotherm was 0.9977, while for the modified Langmuir model was 0.9852, indicating better agreement with Freundlich isotherm. Previous studies that evaluated the adsorption of organics such as lamivudine onto biochar indicated similar results [96,116]. The equilibrium data fitted the Freundlich model better than the Langmuir model, indicating surface heterogeneity of the adsorbent and multilayer adsorption. From Langmuir isotherm, the adsorption capacity was calculated to be 400 mg g⁻¹, indicating that the adsorbent had a significantly high capacity to remove lamivudine from the synthetic solution.

Bagher reported the maximum adsorption capacity of moringa seed powder biochar to be 100 mg g⁻¹ [117]. Likewise, Correa-Navarro and colleagues reported an adsorption capacity of 40.2 mg g⁻¹ and 5.40 mg g⁻¹ for the removal of caffeine and diclofenac onto bagasse biochar, respectively [118]. Also, Jung and colleagues, and other previous researchers reported higher adsorption capacities that are potential for remediation of these contaminants from water and wastewater effluents [119–124]. Table 5 represents the general representation of the graphs and calculations results.

The results of this study provided an RL value between 0 and 1, indicating that the adsorption of lamivudine onto JS biochar is favorable. The adsorption capacity reported in the current study supports its use for the remediation of pharmaceutical contaminants from water and wastewater effluents as similarly reported by a previous study [125]. The K_f value presented in Table 5 indicates that the adsorbent is heterogeneous which varies with the heterogeneity of the adsorbent. The fractional value of 1/n was within (0 < 1/n < 1) indicating that the adsorbent's surface is heterogeneous and hence multilayer adsorption as similarly presented by previous scholars [126–129].

4. Conclusions and the outlook

Recently, on a global scale, there have been increasing reports of diseases both communicable and noncommunicable diseases associated with the use and environmental exposure to active chemicals, their metabolites or transformation products. At very low concentrations, active chemicals can lead to the development and propagation of antimicrobial resistant pathogens and resistant genes in the environment, which in turn accelerates the ecological exposure risks. The result of the current study indicated that JS biochar could remove up to 90% of lamivudine from synthetic solution, indicating a potential future use for environmental remediation of these chemicals. The removal efficiency increased at lower lamivudine concentration and as the adsorbent dose

increased. The materials calcined at 750 °C showed relatively higher lamivudine removal compared to those calcined at 400 °C, 500 °C and 600 °C. It was further observed that JS biochar was able to remove lamivudine at relatively lower concentrations. The results of the current study show that the JS adsorbents may be improved and potentially used for remediation of organics including lamivudine.

Future studies may need to focus on the improvement of the applied adsorbents using various additives and preparing composites that may be used for the removal of lamivudine and other organic contaminants. Also, the use of JS biochar for remediation of organics such as lamivudine in real wastewater effluents is recommended to provide data on the adsorptive behavior of JS biochar from complex matrices.

Consent statement

Not Applicable

Ethical statement

Not Applicable.

Data availability statement

The data generated is available on Mendeley data and cited in this manuscript.

Declaration of competing interest

None.

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