

Optimizing ciprofloxacin removal from water using jamun seed (*Syzygium cumini*) biochar: A sustainable approach for ecological protection

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ABSTRACT

Scientific interest in antimicrobial pollutants, such as ciprofloxacin, has increased. Due to spread of antibiotic-resistant bacteria, resistance genes, and their dissemination to the environment. Therefore, their remediation is necessary to ensure ecological sustainability. The current study aimed to optimise the removal of ciprofloxacin from synthetic water using jamun seed (JS) (*Syzygium cumini*) biochar using a response surface methodology (RSM). Result indicates ciprofloxacin elimination efficiency ranged between 32.46 and 94.95%, indicating the material can be improved and used for remediation of organics. The residual standard error of 4.4% were found for the predicted model, implying that the model is credible and can be used to predict future experimental findings. The R-squared value for the improved Langmuir model's R^2 is 0.9681 which is in close agreement with the Freundlich isotherm, R^2 0.9757. Therefore, JS biochar could be utilized for the remediation of ciprofloxacin from contaminated water and wastewater for ecological safety and sustainability.

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1. Introduction

Antimicrobial pollution refers to the contamination of the environment, particularly water bodies with antimicrobial substances, such as antibiotics, antivirals, and antifungals (Tian et al., 2021; Fernandez et al., 2021). Antimicrobial drugs (Zairina et al., 2023; Da Silva et al., 2023), a class of medications used to combat a wide range of microorganisms, including bacteria, viruses, fungi, and other parasites. These drugs include antibiotics such as ciprofloxacin, penicillin and erythromycin for bacterial infections (Pérez-Legaspi and Rico-Martínez, 2023; Rugumisa et al., 2016), antivirals such as lamivudine for viral infections (Huang et al., 2021), antifungals such as fluconazole for fungal infections (Lu et al., 2021), and antiparasitic drugs such as chloroquine for malaria (Meo et al., 2020). Antimicrobials are crucial in the treatment

and prevention of various infectious diseases (Pérez-Legaspi and Rico-Martínez, 2023; Rugumisa et al., 2016). Fluoroquinolone drug ciprofloxacin is utilized to treat bacterial illnesses like pneumonia and urinary tract infections (Thai et al., 2021), joint infections, prostatitis, typhoid, gastrointestinal infections, lower respiratory tract infections, and sexually transmitted infections (gonorrhoea and chancroid) (Thai et al., 2021; Martenson et al., 2022; Okoye et al., 2022). Once ciprofloxacin in the body undergoes biotransformation via metabolic processes to produce metabolites and when released in the environment through chemical and physical processes undergoes transformation. Some of its selected transformation biproducts are presented in Fig. 1, with which each formed biproduct can have enhanced or lower environmental impacts, thus calling attention.

The environmental concerns regarding the possible negative consequences of antimicrobial use and disposal on human and environmental health have developed over the past decades (André Pereira et al., 2020; Ripanda et al., 2022a; Ripanda et al., 2022; Miraji et al., 2021). Data indicates wastewater treatment in developing countries lacks or has inadequate design for removing antimicrobial contaminants (Miraji et al., 2021; Makokola, et al., 2019; Ripanda et al., 2022b; Ripanda et al., 2022; Ripanda et al., 2022b; Necibi et al., 2021; Li et al., 2020; Kumari and Kumar, 2020; Olabode et al., 2019; Miraji et al., 2023a; Hossein, 2019; Ripanda et al., 2023a; Ripanda et al., 2023; Hossein et al., 2023; Ripanda and Miraji, 2022; Ripanda et al., 2023b; Hossein et al., 2023; Hossein et al., 2022; Miraji et al., 2023b). As a result, antibiotics such as ciprofloxacin, its metabolites and transformation products have been reported to contaminate surface waters, groundwater, and aquatics (Olabode et al., 2019; Hossein et al., 2023; Ripanda et al., 2023b; Zhang et al., 2022a; Zhang et al., 2022b; Yang et al., 2022a; Yang et al., 2022b; Wang et al., 2022; Verinda et al., 2022; Surinaidu et al., 2023). These contaminants could impair ecosystems' sustainability and potentially affect human and ecological health. A study by Martins et al., revealed that ciprofloxacin was toxic to freshwater species tested, however the toxicity levels tested was higher than the environmental amounts (Martins et al., 2012; Ramesh et al., 2021). The toxicity of a drug such as ciprofloxacin goes beyond a limit, because of drug-drug interactions, chronic exposure, combined concentration, a delayed effect due to bioaccumulation, bioconcentration, and biomagnification through the food chain and development and dissemination of drug-resistant microbes, and resistant genes to the ecosystems,

threatening public health. Antibiotics and Antibiotic-resistant genes (ARGs) are contaminants of emerging concern, and routine environmental analysis and monitoring guidelines do not consider these contaminants (Ripanda et al., 2022b; Miraji et al., 2023a; Ripanda et al., 2023a; Ripanda et al., 2023; Hossein et al., 2023; Makaye et al., 2022; Dulio et al., 2018). This necessitates further research to generate data required for legislative changes. Similarly, the need for remediation of these contaminants is inevitable for ecological safety.

Research for innovative ideas is going on to improve or modify the treatment schemes to include the remediation of contaminants such as an antibiotic (Zhang et al., 2022a; Zhang et al., 2022b; Yang et al., 2022a; Yang et al., 2022b; Wang et al., 2022; Verinda et al., 2022; Hosseina et al., 2023; Egbedina et al., 2021; Huang et al., 2020; Iqbal et al., 2022; Iqbal et al., 2021). Several techniques have been utilized for the environmental remediation of antibiotics, including physical, chemical, biological, and their combination (Miraji et al., 2021; Hossein et al., 2023; Sodhi and Singh, 2022; Chaturvedi et al., 2021; Jain et al., 2013; Shenbagavalli and Mahimairaja, 2012; Salari et al., 2019; Pundir et al., 2018; Späth et al., 2021; Andersson, 2017). The potential of biosorbents such as biomass-derived biochar on remediation of emerging pollutants such as antimicrobial has been reported in literature (Palapa et al., 2020; Togue, 2019; Aziz et al., 2018; Shakoore and Nasar, A, 2017; Shahrin et al., 2022; Velusamy et al., 2021; Mao et al., 2019; Ashiq et al., 2019; Li et al., 2018). Biochar is a carbon-rich material that is produced through pyrolysis, which involves heating biomass in the absence of oxygen. Adsorption using biochar derived from seed biomass as adsorbent has shown a promising elimination efficiency for antimicrobials such as ciprofloxacin in water and wastewater effluents. The seeds-based biomass is readily available, produces high biochar yield, do not produce toxic metabolite, it requires a simple design, and has low initial cost for implementation (Miraji et al., 2023b; Sophia and Lima, 2018; Kumar and Bhattacharya, 2020; Moto et al., 2024). Results of previous researchers on other biomass-based adsorbents indicate that adsorbent made of lotus stalk was capable to remediate 83% of norfloxacin, macadamia nutshell lignin managed to remove 70 to 100% of tetracycline, peanut husk removed >70% of tetracycline and amoxicillin, rice husk reached a maximum of 68% upon activation (Li et al., 2019; Martins et al., 2015; Varela et al., 2023; Daffalla et al., 2020). A recent studies by Ripanda et al. indicated that JS biochar calcined at 750C, at pH 8, with an initial lamivudine concentration of 10 ppm and contact time of 30 min indicated a maximum experimental

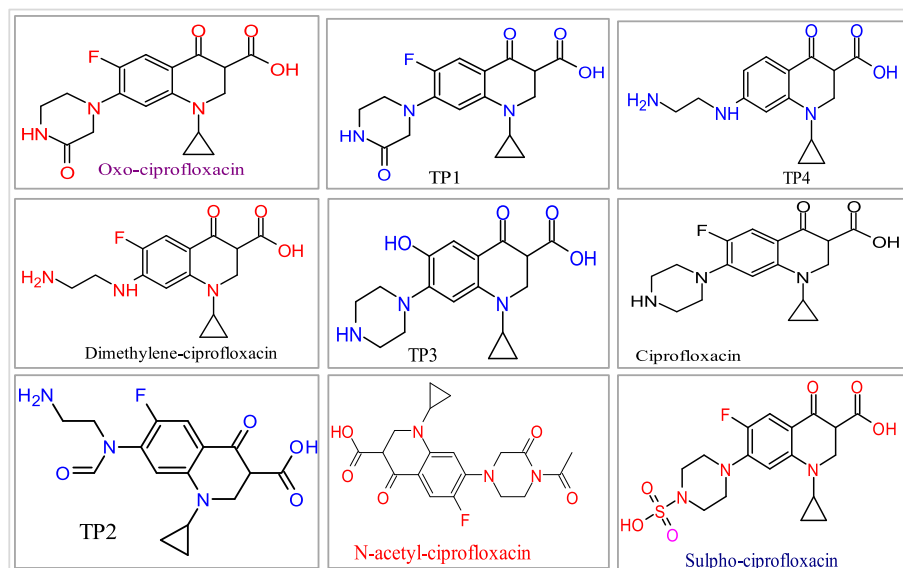


Fig. 1. Structures of ciprofloxacin and its selected metabolite and transformational products that may potentially impair ecological health.

removal efficiency of 84.9% (Ripanda et al., 2023; Ripanda, 2023). The result indicates the potential of JS biochar adsorbent for remediation of other emerging contaminants. Jamun seed adsorbent is rarely utilized for removal of antimicrobials and other organic contaminants (Ripanda et al., 2023; Ripanda, 2023), and there is limited information on the use of JS biochar to remove antimicrobial. The present study aimed to investigate the remediation of ciprofloxacin from the synthetic water using biochar prepared from the jamun seed (*Syzygium cumini*) biomass as an adsorbent and its optimization through RSM.

2. Materials and methods

2.1. Chemicals and equipment

All the chemicals and reagents including methanol, distilled water, hydrochloric acid, sodium hydroxide, and ciprofloxacin were analytical grade.

2.2. Design of experiments and statistical analysis

Response surface methodology is an empirical modelling method for determining the interaction of multiple operating and response variables. It provides a systematic experimentation strategy for building and optimizing an empirical model. In essence, RSM is a combination of mathematical and statistical approaches suitable for modelling and analyzing problems in which the output is affected by input variables and their interactions (Anderson and Whitcomb, 2016; Feroso et al., 2010; Bakari et al., 2020). Furthermore, the RSM reduces the number of experiments, costs, and time spent on physical experiments while providing adequate data for statistically acceptable conclusions (Bakari et al., 2021). In the current study, an RSM based on the optimality design was used to optimise five independent and one response variable. Independent variables studied are adsorbent dosage (50–1000 mg), calcination temperature (400, 500, 600 and 750 °C), residence time (30–300 min), pH (1–14), and pollutant concentration (10–500 ppm), while the observed response was the removal efficiency (%) of ciprofloxacin. This study utilized higher levels of ciprofloxacin concentration than expected levels to be found in environment to get a comprehensive understanding of the effectiveness, limitations, and applicability of JS biochar. The studied variables were selected based on the data available in the literature (Späth et al., 2021; Andersson, 2017; Dou et al., 2022). The D-optimality RSM comprises 55 experimental runs, of which 45 are model points, five are replicate points, and five are lack-of-fit points. The RSM involves five steps: the development of statistically designed experiments, followed by generating an empirical model, statistical analysis of the model, numerical optimisation by using the desirability function and finally, model confirmation. The experimental run was randomized to minimize the error and effect of uncontrolled factors (Salari et al., 2019). The observed responses were used to generate an empirical model conforming to the experimental variables. Experimental results from the 55 runs were used to determine the regression coefficient of the quadratic model using Design-Expert Version 13.0.5 software (Stat-Ease, Inc., Minneapolis, USA).

The coefficient of R-squared established the accuracy of the fitted model, and the significant model terms were evaluated by the probability value (*P*-value) at a 95% confidence level. The contour 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 provides valuable information about the system's behaviour on the variation of the processing parameter within the design space. In this study, A represent pH, B is pollutant concentration, C is adsorbent ratio, and D is contact time. Experimental design conditions and predicted, and actual ciprofloxacin removal efficiency are presented in Table 1.

Table 1

Experimental design conditions and predicted, and actual ciprofloxacin removal efficiency.

Run	Independent variables					Dependent variables	
	A (–)	B (mg/L)	C(g/L)	D (Minutes)	E (°C)	Experimental	Predicted
1	0	10	50	212	250	85.05	87.59
2	0	10	1000	300	300	93.16	92.95
3	0	10	1000	30	500	94.14	92.27
4	0	500	50	165	0	32.7	33.26
5	14	500	1000	30	600	62.27	62.37
6	14	10	50	30	400	92.54	92.27
7	4	255	216	30	0	68.26	69.82
8	14	355	1000	30	750	50.65	47.73
9	14	10	1000	300	500	90.4	92.28
10	0	500	1000	30	400	32.58	32.46
11	7	245	507	158	400	71.16	71.92
12	14	500	1000	300	300	49.15	49.26
13	14	500	1000	30	600	62.27	62.26
14	5	10	50	300	0	79.64	79.22
15	14	10	50	300	300	91.81	92.24
16	3	248	1000	300	600	64.65	64.4
17	14	500	50	30	300	44.82	44.61
18	12	304	50	166	0	52.53	50.35
19	14	10	50	30	500	91.48	92.27
20	0	500	50	300	400	32.52	32.46
21	0	500	744	300	250	36.63	36.37
22	14	500	1000	118	250	40.61	43.15
23	0	10	1000	300	400	93.68	93.56
24	0	10	1000	30	0	93.12	92.26
25	14	500	1000	300	400	32.56	32.5
26	14	500	50	300	750	34.4	35.98
27	14	10	1000	300	250	94.01	92.27
28	14	10	1000	300	750	92.47	93.32
29	0	500	1000	30	300	32.34	32.77
30	14	10	1000	300	750	92.47	92.27
31	0	500	50	300	300	32.88	32.55
32	4	257	810	38	750	67.5	72
33	0	500	50	30	250	34.28	32.54
34	0	10	1000	30	250	92.83	92.29
35	13	110	50	30	600	78.67	78.45
36	0	152	50	300	750	68.45	66.84
37	14	10	311	30	250	93.54	93.28
38	0	10	649	30	600	91.1	91.23
39	0	125	582	207	500	87.98	88.56
40	0	500	50	300	400	32.52	32.46
41	0	500	50	300	500	33.2	34.48
42	14	255	1000	300	0	64.13	65.37
43	14	10	50	30	0	82.98	83.73
44	14	378	558	221	500	57.87	52.87
45	9	500	1000	30	0	37.22	36.56
46	0	500	50	300	600	35.81	35.74
47	0	10	50	30	300	94.84	94.95
48	0	500	50	300	600	35.81	35.96
49	14	500	1000	30	500	33.64	35.98
50	8	10	50	30	750	92.16	91.29
51	14	500	50	300	250	33.31	32.77
52	14	500	1000	300	400	32.56	32.5
53	14	10	1000	30	300	85.26	84.93
54	0	500	1000	184	750	34.02	32.69
55	14	10	538	300	600	83.56	83.73

2.3. Adsorption experiments

The batch equilibrium method was used to investigate the adsorptive elimination of ciprofloxacin from the synthetic water. By dissolving 1 g of ciprofloxacin in deionized water, a stock solution with 1000 mg/L of the antibiotic was created. The recovery experiment indicated 97%, which is acceptable range as reported by Muchakayala et al., (Muchakayala et al., 2022). All the adsorption systems were continuously stirred at 220 rpm for a given a contact time. Following this time frame, samples of the adsorption system solutions were taken. The ciprofloxacin and JS biochar-containing sampling solutions were centrifuged at 500 rpm for 15 min before being filtered through a 0.45 μm syringe filter. The concentration of ciprofloxacin in the grab sample

was then determined spectrophotometrically using a UV–Vis instrument, following recommendations from previous studies (Naveed and Waheed, 2014). The amount of ciprofloxacin uptake at equilibrium (q_t , mg/g) and elimination efficiency (R %) was calculated using eqs. (1) and (2), respectively.

$$\text{Adsorption capacity } q_t = \frac{(C_0 - C_t)V}{m} \quad (1)$$

$$\text{Elimination efficiency } R = \frac{(C_0 - C_t)}{C_0} * 100 \quad (2)$$

Where

C_0 and C_t = concentrations of ciprofloxacin (mg/L) at the beginning and at time t , V = volume of the solution and, m = mass of the adsorbent.

These calculations were done according to procedures reported by Wakejo et al. (Wakejo et al., 2022), and other researchers (Iqbal et al., 2021; Shenbagavalli and Mahimairaja, 2012; Späth et al., 2021; Andersson, 2017; Velusamy et al., 2021; Mao et al., 2019; Ashiq et al., 2019; Li et al., 2018; Anderson and Whitcomb, 2016; Wakejo et al., 2022; Zheng et al., 2021; Carabineiro et al., 2011; Qin et al., 2022; Bezerra et al., 2008). The Langmuir isotherm, given in the linear form by Eq. (3), is valid for monolayer sorption caused by a surface with a limited number of identical sites.

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

Where

C_e = equilibrium ciprofloxacin concentration (mg/L) and, Q_e = amount adsorbed at equilibrium (mg/g).

The Langmuir constants Q_0 (mg/g) and b (L/mg) relate the heat of adsorption and the monolayer adsorption capacity, respectively. The key component of the Langmuir adsorption is represented by the dimensionless constant R_L , also known as the separation factor or equilibrium parameter, which can be used to forecast whether an adsorption system will be advantageous or disadvantageous. The R_L value is calculated using Eq. (4).

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

Adsorption is advantageous if the R_L values fall between 0 and 1. Eq. (5) illustrates a non-linear way in which this model can be expressed.

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

Eq. (6) illustrates the linear version of the Freundlich isotherm.

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

Where

K_f = adsorption capacity and, n = empirical parameter related to the intensity of adsorption.

The favourability of adsorption increases with increasing $1/n$ values. The higher fractional value of $1/n$ ($0 < 1/n < 1$) signifies that the adsorbent's surface is heterogeneous.

3. Results and discussions

Utilizing biochar derived from biomass presents a novel method for tackling antimicrobial pollution such as ciprofloxacin and other emerging contaminants, capitalizing on biochar's adsorptive characteristics to eliminate the contaminants from polluted settings. Further, integrating RSM into the process optimization can amplify remediation efficiency

by pinpointing the ideal parameters for maximizing pollutant such as ciprofloxacin adsorption onto the JS biochar. The results of RSM model indicates B, C, D, E, AB, AD, AE, BE, CD, CE, DE, C^2 are significant model terms and their interactions. Further, ciprofloxacin elimination efficiency ranged between 32.46 and 94.95%, indicating that the material can be improved and used for remediation of ciprofloxacin and other organics. More, the experimental remediation of ciprofloxacin onto JS biochar adsorbent at pH of 7 reached 71.16% and at pH of 5 reached 79.64 indicating agreement with previous studies. These results indicate that jamun seed biochar like other seed biomass-based adsorbents can be used for adsorptive remediation of contaminants for healthier and sustainable ecology.

3.1. Adsorption of ciprofloxacin onto JS biochar

Previous study by Ripanda et al. (Ripanda et al., 2023a), indicated potential properties of JS biochar, including presence of functional groups, porous structures, high surface area, and higher content of carbon. Functional groups on the biochar surface, such as hydroxyl (-OH), carboxyl (-COOH), and aromatic groups, can interact with ciprofloxacin molecules through electrostatic interactions, hydrogen bonding, ion exchange, and hydrophobic interactions. Fig. 2 presents the FTIR spectra of JS biochar calcined at 400 °C, 500 °C, 600 °C, and 750 °C, showing the available functional groups potential for adsorption. The presence of porous structures may contribute to the removal of ciprofloxacin through pore-filling mechanism. These properties are potential for adsorptive remediation of pollutants such as ciprofloxacin.

According to earlier studies (Aziz et al., 2018; Shakoore and Nasar, 2017; Shahrin et al., 2022; Dou et al., 2022; Li et al., 2022; Dritsa et al., 2009; Yu et al., 2023), the removal efficiency of JS biochar was found to rise when the calcination temperature rose from 400 to 600 °C. Similarly, the sorption properties of these materials were relatively similar (Ojeda-López et al., 2021), as reported in the current study. The use of JS biochar for remediation of antimicrobials such as ciprofloxacin is economical and may help create a sustainable future. Previous studies indicated that biomass-based biochar demonstrated excellent reusability, with the ability to be reused for up to four cycles while retaining 50–61% of its original properties after regeneration (Singh et al., 2022; Li et al., 2022). This implies that biochars, which are a type of charcoal produced from biomass, can effectively retain their functionality over

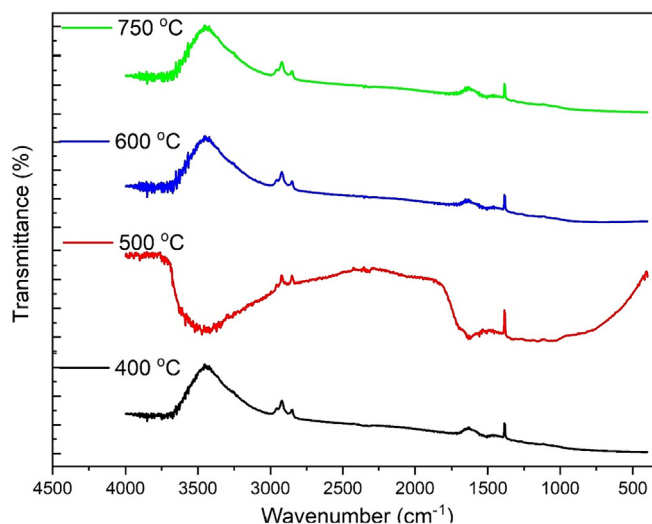


Fig. 2. FTIR spectra of JS biochar calcined at 400 °C, 500 °C, 600 °C, and 750 °C, showing available functional groups potential for adsorption.

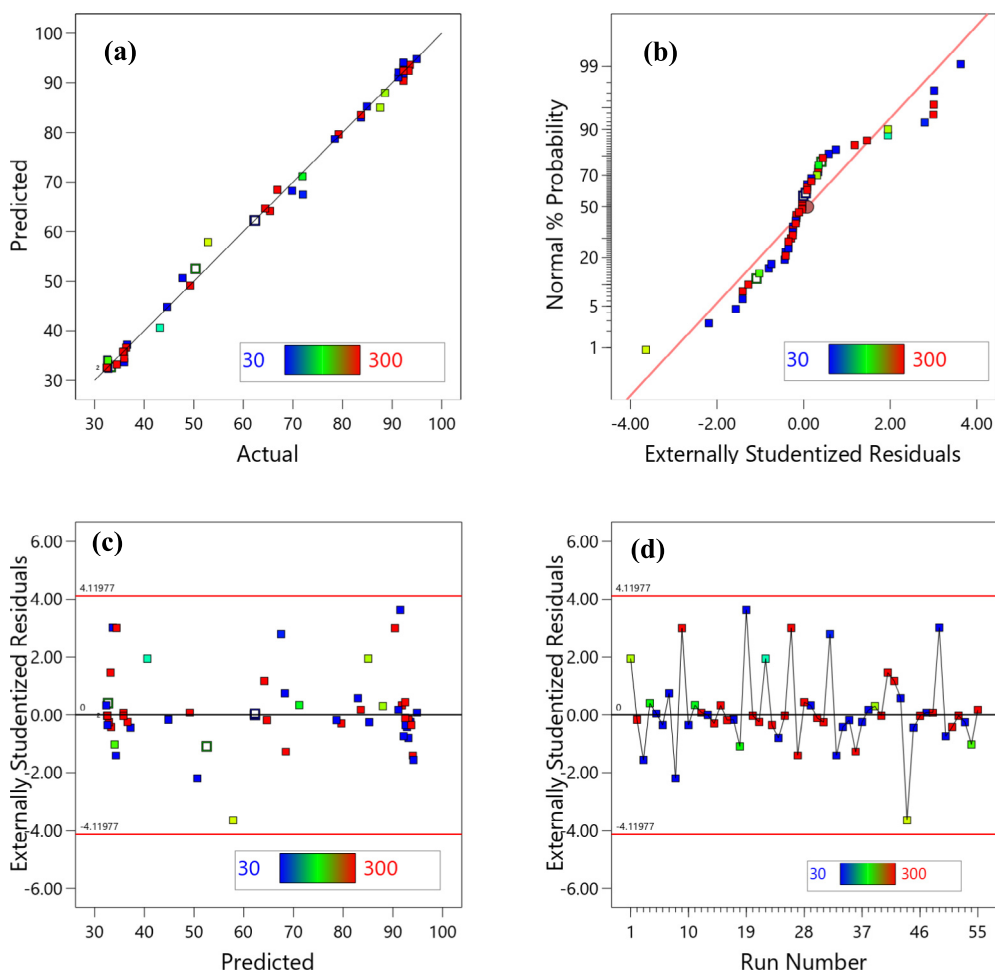
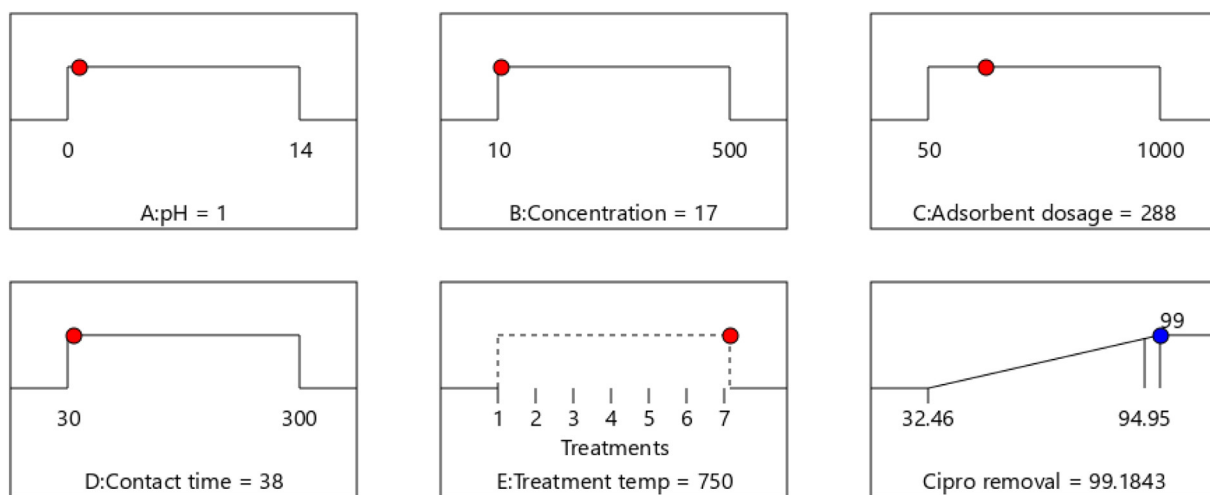


Fig. 3. Model diagnostic plots (a) actual vs predicted (b) normal probability (c) externally studentized residuals vs predicted, and (d) externally studentized residuals vs run number.



Desirability = 1.000
 Solution 1 out of 100

Fig. 4. Perturbation plots employing adsorbents that were calcined at various temperatures that demonstrate the sensitivity of variables on the removal of ciprofloxacin. A is pH, B is pollutant concentration, C is adsorbent ratio, and D is contact time.

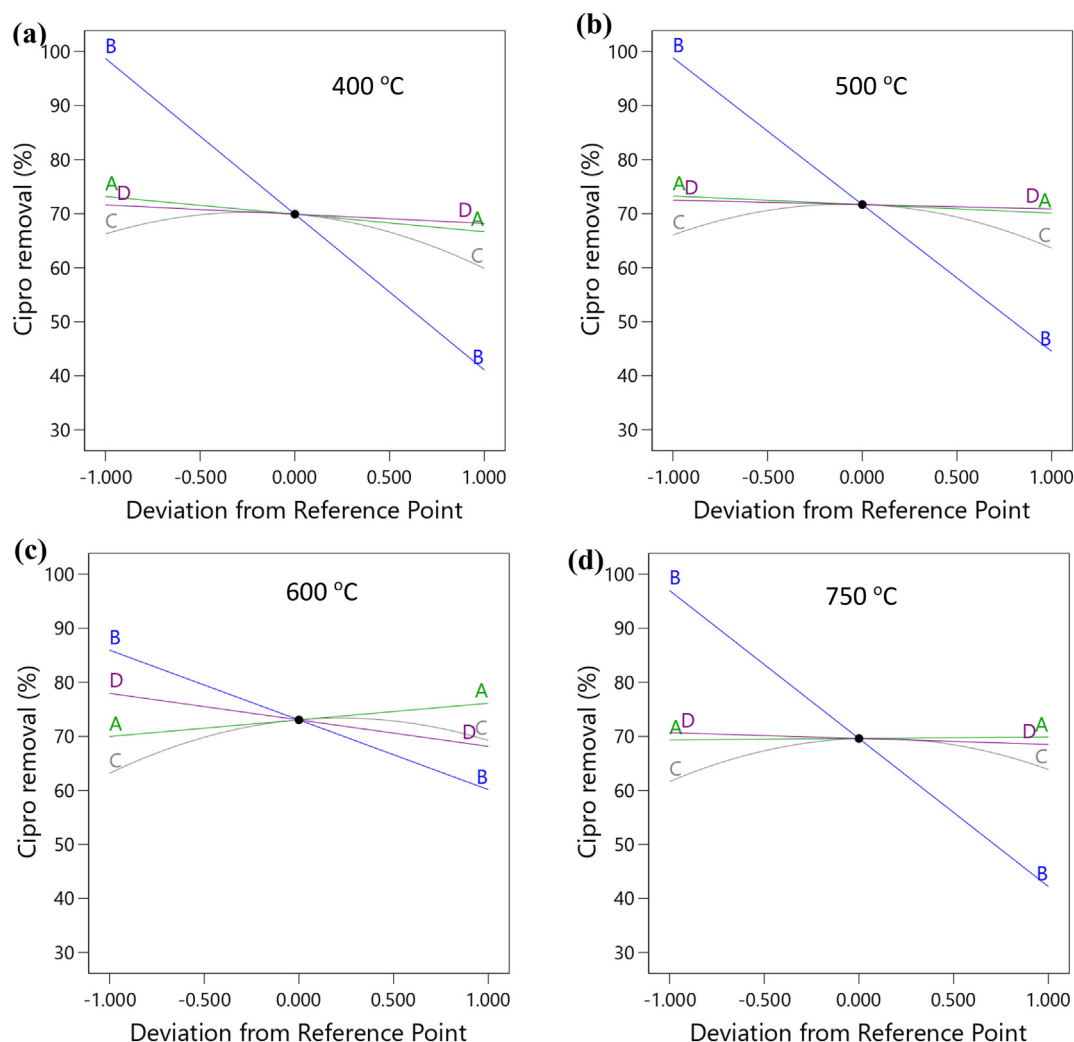


Fig. 5. Contour and 3D plots showing the effect of parameters on the removal of ciprofloxacin from JS biochar calcined at 400 °C. (a) and (b) effect of concentration and pH, (c) and (d) effects of adsorbent dosage and pH, (e) and (f) effects of contact time and pH, and (g) and (h) the effects of adsorbent dosage and initial ciprofloxacin concentration.

multiple cycles of use, making them a potentially sustainable and cost-effective option in various applications such as soil remediation, water filtration, and as a carbon sequestration method.

The results of model fit and statistical analysis was presented in previous work by Ripanda et al. (Ripanda et al., 2023a), the Design-Expert software recommended a quadratic model to fit the obtained findings to a general model. The significance of the model terms was assessed, and if there were a considerable number of inconsequential terms (excluding those needed to support the hierarchy), the model reduction might be used to enhance the model in use, according to other researchers (Ramírez-Malule et al., 2020; Kumar et al., 2022). After trivial terms ($p > 0.1$) were eliminated using the Design-Expert software's p -values backward elimination procedure, the final quadratic equation in terms of the coded variable provided in Eq. (8) was obtained. Therefore, Eq. (8) can be used to describe the ciprofloxacin elimination behaviour, while the negative and positive signs denote the components' respective antagonistic and synergistic effects. Model diagnostic graphs used to determine whether the proposed models are valid are shown in Fig. 3.

The plots for actual against predicted show that the data are well distributed along the line of best fit suggesting that the predicted values are in good agreement with the experimentally obtained values. The residuals are commonly distributed along within the bounds, as seen by the

externally studentized residuals plot, suggesting that errors are distributed normally. The disparity between experimental results and values anticipated by the model is known as the residuals. Additionally, the normal probability plot demonstrates that the model is not anomalous.

$$\begin{aligned} \text{Ciprofloxacin elimination\%} = & 97.403 - 0.5526A - 0.120798B \\ & + 0.0271615C - 0.0416934D \\ & + 0.00129052AB + 0.0015913AD \\ & + 4.28653 \times 10^{-5}CD - 3.03637 \\ & \times 10^{-5}C^2 \end{aligned} \tag{8}$$

3.2. Analysis of variance

The outcomes of the ANOVA analysis for the simplified quadratic model, the model's F -value of 130.43 indicates that it is significant, and noise has a 0.01% probability of causing an F -value this large. The adsorption of ciprofloxacin onto JS biochar may be explained by model terms that have p -values < 0.0500 , which also indicates that they are significant. The fact that the model's R -squared was nearly one ($R^2 = 0.9968$) indicated that the data suited the chosen model well. The rectified R^2 of 0.9891 and the predicted R^2 of 0.8023 are reasonably in agreement, with the difference being < 0.2 . A ratio > 4 is

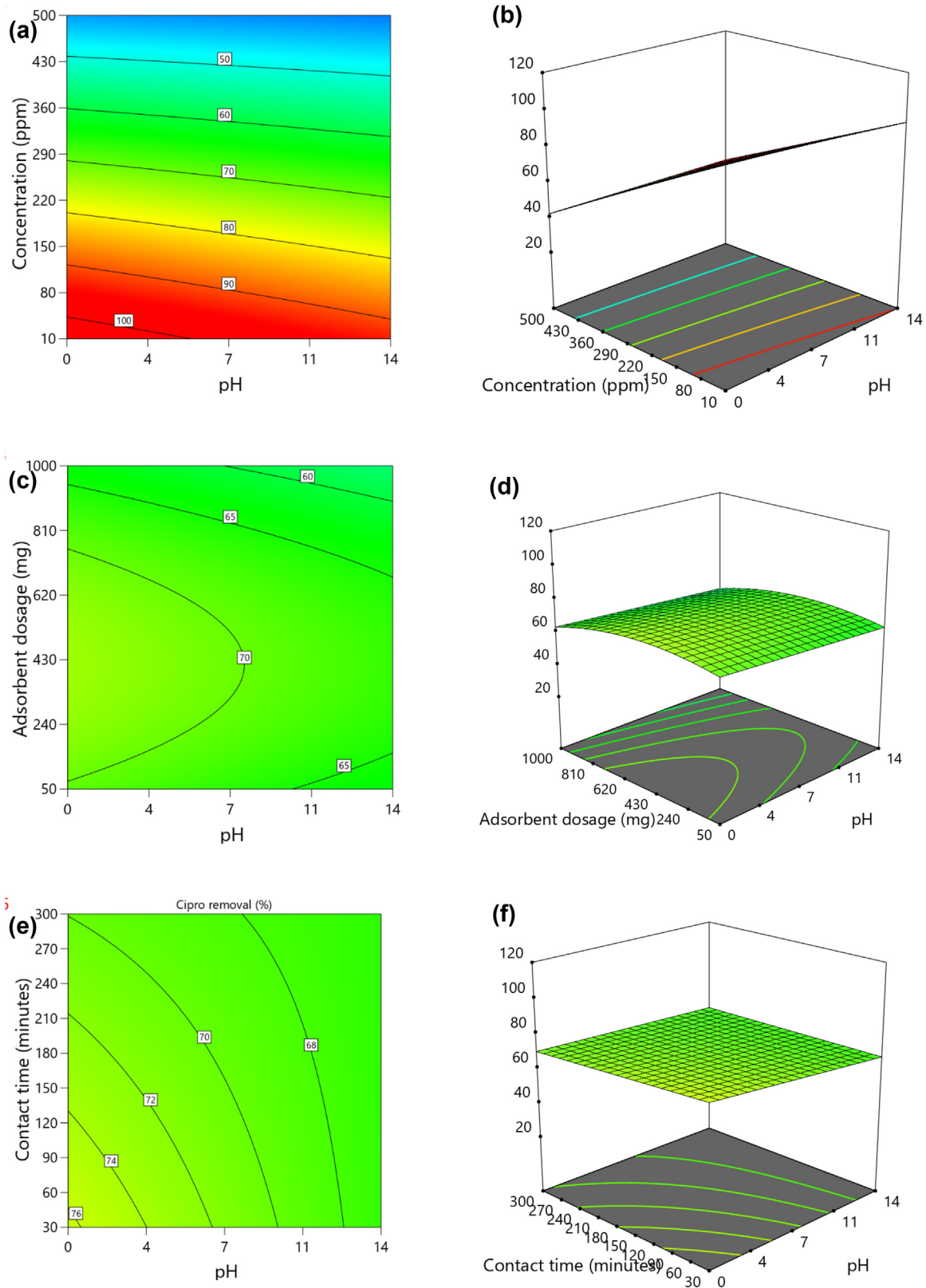


Fig. 6. Contour and 3D plots for the influence of parameters on the removal of ciprofloxacin from JS biochar calcined at 500 °C. (a) and (b) effect of concentration and pH, (c) and (d) the effects of adsorbent dosage and pH, (e) and (f) the effects of contact time and pH, and (g) and (h) the effects of adsorbent dosage and initial ciprofloxacin concentration.

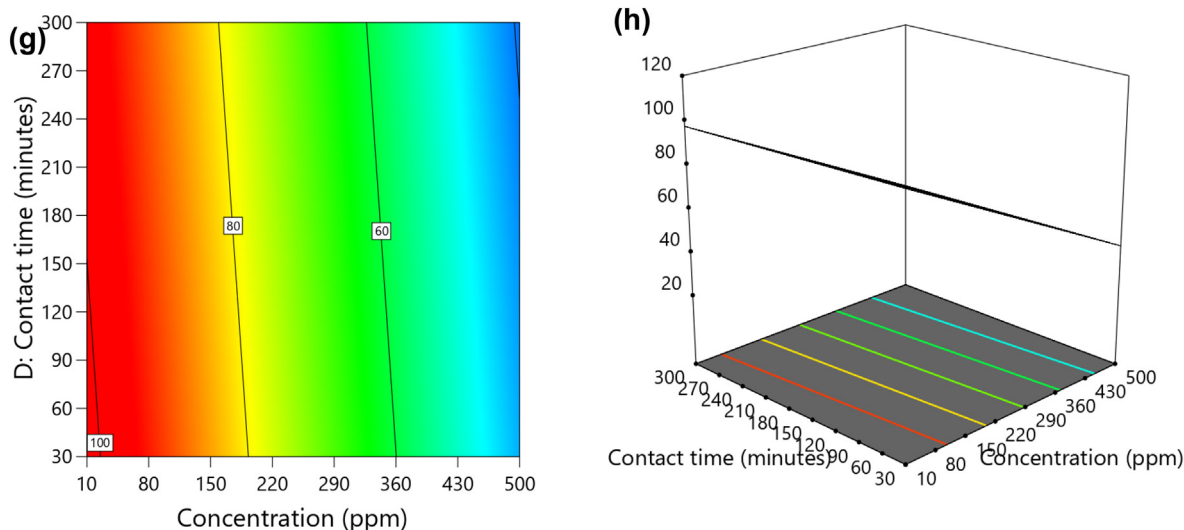


Fig. 6 (continued).

preferred according to the signal-to-noise ratio as measured by Adeq precision. A sufficient signal was suggested by the obtained ratio of 28.377. This model can, therefore, be used to explore the design space. The proposed model has a substantial lack-of-fit (p value 0.05), but its other statistical parameters are significant, and its level of precision is widely regarded as satisfactory, allowing the model to be used for optimization. A similar observation was reported by Dritsa et al., (Dritsa et al., 2009).

3.3. Effect of parameters on adsorption of ciprofloxacin

The perturbation plots were used to examine the impact of individual effects of input factors on the ciprofloxacin elimination efficiency. By changing one element while maintaining the other elements' values, the plots enable the comparison of all the parameters at a specific location in the design space. In general, the operational range of each variable is changed while choosing a reference point to build the perturbation curve. The findings are then plotted against the deviation from the reference point after the reference point has been altered at the center of each factor. The response's slope will be steeper or more curved depending on how sensitive that parameter is. A rather flat line indicates that the response is unaffected by changes in that specific element as reported by previous researchers (Bezerra et al., 2008; Kitamura et al., 2022). Once more, Fig. 4(a–d) demonstrates that the starting concentration and adsorbent dose have a significant impact on the elimination efficiency, whereas the pH and contact time have a much less impact.

3.4. Effect of interactions on adsorption of ciprofloxacin

The results of effects of interactions on adsorptive elimination of ciprofloxacin onto JS biochar, are presented in 3D and contour plots Figs. 5–8. The calcination temperature is an important factor for material adsorption, and results indicate that carbon content increases as the material is calcined to higher temperatures, which is necessary for the adsorption. Further, the results of the adsorption of ciprofloxacin from synthetic solution using JS biochar prepared at 400 °C (Fig. 5), 500 °C (Fig. 6), 600 °C (Fig. 7) and 750 °C (Fig. 8) and all these materials had a similar effectiveness. Previous studies reported a similar trend (Huang et al., 2020; Iqbal et al., 2022; Andersson, 2017; Wakejo et al., 2022; Carabineiro et al., 2011) on biomass-based biochar. A study by Huang and Colleagues prepared and investigated rabbit manure biochar

prepared at 400 °C, 500 °C, 600 °C, and 700 °C (Huang et al., 2020). The carbon content of prepared biochar was observed to increase with increased temperature, the activity of the material increased with temperature, and the material had relatively similar effectiveness (Huang et al., 2020). The results for adsorbent calcined at 400 °C, 500 °C, 600 °C and 750 °C are presented as representative. The elimination of ciprofloxacin ranged from 90 to 100% at lower initial ciprofloxacin concentrations of (10–100 ppm), throughout the pH range.

Ciprofloxacin remediation for material calcined at 400 °C ranged from 80% initial concentration of 200 ppm to 100% initial concentration of 10 ppm, as presented by Fig. 5. This result indicates that JS biochar adsorbent is capable to remediate ciprofloxacin from synthetic water at low levels that are similar to relevant environmental reported levels.

Fig. 6a and 7b indicates that the elimination of ciprofloxacin onto JS biochar calcined at 500 °C reached 100% at pH of 3 and at pH of 7, it reached the elimination of 90%. The elimination of ciprofloxacin decreases as the initial ciprofloxacin concentration increases, at 10 ppm the elimination was 100% and reached 80% at about 200 ppm initial ciprofloxacin concentration.

In addition, the elimination of ciprofloxacin increases as adsorbent dose increases due to increase in surface area available for adsorption. Fig. 6c and d, at an initial adsorbent dose of 50 mg at pH of 7, the elimination reached 66%, which increases to 70% at a dose of about 100 mg. Further increases in adsorbent dose lead to small variation in elimination of ciprofloxacin, this is because the system reached the equilibrium, and the rate of adsorption and desorption is constant. Fig. 6e and f presents the contribution of contact time on the elimination of ciprofloxacin onto JS biochar from synthetic water. The elimination of ciprofloxacin reached 75% at 60 min, further increase in contact time led to small decrease in removal of ciprofloxacin as the system reached equilibrium. Fig. 6g and h indicates that at lower initial ciprofloxacin concentration (10–100 ppm) the elimination reached 90%, as the initial concentration increases elimination of ciprofloxacin decreases throughout the contact time.

Similarly, JS biochar calcined at 600 °C could remove ciprofloxacin from synthetic water as presented in Fig. 7a and b. Results indicates that at pH of 7 and initial ciprofloxacin concentration of (10–100 ppm), ciprofloxacin elimination reached 80% throughout the pH range, and elimination decreased as initial ciprofloxacin concentration increases Fig. 7c and d. Similarly, the elimination of ciprofloxacin decreases as contact time increases, this may be due to saturation of active sites of adsorbent as the system reaches equilibrium Fig. 7e and f.

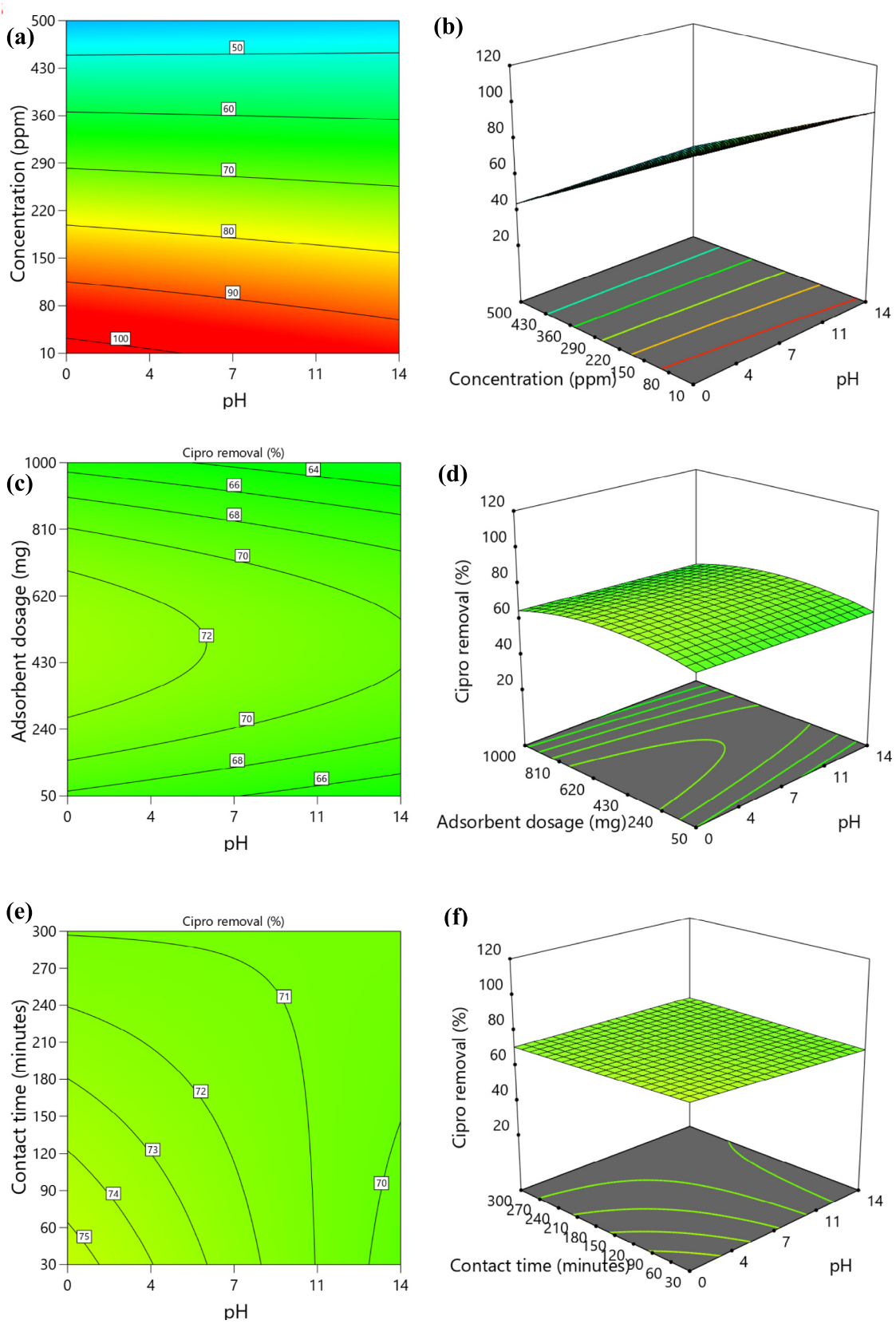


Fig. 7. Contour and 3D graphs showing the influence of parameters on the removal of ciprofloxacin from JS biochar calcined at 600 °C. (a) and (b) the effect of concentration and pH, (c) and (d) the effects of adsorbent dosage and pH, (e) and (f) the effects of contact time and pH, and (g) and (h) the effects of adsorbent dosage and initial ciprofloxacin concentration.

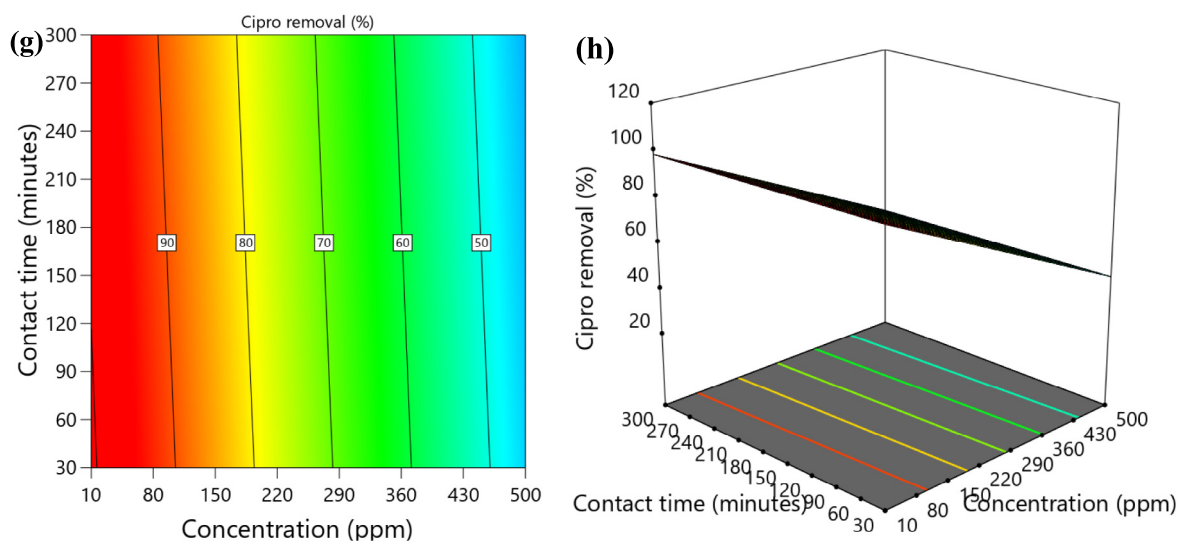


Fig. 7 (continued).

Further, Fig. 7g and 7h, indicate that at lower initial ciprofloxacin concentrations and lower contact time the elimination is higher, this may be because the active sites of an adsorbent are free allowing more ciprofloxacin to adsorb. Results indicate that at 10 ppm of ciprofloxacin and 30 min contact time the elimination reached 90%, but as initial ciprofloxacin concentration reaches 150 ppm the elimination decreased to 80%, indicating the JS biochar may potentially be used for remediation of low levels of contaminants that are threatening the ecosystem health as reported by previous researchers (Ripanda et al., 2023; Yu et al., 2023; Ramírez-Malule et al., 2020; Kumar et al., 2022).

Likewise, JS biochar calcined at 750 °C was capable to remove ciprofloxacin from synthetic water as presented in Fig. 8 a to h. Results indicate that the elimination reached 90% at ciprofloxacin initial concentration of (10 to 80 ppm), when the initial ciprofloxacin concentration reached 150 ppm the elimination decreased to 80%, throughout the pH range, further the elimination decreases as the initial ciprofloxacin concentration increases, as presented in Figs. 8a and b. Further results indicate increased ciprofloxacin elimination as adsorbent dosage increases Fig. 8c and d. But when the adsorbent dosage reached 500 mg, further increase has no significant effect on ciprofloxacin elimination. This might be due to the saturation and the system reached equilibrium, the rate of adsorption is like the rate of desorption, hence no significant difference.

The elimination of ciprofloxacin is higher at lower contact time, but further in time there is no significant elimination as presented in Fig. 8e and f. This may be due to the saturation of active sites of adsorbent as the system reaches equilibrium. The result indicates that at initial ciprofloxacin concentration of 10 to 80 ppm the elimination of ciprofloxacin reached 90%, as the initial concentration of ciprofloxacin reaches 150 ppm the elimination reaches 80%, throughout the contact time, and further the elimination of ciprofloxacin decreases and contact time increases.

3.4.1. Optimization process and validation of the model

Design-Expert software was utilized for numerical optimization, employing integrated algorithms to systematically explore the factor space, evaluating different combinations, and iteratively refining the search process to converge on the optimal solution. The optimization utilized the desirability function to optimise operating parameters within a specific range while maximizing ciprofloxacin removal. The optimum condition observed for the 99.1% ciprofloxacin removal were

pH 1, concentration 17 ppm, adsorbent dosage 288 mg, contact time 38 min, and adsorbent calcination temperature 750 °C, as shown in Fig. 9. Three replicates of confirmation experiments under optimum conditions were conducted to ascertain the validity of the predicted value by the model. The average experimental value was found to be 94.61%, which is slightly lower than the predicted value. Eq. (9) is used to compute the Residual Standard Error (RSE) which is found to be 4.4%, implying an excellent agreement of experimental values with the model-predicted results.

$$\%RSE = \frac{\text{Exp.value} - \text{Pred.value}^*}{\text{Pred.value}} \times 100 \quad (9)$$

(See Fig. 10).

In the current study, the adsorptive elimination of ciprofloxacin was observed to decrease as the initial concentration of ciprofloxacin increased. Furthermore, materials calcined at 500, 600, and 750 °C had similar effects on the elimination of ciprofloxacin from synthetic water. A similar study reported a ciprofloxacin elimination efficiency of 96% from an aqueous solution using a chemically modified bamboo biochar prepared from bamboo sawdust (Wakejo et al., 2022). Moreover, a significant ciprofloxacin elimination reported by other researchers using biochar (Huang et al., 2020; Sodhi and Singh, 2022; Carabineiro et al., 2011) indicated that biochar prepared from biomass (Mtenga and Ripanda, 2022), may be the potential for removing pharmaceutical contaminants. The elimination of ciprofloxacin onto JS biochar is higher at lower contact time and slightly decreases as time increases; the availability of free functional groups may contribute to this onto JS biochar, which gets used as time goes on as reported by previous researchers (Iqbal et al., 2021; Velusamy et al., 2021; Wakejo et al., 2022; Zheng et al., 2021). The dose of adsorbent was found to significantly affecting the elimination of ciprofloxacin from synthetic water, as presented in Figs. 5–7. A similar study by Wakejo and Colleagues reported an increased elimination of ciprofloxacin with increased adsorbent dosage (Wakejo et al., 2022). The elimination of other organic contaminants increased with increased dosage as reported by previous researchers (Späth et al., 2021; Velusamy et al., 2021; Mao et al., 2019; Li et al., 2018; Wakejo et al., 2022; Carabineiro et al., 2011). These results indicate that JS biochar may potentially be used for remediation of contaminants such as ciprofloxacin.

According to Shen et al., ciprofloxacin concentration was positively correlated with cardiac dysfunction in zebrafish, such as decreased

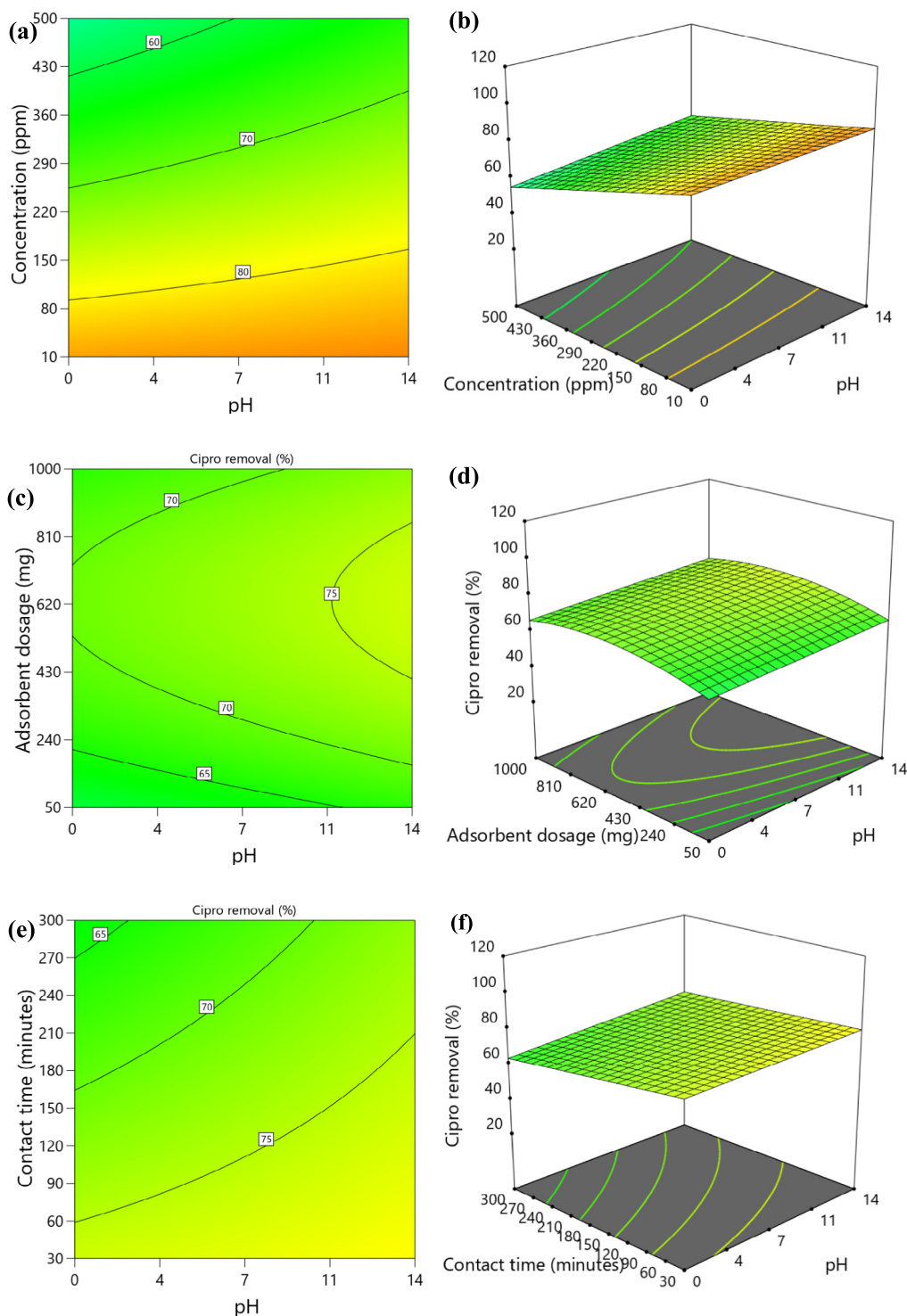


Fig. 8. Contour and 3D graphs for the effect of parameters on the removal of ciprofloxacin from JS biochar calcined at 750 °C. (a) and (b) the interaction of pH and concentration, (c) and (d) adsorbent dosage and pH, (e) and (f) the effects of contact time and pH, and (g) and (h) the effects of adsorbent dosage and initial ciprofloxacin concentration.

heart rate and cardiac output (Shen et al., 2019). Short-term exposure to ciprofloxacin doses of 1, 10, and 100 mg/L had sublethal effects on Neotropical catfish (*Rhamdia quelen*), according to a study by Kitamura et al., (Kitamura et al., 2022). In addition, Ciprofloxacin increased antioxidant system activity catalase in the liver and posterior kidneys (Kitamura et al., 2022). These results indicate that under short-term exposure, ciprofloxacin causes toxic effects in *R. quelen* that require intervention for ecosystem sustainability. The current study found that the JS

biochar adsorbent removed ciprofloxacin 10 to 100 ppm with the best efficiency. Therefore, future improvement and use of the adsorbent in environmental remediation is promising.

3.5. Adsorbent treatment temperature

The calcination temperature is an important factor for preparation of adsorption material. The results indicates that, the number of carbon

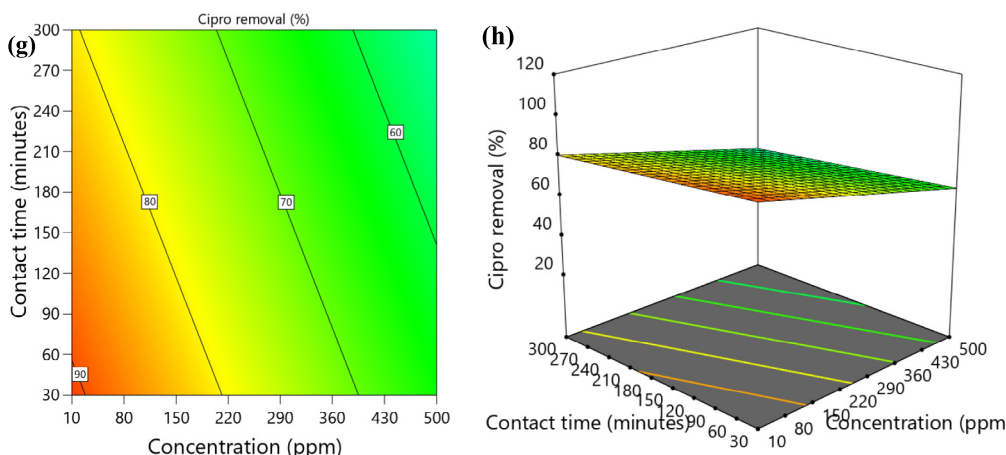


Fig. 8 (continued).

increases as the material is calcined to higher temperatures (from 400 to 600 °C), which is necessary for the adsorption. The adsorption of ciprofloxacin from synthetic water using JS biochar prepared at 400 °C (Fig. 5), 500 °C (Fig. 6), 600 °C (Fig. 7), and 750 °C (Fig. 8), all these materials had similar effectiveness. The past studies reported a similar characteristic (Huang et al., 2020; Iqbal et al., 2022; Andersson, 2017; Wakejo et al., 2022; Carabineiro et al., 2011). A study by Huang et al., prepared the biochar from rabbit manure at 400 °C, 500 °C, 600 °C, and 700 °C (Huang et al., 2020). The carbon content of prepared biochar was observed to increase with increased temperature, the activity of the material increased with temperature, and the material exhibited similar effectiveness (Huang et al., 2020).

3.6. Isotherm studies

The critical factor in the optimisation of adsorbent usage is the quality of ciprofloxacin adsorption onto JS biochar, which is explained by adsorption isotherms, as presented in Fig. 10. To get rid of pollutants like ciprofloxacin, a proper relationship between the equilibrium curve and optimizing the design of the adsorption system is formed (Ashiq et al., 2019; Li et al., 2018; Wakejo et al., 2022). Recently, a published studies made use of the modified Langmuir and Freundlich isotherm models. Freundlich isotherm model best fit the isotherm data for eliminating ciprofloxacin onto JS biochar with R^2 value 0.9757, followed Freundlich isotherm with R^2 of 0.9681. These results, indicates surface heterogeneity of the adsorbent and multilayer adsorption. Previous studies that evaluated the adsorption of ciprofloxacin onto biochar indicated similar results (Li et al., 2018; Wakejo et al., 2022). The results of the Langmuir isotherm show that the calculated adsorption capacity was 555.55 mg/g, indicating that the adsorbent had a significantly high capacity to eliminate ciprofloxacin. A similar study, by Shahrin et al., reported the adsorption of streptomycin, rifampicin, and ibuprofen onto chitosan (Shahrin et al., 2022). Results indicate the maximum estimated adsorption capacities of 11.00 mg/g, 66.91 mg/g and 24.21 mg/g respectively. The current study's adsorption capability supports its usage for removing organic pollutants from water and wastewater effluents, which was also reported by a prior study (Shahrin et al., 2022).

These results provide an R_L value between 0 and 1, indicating that the adsorption of ciprofloxacin onto JS biochar is favourable. Table 2 presents the general representation of the graphs and calculation results.

The K_f value shown in Table 2 shows that the adsorbent is heterogeneous, and that it fluctuates with the adsorbent's

heterogeneity. The fractional value of $1/n$ was within ($0 < 1/n < 1$), indicating that the surface of the adsorbent is heterogeneous and hence a multilayer adsorption, as similarly suggested by prior researchers (Togue Kamga, 2019; Aziz et al., 2018; Shakoore and Nasar, A, 2017; Nassar et al., 2011).

3.7. Adsorption mechanism

Biochar produced by heating biomass in a limited-oxygen environment, has also been investigated as an adsorbent for remediation of antimicrobials (Kumar et al., 2022; Mtenga and Ripanda, 2022). Biochar has a porous structure with the potential for storage of materials to be remediated such as antimicrobials. Similarly, other researchers reported that biochar contained substantial storage pores (0.5–50 μm), accounting for >80% of total pore volume, and small transmission and residual pores (Lu and Zong, 2018). High carbon content provides material stability for adsorption, making it effective in adsorbing a wide range of contaminants, including antimicrobials such as ciprofloxacin and lamivudine (Kumar et al., 2022). Fig. 12 presents SEM images of unused JS biochar prepared at 600 °C, from various orientations, indicating porous nature and rough surface. Adsorptive interactions are accompanied by external mass transfer of organic pollutants from the aqueous solution to the biochar surface, which is followed by internal diffusion of organic molecules into the pores of the adsorbent, as previously reported (Gasim et al., 2022). The primary driving force for adsorption is the Van der Waals forces, which are weak attractive forces between molecules. Physical and chemical processes, such as H-bonding, hydrophobic interactions, electrostatic attraction, Lewis acid-base interactions, pore filling, Coulombic attraction, spectrometer exchange, and acceptor interactions, are involved in the adsorption of the organic pollutant onto the biochar.

Furthermore, results indicated JS biochar had relatively higher surface area, pore size and volume necessary for adsorption (Ripanda et al., 2023a; Ripanda et al., 2023). The larger the surface area provides more space for adsorbent and adsorbate interactions leading to its adsorptive removal. Similarly, the large surface area with a rough, porous structure as presented by Fig. 11, provides ample space for molecules from the fluid to meet the solid surface and adhere to it. The presence of surface functional groups onto JS biochar adsorbent evidenced by Fig. 2, provides potential for the pollutants binding. Indicating that the adsorptive removal of ciprofloxacin onto JS biochar was contributed by the presence of these functional groups and a surface phenomenon. Further, Fig. 12 presents interactions between adsorbent and ciprofloxacin indicating surface process.

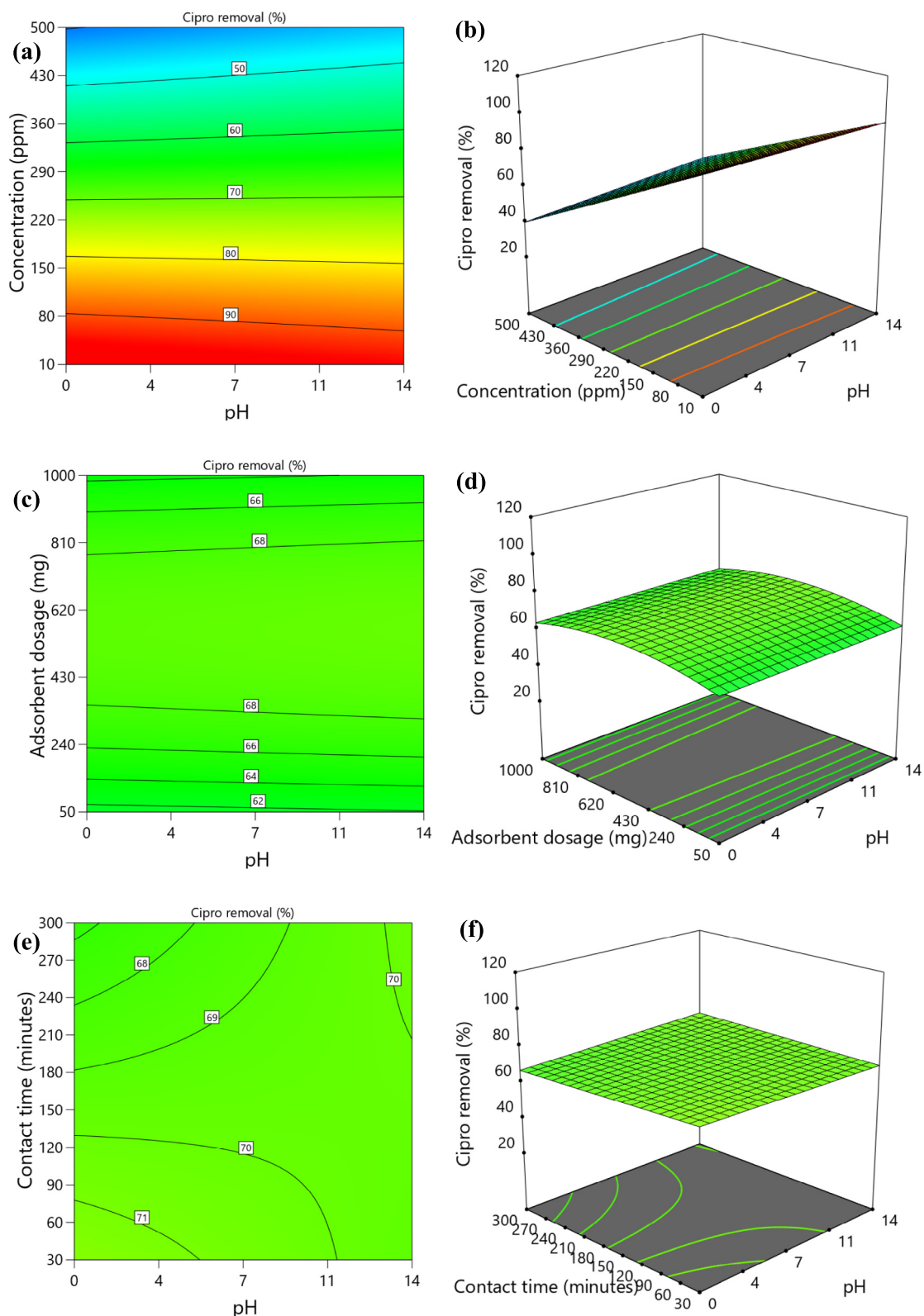
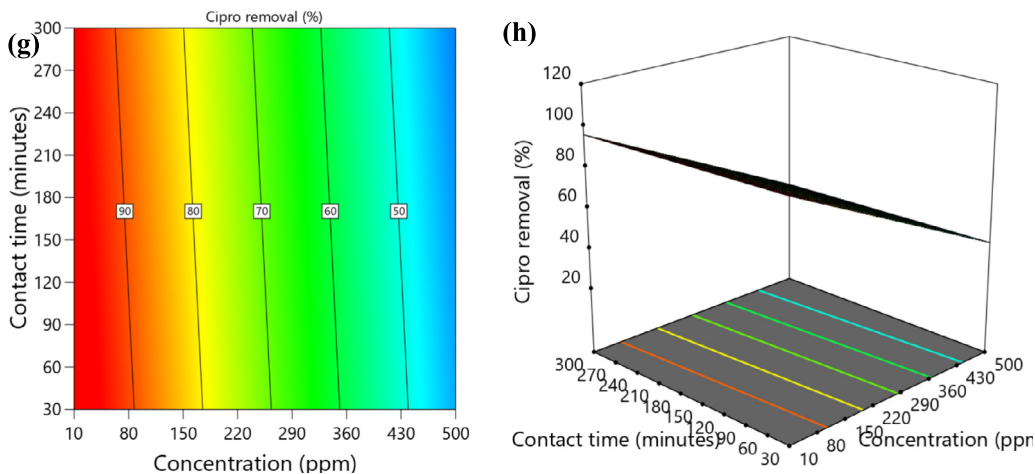


Fig. 9. Desirability ramp plot for the optimisation of ciprofloxacin removal.

The presence of acidic hydroxyl group on the surface of JS biochar adsorbent indicates the possibility of hydrogen bonding a specific type of dipole-dipole interaction, which can also play a role in adsorption.

Molecules that can form hydrogen bonds, like water, can be adsorbed by surfaces with hydrogen bond acceptors or donors resulting to removal. Similarly, electrostatic interactions between charged molecules



in the fluid and charged sites on the adsorbent surface can also contribute to adsorption. This is complimented by results of adsorption isotherm experiments that followed the Freundlich isotherm accounting for multilayer adsorption on heterogeneous surfaces.

4. Conclusion and recommendations

Adsorption is a sustainable and efficient method for the remediation of organic pollutants from waters and effluents. Jamun seed biochar adsorbent, has shown to be a potential adsorbent for a variety of organic pollutants, including ciprofloxacin. Its porous structure, rough surface and high surface area provide ample opportunities for the adsorption of contaminants, while its stability and longevity ensure long-term remediation. In the recent investigation, the proposed model and the adsorption models provided the best descriptions of the ciprofloxacin adsorption process onto the JS biochar adsorbent. Ciprofloxacin adsorption onto the JS biochar provided relatively higher elimination efficiency. The elimination of ciprofloxacin onto biochar increased at low initial concentrations. Reports indicates

Table 2

The general representation of the results obtained from the graph and calculations.

Temp(°C)	Langmuir Isotherm				Freundlich Isotherm		
	Q _e (mg/g)	Q _{max} (mg/g)	b(L/mg)	R ²	n	K _f	R ²
29.85	459.2	555.55	0.018219	0.9681	1.745	3.901	0.9757

that low levels of contaminants including ciprofloxacin are persistent in the environment while creating harm, indicating adsorption using jamun seed biochar adsorbent may be a solution. With continued research and development, the use of biochar has the potential to significantly reduce the negative impacts of organic contaminants on the environment and human health. Future studies should concentrate on the improvement of the material and studying the elimination of ciprofloxacin in real water, wastewater, and effluents from pharmaceutical industries.

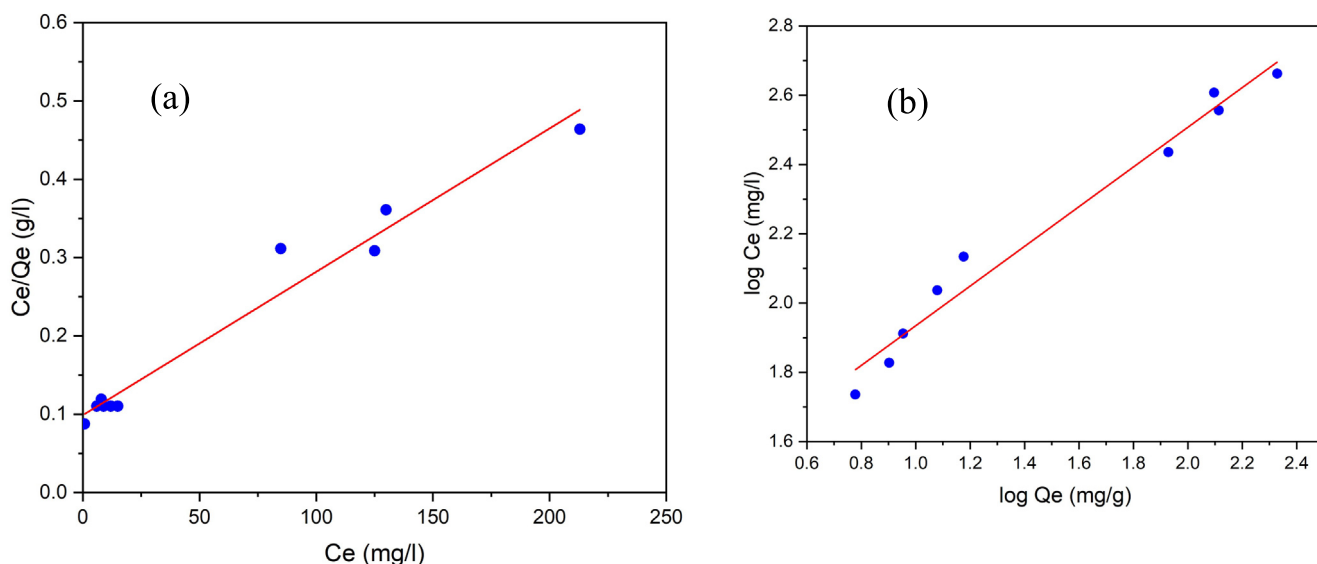


Fig. 10. Represents the isotherm models for the elimination of ciprofloxacin from synthetic water (a) modified Langmuir (b) Freundlich.

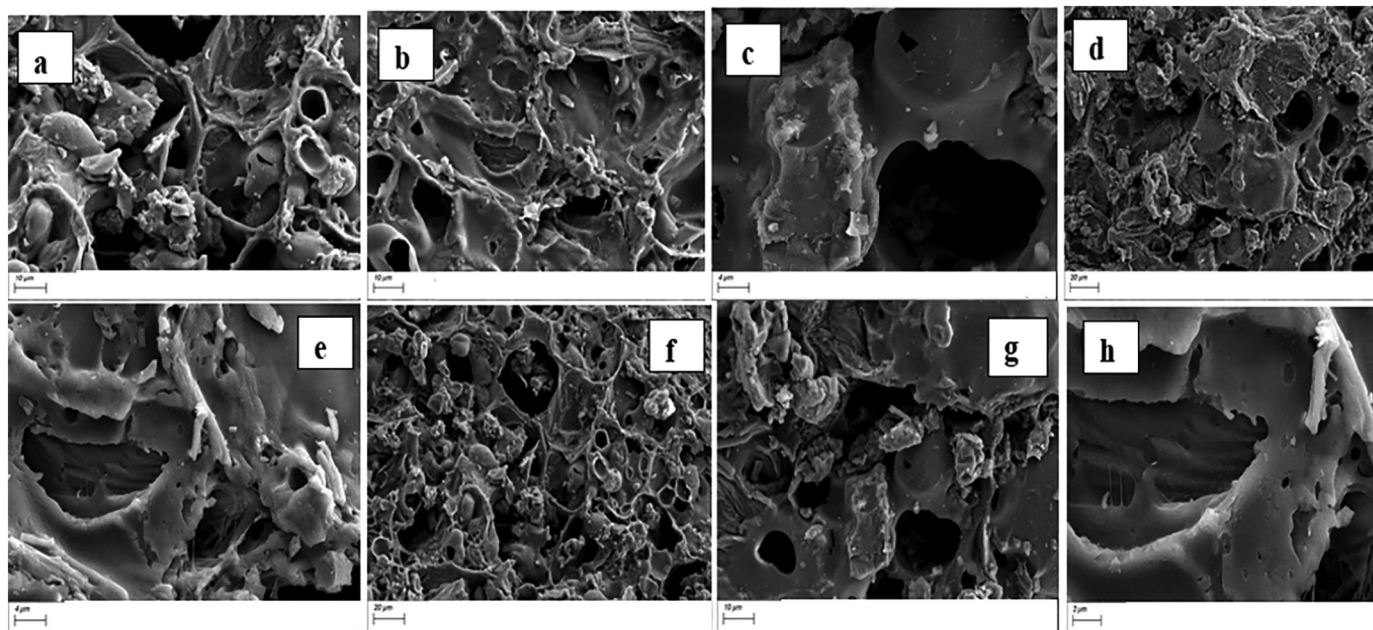


Fig. 11. SEM images of unused JS biochar prepared at 600 °C, images labelled a to h are taken from various orientations, indicating porous nature, and rough surface.

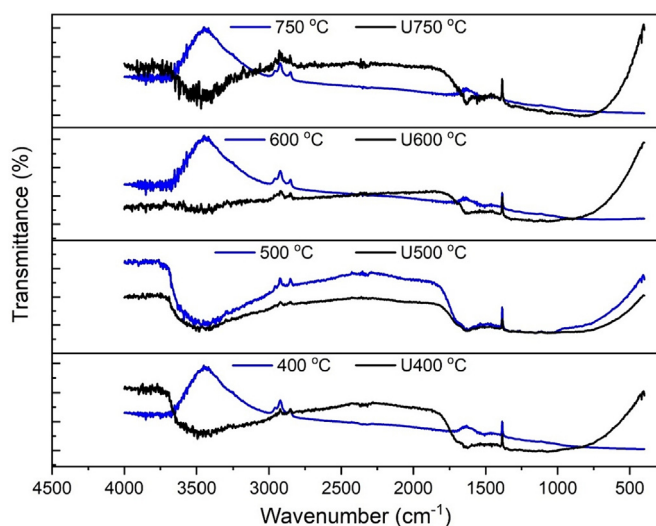


Fig. 12. Comparison of FTIR spectra of JS biochar calcined at 400 °C, 500 °C, 600 °C, and 750 °C used for adsorption of ciprofloxacin (black lines), and unused (blue lines). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

CRediT authorship contribution statement

Asha Ripanda: Writing – original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Mwemezi J. Rwiza:** Writing – review & editing, Visualization, Validation, Supervision, Formal analysis, Data curation, Conceptualization. **Elias Charles Nyanza:** Writing – review & editing, Visualization, Validation, Supervision,

Formal analysis, Data curation, Conceptualization. **Linda Numph Bih:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Miraji Hossein:** Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Ramadhani Bakari:** Writing – original draft, Supervision, Methodology, Formal analysis, Data curation, Conceptualization. **Somit Kumar Singh:** Writing – review & editing, Visualization, Validation, Methodology. **Giridhar Reddy:** Visualization, Validation, Software, Formal analysis, Data curation. **C.R. Ravikumar:** Visualization, Validation, Supervision, Methodology, Formal analysis, Data curation. **H.C. Ananda Murthy:** Writing – review & editing, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Karoli N. Njau:** Writing – review & editing, Visualization, Validation, Supervision, Investigation, Formal analysis, Data curation, Conceptualization. **Said Ali Hamad Vuai:** Writing – review & editing, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Revocatus L. Machunda:** Writing – review & editing, Visualization, Validation, Supervision, Formal analysis, Data curation, Conceptualization.

Data availability statement

The initial data are deposited Mendeley data (<https://data.mendeley.com/drafts/pphv3ygkfk>), and the update deposited Mendeley data (<https://data.mendeley.com/drafts/2bkk7rxntv>). Used secondary data and cited within.

Declaration of competing interest

None.

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