OPTIMIZATION OF A TRICKLING FILTER SYSTEM FOR THE REMOVAL OF CIPROFLOXACIN FROM SIMULATED PHARMACEUTICAL WASTEWATER

UNDER PEER REVIEW

Abstract

The presence of ciprofloxacin, a widely used antibiotic, in pharmaceutical wastewater poses significant environmental challenges due to its persistence and potential to induce antibiotic resistance. This study investigates the optimization of a trickling filter system for the effective removal of ciprofloxacin from simulated pharmaceutical wastewater. Key operational parameters, including pump flow rate, initial concentration and contact time, were systematically varied to assess their impact on removal efficiency of total suspended solids (TSS), total dissolved solids (TDS). Response (RSM) was employed to design experiments and analyze the interactions between variables. The optimized parameter factors conditions achieved contact time at 8hrs, concentration at 277.571mg/l and flow rate at 6.466m3/hr. Also optimized responses were achieved , a total suspended solids (TSS) of a ciprofloxacin removal efficiency of 84.766% (actual 83.5%), total dissolved solids (TDS) of a ciprofloxacin removal efficiency of 49.27% (actual 47.3%) and biological oxygen demand (BOD) of a ciprofloxacin removal efficiency of 33.005% (actual 32.6%), demonstrating the potential of trickling filter systems in mitigating pharmaceutical contaminants in wastewater.

*Corresponding author: Email: ro.ajemba@unizik.edu.ng, cf.okey-onyesolu@unizik.edu.ng;

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1. Introduction

There were different classes of antibiotics based on their mechanism of action, chemical structure, action spectrum, and route of administration (1). The most common classification of that was based on their mode of action. They include fluoroquinolones, quinolones, *β*-lactams, sulfonamides, monobactams, carbapenems, and aminoglycosides. Quinolones (ciprofloxacin, levofloxacin, norfloxacin, and ofloxacin) were broad-spectrum antibiotics that were widely prescribed (1a). Ciprofloxacin (CIP) was a quinolone antibacterial agent classified as second-generation fluoroquinolone with broad-spectrum action that was commonly used to treat human and animal bacterial infections (2) was frequently detected in wastewater due to its widespread use and incomplete removal during conventional treatment processes (3). The presence of CIP or any antibacterial derivative in wastewater and surface water was considered a significant environmental hazard, even at very low concentrations. That was because these products can increase the antibiotic resistance of pathogenic bacteria and generate modifications in the biological balance of aquatic ecosystems (4). The continuous and unregulated discharge of CIP into the aquatic environment posed serious environmental and health problems to man and the aquatic lives. That has result sickness and diseases to mankind and imbalance in ecosystem and consequently the food chain was threatened. Among the various concern on humans and environment includes chronic toxicity, endocrine disruption, and direct toxicity of micro flora, even at low concentrations. The effect of CIP on the water in a low concentration affected the photosynthesis of plants, transforms the morphological structure of the algae, and then disrupts the aquatic ecosystem (5). When CIP contaminated water was consumed by humans, it might cause anger, nausea, vomiting, headache, diarrhea, and tremor. The high concentration of CIP can cause severe kidney failure and increase liver and thrombocytopenic enzymes (6).

Several methods have been employed to remove CIP from aqueous solution. They include adsorption in activated carbon (7); zeolites (8); montmorillonites (9); microalgae intake (10); photo catalytic degradation (11) and electrocoagulation (12); coagulation, sedimentation, biodegradation, photo-transformation, electrochemical, chlorination, ozonation, and Nano filtration through membranes (13;14). There was, therefore, the need to treat the influent streams using eco-friendly and energy efficient methods, prior to discharge into the receiving waters bodies. Trickling filter techniques was simple, reliable, low-cost and effective in treating high concentrations of organic material. However, most of the treatment techniques that have been employed in time past have the issue of efficiency, generation of toxic by-products, high treatment cost, and high energy requirement, (15). Therefore, efficient and low-cost treatment options that do not introduce harmful by-products were required to eliminate antibiotics from water. Due to the inherent limitations of the existing treatment processes, trickling filter present a good alternative since it was simple, reliable, low-cost, effective in treating high concentrations of organic material, relatively low power requirement and requires moderate skill and technical expertise to operate the system (16;17). A trickling filter was an aerobic wastewater treatment process generally used for industrial effluents and domestic sewage treatment. Its operation consists of passing the effluent to be treated over a fixed bed of support medium. Optimizing the operational parameters of trickling filters is crucial to enhance their efficiency in removing specific contaminants such as total suspended solids (TSS) and total dissolved solids (TDS) of ciprofloxacin.

2. Materials and Methods

2.1 Preparation of Ciprofloxacin Simulated Wastewater

1000mg (1g) of ciprofloxacin tablet was dissolved in the 1000 mL standard flask with distilled water, working solutions were prepared from the stock solution and the stock solution was stored in the refrigerator to minimize degradation and prevent it from losing its integrity.

2.2 Trickling Filter Setup

A bioreactor body with cylindrical shape and made of stainless steel was constructed. Its length of the reactor would be 152.4 and its diameter would be about 76.2 cm as shown below in fig 1. Modified PFF would be used as filter media in the trickling filter system for microbial growth. The PFF would be placed vertically in trickling filter system with 129.5 cm in height and diameter varies from 0.5 to 1.0 inches. A distributor would be installed at the top of reactor to spread CIP wastewater uniformly over filter media. Flow rates would be adjusted with the help of control valves. A 6inches depth drainage layer would be constructed at the bottom of reactor for ventilation and for outflow of the wastewater from the reactor tank for final sedimentation. A settling tank would be provided for collecting and settling waste water. The system was operated at flow rate of 2.6-9.3m3/hr and contact time of 2.6-9.3hrs.

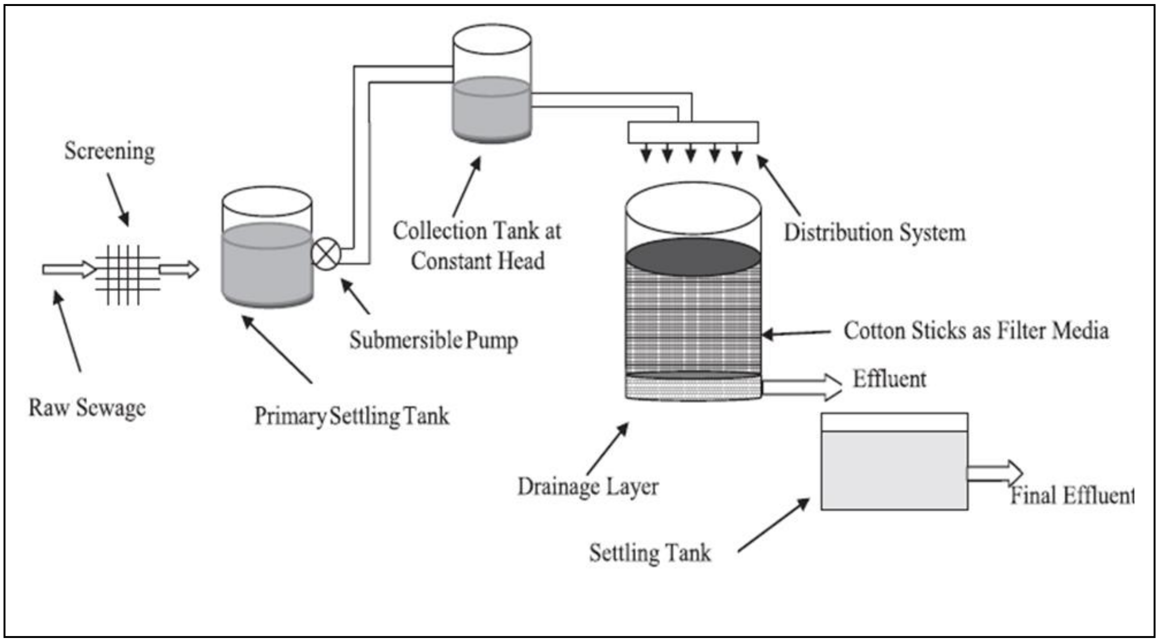


Fig 1: Schematic diagram of Trickling Filter System setup

2.3 Experimental Design and Optimization

Response surface methodology (RSM) was a collection of mathematical and statistical techniques for modelling and analysis of problems involving multivariate experimental design, statistical modelling focusing on three independent variables: flow rate, initial concentration and contact time and process optimization (18:19:20:21) using central composite design (CCD) which was employed to evaluate the effects and interactions of that variables on ciprofloxacin removal efficiency.

2.4 Analytical Methods

1. **Total suspended solids were determined through gravimetric analysis**
2. **Total dissolved solid were determined through gravimetric analysis.**

3. **Biological oxygen demand were measured using standard APHA methods (APHA, 2017).**
4. **Optimization of CIP Degradation Using Response Surface Methodology**

2.5 Performance Evaluation

The removal efficiency was calculated using (1)

Removal Efficiency (%) = co-ct/co × 100 **(1)**

3. Results and Discussion

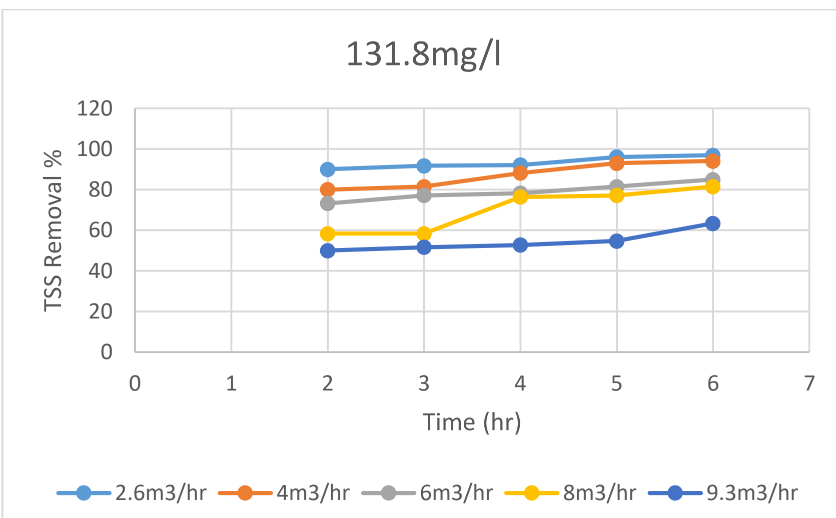
3.1 Batch Biodegradation Studies of the Trickling Filter System Operating Variables

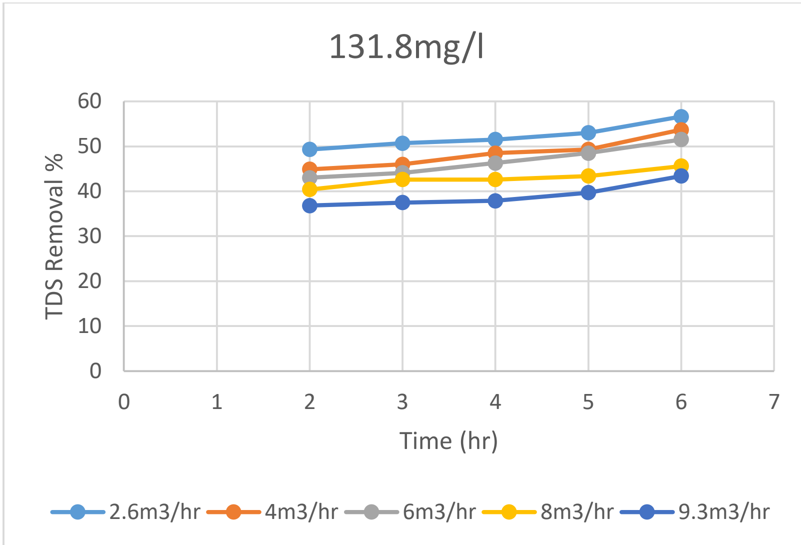
3.1.1 Effect of Combination of Flow rate and Contact time on TSS Removal Efficiency

Flow rate has significant effect on percentage removal of TSS, TDS and BOD from simulated pharmaceutical wastewater treatment. As such, the effect of flow rate on optimization of trickling filter system for the removal of ciprofloxacin from simulated pharmaceutical wastewater treatment was evaluated using five different flow rates which were 2.6m3/hr, 4m3/hr, 6m3/hr, 8m3/hr and 9.3m3/hr, at concentration of 131.8mg/l, 200mg/l, 300mgl, 400mg/l and 468mg/l. It could be seen from fig 2a that the highest percentage removal TSS from simulated pharmaceutical wastewater was obtained at flow rate of 2.6m3/hr and concentration of 131.8mg/l. The highest percentage removal of TSS with decrease in flow rate was due to particle agglomeration and filter media performance. Decreased flow rates ensured efficient mass transfer between the liquid phase (wastewater) and solid phase and stops breakthrough occurrence (22:23:24). Therefore, the percentage removal of TSS obtained at flow rate of 2.6m3/hr at concentration of 131.8mg/l was 99.7%.

There was increase in TDS percentage removal at 2.6m3/hr and concentration of 131.8mg/l in figure 2b. There was increase in TDS percentage removal as flow rates decreases because it allowed for longer contact times between the water and treatment media, potentially enhancing the removal efficiency of TDS from simulated

pharmaceutical wastewater using trickling filter system. Increased flow rates reduced the efficiency of TDS removal due to shorter contact time. However, excessively low flow rates might lead to issues such as increased residence time in the treatment system, which could promote bacterial growth or other undesirable reactions (26). Therefore, TDS percentage removal from simulated pharmaceutical wastewater using trickling filter system at 2.6m3/hr and concentration of 131.8mg/l was 58.7%. From the figure 2c, it was observed that there was increase in BOD percentage efficiency as the flow rate decreases during the biodegradation because slower flow rates can allow contact times between the water and treatment media, which enhances the biological degradation of ciprofloxacin and associated organic matter contributing to BOD. Increase flow rates could reduce the efficiency of biological processes due to shorter contact times between the wastewater and microorganisms which could result to lower BOD percentage efficiency as microorganisms have less time to metabolize organic matter, including ciprofloxacin (30; 31; 32). Therefore, the BOD percentage efficiency of ciprofloxacin was 51%.





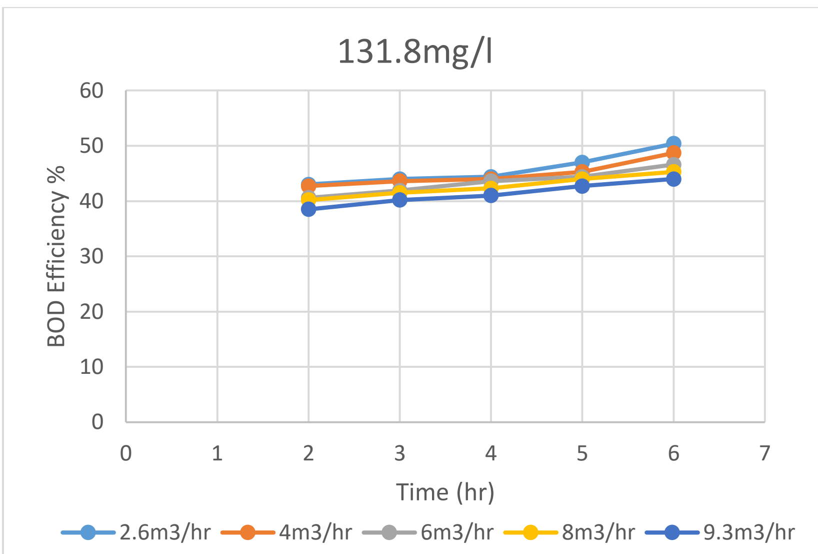
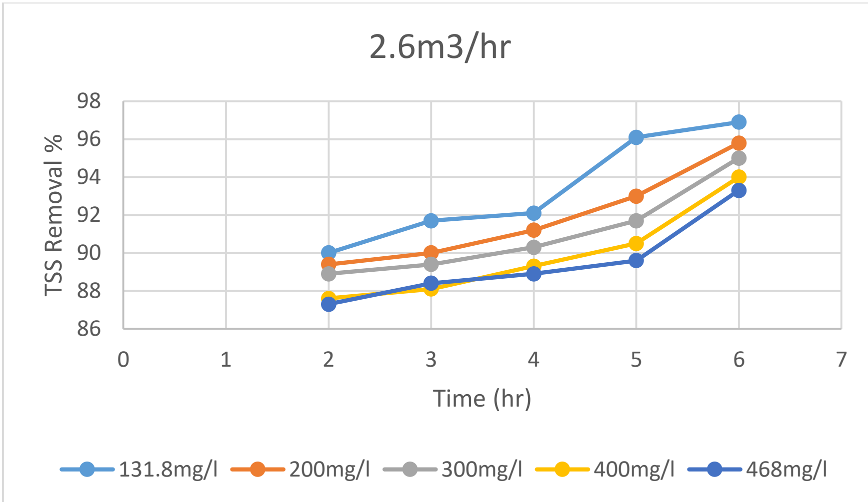


Fig 2 (a-c): Effect combination of flow rate and contact time on TSS removal efficiency from simulated ciprofloxacin pharmaceutical wastewater.

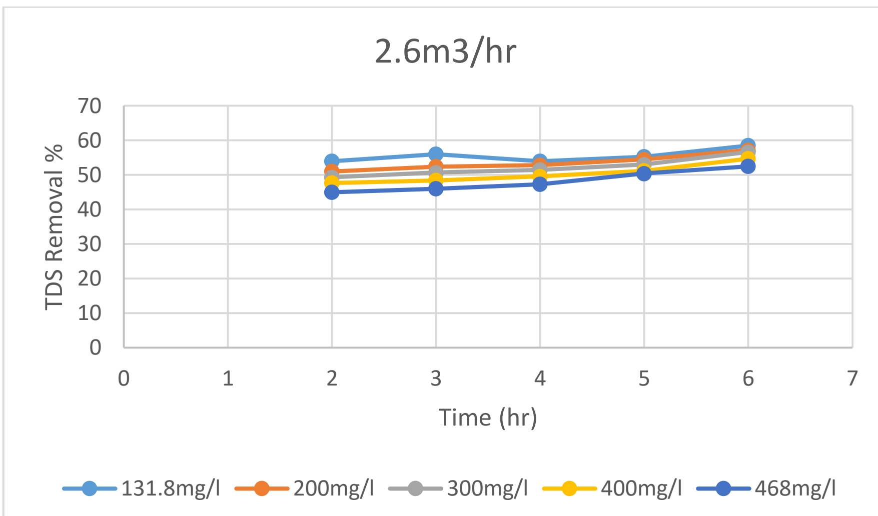
3.1.2 Effect of Combination of Initial Concentration and Contact time on TSS Removal Efficiency

Figure 3a-c below showed the TSS, TDS and BOD percentage removal of ciprofloxacin from simulated pharmaceutical wastewater at different concentration

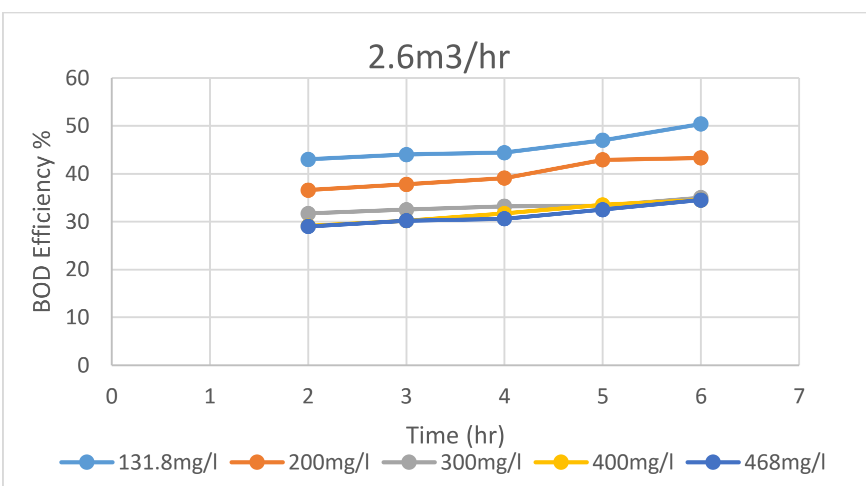
of 131.8mg/l, 200mg/l, 300mg/l, 400mg/l and 468mg/l and contact time of 2.6hr, 4hr, 6hr, 8hr and 9.3hr using five different flow rates of 2.6m3/hr, 4m3/hr, 6m3/hr, 8m3/hr and 9.3m3/hr. From the figure 3a below, it could be observed that TSS percentage removal increases as the concentration of simulated pharmaceutical wastewater decreases at flow rates of 2.6m3/hr and concentration of 131.8mg/l respectively because there was less quantity of ciprofloxacin to be removed. Generally, high concentrations of ciprofloxacin could lead to reduced removal efficiency due to saturation of treatment mechanisms or competition with other contaminants for biodegradation sites (25). In figure 3b, There was increase in TDS percentage removal from simulated pharmaceutical wastewater as the concentration decreases due less competition for reactive sites or biodegradation sites on treatment media (27). Higher concentration of TDS might lead to increased competition for reactive sites or complexation with other ions in the water, which could affect the mechanisms involved in TDS removal (28;29). Therefore, the TDS percentage removal from simulated pharmaceutical wastewater was 58.7% at concentration of 131.8mg/l under flow rate of 2.6m3/hr. It could be seen from fig 3c that the highest BOD efficiency of ciprofloxacin was obtained at concentration of 131.8mg/l in each of the three flow rate. There was increase in BOD percentage removal as the concentration of ciprofloxacin decreases due to lower concentrations of ciprofloxacin in water could lead to lower levels of organic matter, which may contribute to decreased BOD. That lower organic load can potentially overwhelm biological treatment systems, increasing the percentage of BOD efficiency (33; 34; 35).


2.6m3/hr
TSS Removal %
98
96
94
92
90
88
86
0
1
2
3
4
5
6
7
Time (hr)
131.8mg/l
200mg/l
300mg/l
400mg/l
468mg/l

a


2.6m3/hr
TDS Removal %
70
60
50
40
30
20
10
0
0
1
2
3
4
5
6
7
Time (hr)
131.8mg/l
200mg/l
300mg/l
400mg/l
468mg/l

b


2.6m3/hr
BOD Efficiency %
60
50
40
30
20
10
0
0
1
2
3
4
5
6
7
Time (hr)
131.8mg/l
200mg/l
300mg/l
400mg/l
468mg/l

c

Fig 3 (a-c): Effect of combination of initial concentration and contact time on TSS removal efficiency from simulated ciprofloxacin pharmaceutical wastewater using trickling filter system.

3.2 Optimization and Model Validation

3.2.1 Optimization and Modelling Validation of TSS percentage removal efficiency

Table 1 showed the design CCD matrix consisting of number of run, independent factors, actual percentage and predicted percentage as randomized by the Design Expert software (version 8) and the respective response obtained from the experiment.

Table 1: CCD matrix factors and response of TSS percentage removal

Source	Factor 1 Contact Time hrs	Factor 2 Conc. Mg/l	Factor 3 Flow rate m3/hr	Actual %	Predicted %
1	4	200	4	82.4	83.3808
2	8	200	4	88	89.0908
3	4	400	4	83.3	84.9356
4	8	400	4	86.2	85.9656
5	4	200	8	63.7	64.3689
6	8	200	8	77.8	76.5669
7	4	400	8	72.4	71.7317
8	8	400	8	79.8	79.2417
9	2.6	300	6	77.7	76.3429
10	9.4	300	6	86.7	87.4596
11	6	131.8	6	78.3	77.6145
12	6	468.1	6	81.1	81.188
13	6	300	2.6	90.3	88.4269
14	6	300	9.4	65.5	66.7757

15	6	300	6	83.3	83.3171
16	6	300	6	83.3	83.3171
17	6	300	6	83.3	83.3171
18	6	300	6	83.3	83.3171
19	6	300	6	83.3	83.3171
20	6	300	6	83.3	83.3171

3.2.2 Development of model equations of TSS percentage removal efficiency

It was also observed in the table 2 that the quadratic model has relatively high standard deviation of 3.08 and relatively low R^2 (0.8512) in reasonable agreement with adjusted R^2 (0.7174) The TSS percentage removal of ciprofloxacin by optimization of trickling filter system for the removal of ciprofloxacin from simulated pharmaceutical wastewater were obtained with respect to RSM as presented in table 1. Analysis of variance (ANOVA) for quadratic model, equation 3.1 was presented in table 3. If $p > 0.05$ threshold, it indicated significant input variables for the descriptive process models. Furthermore, the lower the model p-value (or the higher F-ratio), the higher the significance of the input variable effect on the response variable (36). From table 3, it was evident that the effects of contact time, flow rate and concentration were significant ($P>0.1000$). Equation 3.1 and 3.2 indicate the regression model prior and after the elimination of insignificant factors.

$$\%TSS = 46.31 + 1.98A - 3.42B - 1.33C - 0.20AB - 0.050AC + 0.18BC + 0.47A^2 + 1.37B^2 - 0.17C^2 \quad (3.1)$$

$$\%TSS = 46.31 + 1.98A - 3.42B - 1.33C - 0.20AB - 0.050AC + 0.18BC + 1.37B^2 - 0.17C^2 \quad (3.2)$$

The model equations selected were further evaluated using ANOVA component of the software. From table 3, response surface quadratic model for removal efficiency has F-value of 60.84 indicating that the model is significant. For the model terms, p-

value less than 0.05 implies that model term was significant (37; 38; 39; 40; 41) and largest F-value signifies the model term having the most significant model significant effect on the response (42; 43). In that case, the significant model terms were A, B, C, AB, AC, BC, B^2, C^2 were significant model terms while A^2 was the insignificant model term. Values greater than 0.1000 indicate the model terms were not significant. The model term having the most significant effect on the response was C with F-value of 360.61.

Table 2: Model Summary Statistics of TSS percentage removal efficiency

Source	Std.dev.	R-squared	Adjusted R-squared	Predicted R-squared	Press
Linear	3.76	0.6451	0.5786	0.3661	403.50
2FI	3.69	0.7221	0.5938	-0.1195	712.64
Quadratic	3.08	0.8512	0.7174	-0.1317	720.41 Suggested
Cubic	2.68	0.9322	0.7853	-13.9420	9511.89 Aliased

Table 3: Quadratic RSM ANOVA for the TSS percentage removal efficiency

Source	Sum of squares	Df	Mean Square	F-Square	P-Value Prob>F
Model	859.24	9	95.47	60.84	<0.0001 Significant
A-Contact Time	149.18	1	149.18	95.07	<0.0001
B-Concentration	15.41	1	15.41	9.82	0.0106
C-Flow rate	565.86	1	565.86	360.61	<0.0001
AB	11.04	1	11.04	7.04	0.0242
AC	21.13	1	21.13	13.46	0.0043
BC	16.82	1	16.82	10.72	0.0084
A^2	3.61	1	3.61	2.30	0.1602
B^2	27.62	1	27.62	17.60	0.0018
C^2	58.85	1	58.85	37.51	0.0001
Residual	15.69	10	1.57		
Lack of fit	15.69	5	3.14		
Pure Error	0.000	5	0.000		
Corr. Total	874.93	19			

3.2.3 Design of experiment of TDS percentage removal efficiency

Table 4 showed the design CCD matrix consisting of number of run, independent factors, actual percentage and predicted percentage as randomized by the Design Expert software (version 8) and the respective response obtained from the experiment.

Table 4: CCD matrix factors and response of TDS percentage removal efficiency

Run	Factor 1 Contact Time hrs	Factor 2 Conc. Mg/l	Factor 3 Flow rate m3/hr	Actual %	Predicted %
1	4	200	4	50	50.9619
2	8	200	4	53.5	55.1285
3	4	400	4	45.1	43.8889
4	8	400	4	48	47.5475
5	4	200	8	47	47.7592
6	8	200	8	50.5	52.0179
7	4	400	8	43	41.6782
8	8	400	8	45.5	45.1369
9	2.63641	300	6	43.5	44.3043
10	9.36350	300	6	52.2	50.9619
11	6	131.821	6	58.5	55.9273
12	6	468.179	6	42.3	44.4389
13	6	300	2.63641	48.3	48.0704
14	6	300	9.36350	43.8	43.5958
15	6	300	6	46.3	46.3124
16	6	300	6	46.3	46.3124
17	6	300	6	46.3	46.3124

18	**6**	**300**	**6**	**46.3**	**46.3124**
19	**6**	**300**	**6**	**46.3**	**46.3124**
20	**6**	**300**	**6**	**46.3**	**46.3124**

3.2.4 Development of model equations of TDS percentage removal efficiency

It was also observed in the table 5 that the quadratic model has relatively high standard deviation of 1.52 and relatively high R^2 (0.9209) in reasonable agreement with adjusted R^2 (0.8497). The TDS removal of ciprofloxacin from simulated ciprofloxacin pharmaceutical wastewater obtained with respect to RSM are presented in table 6. Analysis of variance (ANOVA) for quadratic model equation 3.3 was presented in table 6. If P>0.0500 threshold, it indicated significant for the quadratic process models. Furthermore, the lower the model p-value (or the higher F-value), the higher the significance of the input variable effect on the response variable (36). It was evident that in table 6, the effects of contact time, concentration and flow rate were highly significant (P>0.1000). Table 6 indicated that quadratic coefficient of determination (R^2) was 92.09%. Equation 3.3 and 3.4 indicate the quadratic model prior and after the elimination of insignificant factors.

$$TDS\% = 46.31 + 1.98A - 3.42B - 1.33C - 0.20AB - 0.050AC + 0.18BC + 0.47A^2 + 1.37B^2 - 0.17C^2 \quad (3.3)$$

$$TDS\% = 46.31 + 1.98A - 3.42B + 1.37B^2 \quad (3.4)$$

The model equations selected were further evaluated using ANOVA component of the software. From table 4, response surface quadratic model for removal efficiency has F-value of 12.94 indicating that the model was significant. For the model terms, p-value less than 0.05 implies that model term was significant (37; 38; 39; 40; 41) and largest F-value signifies the model term having the most significant model significant effect on the response (42; 43). Values greater than 0.1000 indicate the model terms

are not significant. The model term having the most significant effect on the response was B with F-value of 69.27.

Table 5 Model Summary Statistics of TDS percentage removal efficiency

Source	Std.dev	R-Squared	Adjusted R-Square	Predicted R-Squared	Press
Linear	1.83	0.8149	0.7802	0.6816	92.63 Suggested
2FI	2.02	0.8149	0.7325	0.4676	154.82
Quadratic	1.52	0.9209	0.8497	0.3991	174.74 Suggested
Cubic	0.28	0.9984	0.9949	0.6427	103.91 Aliased

Table 6: Quadratic RSM ANOVA for the TDS percentage removal efficiency

Source	Sum of squares	Df	Mean square	F-value	P-value. Prob>F
Model	267.81	9	29.76	12.94	0.0002 Significant
A-contact time	53.50	1	53.50	23.26	0.0007
B-conc.	159.32	1	159.32	69..27	<0.0001
C-flow rate	24.17	1	24.17	10.51	0.0088
AB	0.32	1	0.32	0.14	0.7169
AC	0.020	1	0.020	8.695E-003	0.9275
BC	0.25	1	0.25	0.11	0.7509
A^2	3.14	1	3.14	1.37	0.2696
B^2	26.99	1	26.99	11.73	0.0065
C^2	0.41	1	0.41	0.18	0.6804

Residual	**23.00**	**1**	**2.30**		
Lack of fit	**23.00**	**5**	**4.60**		
Pure error	**0.000**	**5**	**0.000**		
Cor total	**290.81**				

3.2.5 Design of experiment of BOD percentage efficiency

Table 7 showed the design CCD matrix consisting of number of run, independent factors, actual percentage and predicted percentage as randomized by the Design Expert software (version 8) and the respective response obtained from the experiment.

Table 7: CCD matrix of factors and response of BOD percentage efficiency

Run	Factor1 Time hr	Factor2 Conc. Mg/l	Factor 3 Flow rate m3/hr	Actual %	Predicted %
1	4	200	4	36.6	37.0233
2	8	200	4	41.2	40.766
3	4	400	4	29	28.9729
4	8	400	4	32.3	31.5657
5	4	200	8	32.8	33.6125
6	8	200	8	36.1	36.2053
7	4	400	8	26	26.5122
8	8	400	8	28.3	27.9549
9	2.63641	300	6	30	29.0145
10	9.36350	300	6	32.5	33.3749
11	6	131.821	6	43.6	43.0983
12	6	468.179	6	29	29.3911
13	6	300	2.63641	33.2	33.6968
14	6	300	9.36350	28.4	27.7926
15	6	300	6	31.1	31.1032
16	6	300	6	31.1	31.1032
17	6	300	6	31.1	31.1032
18	6	300	6	31.1	31.1032
19	6	300	6	31.1	31.1032
20	6	300	6	31.1	31.1032

3.2.6 Development of model equation of BOD percentage efficiency

Table 8 and 9 showed that the quadratic model for TSS percentage removal efficiency response was aliased. It was also observed in the table that the quadratic model has relatively high standard deviation of 0.69 and relatively high R^2 (0.9864) in reasonable agreement with adjusted (0.9742). From table 9 the model F-value of 80.74 implies that model was significant. Values of "pro>F" less than 0.0500 or <0.0001 indicates that the model terms were significant. In this case A, B, C, B^2 were significant model terms. From table 9, it was evidence that the effects of contact time, concentration and flow rate were highly significant. Equations 3.5 and 3.6 indicate the quadratic model prior and after the elimination of insignificant factors.

$$\%BOD=3.10 +1.30A -4.08B -1.76C -0.29AB -0.29AC +0.24BC+0.032A^2 +1.82B^2 -0.13C^2 \quad (3.5)$$

$$\%BOD=3.10 +1.30A -4.08B -1.76C -0.29AB -0.29AC +0.24BC +1.82B^2 \quad (3.6)$$

The model equations selected were further evaluated using ANOVA component of the software. From table 9, response surface quadratic model for removal efficiency has F-value of 80.74 indicating that the model was significant. For the model terms, p-value less than 0.05 implies that model term was significant (37; 38; 39; 40; 41) and largest F-value signifies the model term having the most significant model significant effect on the response (42; 43). Values greater than 0.1000 indicate the model terms were not significant. The model term having the most significant effect on the response was B with F-value of 480.75

Table 8 Model Summary Statistic for BOD percentage efficiency

Source	Std. dev.	R-Squared	Adjusted R-Squared	Predicted R-Squared	PRESS
Linear	1.87	0.8397	0.8097	0.7255	95.41
2FI	2.04	0.8448	0.7732	0.6653	116.31
Quadratic	0.69	0.9864	0.9742	0.8968	35.85 Suggested
Cubic	0.071	0.9999	0.9997	0.6	6.75 Aliased

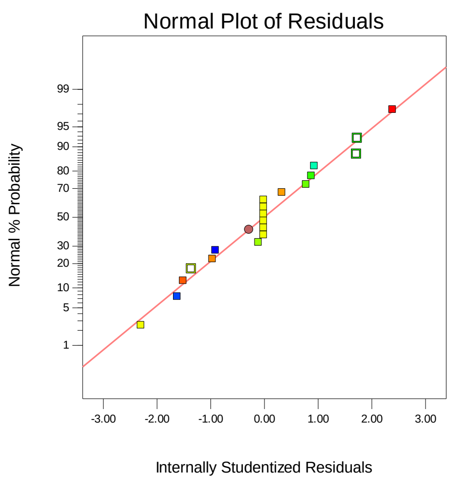
Table 9: Quadratic RSM ANOVA for the BOD percentage removal efficiency

Source	Sum of Squares	Df	Mean Square	F-Value	P-value. Prob>F
Model	342.81	9	38.09	80.74	<0.0001 Significant
A-Contact Time	22.95	1	22.95	48.65	<0.0001
B-Concentration	226.80	1	226.80	480.75	<0.0001
C-Flow rate	42.08	1	42.08	89.20	<0.0001
AB	0.66	1	0.66	1.400	0.2638
AC	0.66	1	0.66	1.400	0.2638
BC	0.45	1	0.45	0.96	0.3511
A^2	0.015	1	0.015	0.032	0.8616
B^2	47.62	1	47.62	100.94	<0.0001
C^2	0.23	1	0.23	0.49	0.4996
Residual	4.72	10	0.47		
Lack of Fit	4.72	5	0.94		

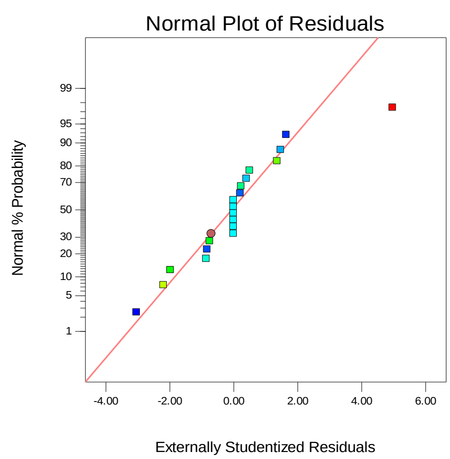
Pure Error	**0.000**	**5**	**0.000**		
Cor.Total	**347.53**	**19**			

<u>3.2.7 Comparison of actual (experimental) and predicted values of TSS, TDS and BOD percentage removal efficiency</u>

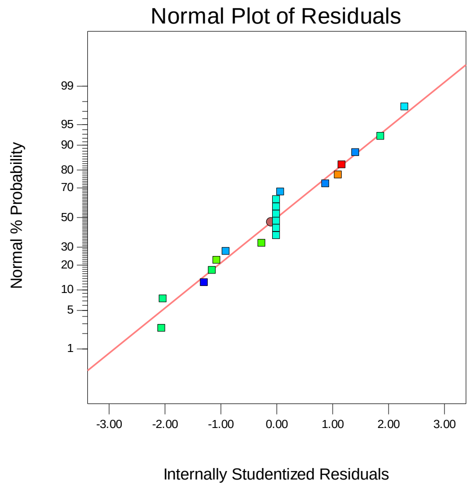
The plot of actual vs predicted values for response TSS, TDS and BOD percentage removal efficiency as observed in Fig 4a-c showed very minimal divergence of points from the diagonal indicating that response surface model equations could be used to adequately represent the interaction of the three factors. Figure 4a-c showed that the ANOVA results were valid because of normal distribution of the experimental data. Thus, the values of the response predicted from the model in TSS, TDS and BOD were in line with actual values over the range of the selected operating variables of contact time, concentration and flow rate with relatively high coefficient of determination R^2 (96.59), (92.09%) and (98.64%) respectively.



a



b



c

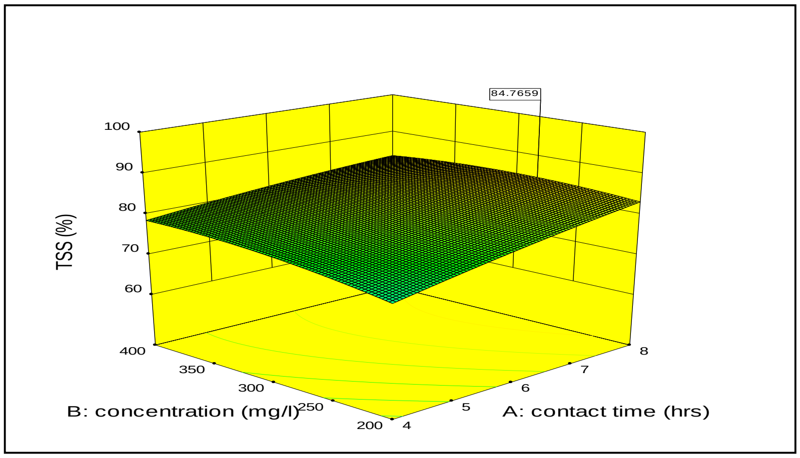
Fig 4 **(a-c) Normal probability plot of residuals for TSS, TDS and BOD**

3.3 Combined effect of variable independent factors

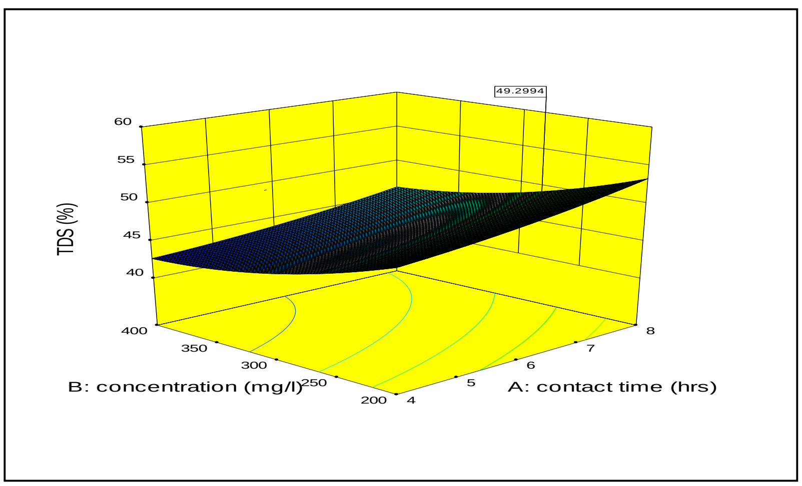
<u>3.3.1 Combined effect of Contact time and Concentration on TSS, TDS and BOD percentage removal efficiency</u>

The fig 5a, showed the relationship between contact time, concentration and removal percentage. The surface appears mostly flat, suggesting consistent removal efficiency across different concentrations and contact times. Some color variations indicate slight changes in removal efficiency. Therefore, there was increase in TSS percentage removal as the contact time increases at lower concentration. From the figure below, the predicted TSS percentage removal was 84.76%. From fig 5b, the surface plot showed a peak, indicating the optimal TDS percentage removal. That peak occurred at specific concentration levels and contact times. The yellow region of the surface slopes downward which suggested that as concentration of ciprofloxacin increases with contact time increases, the TDS percentage removal decreases. Also, as the concentration of ciprofloxacin decreases with contact time increases, the TDS percentage removal increases. The optimal TDS percentage removal efficiency was 49.2994%. In fig 5c, the peak of black plot was at contact

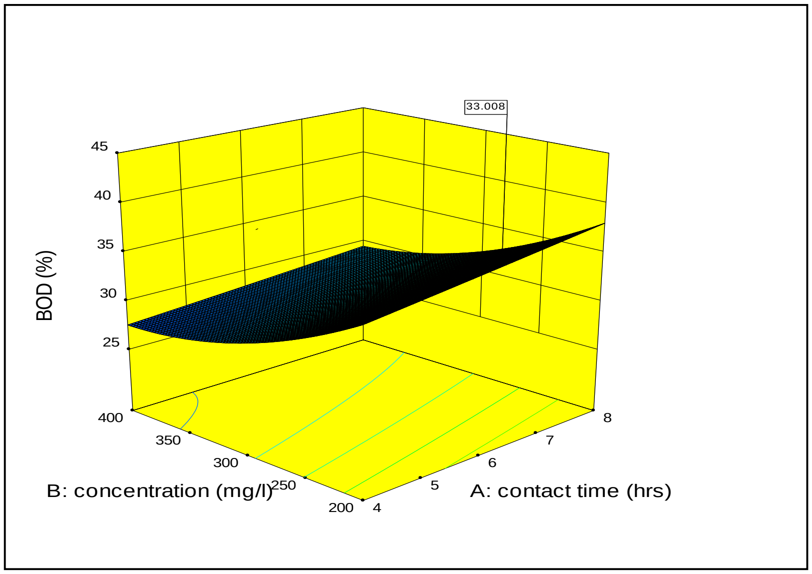
time of 8hrs, although the contact time does not show serious effect as the concentration does in the percentage efficiency of BOD. BOD percentage efficiency increases at contact time of 8hrs with decrease in concentration. The predicted BOD percentage efficiency was 33.008%.



a



b

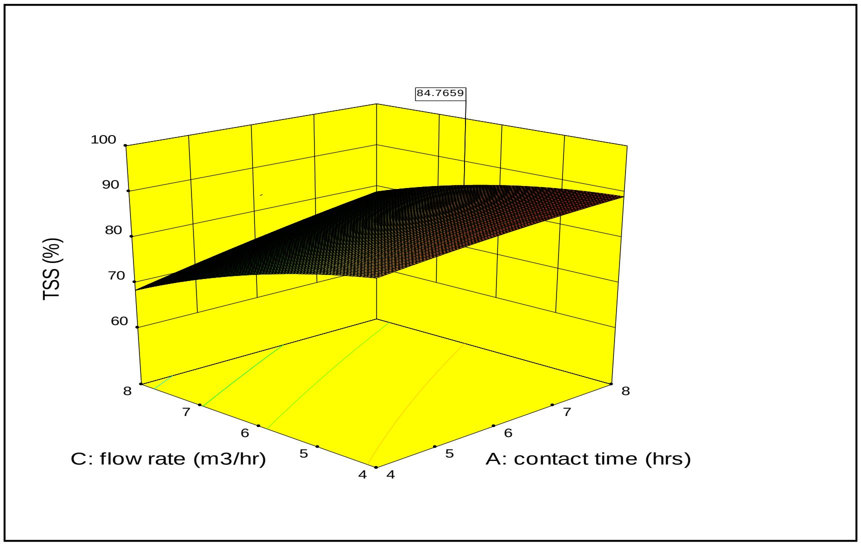


c

Fig 5 (a-c): Combined effect of contact time and conc. on TSS, TDS and BOD percentage removal efficiency

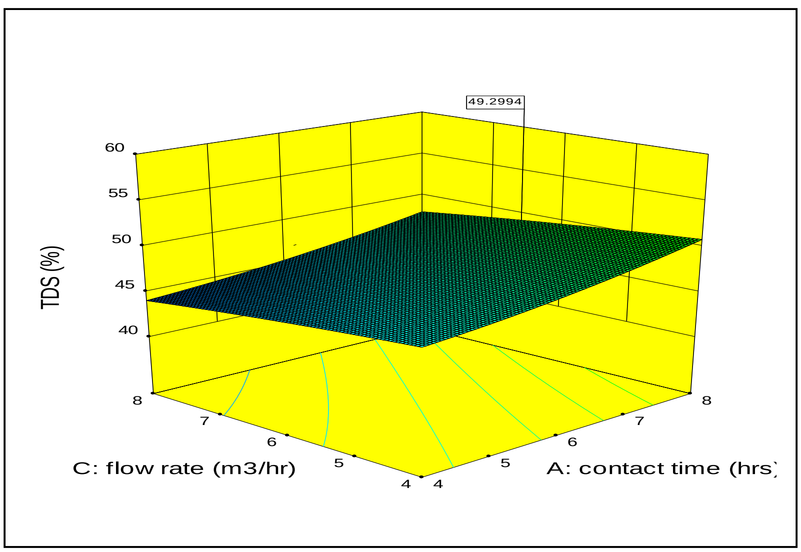
<u>3.3.2 Combined effect of Flow rate and Contact time on TSS percentage removal efficiency</u>

The fig 6a demonstrated how different flow rates and contact times affect the TSS percentage removal. The color transit from black at higher elevations to yellow at lower elevations. Therefore, there was increase in TSS percentage removal at flow rate of 6.4m3/hrs and contact time of 8hrs. The predicted TSS percentage removal was 84.7659%. In the figure 6b, the bars increased in height along the "C flow rate" axis (from left to right), indicating higher percentage removal of TDS with lower flow rate. That suggested that certain flow rates lead to better TDS removal. Interestingly, the bars along the "A contact time axis" increases at 8hrs. The predicted TDS percentage removal was 49.2994%. The figure 6c was a 3D graph, plotted on a three axis system. The x-axis was labelled " A: contact time (hrs), the y-axis was labelled "B:BOD% and the z-axis was labelled "C: flow rate (m3/hr). The surface plot in yellow represent some form of interaction or relationship between the variables' contact time; BOD; and flow rate. Therefore, the flow rate affected/ determined the increase in BOD percentage efficiency at 8hrs.



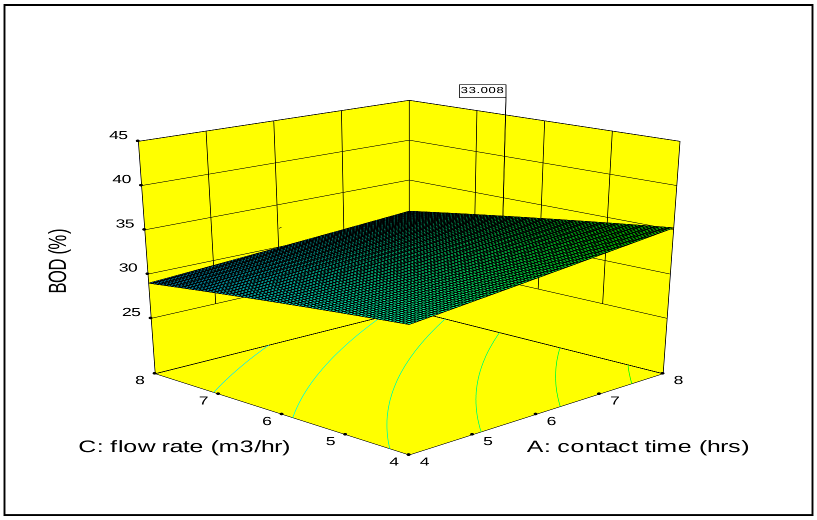
84.7659
100
90
80
70
60
TSS (%)
8
7
6
5
4
C: flow rate (m3/hr)
4
5
6
7
8
A: contact time (hrs)

a



49.2994
60
55
50
45
40
TDS (%)
8
7
6
5
4
C: flow rate (m3/hr)
4
5
6
7
8
A: contact time (hrs)

b

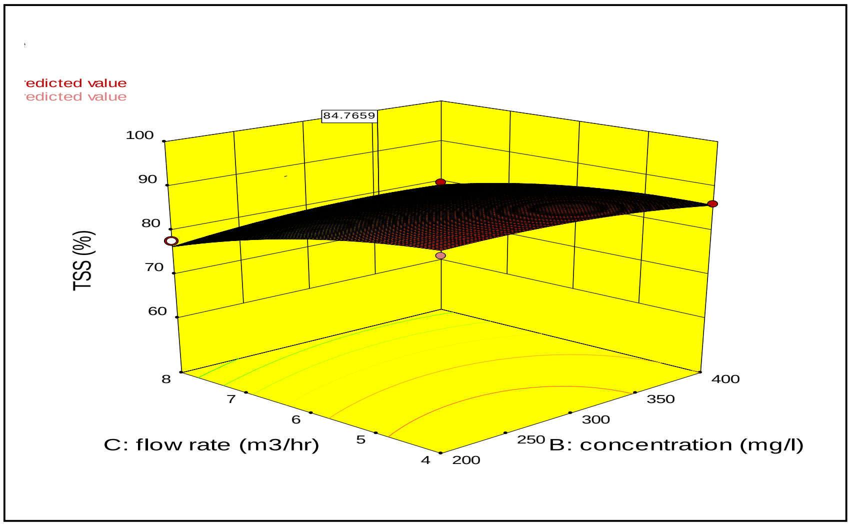


c

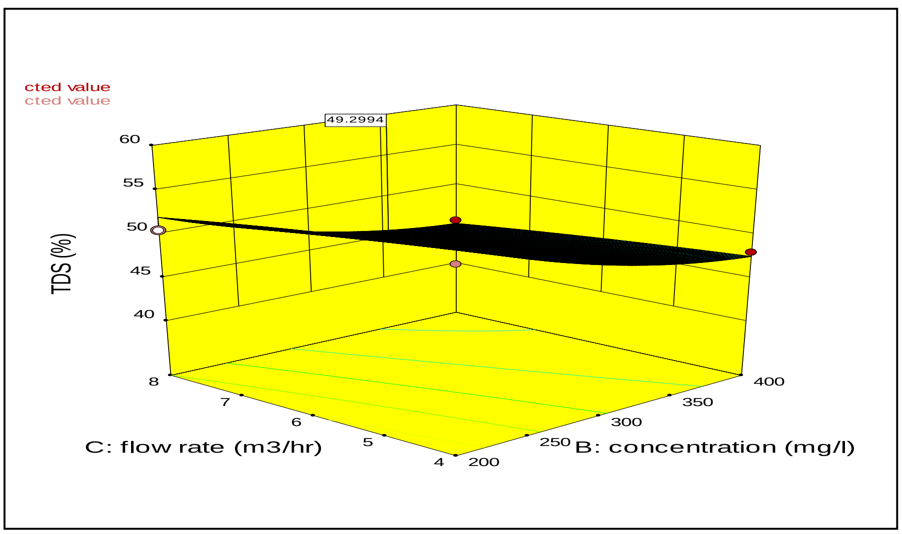
Fig 6 (a-c): Combined effect of flow rate and contact time for TSS percentage removal efficiency

<u>3.3.3 Combined effect of Flow rate and Concentration on TSS, TDS and BOD percentage removal efficiency</u>

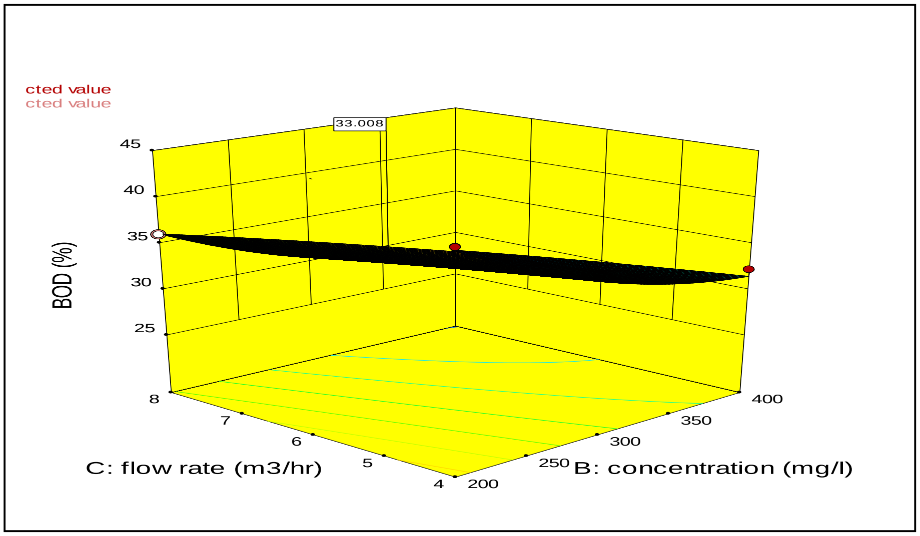
In fig 7a, the surface plots showed a peak, indicating the maximum removal percentage. That peak occurred around flow rate of 6.4m3/hrs and concentration of 277mg/l.. Below the peak, there was yellow shaded regions. That area represented significant removal percentages over flow rate and concentration. It suggested that certain combinations of flow rate and ciprofloxacin concentration lead to effective percentage removal of TSS. The optimum efficiency TDS removal occur at the predicted TDS percentage removal of 49.2994% by adjusting flow rate at 6.4m3/hrs and concentration of 277mg/l. Figure 7b showed that TDS percentage removal efficiency increases at lower concentration as the flow rates decreases. The figure 7c showed higher and lower predicted values. The optimum BOD efficiency was contribute mainly by the concentrations of ciprofloxacin, although the flow rates also contribute. The optimum BOD percentage efficiency was achieved at 277mg/l.


edicted value
edicted value
84.7659
100
90
80
70
60
TSS (%)
8
7
6
5
4
C: flow rate (m3/hr)
200
250
300
350
400
B: concentration (mg/l)

a


cted value
cted value
49.2994
60
55
50
45
40
TDS (%)
8
7
6
5
4
C: flow rate (m3/hr)
200
250
300
350
400
B: concentration (mg/l)

b



c

Fig 7 (a-c): Combined effect of conc. and flow rate on TSS, TDS and BOD percentage removal

3.4 Optimization

Optimization of three responses under the optimum operating conditions for the removal of ciprofloxacin from simulated pharmaceutical wastewater by application of modified palm fruit fiber using trickling filter system are:

Factors

A-Contact time =8hrs

B-Concentration =277.571mg/l

C-Flow rate =6.468m3/hr

Responses

TSS =84.766% (Actual =83.5%)

TDS =49.297% (Actual =47.3%)

BOD =33.005% (Actual =32.6%)

4. Conclusion

The study demonstrated that trickling filter systems, when optimized for key operational parameters, can effectively remove ciprofloxacin from simulated pharmaceutical wastewater. The application of response surface methodology proved valuable in identifying the optimal conditions for maximum removal efficiency. These findings suggested that trickling filters represent a viable option for enhancing the treatment of pharmaceutical wastewater.

DISCLAIMER

Author (s) hereby declare that NO generative AI technologies and text-to-image generators have been used during writing or editing of manuscripts.

COMPETING INTERESTS DISCLAIMER:
Authors have declared that they have no known competing financial interests OR non-financial interests OR personal relationships that could have appeared to influence the work reported in this paper.

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