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Dosage and settling time course optimization of *Moringa oleifera* in municipal wastewater treatment using response surface methodology

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ABSTRACT

The efficacy of the *Moringa oleifera* (MO) in wastewater treatment has been well studied and documented. However, there exists a significant gap in exploring the operating conditions to optimize the wastewater treatment process. This study investigated and optimized the removal efficiency of turbidity, biochemical oxygen demand (BOD), and chemical oxygen demand (COD) from municipal wastewater under the operating conditions of pH (5–7), MO dosage (50–200 mg/L), and settling time courses (60–240 min) using face-centered central composite design (FCCCD) of response surface methodology. The experimental results of FCCCD were fitted to the second-order quadratic model to approximate the effects of each variable factor and their interactions on the responses of interest in a mathematical relationship and consequently, predict the process responses. The obtained results revealed that under the optimum operating conditions of pH, MO dosage, and settling time of 6.01, 182.74 mg/L, and 228.08 min, respectively, the predicted values of turbidity, BOD, and COD removal efficiencies were 98.20%, 92.96%, and 78.82%, respectively, with the desirability of 1.000. This study demonstrated the effectiveness of FCCCD with a desirability function to optimize the process conditions (pH, MO dosage, and settling time) of coagulation for the turbidity, BOD, and COD removal efficiencies.

Keywords: Moringa oleifera; Wastewater treatment; Face-centered central composite design; RSM

1. Introduction

Globally, water pollution is a critical issue that has contributed to the adverse environmental and human health effects which demand keen attention [1,2]. The management and treatment technology of water needs to be affordable, effective, and sustainable to provide safe water in the developing world [3–5]. Although many water treatment techniques such as precipitation, ion exchange, electrodialysis, and membrane processes have been invented, most of them require higher expenditure and may also generate secondary waste which is difficult to treat [6]. A conventional technique of wastewater treatment involves the use of chemical coagulants such as aluminum sulphate and iron salts [7,8]. While the effectiveness of chemical coagulants has been well documented [9,10], there are, nonetheless,

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many side effects associated with usage of these coagulants. These include production of large quantity of sludge [11], relatively high procurement and operational costs [12], detrimental effects on human health [13], including a strong evidence linking aluminium-based coagulants to the development of Alzheimer's or other neurodegenerative diseases in human beings [14]. Thus, natural materials that are cost-effective and can efficiently be utilized within the environmentally permissible limit will provide a sustainable alternative for wastewater purification, specifically in the treatment of municipal wastewater with a high level of pollution before discharging into the environment. *Moringa oleifera* (MO) seeds are one of those coagulants, as they contain water-soluble extract (proteins) that can be applied either in the treatment of drinking water or wastewater [15–18].

Moringa oleifera is a versatile plant frequently referred to as the 'miracle tree' [19]. It is the most prevalent species, which grows rapidly in equatorial regions of the world where human health is at risk by unsuitable drinking water [20]. It has been employed extensively in the developing countries as a source of vegetable oil, as a medicinal plant, and for the treatment of hypertension and inflammation [21,22]. It has been the most favoured plant to grow in the surroundings of homes for its aesthetic beauty, as a fence and for providing shading. The MO seed has been found to exhibit coagulating characteristic for treatment of alkalinity, turbidity, hardness, and total dissolved solids [23], due to its biodegradable, non-toxic, and easy to use properties [24]. Numerous studies have shown the non-toxic effects of MO seed in wastewater treatment [25,26], and its effective coagulation properties and antimicrobial characteristics [27,28]. Through oil extraction from the MO seeds, the natural coagulant in the form of seed cake residue can be derived [29]. The derived cake can then be used directly in wastewater treatment without further processing MO [23]. The extracted part of MO seed inhibits the growth of coliforms and pathogens [29], reduces colour, turbidity, and microorganisms from raw waters [30,31], helps in water softening [28], and also in sludge conditioning [32]. The process implies the reduction of the disinfection requirements. The MO seeds also help to remove reactive dye [33], bacteria and fungi [29,34] and its pod act as sorbent for the removal of organics such as benzene and ethylbenzene [35], atrazine [36], and heavy metals such as lead, cadmium, manganese, copper, iron, magnesium, and zinc [35,37] as a result of being cheap and efficient in pollutants removal which ultimately reduce the exorbitant cost of water treatment. The utilization of a natural coagulant such as *Moringa oleifera* has demonstrated significant advantages over other processes of wastewater treatment including biological treatment process (aerobic and anaerobic) of wastewater. These advantages include simplicity in the treatment processes, low investment costs, relatively production of low volume of sludge, and the achievability of gravitational settling and clarification which are highly important in developing countries, especially in the rural areas [22,32,35]. Biological treatment of wastewater is often employed as a secondary treatment process to remove remaining pollutants after primary treatment [12,19]. In fact, biological wastewater treatments are often supplemented with other treatment processes including carbon filtration and chlorination.

As a common practice, coagulants including MO are applied in quantity experimentally determined by jar test. However, due to lack of a general standard method for conducting the jar test as pointed out by Ndabigengesere et al. [22], the operating conditions of the wastewater treatment using MO to maximize the target response has been opened for investigation. One of the statistical methods for investigating and optimizing the process parameters is response surface methodology (RSM), due to its sophistication to predicting the process responses using the most efficient variable factors interactions and best process time [38]. RSM has been successfully utilized to optimize some operating variable factors interaction of wastewater treatment using MO [39–42]. However, as important as the influence of settling time is on the removal efficiency of pollutants in wastewater treatment [43], it has rarely been considered for optimization as one of the variable operating factors in municipal wastewater treatment using MO coagulant. In fact, settling time of the flocs is considered to have high influence on the overall cost of removal efficiency of turbidity, chemical oxygen demand (COD), and biochemical oxygen demand (BOD) in coagulation–flocculation process [44]. Therefore, the optimal operating conditions of MO coagulant for the treatment of municipal wastewater need to be investigated to ensure a considerable improvement on the effectiveness and efficiency of pollutants removals from the wastewater before the final discharge into the environment or waterbodies. This study investigates and optimizes the operating conditions including pH, MO dosage, and settling time courses in municipal wastewater treatment to maximize the turbidity, BOD, and COD removal efficiency using the face-centered central composite design (FCCCD) of RSM with desirability function methodology.

2. Materials and methods

2.1. Sample preparation of MO seeds

The MO seeds used in this study were from matured dried pods of *Moringa oleifera* tree obtained from the Kwara State Ministry of Agriculture, Ilorin, Nigeria. MO pods (30–60 cm length) were checked for any crack before plucking. The process is to ensure that the seeds are appropriately matured and dried. The MO seeds were prepared using the method as described by Delelegn et al. [45] and the oil extraction from MO powder as proposed by Muyibi et al. [46]. Concisely, the matured dried pods were shelled to obtain the MO seeds, which were air-dried for 2 d to maintain uniform weight. The shells were removed from the seed kernels using a knife before they were ground into the powdered form using an electric grinder (Eurosonic Model ES-242) and the powered MO obtained was sieved using a sieve of 0.5 mm mesh screen size to get a fine powder of MO.

Oil was extracted from the 60 g weighed of MO powder mixed with 170 mL of hexane solvent, using an electrothermal Soxhlet extractor at 70°C. Extraction of oil from the MO powder is essential to enhancing the coagulation potential of the MO seed powder [46]. The resulting MO residue after the oil extraction of 35% w/w was oven-dried at 50°C overnight to obtain the MO cake. The MO cake was used in this study.

2.2. Wastewater sampling and characterization

The wastewater samples were collected from the sewer outlet of the main campus of the University of Ilorin, Nigeria, where wastewaters around the campus meet. The wastewater pollution at this outlet is considered very important owing to the enormous volume of effluents from the campus, and the only significant source of pollution of the nearby receiving stream. The samples were collected and transported to the laboratory within 30 min and subsequently characterized after 1 h of collection. The wastewater sampling and characterization (turbidity, BOD, and COD) were carried out following the Standard Methods for Water and Wastewater Examination [47]. The characteristics of municipal wastewater used in this study were presented in Table 1.

Table 1
Characteristics of municipal wastewater

Parameters	Value
pH	7.26 ± 1.09
Turbidity, NTU	76.74 ± 3.69
BOD, mg/L	147.45 ± 12.87
COD, mg/L	474.18 ± 11.35
TSS, mg/L	111.93 ± 15.23
Total solids	474.18 ± 10.09
Alkalinity, mg/L	385
Total hardness, mg/L	128.50 ± 4.36

2.3. Coagulation process and wastewater treatment

Jar test has been recognized as a widely used method to monitor coagulation–flocculation processes. However, depending on the needs of the individual test to be performed, there is no standard method for conducting the jar test [22]. The jar test used in our study was Janke and Kunkel (Lovibond ET 730) apparatus consisting of four beakers of 1,000 mL capacity each with four paddles rotation. Each of the beakers was filled with 1,000 mL of wastewater sample after which varying dosages (50, 125, and 200 mg/L) of de-oiled powder produced from the MO seeds were added to the beakers in order of their sizes, while the last beaker was kept as a control without any treatment. The pH of the wastewater sample was adjusted between the range of 5 and 7 using 0.5 M of NaOH or 0.5 M of HCl, after adding the MO dosage. The mixture of wastewater samples and MO dosage in the jar was stirred rapidly at 100 rpm for 2 min; followed by 20 min of gentle mixing at 40 rpm to aid in sludge formation. Subsequently, the samples were gently transferred and left to sediment without disturbance for 60, 150, and 240 min in the sedimentation cones (Imhoff). The sampling bottles of 250 mL were used for the collection of the treated wastewater at each settling interval. The supernatants formed were then decanted and analyzed for turbidity, COD, and BOD at each collection, in triplicate. The percentage removal of the contaminants was calculated using Eq. (1).

$$Y(\%) = \left[\frac{C_i - C_f}{C_i}\right] \times 100 \tag{1}$$

where Y is the pollutant removal efficiency; C_i and C_f are the initial and final concentrations of the pollutants in the wastewater, respectively.

Furthermore, the amount of sludge that settles at the bottom of the sedimentation cones was used for estimating the volume of the sludge generated at varying MO dosage and under different settling time at a pH value of 7 in the coagulation process.

2.4. Experimental design

The response surface methodology based FCCCD was applied to assess and optimize the interactions among the independent variables; pH (X_1), dosage of MO (X_2), and settling time (X_3), for the response processes; turbidity, BOD, and COD removals. The range values of the factor parameters used are presented in Table 2.

The FCCCD employed in this study consists of eight factorial points, six axial (star) points, and five replicates at the center point, with a total number of 19 experimental runs. Moreover, the FCCCD is suitable for fitting quadratic response surface and optimization of response processes [48–50]. The experimental response surface FCCCD data were fitted into the quadratic model to establish the relationship between the variable factors (X_1, X_2, and X_3) and the response (Y), using the generalized form of second-order multiple regression Eq. (2).

$$Y(\%) = \beta_0 + \sum_{i=1}^{n}\beta_i X_i + \sum_{i=1}^{n}\beta_{ii} X_i^2 + \sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\beta_{ij} X_i X_j \tag{2}$$

where Y (%) represents the response model (percentage turbidity, BOD, and COD removal); β_0, β_i, β_{ii}, β_{ij} are the coefficients for the intercept, linear term, quadratic term, and interaction effect term, respectively; X_i and X_j are the independent variables in coded form, n is the number of independent variables.

Table 2
Experimental independent variables with factor levels

Independent variables	Symbols	Range and levels		
		Low level (–1)	Center level (0)	High level (+1)
pH	X_1	6	7	8
Dosage, mg/L	X_2	50	125	200
Settling time, min	X_3	60	150	240

Statistical analysis in the form of analysis of variance (ANOVA) was employed to assess the interactions between the variable factors and their respective responses. The quality of fit of the second-order polynomial model was checked using the correlation coefficient (R^2) and the lack of fit value (*p*-value) at 95% confidence level. The fitted models were used to construct contour plots to predict the greater removal efficiency of the desired pollutants (turbidity, BOD, and COD) through the optimization of the variable parameters (pH, MO dosage, and settling time) within the specified value range. Design-Expert 11.1.2 (Stat-Ease Inc., Minneapolis, USA) statistical software package was used for all the analyses and graphical presentations of the results.

3. Results and discussion

3.1. Model development

The results of the FCCCD experimental data showing the number of runs and the associated factor variables (pH, MO dosage, and settling time), and responses (turbidity, BOD, and COD) are presented in Table 3. To select the appropriate model, from several available response models that describe the interactions of the variable factors and the process responses including linear, two-factor interaction (2FI), quadratic, and cubic, the model quality was evaluated using the statistical indices such as coefficient of determination (R^2, adjusted R^2, and predicted R^2) and lack-of-fit tests, as shown in Table 4. The R^2 and predicted R^2 values were 0.9921 and 0.9468 for turbidity, 0.9942 and 0.9603 for BOD, and 0.9818 and 0.9242 for COD, which are very close to 1, and observed reasonable agreement of difference of less than 0.2 between predicted R^2 values and the adjusted R^2 values of 0.9841, 0.9885, 0.9635 for turbidity, BOD, and COD removals, respectively, in each of the response quadratic model equations. Similarly, the adequate precision values of 39.92, 41.88, and 27.71 for turbidity, BOD, and COD removals, respectively, which are higher than 4, show the ability of each of the models to navigate the design space. These results, therefore, indicate the reliability of the experimental data and predictability of the models to estimate the turbidity, BOD, and COD removals [38,48]. It also shows that FCCCD models can explain 99%, 99%, and 98% of the total variations in turbidity, BOD, and COD removals. Furthermore, the lack-of-fit *F*-test, which describes the data variations around the developed model, was checked. The probability value (*p*-value) of lack-of-fit exceeding 0.05 signifies insignificant *p*-value and ability of the model to fit the experimental data accurately [51,52]. The statistically insignificant lack-of-fit *p*-values of 0.2192, 0.1064, and 0.3898 were observed for turbidity, BOD, and COD removals, respectively. This implies that the chances of lack-of-fit occurring due to noise are 22%, 11%, and 39% for turbidity, BOD, and COD removals, respectively. This further confirmed the suitability of the models to fit the experimental data, and also make good predictions. Hence, the quadratic model was found to give the best fit to the FCCCD experimental data, with good correlations between the variable factors and their respective responses. The final quadratic polynomial equations in coded units and real values that

Table 3
FCCCD matrix for variable factors and investigated responses

Run number	Coded variables			Actual variables			Responses					
	X_1	X_2	X_3	pH	Dosage (mg/L)	Settling time (min)	Turbidity removal (%)		BOD removal (%)		COD removal (%)	
							Measured	Predicted	Measured	Predicted	Measured	Predicted
1	–1	1	1	6	200	240	98.18	97.37	91.42	91.81	78.25	78.92
2	1	1	1	8	200	240	91.29	91.75	88.46	87.13	71.64	70.75
3	0	0	0	7	125	150	89.74	87.51	86.25	84.58	66.03	63.566
4	0	0	0	7	125	150	86.63	87.51	85.19	84.58	65.84	63.56
5	0	0	0	7	125	150	88.65	87.51	84.05	84.58	65.88	63.56
6	0	0	0	7	125	150	89.74	87.51	84.11	84.58	65.81	63.56
7	1	–1	–1	8	50	60	46.24	46.43	42.79	42.06	28.75	27.28
8	–1	–1	1	6	50	240	86.17	86.60	79.91	79.38	70.26	70.30
9	0	1	0	7	200	150	90.82	91.54	87.24	87.63	67.03	68.83
10	0	–1	0	7	50	150	74.51	76.27	72.38	73.32	53.21	54.61
11	1	–1	1	8	50	240	76.23	74.92	80.29	79.61	68.73	68.67
12	1	0	0	8	125	150	81.70	83.41	79.41	81.95	57.94	61.20
13	1	1	–1	8	200	60	67.26	66.21	58.04	58.24	47.96	47.12
14	0	0	–1	7	125	60	62.22	63.47	58.26	57.44	40.06	43.02
15	0	0	0	7	125	150	87.74	87.51	85.94	84.58	60.67	63.56
16	–1	0	0	6	125	150	90.21	90.98	86.51	85.30	69.39	69.33
17	–1	–1	–1	6	50	60	57.01	55.93	43.09	44.09	35.28	35.37
18	–1	1	–1	6	200	60	68.96	69.65	64.84	65.19	62.47	61.73
19	0	0	1	7	125	240	90.35	60.22	87.38	89.53	72.05	72.30

Table 4
Summary statistics of model selection for turbidity, BOD, and COD removals

Source	Standard deviation	R^2	Adjusted R^2	Predicted R^2	PRESS	Lack-of-fit p-value	Remark
Summary statistics for turbidity removal model							
Linear	7.34	0.7699	0.7239	0.6098	1,368.77	0.0014	
2FI	8.07	0.7770	0.6655	–0.3843	4,856.05	0.0008	
Quadratic	1.76	0.9921	0.9841	0.9468	186.60	0.2192	Suggested
Cubic	2.10	0.9937	0.9773	–4.5280	19,392.61	0.0449	Aliased
Summary statistics for BOD removal model							
Linear	8.45	0.7442	0.6931	0.5516	1,876.21	0.0003	
2FI	9.21	0.7567	0.6350	–0.6154	6,759.44	0.0002	
Quadratic	1.64	0.9942	0.9885	0.9603	165.94	0.1064	Suggested
Cubic	1.30	0.9980	0.9928	–0.3390	5,602.88	0.1103	Aliased
Summary statistics for COD removal model							
Linear	5.43	0.8644	0.8373	0.7448	830.97	0.0376	
2FI	4.49	0.9257	0.8886	0.7141	930.81	0.0677	
Quadratic	2.57	0.9818	0.9635	0.9242	246.70	0.3898	Suggested
Cubic	3.06	0.9857	0.9483	–8.9497	32,395.77	0.0996	Aliased

2FI, two-factor interaction.

express the empirical relationships between the factor variables and responses are presented in Eqs. (3)–(5).

$$Y_1 = 87.51 - 3.78X_1 + 7.63X_2 + 14.05X_3 + 1.52X_1X_2 - 0.54X_1X_3 - 0.74X_2X_3 - 0.31X_1^2 - 3.60X_2^2 - 9.98X_3^2 \quad (3)$$

$$Y_2 = 84.58 - 1.68X_1 + 7.15X_2 + 16.04X_3 - 1.23X_1X_2 + 0.57X_1X_3 - 2.16X_2X_3 - 0.95X_1^2 - 4.10X_2^2 - 11.09X_3^2 \quad (4)$$

$$Y_3 = 63.56 - 4.06X_1 + 7.11X_2 + 14.64X_3 - 1.63X_1X_2 + 1.61X_1X_3 - 4.44X_2X_3 + 1.70X_1^2 - 1.84X_2^2 - 5.91X_3^2 \quad (5)$$

where, Y_1, Y_2, and Y_3 are percentage removal of turbidity, BOD, and COD in coded units, respectively; X_1, X_2, and X_3 are pH, MO dosage, and settling time in coded units, respectively.

The adequacy of the developed quadratic models to replicate the approximation of the real systems was validated using diagnostic plots. The normal probability plots in Figs. 1a–c evaluate the residuals normality of the model distribution, while the predicted vs. actual values plots in Figs. 2a–c show the correlations of actual (experimental) values and predicted values in response to the turbidity, BOD, and COD removal. The clustering of data around the straight line in the plots of normal percentage probability values against the internally studentized residuals shows that the normality of the distribution is well defined and no variance deviation [53,54]. Similarly, the spread of the data points for each of the response in the plots of predicted vs. measured values (Figs. 3a–c) and their closeness to each other indicate satisfactory agreement between the experimental data and the predicted values for turbidity, BOD, and COD removals.

3.2. Analysis of variance

The statistical significance of model terms and their interactions was evaluated using the analysis of variance (ANOVA). The significance of the model coefficients for each of the three responses was assessed based on their corresponding F and p-values at 5% confidence level, as shown in Table 5. The ANOVA results revealed the high significance of the quadratic models, as evident from the large model F-values of 124.97, 172.42, and 53.85 for turbidity, BOD, and COD removals, respectively. Similarly, X_1, X_2, X_3, and X_3^2 are the common significant model terms with a p-value of less than 0.05 for turbidity, BOD, and COD removal models. Other significant model terms are X_1X_2 and X_2^2 for turbidity removal model, X_1X_3 and X_2^2 for BOD removal model, and X_2X_3 for COD removal model.

Furthermore, the values of the coefficient of variance (CV) that measure the reproducibility of the model are 2.19%, 2.15%, and 4.25% for turbidity, BOD, and COD removals, respectively. The CV value of less than 10% is considered appropriate for the reproducibility of any model [48]. The signal-to-noise ratio of the adequate precision measures for the response models were 39.92, 41.88, and 27.71 for turbidity, BOD, and COD removals. The adequate precision value higher than 4 is appropriate and shows that the regression model equation can be employed within the range of factors in the design space [49]. These two indices, CV and adequate precision, suggested that the models are reproducible, and have a high degree of reliability and accuracy of the experiments.

3.3. Coagulation process of variable factors on response parameters

The combined interaction effects of different levels of variable factors on the turbidity, BOD, and COD removals

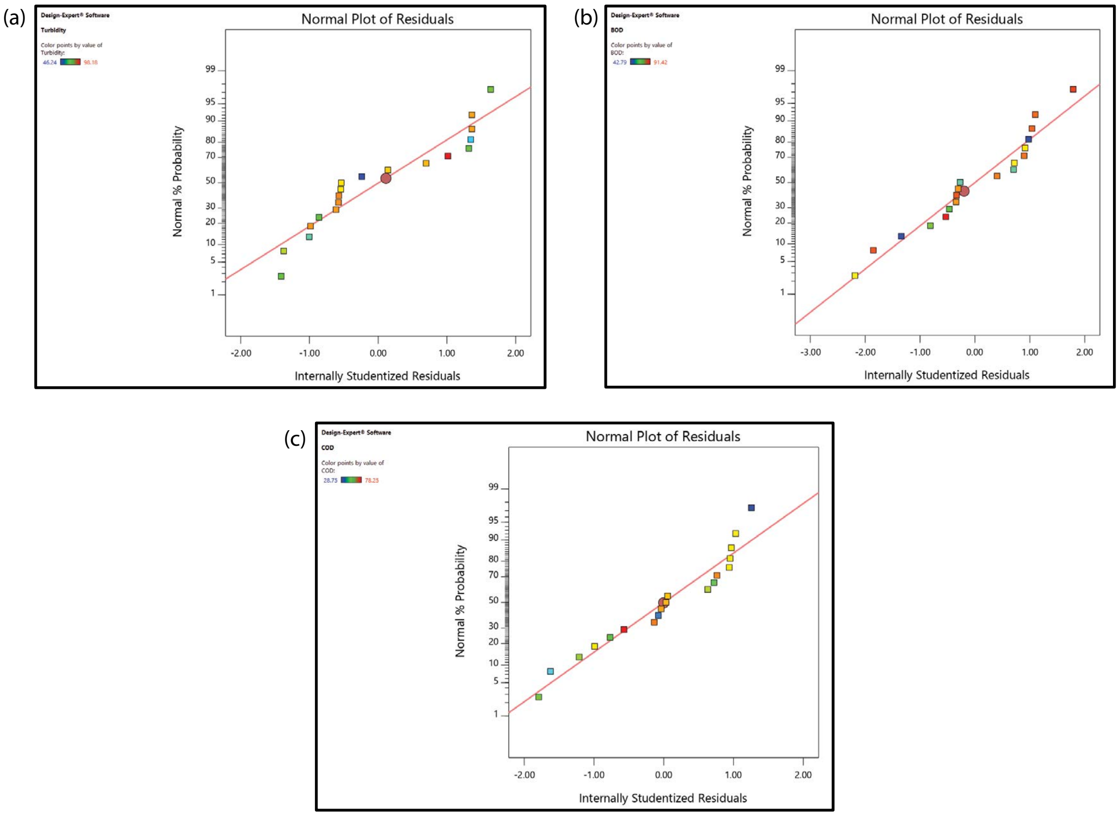

Fig. 1. Normal probability plot of the internally studentized residuals for (a) turbidity removal, (b) BOD removal, and (c) COD removal.

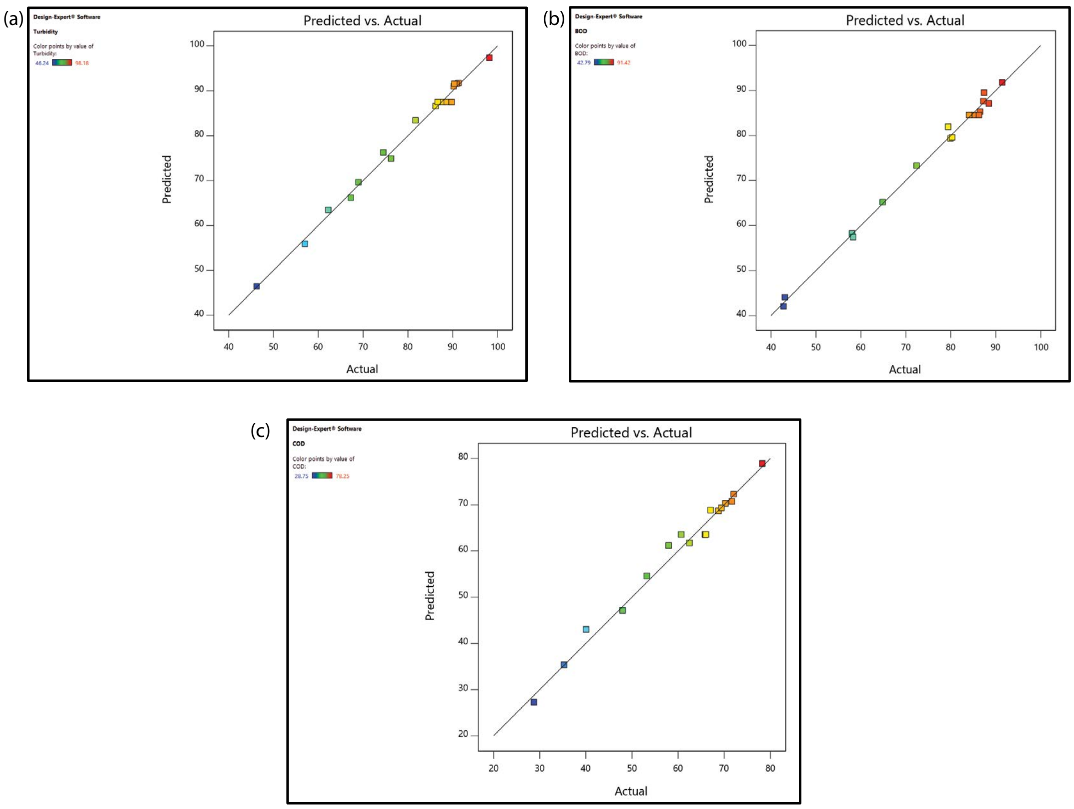

Fig. 2. Predicted vs. observed data plots for (a) turbidity removal (%), (b) BOD removal (%), and (c) COD removal (%).

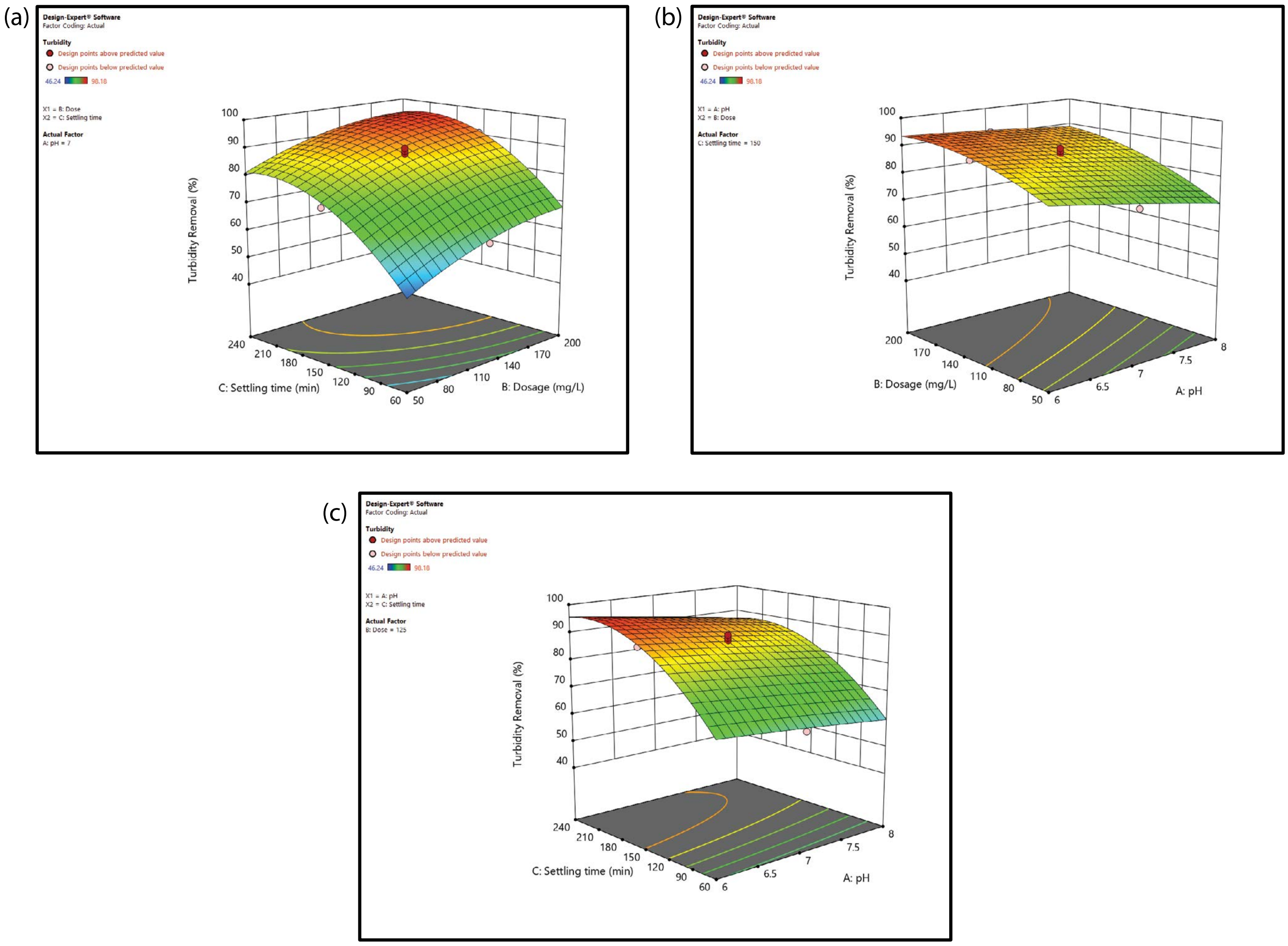

Fig. 3. Response 3D surface and contour plots for the effects of (a) dosage and settling time, (b) dosage and pH, and (c) settling time and pH on the turbidity removal.

from municipal wastewater were assessed using three-dimensional graphic plots. The 3D plots and the contour plots show variations in the removal of turbidity, BOD, and COD as a function of pH, MO dosage concentrations, and settling time as used during the experimentation. The response surface plots in Figs. 3–5 were generated by concurrent variations of the three variable factors (pH, dosage, and settling time) from 6 to 8, 50 to 200 mg/L, and 60 to 240 min, respectively.

The trends of removal efficiencies of turbidity, BOD, and COD against varying MO dosage and settling time at a fixed pH of 7 are shown in Figs. 3a, 4a, and 5a, respectively. The removal efficiencies were found to increase proportionally with increasing MO dosage. The removals of turbidity, BOD, and COD were notably enhanced as the settling time increased between 60 and 220 min. However, further increase in settling time indicated negligible effects. This phenomenon demonstrates that increase in coagulant dosage and settling time enhances the effectiveness of active coagulant compounds such as cationic peptides in the MO [54]. This leads to the formation of large, concentrated, and closely packed floc units through bridging mechanism [55]. Expressly, more quantity of MO with increased settling time provides more room for the effective treatment process, as such, enhances the removal efficiencies of turbidity, BOD, and COD. Adeniran et al. [56] investigated the treatment potential of bio-coagulant protein from MO seed. The authors found that the higher the quantity of extracted MO bio-coagulant protein applied to sewage water, the better the treatment efficiency of COD, and BOD removals. Camacho et al. [16] observed similar results in the removal of soil water turbidity using different dosages of MO (0–100 mg/L) at different settling time (0–120 min). Both dosage and settling time have significant effects on COD removal as obtained from a similar study conducted on paper mill effluent by Boulaadjoul et al. [57]. The COD removal from wastewater is associated with suspended organic materials [58]. Consequently, MO has the high potential to bind such organic materials together [57].

The combined effects of MO dosage and pH level on turbidity, BOD, and COD removal efficiencies, at a fixed settling time of 150 min are presented in Figs 3b, 4b, and 5b, respectively. The results show that the removal efficiencies of turbidity and COD were strongly associated with the MO dosage and pH value. However, the varying level of pH is less pronounced in the removal efficiency of BOD. The removal efficiencies of the three responses were found

Table 5
ANOVA results for turbidity, BOD, and COD removal models

Response	Source	Sum of squares	*df*	Mean square	*F*-value	*p*-value (Prob. > *F*)
Turbidity	Model	3,480.20	9	386.69	124.97	<0.0001
	X_1	142.96	1	142.96	46.20	<0.0001
	X_2	582.93	1	582.93	188.40	<0.0001
	X_3	1,974.87	1	1,974.87	638.25	<0.0001
	X_1X_2	18.36	1	18.36	5.93	0.0376
	X_1X_3	2.38	1	2.38	0.7680	0.4036
	X_2X_3	4.35	1	4.35	1.41	0.2660
	X_1^2	0.27	1	0.27	0.09	0.7749
	X_2^2	35.48	1	35.48	11.47	0.0080
	X_3^2	272.34	1	272.34	88.02	<0.0001
	Residual	27.85	9	3.09		
	Lack-of-fit	20.68	5	4.14	2.31	0.2192
	Pure error	7.17	4	1.79		
	Cor total	3,508.04	18			
BOD	Model	4,160.18	9	462.24	172.42	<0.0001
	X_1	28.16	1	28.16	10.50	<0.0001
	X_2	511.80	1	511.80	190.90	<0.0001
	X_3	2,574.10	1	2,574.10	960.14	<0.0001
	X_1X_2	12.10	1	12.10	4.51	0.0626
	X_1X_3	2.55	1	2.55	0.95	0.3546
	X_2X_3	37.50	1	37.50	13.99	0.0046
	X_1^2	2.46	1	2.46	0.92	0.3631
	X_2^2	45.91	1	45.91	17.12	0.0025
	X_3^2	335.98	1	335.98	125.32	<0.0001
	Residual	24.13	9	2.68		
	Lack-of-fit	20.01	5	4.00	3.89	0.1064
	Pure error	4.12	4	1.03		
	Cor total	4,184.31	18			
COD	Model	3,196.60	9	355.18	53.85	<0.0001
	X_1	165.08	1	165.08	25.03	0.0007
	X_2	505.81	1	505.81	76.69	<0.0001
	X_3	2,143.59	1	2,143.59	324.99	<0.0001
	X_1X_2	21.32	1	21.32	3.23	0.1057
	X_1X_3	20.80	1	20.80	3.15	0.1095
	X_2X_3	157.53	1	157.53	23.88	0.0009
	X_1^2	7.92	1	7.92	1.20	0.3016
	X_2^2	9.28	1	9.28	1.41	0.2660
	X_3^2	95.36	1	95.36	14.46	0.0042
	Residual	59.36	9	6.60		
	Lack-of-fit	37.53	5	7.51	1.38	0.3898
	Pure error	21.83	4	5.46		
	Cor total	3,255.96	18			

Parameters	Turbidity removal	BOD removal	COD removal
Standard deviation	1.76	1.64	2.57
Mean	80.19	76.08	60.38
Coefficient of variation (%)	2.19	2.15	4.25
PRESS	186.60	165.94	246.70
Adequate precision	39.92	41.88	27.71

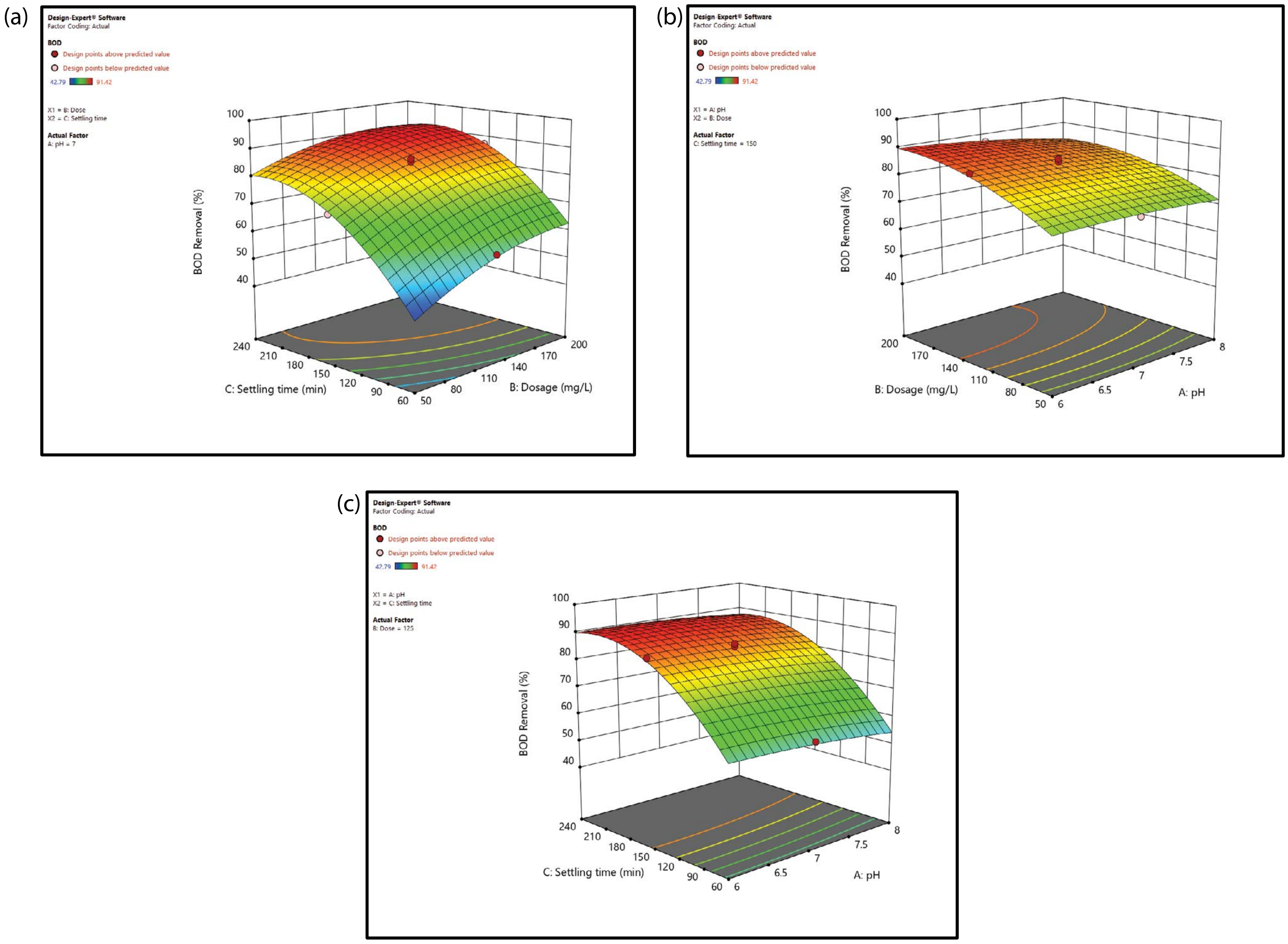

Fig. 4. Response 3D surface and contour plots for the effects of (a) dosage and settling time, (b) dosage and pH, and (c) settling time and pH on the BOD removal.

to increase with increasing MO dosage and decreasing pH level. The effect of pH was observed to be more significant at high level of MO dosage, hence maximum removals of turbidity, BOD, and COD occurred. Dotto et al. [59] reported that the highest contaminants reduction efficiency was achieved at a pH acidic medium of MO coagulant in the textile wastewater. Dehghani and Alizadeh [60] found the optimum pH of 6 for the removal efficiency of turbidity, COD, and total suspended solids using MO in refinery wastewater.

The effect of concurrent variations of pH and settling time on the removal of turbidity, BOD, and COD is presented in Figs. 3c, 4c, and 5c, respectively. The results indicate that an increase in settling time with lower pH value led to high efficiencies of turbidity, BOD, and COD removals. As the pH level decreases within the optimum range (6 to 8) required for coagulation process of MO [61], the positive charges tend to predominate on the surface of active molecules, thereby causing the formation of colloids and subsequent removal of contaminants [16]. These results are in good agreement with Boulaadjoul et al. [57]. The researchers reported that 97% of turbidity was removed at pH range of 6–8 after 30 min settling time in paper mill effluent. Al-Gheethi et al. [62] reported the need to allow more than 1 h settling time so as to enhance the treatment effect of MO under reduced pH value.

Furthermore, the volume of sludge production was found to vary with the coagulant dosage and settling time (Table 6). The increase in MO dosage from 50 to 200 mg/L, at a settling time of 60 min, resulted in a 15% reduction in sludge volume generated. However, for the same range of 50–200 mg/L dosage of MO, at a different settling time of 240 min, a 27% reduction of sludge volume was produced. Similarly, increasing MO dosage from 50 to 125 mg/L resulted in 18% reduction in sludge volume at a settling time of 240 min. However, 27% reduction in sludge volume was observed for 50–200 mg/L increase of MO dosage under the same settling time of 240 min. These results are in agreement with a study conducted by Muyibi et al. [63]. In their study, similar reductions in the volume of sludge were observed when the coagulant dosages were increased at constant settling time of 30 min.

3.4. Optimization of response parameters

The numerical optimization analysis of the process parameters was implemented using the Derringer's desirability function in the Design-Expert software. A desirability

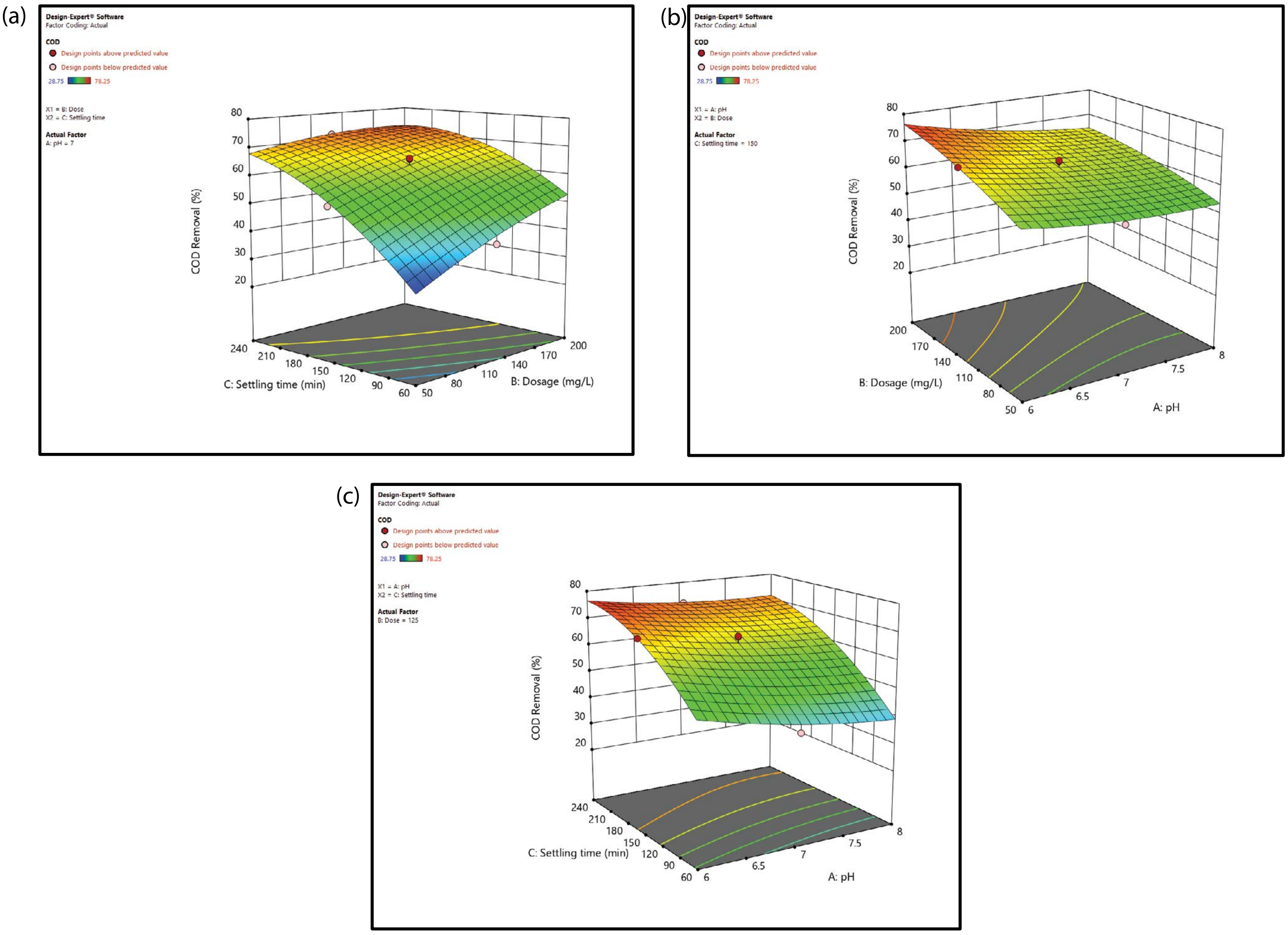

Fig. 5. Response 3D surface and contour plots for the effects of (a) dosage and settling time, (b) dosage and pH, and (c) settling time and pH on the COD removal.

Table 6
Sludge volumes with varying pH, coagulant dosage, and settling time

Moringa oleifera dosage (mg/L)	Settling time (min)	Volume of sludge (ml/L wastewater)
	60	5.2
50	150	4.9
	240	4.5
	60	4.7
125	150	4.3
	240	3.7
	60	4.4
200	150	3.9
	240	3.3

function is a technical approach in a statistical design that simultaneously measures the optimal settings of input parameters to produce optimum performance levels of one or more output variables or responses [64]. The desirability scale varies over the range of 0 to 1, with zero value indicating undesirable for the particular variable of concern and the value of one denotes completely desirable. Numerical optimization allows the selection of a desirable value in the form of a range, target, minimum, or maximum value for each input variable factor and response. For maximum desirability, pH, MO dosage, and settling time were set in the range of 6–8, 50–200 mg/L, and 60–240 min, respectively, with the maximum level of turbidity, BOD, and COD removals. Similar values within these ranges of pH, coagulant/flocculant dosage and settling time have been applied in the previous studies of wastewater treatments [34,65,66]. The FCCCD experimental results, with the stated defined set goal conditions, produced the optimum process conditions of pH, MO dosage, and settling time as 6.07, 189.69 mg/L, and 215.42 min, respectively, for maximum removal of turbidity, BOD, and COD. Under the selected conditions, the predicted removal efficiencies of turbidity, BOD, and COD were found to be 98.28, 93.48, and 78.52, respectively, with the desirability value of 1.000.

To validate the results of the optimization, a confirmation experiment, with three replications, using the optimum conditions obtained from the FCCCD model was conducted. The turbidity, BOD, and COD removal efficiencies at optimal process conditions were 97.37% ± 1.76%, 91.81% ± 1.63%, and 78.92% ± 2.57%, respectively. The results showed a strong agreement with the obtained predictive model values of 98.28%, 93.48%, and 78.52% for turbidity, BOD, and COD, respectively. The results demonstrated the effectiveness of FCCCD with desirability function to optimize the process conditions (MO dosage and settling time) of coagulation for the turbidity, BOD, and COD removal efficiencies.

4. Conclusion

This study explored the operating process conditions, namely: pH, dosage of MO, and treatment settling time in municipal wastewater treatment using FCCCD of response surface methodology. The combined effects of process variable factors on responses of interest, including turbidity, BOD, and COD removal efficiencies were investigated using FCCCD experimental results. The obtained results fitted well to the second-order polynomial regression model based on the coefficients of determination and lack-of-fit tests. Similarly, the statistical ANOVA showed that the two considered variable factors have both individual and combined influence on the three responses of interest. The optimum process settings to maximize the responses of turbidity, BOD, and COD removal efficiencies were achieved using the Derringer's desirability function. The optimum operating conditions with the desirability value of 1.000 were found to be 6.07 of pH, 189.69 mg/L of MO, and 215.42 min of settling time, which achieved the predicted 98.28%, 93.48%, and 78.52% of turbidity, BOD, and COD removals, respectively.

References

[1] R. Cheng, M. Kang, S. Zhuang, S. Wang, X. Zheng, X. Pan, L. Shi, J. Wang, Removal of bacteriophage f2 in water by Fe/Ni nanoparticles: optimization of Fe/Ni ratio and influencing factors, Sci. Total Environ., 649 (2019) 995–1003.

[2] O.S. Bello, B.M. Lasisi, O.J. Adigun, V. Ephraim, Scavenging Rhodamine B dye using *Moringa oleifera* seed pod, Chem. Speciation Bioavailability, 29 (2017) 120–134.

[3] A.N. Jones, J. Bridgeman, Disinfection ability of hibiscus seeds in water treatment, Proc. Inst. Civ. Eng. Water Manage., 171 (2018) 216–222.

[4] B. Xiong, B. Piechowicz, Z. Wang, R. Marinaro, E. Clement, T. Carlin, A. Uliana, M. Kumar, S.B. Velegol, *Moringa oleifera f*-sand filters for sustainable water purification, Environ. Sci. Technol. Lett., 5 (2018) 38–42.

[5] F.O. Ajibade, J.R. Adewumi, A.M. Oguntuase, Sustainable approach to wastewater management in the Federal University of Technology, Akure, Nigeria, Niger. J. Technol. Res., 9 (2014) 27-36.

[6] S. Ali, S.A.U. Rehman, H.Y. Luan, M.U. Farid, H. Huang, Challenges and opportunities in functional carbon nanotubes for membrane-based water treatment and desalination, Sci. Total Environ., 646 (2019) 1126–1139.

[7] H. Faghihian, E. Bahramian, Synthesis of polymeric Al-bentonite and polymeric Fe-bentonite for simultaneous removal of turbidity, Sr^{2+}, and Pb^{2+} from colloidal aqueous wastes, Chem. Eng. Commun., 201 (2014) 1417–1430.

[8] P.K. Gkotsis, E.L. Batsari, E.N. Peleka, A.K. Tolkou, A.I. Zouboulis, Fouling control in a lab-scale MBR system: comparison of several commercially applied coagulants, J. Environ. Manage., 203 (2017) 838–846.

[9] W. Chen, H. Zheng, H. Teng, Y. Wang, Y. Zhang, C. Zhao, Y. Liao, Enhanced coagulation-flocculation performance of iron-based coagulants: effects of PO_4^{3-} and SiO_3^{2-} modifiers, PLoS One, 10 (2015) 1–20.

[10] L. Liao, P. Zhang, Preparation and characterization of polyaluminum titanium silicate and its performance in the treatment of low-turbidity water, Processes, 6 (2018) 125.

[11] L. Semerjian, G.M. Ayoub, High-pH-magnesium coagulation-flocculation in wastewater treatment, Adv. Environ. Res., 7 (2003) 389–403.

[12] J. Keeley, P. Jarvis, S.J. Judd, Coagulant recovery from water treatment residuals: a review of applicable technologies, Crit. Rev. Env. Sci. Technol., 44 (2014) 2675–2719.

[13] F. Ghafar, A. Mohtar, N. Sapawe, N.N. Hadi, M.R.M. Salleh, Chemically modified *Moringa oleifera* seed husks as low cost adsorbent for removal of copper from aqueous solution, AIP Conference Proceedings, 1901 (2017), https://doi.org/10.1063/1.5010524.

[14] V. Rondeau, H. Jacqmin-Gadda, D. Commenges, C. Helmer, J.F. Dartigues, Aluminum and silica in drinking water and the risk of Alzheimer's disease or cognitive decline: findings from 15-year follow-up of the PAQUID cohort, Am. J. Epidemiol., 169 (2009) 489–496.

[15] G.A.P. Mateus, M.P. Paludo, T.R.T. dos Santos, M.F. Silva, L. Nishi, M.R. Fagundes-Klen, R.G. Gomes, R. Bergamasco, Obtaining drinking water using a magnetic coagulant composed of magnetite nanoparticles functionalized with *Moringa oleifera* seed extract, J. Environ. Chem. Eng., 6 (2018) 4084–4092.

[16] F.P. Camacho, V.S. Sousa, R. Bergamasco, M.R. Teixeira, The use of *Moringa oleifera* as a natural coagulant in surface water treatment, Chem. Eng. J., 313 (2017) 226–237.

[17] A.T.A. Baptista, M.O. Silva, R.G. Gomes, R. Bergamasco, M.F. Vieira, A.M.S. Vieira, Protein fractionation of seeds of *Moringa oleifera* lam and its application in superficial water treatment, Sep. Purif. Technol., 180 (2017) 114–124.

[18] K.A. Ghebremichael, K.R. Gunaratna, H. Henriksson, H. Brumer, G. Dalhammar, A simple purification and activity assay of the coagulant protein from *Moringa oleifera* seed, Water Res., 39 (2005) 2338–2344.

[19] M. Daba, Miracle tree: a review on multi-purposes of *Moringa oleifera* and its implication for climate change mitigation, J. Earth Sci. Clim. Change, 7 (2016) 3–8.

[20] S.D. Young, E.J. Joy, E.L. Ander, M.R. Broadley, D.W. Odee, D.B. Kumssa, Variation in the mineral element concentration of *Moringa oleifera* Lam. and M. stenopetala (Bak. f.) Cuf.: role in human nutrition, PLoS One, 12 (2017) e0175503.

[21] U.H. Joshi, T.H. Ganatra, P.N. Bhalodiya, T.R. Desai, P.R. Tirgar, Comparative review on harmless herbs with allopathic remedies as anti-hypertensive, Res. J. Pharm. Biol. Chem. Sci. Rev., 3 (2012) 673–687.

[22] A. Ndabigengesere, K.S. Narasiah, B.G. Talbot, Active agents and mechanism of coagulation of turbid waters using *Moringa oleifera*, Water Res., 29 (1995) 703–710.

[23] E. Arnoldsson, M. Bergman, N. Matsinhe, K. Persson, Assessment of drinking water treatment using *Moringa oleifera* natural coagulant, J. Water Manage. Resour., 64 (2008) 137–150.

[24] A.P. Meneghel, A.C. Gonçalves, F. Rubio, D.C. Dragunski, C.A. Lindino, L. Strey, Biosorption of cadmium from water using moringa (*Moringa oleifera* Lam.) seeds, Water Air Soil Pollut., 224 (2013) 1–13.

[25] V. Nand, M. Maata, K. Koshy, S. Sotheeswaran, Water Purification using *Moringa oleifera* and other locally available seeds in Fiji for heavy metal removal, Int. J. Appl. Sci. Technol., 2 (2012) 125–129.

[26] S.M. Mangale, S.G. Chonde, P.D. Raut, Use of *Moringa oleifera* (drumstick) seed as natural absorbent and an antimicrobial agent for ground water treatment, Res. J. Recent Sci., 1 (2012) 31–40.

[27] J. Sánchez-Martín, J. Beltrán-Heredia, J.A. Peres, Improvement of the flocculation process in water treatment by using *Moringa oleifera* seeds extract, Braz. J. Chem. Eng., 29 (2012) 495–501.

[28] S.A. Muyibi, L.M. Evison, Optimizing physical parameters affecting coagulation of turbid water with *Morninga oleifera* seeds, Water Res., 29 (1995) 2689–2695.

[29] N.A. Eman, C.S. Tan, E.A. Makky, N.A. Eman, Impact of *Moringa oleifera* cake residue application on waste water treatment: a case study, J. Water Resour. Prot., 6 (2014) 677–687.

[30] T.C. Shan, Manaf Al Matar, E. A. Makky, E.N. Ali, The use of *Moringa oleifera* seed as a natural coagulant for wastewater treatment and heavy metals removal, Appl. Water Sci., 7 (2017) 1369–1376.

[31] A.M.S. Vieira, M.F. Viera, G.F. Silva, Á.A. Araújo, M.R. Fagunles-Klen, M.T. Veit, R. Bergamasco, Use of *Moringa oleifera* seed as a natural adsorbent for wastewater treatment, Water Air Soil Pollut., 206 (2010) 273–281.

[32] J.O. Ademiluyi, Sludge conditioning with Moringa seed, Environ. Int., 14 (1988) 59–63.

[33] B.S. dos Santos, E. Eyng, P.R.S. Bittencourt, L.M. Frare, É.L. de M. Flores, M.B. Costa, Electro-flocculation associated with the extract of *Moringa oleifera* Lam as natural coagulant for the removal of reactive blue 5G dye, Acta Sci. Technol., 38 (2016) 483.

[34] C.M.L.L. Teixeira, F.V. Kirsten, P.C.N. Teixeira, Evaluation of *Moringa oleifera* seed flour as a flocculating agent for potential biodiesel producer microalgae, J. Appl. Phycol., 24 (2012) 557–563.

[35] Z. Shirani, C. Santhosh, J. Iqbal, A. Bhatnagar, Waste *Moringa oleifera* seed pods as green sorbent for efficient removal of toxic aquatic pollutants, J. Environ. Manage., 227 (2018) 95–106.

[36] I.M. Reck, R.M. Paixão, R. Bergamasco, M.F. Vieira, A.M.S. Vieira, Removal of tartrazine from aqueous solutions using adsorbents based on activated carbon and *Moringa oleifera* seeds, J. Cleaner Prod., 171 (2018) 85–97.

[37] F.O. Tavares, L.A. de M. Pinto, F. de J. Bassetti, M.F. Vieira, R. Bergamasco, A.M.S. Vieira, Environmentally friendly biosorbents (husks, pods and seeds) from *Moringa oleifera* for Pb(II) removal from contaminated water, Environ. Technol., 38 (2017) 3145–3155.

[38] M.H. Alhaji, K. Sanaullah, S.F. Lim, A.R.H. Rigit, A. Hamza, A. Khan, Modeling and optimization of photocatalytic treatment of pre-treated palm oil mill effluent (POME) in a UV/TiO_2 system using response surface methodology (RSM), Cogent Eng., 4 (2017) 1–17.

[39] I.R. Eri, W. Hadi, A. Slamet, Clarification of pharmaceutical wastewater with *Moringa Oleifera*: optimization through response surface methodology, J. Ecol. Eng., 19 (2018) 126–134.

[40] H.M. de Paula, M.S. de Oliveira Ilha, A.P. Sarmento, L.S. Andrade, Dosage optimization of *Moringa oleifera* seed and traditional chemical coagulants solutions for concrete plant wastewater treatment, J. Cleaner Prod., 174 (2018) 123–132.

[41] P.A. Bedekar, B.N. Bhalkar, S.M. Patil, S.P. Govindwar, *Moringa oleifera*-mediated coagulation of textile wastewater and its biodegradation using novel consortium-BBA grown on agricultural waste substratum, Environ. Sci. Pollut. Res., 23 (2016) 20963–20976.

[42] J. Beltrán-Heredia, J. Sánchez-Martín, A. Muñoz-Serrano, J.A. Peres, Towards overcoming TOC increase in wastewater treated with *Moringa oleifera* seed extract, Chem. Eng. J., 188 (2012) 40–46.

[43] K. Thirugnanasambandham, V. Sivakumar, J.P. Maran, S. Kandasamy, Chitosan based grey wastewater treatment — a statistical design approach, Carbohydr. Polym., 99 (2014) 593–600.

[44] M.I. Aguilar, J. Sáez, M. Llorens, A. Soler, J.F. Ortuño, V. Meseguer, A. Fuentes, Improvement of coagulation-flocculation process using anionic polyacrylamide as coagulant aid, Chemosphere, 58 (2005) 47–56.

[45] A. Delelegn, S. Sahile, A. Husen, Water purification and antibacterial efficacy of *Moringa oleifera* Lam, Agric. Food Secur., 7 (2018) 1–10.

[46] S.A. Muyibi, M.J.M.M. Noor, T.K. Leong, L. H. Loon, Effects of oil extraction from *Moringa oleifera* seeds on coagulation of turbid water, Int. J. Environ. Stud., 59 (2002) 243–254.

[47] APHA, Standard Methods for the Examination of Water and Wastewater, 22nd ed., APHA, AWWA, Water Environment Federation, Washington, D.C., 2012.

[48] Z. Salehi, F. Vahabzadeh, M. Sohrabi, S. Fatemi, H.T. Znad, Statistical medium optimization and biodegradative capacity of *Ralstonia eutropha* toward *p*-nitrophenol, Biodegradation, 21 (2010) 645–657.

[49] A.S. Dawood, Y. Li, Modeling and optimization of new flocculant dosage and pH for flocculation: removal of pollutants from wastewater, Water (Switzerland), 5 (2013) 342–355.

[50] M.M. Alam, M.Z. Alam, M.S. Jami, M.K. Amosa, M. Iwata, Study of the effects of process parameters on electroforced sedimentation in solid–liquid separation using response surface methodology, Waste Biomass Valorization, 7 (2016) 583–591.

[51] S. Demim, N. Drouiche, A. Aouabed, T. Benayad, O. Dendene-Badache, S. Semsari, Cadmium and nickel: assessment of the physiological effects and heavy metal removal using a response surface approach by *L. gibba*, Ecol. Eng., 61 (2013) 426–435.

[52] A.O.J. Cramer, D. van Ravenzwaaij, D. Matzke, H. Steingroever, R. Wetzels, R.P.P.P. Grasman, L.J. Waldorp, E. Wagenmakers, Hidden multiplicity in exploratory multiway ANOVA: prevalence and remedies, Psychon. Bull. Rev., 23 (2016) 640–647.

[53] S. Pal, S. Mukherjee, S. Ghosh, Optimum phenol sorption in peat by the response surface method, Environ. Geotech., 1 (2014) 142–151.

[54] K. Thirugnanasambandham, V. Sivakumar, J.P. Maran, Performance evaluation and optimization of electrocoagulation process to treat grey wastewater, Desal. Wat. Treat., 55 (2015) 1703–1711.

[55] L.O.R. Moreti, P.F. Coldebella, F.P. Camacho, M.C. Bongiovani, A.H.P. de Souza, A.K. Gohara, M. Matsushita, M.F. Silva, L. Nishi, R. Bergamasco, Removal of Anabaena flos-aquae in water treatment process using *Moringa oleifera* and assessment of fatty acid profile of generated sludge, Environ. Technol. (United Kingdom), 37 (2016) 1408–1417.

[56] K.A. Adeniran, T.D. Akpenpuun, B.A. Akinyemi, R.A. Wasiu, Effectiveness of *Moringa oleifera* seed as a coagulant in domestic wastewater treatment, Afr. J. Sci. Technol. Innovation Dev., 9 (2017) 323–328.

[57] S. Boulaadjoul, H. Zemmouri, Z. Bendjama, N. Drouiche, A novel use of *Moringa oleifera* seed powder in enhancing the primary treatment of paper mill effluent, Chemosphere, 206 (2018) 142–149.

[58] M.F. Hamoda, I. Al-Ghusain, N.Z. AL-Mutairi, Sand filtration of wastewater for tertiary treatment and water reuse, Desalination, 164 (2004) 203–211.

[59] J. Dotto, M.R. Fagundes-Klen, M.T. Veit, S.M. Palácio, R. Bergamasco, Performance of different coagulants in the coagulation/flocculation process of textile wastewater, J. Cleaner Prod., 208 (2019) 656–665.

[60] M. Dehghani, M.H. Alizadeh, The effects of the natural coagulant *Moringa oleifera* and alum in wastewater treatment at the Bandar Abbas Oil Refinery, Environ. Health Eng. Manage., 3 (2016) 225–230.

[61] N.T. Hoa, C.T. Hue, Enhanced water treatment by *Moringa oleifera* seeds extract as the bio-coagulant: role of the extraction method, J. Water Supply Res. Technol. AQUA, 67 (2018) 634–647.

[62] A. Al-gheethi, R. Mohamed, A. Wurochekke, N. Nurulainee, J.M. Rahayu, M.A. Hashim, Efficiency of *Moringa oleifera* water, MATEC Web of Conferences, EDP Sci., 103 (2017) 1–8.

[63] S.A. Muyibi, M.J.M.M. Noor, D.T. Ong, K.W. Kai, *Moringa oleifera* seeds as a flocculant in waste sludge treatment, Int. J. Environ. Stud., 58 (2001) 185–195.

[64] M. Mourabet, A. El Rhilassi, H. El Boujaady, M. Bennani-Ziatni, A. Taitai, Use of response surface methodology for optimization of fluoride adsorption in an aqueous solution by Brushite, Arabian J. Chem., 10 (2017) S3292–S3302.

[65] C. Kane, A. Bâ, S.A.M. Mahamat, N. Ayessou, M.K. Mbacké, C.G. Mar Diop, Combination of alum and extracted *Moringa oleifera* bioactive molecules powder for municipal wastewater treatment, Int. J. Biol. Chem. Sci., 10 (2017) 1918–1929.

[66] T.R.T. dos Santos, M.F. Silva, M.B. de Andrade, M.F. Vieira, R. Bergamasco, Magnetic coagulant based on *Moringa oleifera* seeds extract and super paramagnetic nanoparticles: optimization of operational conditions and reuse evaluation, Desal. Wat. Treat., 106 (2018) 226–237.