Desalination and Water Treatment
www.deswater.com
doi:10.5004/dwt.2017.20998

82 (2017) 26–38
July

Optimization of the operational parameters in a submerged membrane bioreactor using Box Behnken response surface methodology: membrane fouling control and effluent quality

Muhammad Bilal Asif[a,*], Rasikh Habib[a], Sidra Iftekhar[b], Zahiruddin Khan[c], Nadeem Majeed[d]

[a]*Department of Environmental Engineering, University of Engineering & Technology, Taxila-47050, Pakistan, Tel. +61 91 82850, email: mba409@uowmail.edu.au (M.B. Asif), rasikh.habib@uettaxila.edu.pk (R. Habib)*
[b]*Laboratory of Green Chemistry, School of Engineering Science, Lappeenranta University of Technology, Sammonkatu 12, FI-50130 Mikkeli, Finland, email: sidra.iftekhar@lut.fi (S. Iftekhar)*
[c]*Institute of Environmental Engineering and Research, University of Engineering and Technology, Lahore-54890, Pakistan, email: zahirkhan61@gmail.com (Z. Khan)*
[d]*Department of Software Engineering, University of Engineering & Technology, Taxila-47050, Pakistan, email: nadeem.majeed@uettaxila.edu.pk (N. Majeed)*

Received 20 November 2016; Accepted 31 May 2017

ABSTRACT

Various strategies have been assessed over the years to minimize membrane fouling in MBR. Backwashing and relaxation, the integral part of MBR operation, have been considered to be effective for the control of membrane fouling. In this study, Box–Behnken design (BBD) method was employed to investigate the combined influence of permeate flux, backwashing and relaxation duration on the performance of MBR. Based on the experimental results, quadratic models for effluent quality parameters, namely COD, NH_4–N, TN and TP removals were developed and the significance of these models was analyzed using the analysis of variance (ANOVA). Trans-membrane pressure (TMP) was monitored as the indicator of membrane fouling, and the increase in TMP was used to generate the regression model. Quadratic models based on the experimental results depicted that high permeate flux, backwashing and relaxation durations can negatively affect the performance of MBR. On the other hand, backwashing and relaxation durations were effective to minimize membrane fouling and the contribution of each tested variable was as follows: backwashing duration > permeate flux > relaxation duration. Since backwashing and relaxation durations negatively affected the removal of bulk organic and nutrients, optimization of these variable is vital for efficient performance of MBR. Based on the optimization of the variables, optimal backwashing and relaxation durations were 25 and 100 s, respectively, at a constant filtration duration of 8 min, while optimum permeate flux was 18 LMH.

Keywords: Box–Behnken design (BBD); Membrane fouling control; Optimization; Membrane bioreactor; Response surface methodology

1.Introduction

Membrane bioreactor (MBR) has been studied extensively over the years for wastewater treatment and reuse. MBR is superior to conventional wastewater treatment processes due to: (i) its compact size; (ii) exceptional quality of final product; and (iii) lower sludge yield [1–3]. However, membrane fouling has been the major constraint in the commercial applications of MBR because it abridges the life of membrane and also reduces the permeate flux [4–7]. Various physical and chemical fouling control strategies such as relaxation, backwashing, air scouring and chemical clean-

*Corresponding author.

ing have been studied over the last few years [1,8–11]. Since chemical cleaning and air scouring increase the operating cost of the MBR by up to 35 and 10%, respectively [12,13], relaxation, backwashing or combination of relaxation and backwashing seems to be an efficient as well as a cost-effective fouling minimization strategy.

Relaxation is the temporary cessation of membrane filtration that reduces the concentration of foulants on membrane surface [12,14,15]. It has been reported that short filtration time or frequent relaxation is better to minimize membrane fouling. However, frequent relaxation can cause critical fouling due to the high instantaneous flux [16,17]. Backwashing, another physical method to control membrane fouling, is a process in which permeate is reversed back into the membrane [18]. Backwashing is suitable to reduce internal fouling of membrane [19]. Backwashing durations can be divided in two categories: (i) long backwash duration but less frequent; and (ii) short backwash duration but more frequent [9,20,21]. Notably, backwashing with water or permeate has been observed to be more effective than aeration [22]. In addition, relaxation and backwashing durations has been optimized using a hit and trail method without incorporating the influence of other factors such as permeate flux (Supplementary Data Table S1 and S2).

Different experimental design approaches such as factorial design approach or fractional factorial models have been applied to optimize the operational parameters of wastewater treatment plants [23,24]. Since the influence of only two parameters can be assessed in fractional factorial models, it is vital to develop three level factorial models [1,25]. Response surface methodology (RSM), a statistical method for the development of an empirical model based on Box–Behnken design (BBD), can be an effective approach to: (i) evaluate the performance of composite systems; (ii) understand the interaction of parameters; (iii) analyze the relationship of an input with an output; and (iv) optimize the input parameters [26,27]. In addition, BBD is considered to be an efficient technique due to its rotatable design features [28].

BBD requires fewer experiments to study and analyze interaction of different independent variables and it has been successfully implemented to optimize different wastewater treatment processes [1,29,30]. For instance, Shim et al. [10] optimized the aeration rate as well as the size of beads for the physical cleaning of the membrane in MBR using BBD model. On the other hand, Fu el al. [1] optimized the effect of aeration rate, position and duration to minimize membrane fouling in MBR. As per our literature survey, effect of permeate flux, relaxation and backwashing durations on MBR performance and membrane fouling has not been evaluated or referred using the BBD model. Moreover, the studies outlined in Supplementary Data Table S1 and S2 reported the optimized relaxation and backwashing durations based on multiple experiments, and the experiment with less fouling was selected as the best option which is not a scientific approach.

In the view of the research gaps discussed above, the aim of this study was to analyze and evaluate the combined influence of different operational parameters such as relaxation, backwashing and permeate flux on effluent quality and membrane fouling. RSM using Box Behnken experimental design was used to evaluate and optimize the combined influence of different operational parameters. Since real domestic wastewater was used in this study, it is also expected that the optimized condition will serve as a reference for full scale installations of MBR.

2.Materials and methods

2.1. MBR Setup

Lab scale submerged MBR setup was installed to conduct this study as shown in supplementary Data Fig. S3. The MBR was comprised of a bioreactor (30 L) and a polyvinylidene difluoride (PVDF) hollow fiber membrane (Hinada, China) having a pore size of 0.2 μm and a surface area of 0.68 m^2. A sedimentation tank (60 min detention time) was provided to remove large suspended impurities from the real wastewater before its introduction into the bioreactor. Mixed liquor suspended solid (MLSS) concentration was kept at 8–9 g/L during all experiments. The bioreactor was also equipped with an air diffuser. Air diffuser was placed at the bottom of the membrane module to keep the dissolved oxygen concentration at >4 mg/L. A mechanical aerator was also provided to completely mix the contents of the bioreactor. The MBR was operated at the room temperature and a digital manometer (Model 840086, Sper Scientific, USA) was used to measure the trans-membrane pressure (TMP). A peristaltic pump (V-FLO, Model BT100M, China) was used to maintain the desired permeate flux during membrane filtration and backwashing.

2.2. Analytical methods

Domestic wastewater was collected from the disposal point located at the University of Engineering and Technology, Taxila. Composite samples of the domestic wastewater were tested for the determination of total suspended solids (TSS), total solids (TS), total dissolved solids (TDS), biological oxygen demand (BOD_5), chemical oxygen demand (COD), total nitrogen (TN), total phosphorous (TP), ammonium nitrogen (NH_4^+–N) and pH on daily basis as per the procedures described in the Standard Methods for the Examination of Water and Wastewater [31]. Characteristics of domestic wastewater are presented in Table 1. Perfor-

Table 1
Characteristics of domestic wastewater used in this study

Parameter	Nos.	Values ± standard-deviation
pH	90	7.5 ± 0.1
Total solids (TS), mg/L	90	850 ± 25
Total suspended solids (TSS), mg/L	90	140.5 ± 26
Biological oxygen demand (BOD_5), mg/L	90	260 ± 43
Chemical oxygen demand (COD), mg/L	90	398.6 ± 20
Ammonium nitrogen (NH_4^+–N), mg/L	90	31.9 ± 1.5
Total nitrogen (TN), mg/L	90	52.3 ± 2.1
Total phosphorus (TP), mg/L	90	9.1 ± 0.4

mance of the MBR was evaluated by measuring the permeate COD, TN, TP and NH_4^+–N on daily basis for a period of 5 d. Membrane fouling was measured based on TMP by recording its value at the start and the end of each run. The bioreactor was seeded with the activated sludge collected from I-9 sewage treatment plant (Islamabad, Pakistan) after the acclimatization period of 30 d. MLSS concentration in bioreactor was kept at 8–9 g/L.

2.3. Box Behnken experimental design

The impact of permeate flux, relaxation and backwashing durations on MBR performance and membrane fouling was examined at three levels (Table 2), based on the literature survey presented in Supplementary Data Table S1 and S2. Filtration duration (8 min) and backwashing flux (30 LMH) were kept constant during all experiments. Quadratic models were developed after 16 experiments *i.e.*, 12 trial experiments and 4-center experiments. Purpose of experiments at central point was to assess innate variability and stability of the process [28]. Furthermore, all the experiments were randomized to eliminate systematic errors (if any).

After each experiment, the membrane was cleaned using three step protocols; (i) cleaning with 40 mL deionized water; (ii) backwashing with 40 mL deionized water at a flux of 30 LMH; and (iii) desorbing in a 2% NaOH solution with effective chlorine strength of 2.5 g/L for 12 h.

Design Expert (7.0.0.), an experimental design software, was used to create, evaluate and optimize the experimental results. Responses were fit in a quadratic polynomial model as defined below:

$$Y = \beta_\circ + \sum \beta_i X_i + \sum \beta_{ij} X_i X_j + \sum \beta_{ii} X_{ii}^2 \quad (1)$$

In Eq. (1), predicted response and constant model coefficient are represented by Y and β_o, respectively. Linear, quadratic and interactional impacts of the model are expressed by β_i, β_{ii} and β_{ij}, respectively. All model coefficients were projected by multiple regression analysis. Coefficient of determination (R^2) was used to assess the fitting quality of polynomial equation(s).

Table 2
Description of independent variables used in BBD

Independent variables	Symbol	Levels		
		Low (–1)	Middle (0)	High (+1)
Permeate flux (LMH)	A	15	20	25
Backwashing period (s)	B	0	30	60
Relaxation period (s)	C	90	120	150

3. Results and discussion

3.1. Performance of MBR

To evaluate the performance of MBR, removals of COD, NH_4^+–N. TN and TP were measured on daily basis. Experimental results of these removals were put in the model to assess the predicted response (Table 3). Based on the experimental results, 2nd order quadratic equations for COD, NH_4^+–N, TN and TP removal efficiencies were developed as shown in Eqs. (2)–(5).

Table 3
Experimental and predicted removal efficiencies

Run	Variable			Experimental and predicted responses							
	A	B	C	COD Removal (%)		NH_4–N Removal (%)		TN Removal (%)		TP Removal (%)	
				Experimental	Predicted	Experimental	Predicted	Experimental	Predicted	Experimental	Predicted
E1	0	–1	–1	85.67 ± 2.1	86.16	72.6 ± 0.7	72.13	82.3 ± 1.4	82.59	64.1 ± 1.9	60.102
E2	–1	0	1	84.88 ± 1.1	84.99	74.4 ± 0.9	74.27	78.34 ± 1.7	78.92	62.67 ± 1.1	62.458
E3	0	0	0	81.03 ± 2.7	81.33	69.8 ± 0.45	69.45	76.4 ± 1.3	76.87	60.76 ± 1.2	60.93
E4	0	–1	1	83.07 ± 1.7	83.64	71.9 ± 1.3	72.13	81.89 ± 1.5	81.97	64.89 ± 1.25	61.058
E5	1	–1	0	80.05 ± 1.5	79.65	65.98 ± 1.5	66.31	78.4 ± 1.1	78.69	62.8 ± 1.8	59.27
E6	–1	0	–1	88.02 ± 3.3	88.21	73.45 ± 0.88	73.99	79.15 ± 0.8	79.52	62.2 ± 2.9	62.142
E7	0	0	0	81.57 ± 3.4	81.33	70.11 ± 1.5	69.45	76.45 ± 3.2	76.87	60.98 ± 1.3	60.93
E8	1	1	0	74.09 ± 4.1	74.77	63.44 ± 2.1	63.53	74.1 ± 2.7	74.77	58.13 ± 0.7	54.09
E9	1	0	1	74.95 ± 2.5	74.77	64.8 ± 1.4	64.25	75.4 ± 1.9	75.04	57.44 ± 2.7	57.498
E10	0	1	1	77.11 ± 2.65	76.62	67.89 ± 0.6	68.35	76.22 ± 1.1	75.93	58.9 ± 2.5	55.578
E11	0	0	0	80.77 ± 1.4	81.33	68.12 ± 0.65	69.45	77.68 ± 0.5	76.87	61.23 ± 1.4	60.93
E12	0	1	–1	82.07 ± 1.22	81.5	69.3 ± 2.2	69.07	77.67 ± 0.33	77.59	60.3 ± 1.9	56.822
E13	–1	–1	0	91.03 ± 2.45	90.35	76.4 ± 1.3	76.33	84.3 ± 1.4	83.63	66.23 ± 1.2	62.97
E14	1	0	–1	79.05 ± 3.1	78.95	65.11 ± 1.1	65.25	77.3 ± 1.7	76.72	57.88 ± 1.88	58.102
E15	0	0	0	81.97 ± 1.9	81.33	69.76 ± 0.95	69.45	76.97 ± 3.1	76.87	60.74 ± 2.5	60.93
E16	–1	1	0	83.17 ± 4.5	83.55	72.6 ± 0.87	72.27	76.8 ± 1.05	76.51	63.15 ± 0.4	59.39

$$Y_{COD} = +81.33 - 4.87A - 2.92B - 1.85C + 0.48AB -0.24AC - 0.59BC + 0.25A^2 + 0.50B^2 + 0.15C^2 \quad (2)$$

$$Y_{NH4-N} = +69.45 - 4.69A - 1.71B - 0.81C + 0.32AB -0.32AC - 0.18BC - 0.41A^2 + 0.57B^2 + 0.40C^2 \quad (3)$$

$$Y_{TN} = +76.87 - 1.67A - 2.76B - 0.57C + 0.80AB -0.27AC - 0.26BC - 0.22A^2 + 1.75B^2 + 0.90C^2 \quad (4)$$

$$Y_{TP} = +60.93 - 2.25A - 2.19B - 0.072C - 0.40AB -0.23AC - 0.55BC - 0.17A^2 - 1.83B^2 - 0.71C^2 \quad (5)$$

In Eqs. (2)-–(5), predicted removals of COD, NH_4^+–N, TN and TP are represented by Y_{COD}, Y_{NH_4-N}, Y_{TN} and Y_{TP}, respectively, while coded values of permeate flux (LMH), backwashing (s) and relaxation durations (s) are denoted by A, B and C, respectively. It can be observed from Eqs. (2)–(5) that all three independent variables have negative impact on performance of MBR. Similarly, the interaction of: (i) permeate flux with relaxation duration; and (ii) backwashing duration with relaxation duration negatively affected the removal of COD and nutrients.

Response surface 3D plots for COD, NH_4–N, TN and TP removal models as a function of permeate flux, backwashing and relaxation durations provide useful insight on the performance of MBR (Fig. 1). The influence of permeate flux and backwashing durations was significant on COD removal. COD removal increased with the decrease in backwashing duration, while it decreased with the increase in permeate flux (Fig. 1a-I). Similarly, the increase in relaxation durations and permeate flux also negatively affected COD removal (Fig. 1a-II). Moreover, increase in backwashing and relaxation durations reduced COD removal (Fig. 1a-III). Since hydraulic retention time (HRT) reduces with the increase in the permeate flux, backwashing and relaxation durations, slightly low COD can be expected [11,32].

On the other hand, the influence of permeate flux and backwashing duration was also significant on NH_4–N removal. NH_4–N removal increased with the decrease in backwashing period and it decreased with the increase in permeate flux (Fig. 1b-I). Similarly, the increase in the relaxation duration and permeate flux also negatively affected NH_4–N removal. However, there is no significant difference in minimum and maximum removal of NH_4–N (Fig. 1b). Notably, increase in permeate flux, relaxation and backwashing durations can deteriorate TN and TP removals in MBR (Fig. 1c,d). Tested variables and their interaction can negatively affect the performance of MBR because the time lost to relaxation and backwashing results in high instantaneous flux that results in short HRT and high organic loading rate. Indeed, Habib et al. [11] observed a 5–10% reduction in COD removal following an increase in the relaxation duration from 0 to 2 min. Similarly, Mohd et al. [33] achieved approximately 5 and 15% less removal of COD and TP, respectively, at a HRT of 4 h compared to that achieved at a HRT of 12 h. In another study by Wang et al. [32], TP removal was reduced by 30% at a low HRT of 6 h compared to TP removal observed at 8 h.

Backwashing has been reported to effectively minimize the formation of cake-layer on the surface of the membrane [19]. It may adversely affect the growth of slow-growing autotrophic bacteria (*e.g.* ammonia oxidizing bacteria), that required anoxic/anaerobic conditions, because their abundance was reported to be higher (60%) in the inner part of the cake layer compared to the bulk activated sludge [34]. In line with this, quadratic models correctly predicted the negative influence of backwashing duration on the removal of NH_4^+–N, TN and TP (Fig. 1).

3.1.1. Statistical analysis of models

Impacts and statistical significance of independent variable and their interactions on responses (Table 4) were analyzed by using the coefficient of determination (R^2) and the F-test [10,35]. The regression models of COD, NH_4–N, TN and TP were highly significant (p-value < 0.01). Significance of all quadratic models can also be confirmed by comparing the lack of fit and the pure error (Table 4). It was observed that the lack of fit was greater than 0.05, meaning that all models were statistically significant (Table 4). Similarly, adjusted R^2 of TP (0.977), COD (0.972), NH_4–N (0.954) and TN (0.925) removals further validated the significance of the model and suggested that the quadratic models could not explain only 2.26, 2.83, 4.62 and 7.52% variations, respectively [36].

Adjusted R^2 for each model was compared with coefficient of determination (R^2) and a strong correlation was found. Correlation between R^2 and adjusted R^2 indicates the presence of insignificant terms in all quadratic models [37]. R values should be close to unity for better correlation between experimental and predicted results [38,39]. Coefficient of determination (R^2) for COD, NH_4–N, TN and TP removal were 98.87, 98.15, 96.99 and 99.10%, respectively. Thus, it can be concluded that experimental and predicted results were in agreement.

p-values provide information about the significance of each coefficient and their interaction. A coefficient is significant if the p-value is less than 0.05, while p-value less than 0.01 is for extremely significant coefficient [37]. For COD and TN removal models, all the variables (A, B and C) showed significant impact *i.e.*, p < 005. On the other hand, independent variable B was not significant for NH_4–N and TP removal models (Table 4). Interaction of A with B, A with C and B with C were also not significant for COD, TP and TN removal models, respectively. Notably, linear, cubic and 2FI models were also significant (Table 4).

It has been recommended that the adequate precision (AP), a ratio of predicted results and the standard deviation among predicted results, must be greater than 4 to ensure model significance and authority [37,40]. In this study, AP values for TP, COD, NH_4–N and TN removal models were 30.14, 26.55, 20.27 and 15.25, respectively (Table 4). Coefficient of variance (CV) indicates the reproducibility of the model and its value of less than 10% is desirable [40]. All the models in this study were reproducible because CV values were 0.62, 0.91, 0.94 and 1.14% for TP, COD, TN and NH_4–N models, respectively.

Probability distribution plots of residuals as well as plots of predicted and actual results are good indicators to check the significance and adequacy of any model [41]. In this study, probability plots (Supplementary Data Fig. S4 and S5) suggested that the predicted and experimental results were in strong agreement for all models, thereby validating the significance of quadratic models presented in Eq. (2-5).

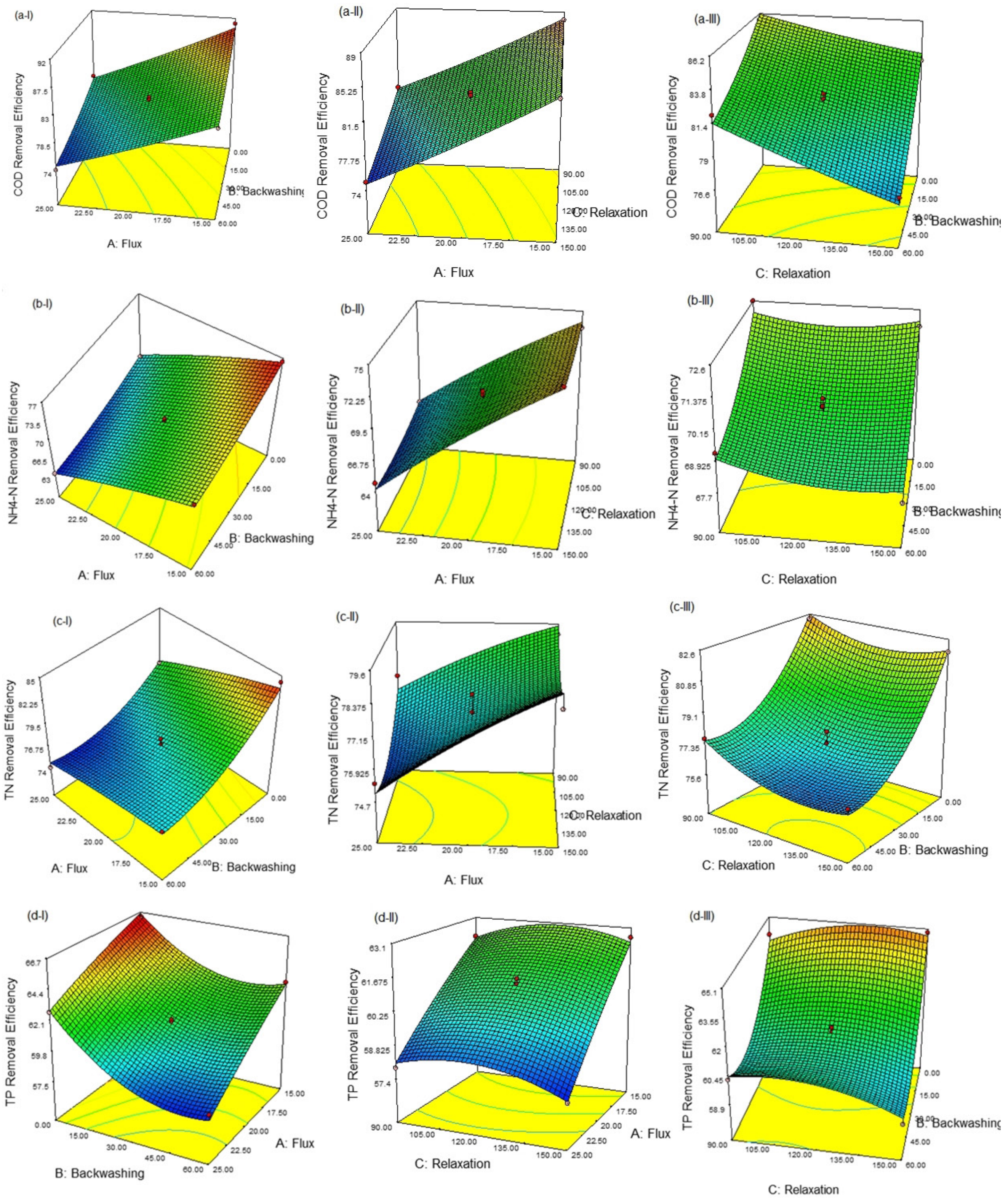

Fig. 1. Response Surface 3D plots for the removal of: (a) COD; (b) NH_4–N; (c) TN; and (d) TPas a function of: (I) permeate flux and backwashing durations at a relaxation duration of 120 s; (II) permeate flux and relaxation durations at a back washing duration of 30 s; and (III) backwashing and relaxation durations at a permeate Flux of 20 LMH.

Table 4
Analysis of variance (ANOVA) for Y_{COD}, Y_{NH_4-N}, Y_{TN} and Y_{TP}

Variables	Y_{COD}					Y_{NH_4-N}					Y_{TN}					Y_{TP}				
	SS	df	MS	F	p	SS	df	MS	F	p	SS	df	MS	F	p	SS	df	MS	F	p
A – Permeate flux	189.579	1	189.579	344.04*	0.000	175.969	1.000	175.969	276.23*	0.000	22.412	1.000	22.412	41.383**	0.0007	40.500	1.000	40.500	276.097*	0.000
B – Backwash period	68.363	1	68.363	124.06*	0.000	23.290	1.000	23.290	36.561*	0.0009	61.051	1.000	61.051	112.732*	0.0001	38.456	1.000	38.456	262.166*	0.000
C – Relaxation period	27.358	1	27.358	49.65*	0.0004	0.270	1.000	0.270	0.424^{NS}	0.5391	2.611	1.000	2.611	4.821**	0.0705	0.042	1.000	0.042	0.287^{NS}	0.6116
AB	0.908	1	0.908	1.65^{NS}	0.2466	0.397	1.000	0.397	0.623**	0.0460	2.560	1.000	2.560	4.727**	0.0726	0.632	1.000	0.632	4.309**	0.0832
AC	0.231	1	0.231	0.42**	0.041	0.397	1.000	0.397	0.623**	0.0460	0.297	1.000	0.297	0.548**	0.0569	0.207	1.000	0.207	1.411^{NS}	0.2797
BC	1.388	1	1.388	2.52**	0.0163	0.126	1.000	0.126	0.198^{NS}	0.6721	0.270	1.000	0.270	0.499^{NS}	0.5063	1.199	1.000	1.199	8.174**	0.0288
A^2	0.246	1	0.246	0.45**	0.0593	0.681	1.000	0.681	1.068^{NS}	0.3412	0.200	1.000	0.200	0.370^{NS}	0.5654	0.122	1.000	0.122	0.835^{NS}	0.3960
B^2	1.013	1	1.013	1.84^{NS}	0.2239	1.300	1.000	1.300	2.040^{NS}	0.2031	12.233	1.000	12.233	22.587*	0.0032	13.323	1.000	13.323	90.822*	0.000
C^2	0.084	1	0.084	0.15^{NS}	0.7091	0.656	1.000	0.656	1.030^{NS}	0.3493	3.213	1.000	3.213	5.933**	0.0508	1.988	1.000	1.988	13.553**	0.0103
Model	289.171	9	32.130	58.31*	0.000	203.085	9.000	22.565	35.422*	0.0002	104.847	9.000	11.650	21.511*	0.0007	96.470	9.000	10.719	73.073*	0.000
Linear	285.300	3	95.100	159.020*	0.000	199.529	3.000	66.510	108.170*	0.000	86.073	3.000	28.691	15.634*	0.0002	78.999	3.000	26.333	17.219*	0.000
Cubic	291.610	12.000	24.301	84.079*	0.0019	204.485	12.000	17.040	21.098**	0.0144	107.033	12.000	8.919	25.165**	0.0111	97.192	12.000	8.099	154.298*	0.0008
2FI	287.828	6.000	47.971	92.863*	0.000	200.449	6.000	33.408	46.555*	0.000	89.201	6.000	14.867	7.081**	0.0051	81.037	6.000	13.506	7.451**	0.0043
Residual	3.306	6.000	0.551	–	–	3.822	6.000	0.637	–	–	3.249	6.000	0.542	–	–	0.880	6.000	0.147	–	–
Lack of fit	2.439	3.000	0.813	2.813^{NS}	0.2091	1.399	3.000	0.466	0.577^{NS}	0.6685	2.186	3.000	0.729	2.056^{NS}	0.2845	0.723	3.000	0.241	4.589^{NS}	0.1214
Pure error	0.867	3.000	0.289	–	–	2.423	3.000	0.808	–	–	1.063	3.000	0.354	–	–	0.157	3.000	0.052	–	–
Core error	292.477	15.000		–	–	206.908	15.000		–	–	108.096	15.000		–	–	97.350	15.000		–	–
Adequate precision	26.554					20.274					15.250					30.137				
CV (%)	0.91					1.14					0.94					0.62				

SS: sum of squares; df: degree of freedom; MS: mean square; CV: coefficient of variance
*significant at 1% level, **significant at 5% level, NS: not significant
"–": not applicable

3.2. Membrane fouling

Effect of permeate flux, backwashing and relaxation durations was assessed and TMP was monitored as an indicator of membrane fouling (Table 5). Based on the experiment, a 2nd order quadratic equation was established accordingly as follows;

$$Y_{TMP} = +16.73 + 1.03A - 5.09B - 0.34C + 0.00AB \\ +0.15AC + 0.38BC - 0.55A^2 + 4.32B^2 + 0.37C^2 \quad (6)$$

Table 5
Experimental and predicted trans-membrane pressure

Run	Variable			Experimental and predicted responses	
	A	B	C	TMP (KPa)	
				Experimental	Predicted
E1	0	–1	–1	27.00	27.23
E2	–1	0	1	15.00	15.03
E3	0	0	0	16.80	16.73
E4	0	–1	1	25.60	25.79
E5	1	–1	0	26.80	26.62
E6	–1	0	–1	16.00	16.01
E7	0	0	0	16.56	16.73
E8	1	1	0	16.20	16.44
E9	1	0	1	17.40	17.39
E10	0	1	1	16.60	16.37
E11	0	0	0	17.20	16.73
E12	0	1	–1	16.50	16.29
E13	–1	–1	0	24.80	24.56
E14	1	0	–1	17.80	17.77
E15	0	0	0	16.36	16.73
E16	–1	1	0	14.20	14.38

In above equation, predicted TMP is represented by Y_{TMP}, whereas coded values of operational parameters such as permeate flux (LMH), backwashing (s) and relaxation durations (s) are denoted by A, B and C, respectively. It can be assessed from Eq. (6) that the increase in permeate flux showed negative impact on membrane fouling. High permeate flux has been linked with rapid membrane fouling [42,43]. In line with previous studies[8,11,19], backwashing and relaxation exhibited positive impact on membrane fouling. On the other hand, the interaction of permeate flux with backwashing period did not show any influence on membrane fouling.

The significance of the TMP quadratic model was analyzed using F-test and ANOVA (Table 6). The regression model of TMP was observed to be highly significant ($p < 0.01$). Adjusted R^2 (99.36%) and R^2 (99.74%) values were compared and found to be in good agreement. It was also observed that only 0.64% variations cannot be explained by TMP model. p-values can be used to further validate the significance of model coefficients and variables. All the regression variables were significant except for the interaction of flux and backwashing. Based on the results (Table 6), backwashing duration was the most influencing variable to control membrane fouling compared to other parameters. Influence of independent variables on TMP was as follows: backwashing duration (B) >permeate flux (A) >relaxation

Table 6
Statistical analysis (ANOVA) of TMP model

Variables	YTMP				
	SS	df	MS	F	p
A – Permeate flux	8.405	1.000	8.405	66.434*	0.0002
B – Backwash period	207.061	1.000	207.061	1636.632*	0.000
C – Relaxation period	0.911	1.000	0.911	7.203**	0.0364
AB	0.000	1.000	0.000	0.000^{NS}	1.0000
AC	0.090	1.000	0.090	0.711^{NS}	0.4313
BC	0.563	1.000	0.563	4.446*	0.0795
A^2	1.221	1.000	1.221	9.651**	0.0209
B^2	74.736	1.000	74.736	590.721*	0.000
C^2	0.555	1.000	0.555	4.387**	0.0811
Model	293.5421	9	32.61579	257.7983*	0.000
Linear	216.3775	3	72.12583	11.10715*	0.0009
Cubic	293.910	12.000	24.492	187.6338*	0.0006
2FI	217.030	6.000	36.172	4.21302**	0.0269
Residual	77.271	9.000	8.586	–	–
Lack of fit	76.880	6.000	12.813	98.160^{NS}	0.0016
Pure error	0.392	3.000	0.131	–	–
Core error	294.301	15.000	–	–	–
Adequate precision	45.653				
CV (%)	1.89				

SS: sum of squares; df: degree of freedom; MS: mean square; CV: coefficient of variance
*significant at 1% level, **significant at 5% level, NS: not significant
"–": not applicable

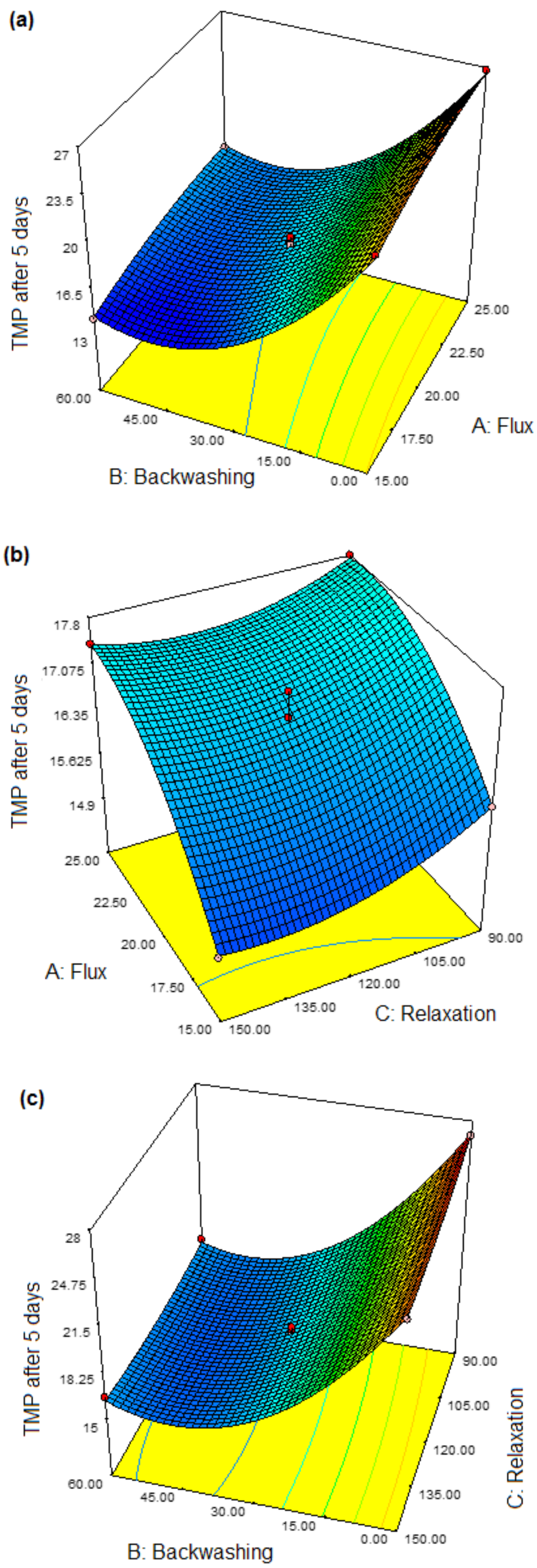

Fig. 2. Response Surface 3D plots for TMP as a function of (a) permeate flux and backwashing durations at a relaxation duration of 120 s; (b) permeate flux and relaxation durations at a backwashing duration of 30 s; and (c) backwashing and relaxation durations at a permeate Flux of 20 LMH.

duration (C). Adequate precision and CV values of 45.69 units and 1.89%, respectively further validated the significance of the TMP model.

Probability distribution plots of residuals (Supplementary Data Fig.S6a) in addition to the plots of predicted and actual results (Supplementary Data Fig.S6b) were developed to check the adequacy of the TMP model. It is clear from probability plots that the experimental and predicted TMP were close to the straight line, thereby proving the normal distribution of results.

Response surface 3D plots for TMP model show that backwashing and relaxation durations caused rapid membrane fouling after a certain duration (Fig. 2). Moreover, TMP increased abruptly beyond the permeate flux of 20 LMH (Fig. 2a). Notably, TMP reduced with the decrease in permeate flux but the increase in relaxation period initially showed positive effect on TMP (Fig. 2b). Interestingly, increase in backwashing duration and decrease in relaxation duration reduced resultant TMP (Fig. 2c). It is not possible to compare the results of this study with the previous studies because integrated effects of permeate flux, backwashing and relaxation durations on membrane has not been studied.

3.3. Model optimization

Based on earlier discussion, it is clear that each variable and their interactions may have a distinct influence over each response. Hence, permeate flux, backwashing and relaxation durations were optimized to achieve: (i) optimal treatment efficiency; and (ii) efficient reduction in membrane fouling (Table 7). All effluent quality parameters were given equal significance and optimized for maximum removal efficiency. TMP was the indicator of membrane fouling and its value was set at minimum for the purpose of optimization (Table 7). TMP was given the highest significance compared to other parameters. Recommended permeate flux, backwashing and relaxation durations based on model optimization were 18 LMH, 25 and 100 s, respectively. The predicted optimized responses for TMP, COD, NH_4–N, TN and TP removal efficiency were 17.47 KPa, 88.87%, 74.44%, 80.36%, and 62.52%, respectively.

Predicted values of input variables were also examined experimentally to validate the predicated results (Table 8). It was observed that the predicted and experimental values

Table 7
Description of responses for optimization

Parameters	Target	Lower limit	Upper limit	Significance
COD removal efficiency (%)	Maximum	85	90	+++
NH_4–N removal efficiency (%)	Maximum	70	76.4	+++
TN removal efficiency (%)	Maximum	78	84	+++
TP removal efficiency (%)	Maximum	60	66	+++
TMP (KPa)	Minimum	15	20	+++++

Table 8
Predicted and experimental responses based on optimized variables

Optimized variables	Values	COD (%)	NH_4–N (%)	TN (%)	TP (%)	TMP (KPa)
Permeate flux = 18 LMH	Predicted	88.87	74.44	80.36	62.52	17.47
Backwashing duration = 25 s						
Relaxation duration = 100 s	Experimental	89.5 ± 1.7	72.38 ± 1.4	81.8 ± 0.9	56.2 ± 3.7	16.37

were in agreement except TP removal efficiency. Interestingly, TMP was further reduced to 16.37 KPa during the experimental validation of predicted conditions. Therefore, it can be concluded that response surface methodology was successful for the optimization of permeate flux, backwashing and relaxation duration that are integral operating parameters of MBR.

4. Conclusion

Permeate flux, backwashing and relaxation durations are important operating parameter of MBR because of their influence on the performance of MBR as well as on membrane fouling. These process parameters were optimized successfully using Response Surface Methodology (RSM). Box–Behnken design (BBD) was employed to: (i) predict responses; and (ii) evaluate the influence of permeate flux, backwashing and relaxation durations on membrane fouling. All independent variables negatively affected the performance of MBR, while backwashing and relaxation durations were found to be effective for membrane fouling control. All models were statistically significant as validated through F-test, p-value, adequate precision, coefficient of variance (CV) and coefficient of determination (adjusted R^2 and R^2). Since all models were significant and reproducible, predicted responses were optimized. Optimized values for permeate flux, backwashing and relaxation durations were 18 LMH, 25 s and 100 s, respectively. Since this study was conducted on a lab scale submerged MBR, testing of this model for pilot scale MBR is recommended for future work.

Acknowledgement

This research is funded by the University of Engineering and Technology, Taxila, Pakistan through Faculty Research Projects (ASR&TD - 18/2015).

References

[1] H.-Y. Fu, P.-C. Xu, G.-H. Huang, T. Chai, M. Hou, P.-F. Gao, Effects of aeration parameters on effluent quality and membrane fouling in a submerged membrane bioreactor using Box–Behnken response surface methodology, Desalination, 302 (2012) 33–42.
[2] B. Marrot, A. Barrios-Martinez, P. Moulin, N. Roche, Industrial wastewater treatment in a membrane bioreactor: A review, Environ. Prog., 23 (2004) 59–68.
[3] Y. Rahimi, A. Torabian, N. Mehrdadi, M. Habibi-Rezaie, H. Pezeshk, G.-R. Nabi-Bidhendi, Optimizing aeration rates for minimizing membrane fouling and its effect on sludge characteristics in a moving bed membrane bioreactor, J. Hazard. Mater., 186 (2011) 1097–1102.
[4] C. Brepols, K. Drensla, A. Janot, M. Trimborn, N. Engelhardt, Strategies for chemical cleaning in large scale membrane bioreactors, Water Sci. Technol., 57 (2008) 457–463.
[5] T. Mukai, K. Takimoto, T. Kohno, M. Okada, Ultrafiltration behaviour of extracellular and metabolic products in activated sludge system with UF separation process, Water Res., 34 (2000) 902–908.
[6] X. Zheng, M. Ernst, M. Jekel, Stabilizing the performance of ultrafiltration in filtering tertiary effluent—Technical choices and economic comparisons, J. Membr. Sci., 366 (2011) 82–91.
[7] F.I. Hai, K. Yamamoto, K. Fukushi, F. Nakajima, Fouling resistant compact hollow-fiber module with spacer for submerged membrane bioreactor treating high strength industrial wastewater, J. Membr. Sci., 317 (2008) 34–42.
[8] T. Maqbool, S.J. Khan, C.-H. Lee, Effects of filtration modes on membrane fouling behavior and treatment in submerged membrane bioreactor, Bioresour. Technol., 172 (2014) 391–395.
[9] Z. Wang, J. Ma, C.Y. Tang, K. Kimura, Q. Wang, X. Han, Membrane cleaning in membrane bioreactors: A review, J. Membr. Sci., 468 (2014) 276–307.
[10] S.N. Shim, S.-R. Kim, S.J. Jo, K.-M. Yeon, C.-H. Lee, Evaluation of mechanical membrane cleaning with moving beads in MBR using Box–Behnken response surface methodology, Desal. Water Treat., 56 (2015) 2797–2806.
[11] R. Habib, M.B. Asif, S. Iftekhar, Z. Khan, K. Gurung, V. Srivastava, M. Sillanpää, Influence of relaxation modes on membrane fouling in submerged membrane bioreactor for domestic wastewater treatment, Chemosphere, 181 (2017) 19–25.
[12] S. Judd, The MBR Book: Principles and Applications of Membrane Bioreactors for Water and Wastewater Treatment, Elsevier, Oxford, 2010.
[13] F.I. Hai, K. Yamamoto, C.H. Lee, Membrane Biological Reactors: Theory, Modeling, Design, Management and Applications to Wastewater Reuse, IWA Publishing, London, 2014.
[14] A. Yuniarto, Z.Z. Noor, Z. Ujang, G. Olsson, A. Aris, T. Hadibarata, Bio-fouling reducers for improving the performance of an aerobic submerged membrane bioreactor treating palm oil mill effluent, Desalination, 316 (2013) 146–153.
[15] H. Monclus, S. Zacharias, A. Santos, M. Pidou, S. Judd, Criticality of flux and aeration for a hollow fiber membrane bioreactor, Sep. Sci. Technol., 45 (2010) 956–961.
[16] J. Wu, P. Le-Clech, R.M. Stuetz, A.G. Fane, V. Chen, Effects of relaxation and backwashing conditions on fouling in membrane bioreactor, J. Membr. Sci., 324 (2008) 26–32.
[17] D.-Y. Zuo, H.-J. Li, H.-T. Liu, G.-P. Wu, A study on submerged rotating MBR for wastewater treatment and membrane cleaning, Korean J. Chem. Eng., 27 (2010) 881–885.
[18] E.H. Bouhabila, R. Ben Aïm, H. Buisson, Fouling characterisation in membrane bioreactors, Sep. Purif. Technol., 22–23 (2001) 123–132.
[19] Z. Yusuf, N. Abdul-Wahab, S. Sahlan, Fouling control strategy for submerged membrane bioreactor filtration processes using aeration airflow, backwash, and relaxation: a review, Desal. Water Treat., 57 (2016) 17683–17695.
[20] H. Itokawa, C. Thiemig, J. Pinnekamp, Design and operating experiences of municipal MBRs in Europe, Water Sci. Technol., 58 (2008) 2319–2327.
[21] T. Jiang, M.D. Kennedy, B.F. Guinzbourg, V.P. A., J.C. Schippers, Optimising the operation of a MBR pilot plant by quantitative analysis of the membrane fouling mechanism, Water Sci. Technol., 51 (2005) 19–25.

[22] T.M. Qaisrani, W.M. Samhaber, Impact of gas bubbling and backflushing on fouling control and membrane cleaning, Desalination, 266 (2011) 154–161.

[23] W. Peng, I.C. Escobar, D.B. White, Effects of water chemistries and properties of membrane on the performance and fouling—a model development study, J. Membr. Sci., 238 (2004) 33–46.

[24] W.-L. Lai, L.-F. Chen, J.-J. Chen, S.-W. Liao, Effects of the operational parameters on carbon recovery and water flux in ultrafiltration using fractional factorial design, Desalination, 249 (2009) 1365–1370.

[25] M. Raffin, E. Germain, S. Judd, Optimising operation of an integrated membrane system (IMS) — A Box–Behnken approach, Desalination, 273 (2011) 136–141.

[26] W.-Q. Guo, Z.-H. Meng, N.-Q. Ren, Z.-P. Zhang, F.-Y. Cui, Optimization of key variables for the enhanced production of hydrogen by Ethanoligenens harbinense W1 using response surface methodology, Int. J. Hydrogen Energy, 36 (2011) 5843–5848.

[27] D. Baş, İ.H. Boyacı, Modeling and optimization I: Usability of response surface methodology, J. Food Eng., 78 (2007) 836–845.

[28] M.A. Bezerra, R.E. Santelli, E.P. Oliveira, L.S. Villar, L.A. Escaleira, Response surface methodology (RSM) as a tool for optimization in analytical chemistry, Talanta, 76 (2008) 965–977.

[29] G. Hanrahan, K. Lu, Application of factorial and response surface methodology in modern experimental design and optimization, Crit. Rev. Anal. Chem., 36 (2006) 141–151.

[30] Z. Zhang, H. Zheng, Optimization for decolorization of azo dye acid green 20 by ultrasound and H_2O_2 using response surface methodology, J. Hazard. Mater., 172 (2009) 1388–1393.

[31] E.W. Rice, R.B. Baird, A.D. Eaton, L.S. Clesceri, Standard Methods for the Examination of Water and Wastewater, 22nd ed., American Public Health Association, Washington, DC, 2012.

[32] Y.-K. Wang, X.-R. Pan, Y.-K. Geng, G.-P. Sheng, Simultaneous effective carbon and nitrogen removals and phosphorus recovery in an intermittently aerated membrane bioreactor integrated system, Sci. Rep., 5 (2015) (DOI: 10.1038/srep16281).

[33] M. Idris, A. Isma, A. Idris, R. Omar, A. Razak, P. Razreena, Effects of SRT and HRT on treatment performance of MBR and membrane fouling, Int. J. Chem. Mol. Nuc. Mat. Met. Eng., 8 (2014) 468–472.

[34] J. Choi, E.-S. Kim, Y. Ahn, Microbial community analysis of bulk sludge/cake layers and biofouling-causing microbial consortia in a full-scale aerobic membrane bioreactor, Bioresour. Technol., 227 (2017) 133–141.

[35] A.T. Nair, M.M. Ahammed, The reuse of water treatment sludge as a coagulant for post-treatment of UASB reactor treating urban wastewater, J. Cleaner Prod., 96 (2015) 272–281.

[36] G.E.P. Box, N.R. Draper, Empirical Model Building and Response Surfaces, John Wiley, New York, 1987.

[37] T.K. Trinh, L.S. Kang, Response surface methodological approach to optimize the coagulation-flocculation process in drinking water treatment, Chem. Eng. Res. Des., 89 (2011) 1126–1135.

[38] P. Tripathi, V.C. Srivastava, A. Kumar, Optimization of an azo dye batch adsorption parameters using Box–Behnken design, Desalination, 249 (2009) 1273–1279.

[39] V. Pujari, T.S. Chandra, Statistical optimization of medium components for enhanced riboflavin production by a UV-mutant of Eremothecium ashbyii, Process Biochem., 36 (2000) 31–37.

[40] S. Ghafari, H.A. Aziz, M.H. Isa, A.A. Zinatizadeh, Application of response surface methodology (RSM) to optimize coagulation–flocculation treatment of leachate using poly-aluminum chloride (PAC) and alum, J. Hazard. Mater., 163 (2009) 650–656.

[41] M.J. Anderson, P.J. Whitcomb, RSM Simplified: Optimizing Processes Using Response Surface Methods for Design of Experiments, Productivity Press, New York, 2005.

[42] Z. He, D.J. Miller, S. Kasemset, D.R. Paul, B.D. Freeman, The effect of permeate flux on membrane fouling during microfiltration of oily water, J. Membr. Sci., 525 (2017) 25–34.

[43] H. Chang, F. Qu, H. Liang, R. Jia, H. Yu, S. Shao, K. Li, W. Gao, G. Li, Correlating ultrafiltration membrane fouling with membrane properties, water quality, and permeate flux, Desal. Water Treat., 56 (2015) 1746–1757.

Supplementary Data

Table S1
Relaxation scenarios used by different researchers

Installation type	Membrane	Relaxation/Filtration	Permeate flux	Author(s)
Lab Scale	Hollow Fiber	20 s / 440 s	21 LMH	Wu et al. 2008
Lab Scale	Hollow Fiber	0.5 min / 8 min	20 LMH	Annop et al. 2014
Lab Scale	Hollow Fiber	15 min / 145 min	Variable	Hong et al. 2002
Lab Scale	Rotating	2 min / 8 min	47.5 LMH	Zuo et al. 2010
Pilot Scale	Hollow Fiber	1 min / 9 min	17.5–25.7 LMH	Oh et al. 2012
Pilot Scale	Flat Sheet	1 min / 9 min	10–15 LMH	Guglielmi et al. 2008
Full Scale	Flat Sheet	1 min / 9 min	15–25 LMH	Dalmau et al. 2015

Table S2
Backwash scenarios used by different researchers

Installation type	Membrane	Backwash/Filtration	Permeate flux	Author(s)
Lab Scale	Hollow Fiber	0.66 min / 8 min	24.5 LMH	Wu et al. 2008
Lab Scale	Hollow Fiber	0.5 min / 60 min	73.5 LMH	Ye et al. 2010
Lab Scale	Flat Sheet	0.5 min / 8 min	Variable	Qaisrani and Samhaber 2011
Lab Scale	Ceramic	2 min / 30 min	Variable	Hwang et al. 2009)
Pilot Scale	Hollow Fiber	0.75 min / 10 min	25 LMH	(Jiang et al. 2005)
Pilot Scale	Hollow Fiber	1.25 min / 15 min	30–50 LMH	(Raffin et al. 2011)
Full Scale	Hollow Fiber	0.5–1 min / 6–12 min	10–25 LMH	(Zsirai et al. 2012)

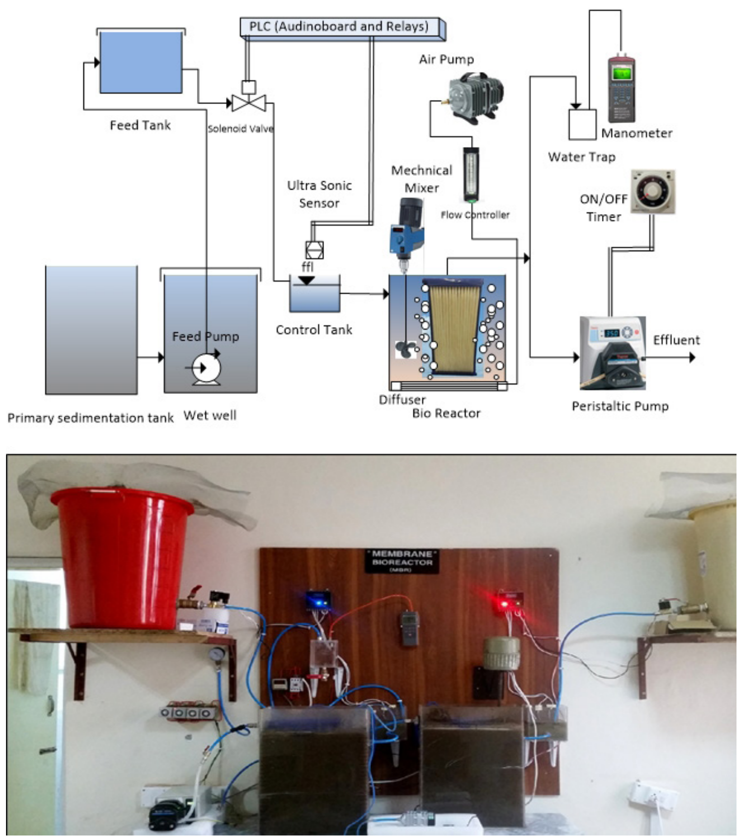

Fig. S3. The lab-scale MBR setups.

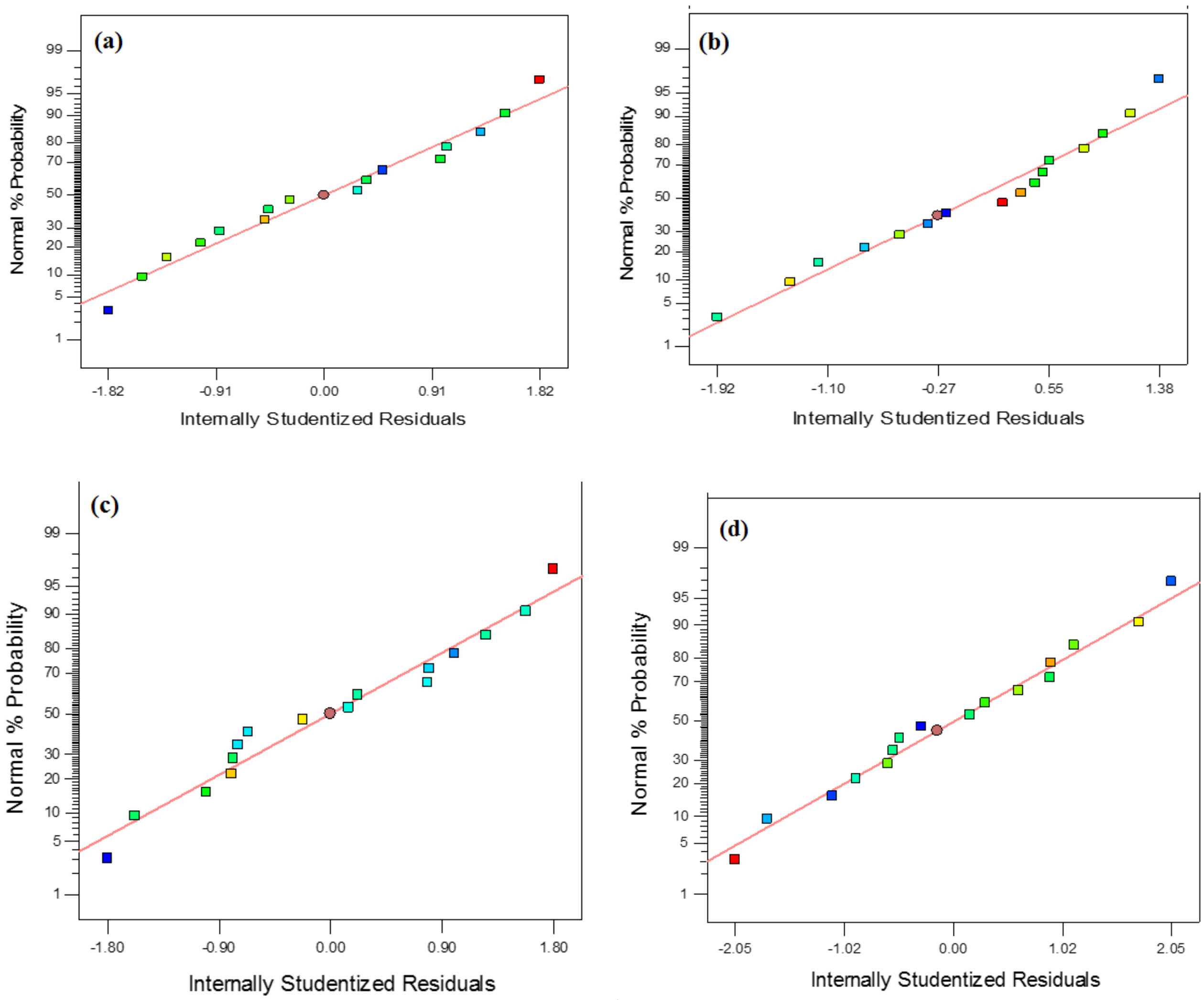

Fig. S4. Probability plots of residuals for (a) COD removal efficiency, (b) NH_4–N removal efficiency, (c) TN removal efficiency, and (d) TP removal efficiency.

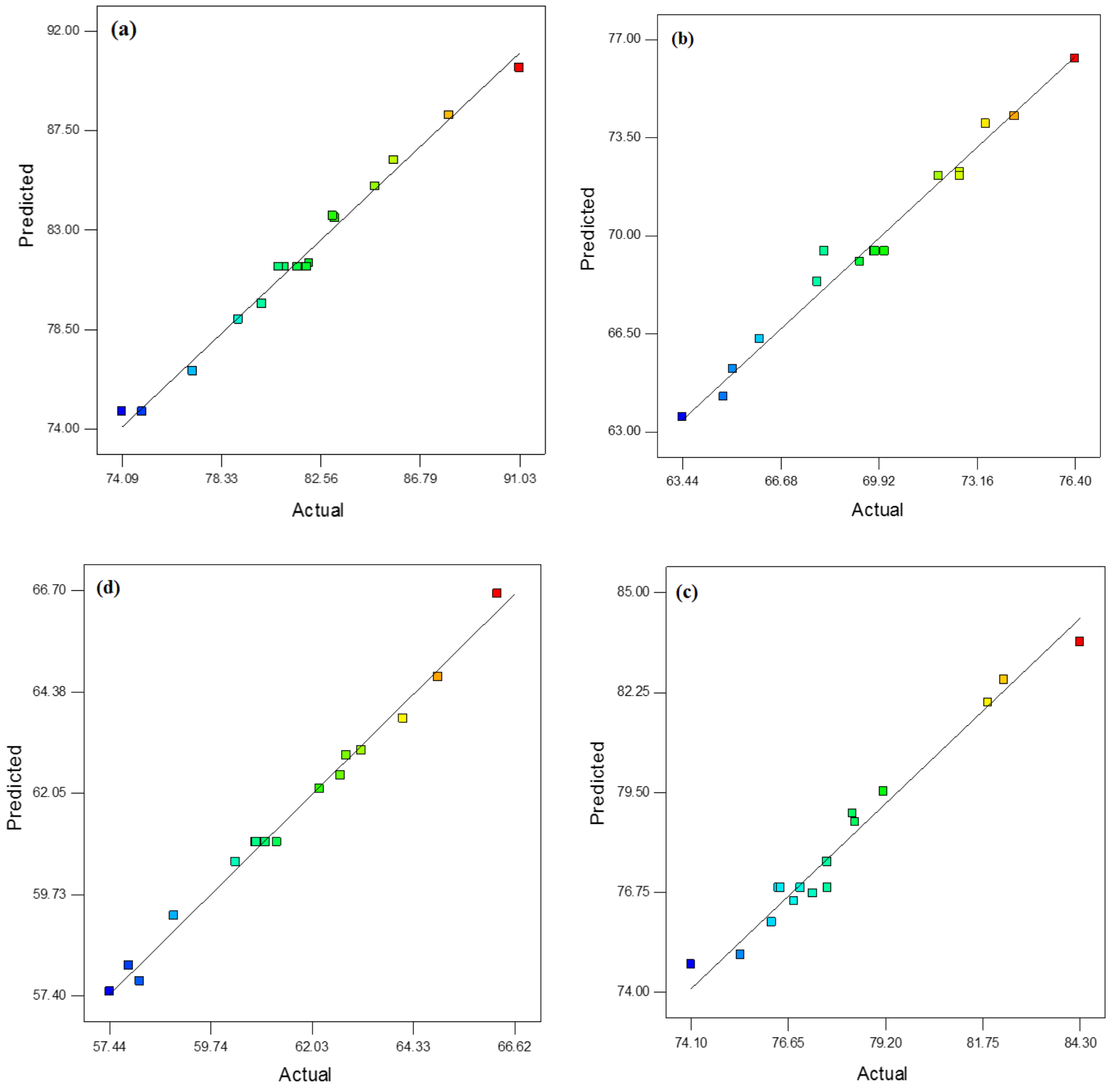

Fig. S5. Correlation between actual and predicted results for (a) COD removal efficiency, (b) NH_4–N removal efficiency, (c) TN removal efficiency, and (d) TP removal efficiency.

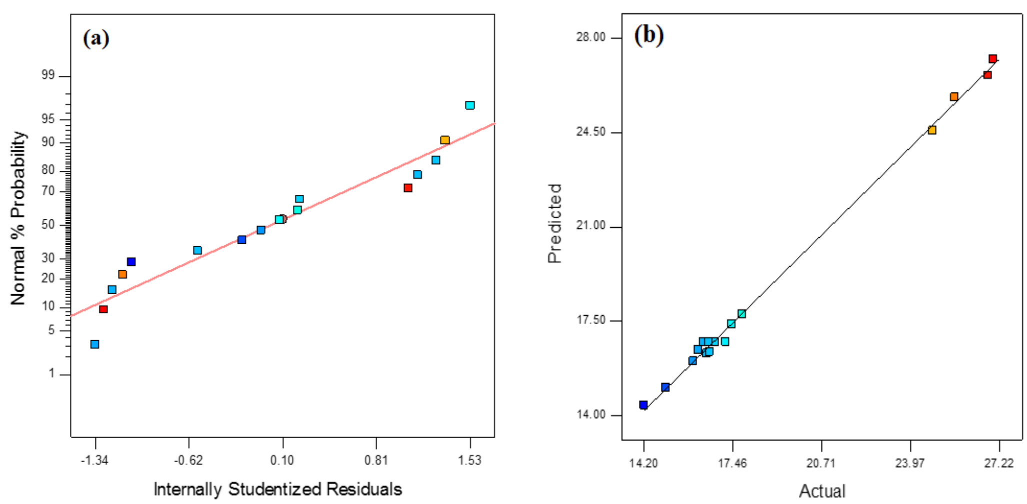

Fig. S6. Diagnostic plots (a) Probability plots of residuals for TMP model, (b) Predicted versus Actual results.