

## Performance, evaluation, and modeling of an integrated petroleum refinery wastewater treatment system using multi-layer perceptron neural networks

Habib A. Mokhtari\*, Sayed Ahmad Mirbagheri, Nazli Rafei Dehkordi

*Department of Environmental Engineering, Faculty of Civil Engineering, K. N. Toosi University of Technology, Tehran, Iran, Tel. +98 9133700074; emails: h.mokhtari@mail.kntu.ac.ir/habib.al.mokhtari@gmail.com (H.A. Mokhtari), Tel. +98 9121374357; emails: mirbagheri@kntu.ac.ir (S.A. Mirbagheri), nazli.rafeidehkordi@yahoo.com (N. Rafei Dehkordi)*

Received 20 May 2020; Accepted 27 September 2020

---

### ABSTRACT

This study investigates the performance of an integrated petroleum refinery wastewater treatment system. The proposed system attempts to improve the performance of an activated sludge system by using immersed vertical rotating biological contactors in the aeration basin of the system. This system is an innovative method of biological petroleum refinery wastewater treatment with a hybrid growth process. A sand filter column was used in the last part of the treatment process. Also, a multi-layer perceptron neural network (MLPNN) was applied to predict pollutants in the effluent. Overall treatment efficiencies of chemical oxygen demand (COD), total suspended solids (TSS), oil, ammonia (NH<sub>3</sub>), and turbidity were 94%, 90%, 88%, 93%, and 92%, respectively. According to the findings, the removal efficiencies of pollutants in our integrated system were superior to conventional activated sludge systems. Training procedures of all effluent quality parameters were successful for the MLPNN model. The training model showed an almost acceptable match between the experimental and predicted values. For all models predicting effluent COD, TSS, oil, NH<sub>3</sub>, and turbidity, the correlation coefficient was higher than 0.90, and the mean squared error varied from 0.0001 to 0.234 for the measured parameters. The results confirmed the effectiveness of the integrated system in achieving high removal efficiencies.

*Keywords:* Petroleum refinery wastewater treatment; Activated sludge process; Rotating biological contactors; Sand filter; Multi-layer perceptron neural networks; Prediction

---

### 1. Introduction

In recent years, the amount of produced wastewater by different industries, especially oil refineries, has drastically increased. Petroleum refineries and petrochemical industries generate and release a lot of hazardous materials, including sewage into the environment [1]. Therefore, it is crucial to collect and treat wastewater to achieve a healthy environment. Also, the reuse and reclamation of wastewater are needed, especially in the oil-producing arid regions, which are plagued with water scarcity. Wastewater treatment systems are usually the combination of physical, chemical, and biological processes. In biological wastewater

treatment systems, microorganisms, which are suspended in the reactors or attached to different media, are responsible for the removal of pollutants [2]. The hybrid treatment processes commonly use attached and suspended growth within a reactor; hence, they are more advantageous than single processes [3].

Activated sludge systems are suspended growth biological wastewater treatment systems. In other words, a bacterial biomass suspension is responsible for the treatment process [4]. Activated sludge systems have been successfully applied to treat various wastewaters with promising removal efficiencies. Recently, several types of research have been conducted to improve performance

---

\* Corresponding author.

and decrease the cost of activated sludge systems for treating different wastewaters. As an example, Pala and Tokat [5] used an activated sludge pilot for the treatment of the cotton textile industry wastewater. That study indicated that the performance of activated sludge systems can be improved by adding some materials, including powdered activated carbon, into the system. Martínez-Alcalá et al. [6] investigated biological degradation, sorption, and mass balance determination in a conventional activated-sludge wastewater treatment plant. In another study, Jung et al. [7] used batch activated sludge systems for treating dairy wastewater. They could improve the performance of the activated sludge system by using the enzymatic pool produced by fungus for the biological treatment of wastewaters with high oil and grease contents. Tellez et al. [8] used a field continuous-flow activated sludge system for removing petroleum hydrocarbons from produced water. Field-scale test results showed that the activated sludge system can successfully remove total petroleum hydrocarbon from produced water. In a study, Raper et al. [9] applied a pilot-scale activated sludge process to treat coke-making wastewater. According to the findings, the addition of powdered activated carbon to the activated sludge process can effectively improve the removal efficiencies. In another study, Cardete et al. [10] used a pilot-scale activated sludge process under different conditions for treating petrochemical wastewater.

The performance of suspended growth processes in wastewater treatment can be improved by using different kinds of packing materials in the wastewater treatment reactors [11]. In other words, packing materials are apt for improving overloaded suspended growth systems such as sequencing batch reactors and activated sludge systems because they can convert unused volumes into biofilm reactors [12]. The application of biofilm media, for instance, rotating biodisks in the aeration basin of activated sludge systems, might improve the performance of these suspended growth systems. In a study, Zaoyan et al. [13] combined rotating biological contactors (RBCs) with the activated sludge system for treating dye wastewater. The results stated that the combined system could effectively remove color. In another study, You et al. [14] used the combination of RBCs with an activated sludge system for increasing the performance of the wastewater treatment system. According to the results, RBCs as biofilms could promote nitrifying activity, which contributed to the nitrification performance. Di Trapani et al. [15] also used an integrated biofilm/activated sludge pilot to investigate the organic removal efficiency of the pilot in different values of the mixed liquor sludge retention time and temperatures. The results showed that the integrated system could successfully treat municipal wastewater in low mixed liquor sludge retention time values and with low temperatures. Park and Lee [16] utilized an activated sludge system with a polyurethane fluidized bed biofilm to treat dyeing wastewater. The chemical oxygen demand (COD) removal in the pilot was efficient in different organic loading rates. In a study, the COD removal for the edible oil wastewaters by an activated sludge system was investigated. Based on the results, the system could remove approximately 80% of COD in 5 d [17]. In another study, Gebara [18] used plastic nets as biofilm media inside the aeration tank of a

conventional activated sludge system. The results displayed that the nets could improve biochemical oxygen demand ( $BOD_5$ ) removal efficiency for treating synthetic wastewater. Su and Ouyang [19] investigated nutrient removal using a combined process with activated sludge and fixed biofilms. According to the results, the integration of packing materials and activated sludge systems can be effectively used for upgrading conventional activated sludge systems. Tang et al. [20] combined an activated sludge pilot with biofilm carriers for municipal wastewater treatment. In addition to the significant removal of organic matter and nutrients from municipal wastewater, the system displayed removal capacity for pharmaceuticals. Hassan et al. [21] monitored an upgraded hybrid moving bed biofilm reactor-conventional activated sludge wastewater treatment plant at low high retention time (HRT) and high C/N ratio for 12 months. The hybrid reactor showed high removal efficiency for  $BOD_5$  and COD. Dang et al. [22] employed loofah sponges in a pilot-scale integrated fixed-film activated sludge system for municipal wastewater treatment. The activated sludge system with modified loofah sponges was effective in organic removal and total nitrogen removal.

Generally, mathematical solutions are capable of solving every problem with hardware available at the moment, but some problems need significantly high computational capacities. Consequently, several algorithms, such as genetic algorithms, ant colony optimization algorithms, neural networks algorithms, and so forth, have been developed to offer a sufficiently promising solution [23]. Recently, artificial neural networks (ANNs) have become popular for prediction in different areas, including medicine, water resources, and environmental engineering [24]. Also, neural networks have been effectively used for monitoring and predicting diverse parameters in water and wastewater treatment systems [25–27]. In a study, Mokhtari et al. [28] applied a neural network model to predict effluent COD, TP, and total suspended solids (TSS) in a hybrid municipal wastewater treatment system. Based on the findings, the training procedure of the NN model was successful, and almost a perfect match was achieved between experimental values and predicted values. In another study, Shokri Dariyan et al. [29] employed an ANN model to predict optimum retention time for dairy wastewater treatment in a hybrid activated sludge system. The results showed that the neural network model could provide an acceptable prediction for the retention time in biological wastewater treatment processes. In addition to the neural networks model, Noroozi et al. [30] used the modified Stover–Kincannon and Grau models to predict the bio kinetic coefficients of COD removal in a hybrid activated sludge process. According to the results, the bioreactor follows the models with 98%–99% correlation coefficients. Neural networks generally predict output values from input values by some internal calculations [31]. Thus, a multi-layer perceptron neural network (MLP-NN) was applied to predict wastewater characteristics at the effluent of the system in our study.

This study attempts to assess the removal efficiency of organic pollutants in petroleum refinery wastewater via the integration of attached and suspended growth by using RBCs in the aeration basin of an activated sludge system. In other words, an activated sludge system was combined

with four fully immersed vertical RBCs in the aeration basin of the system. Additionally, a sand filter column was used as a tertiary treatment system to meet the improved standards for the effluent of the system. According to our knowledge, this integrated system for the treatment of petroleum refinery wastewater has not been previously studied or reported in the literature.

**2. Materials and methods**

*2.1. Wastewater characteristics*

The pilot plant was located in the Tehran Oil Refining Company in the city of Tehran, Iran, which has been operating since 1968. The actual petroleum refinery wastewater from the Tehran refinery wastewater treatment plant was used as the influent of our integrated wastewater treatment system. Influent wastewater analysis for the refinery wastewater treatment plant was carried out for 4 months before designing the pilot. We used the effluent of the dissolved air flotation (DAF) unit in the refinery wastewater treatment plant as raw wastewater in this study. Table 1 shows the minimum, average, and maximum values of the influent characteristics.

*2.2. Pilot plant*

The integrated activated sludge system consisted of a feeding tank, an aeration basin, which was composed of four fully immersed vertical RBCs, a settling tank, and a sand filter column for tertiary treatment (Fig. 1). The configuration of the pilot plant is given in Table 2. The feeding tank was made of plastic and was 1.7 m above the ground level in order to establish a continuous flow. The RBCs were constructed of Plexiglas acrylic sheets. To facilitate both microorganism’s growth and organic matter biodegradation, a layer of polyurethane foam (PUF) was attached to both sides of each biodisk. The PUF serves as suitable media due to having high porosity and specific surface area [32]. Table 3 presents the characteristics of RBCs used in this study. The disks were placed in the aeration basin with a volume of

50 L. The disks were connected via a stainless steel shaft. An induction motor was utilized for rotating the shaft and disks. While biofilm media can improve the efficacy of biological wastewater treatment systems, the detachment of biomass from biofilm media is a crucial issue among these attached growth systems [33]. High rotational speed might result in biofilm detachment, which can decrease biomass concentration in RBCs [34]. For this purpose, the rotational speed of 4 rotations per minute (rpm) was chosen for the rotating disks in the aeration basin. Fig. 2 shows the photos of RBCs before and after usage in the wastewater treatment system. An air compressor supplied airflow with a 30 L/min flow rate to the wastewater treatment system through diffusers installed in the aeration basin to provide oxygen for the aeration basin and also ensure mixing in the reactor. In order to maintain the temperature at 30°C, two aquarium heaters were used with temperature variations of 25°C–35°C. The Plexiglas settling tank had a trapezoidal shape part as this can help sludge and suspended solids settle swiftly. To return settled sludge to the aeration basin with a specific flow, a pump was installed at the bottom of the settling tank. A cylindrical tank was used for building the sand filter column. A 20 cm layer of gravel with two parts

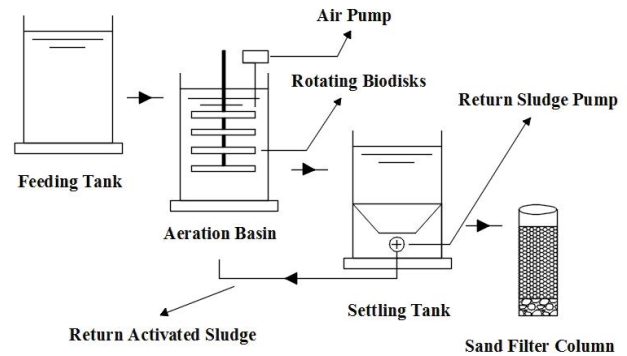


Fig. 1. Configuration of the pilot plant and the process used in this study.

Table 1  
Petroleum refinery wastewater characteristics

Parameter	COD (mg/L)	BOD <sub>5</sub> (mg/L)	TDS (mg/L)	TSS (mg/L)	Oil (mg/L)	NH <sub>3</sub> (mg/L)	Turbidity (NTU)	pH	DO (mg/L)
Maximum	330	83	2,100	67	87	15	29.9	8.2	1.5
Average	226	56.5	1,702	47.5	55	9	23.8	7.6	0.85
Minimum	122	30	1,305	28	23	3	17.7	7.1	0.2

Table 2  
Configuration of pilot plant

	Length (cm)	Width (cm)	Height (cm)	Radius (cm)	Volume (L)
Feeding tank	–	–	96	32	300
Aeration basin	35	35	40	–	50
Settling tank	30	16	25	–	12
Sand filter column	–	–	90	16	18

was placed at the bottom of the sand filter to allow drainage. To support the filtrating sand and prevent it from escaping through the drainage layer, a layer of sand was added above the gravel layer. The main layer of the sand filter was composed of 98% pure silica sand with a uniformity coefficient of 1.7 and an effective size of 0.18 mm.

### 2.3. Operating conditions

After installing rotating contactors in the aeration basin, half of the effective volume of the aeration basin was filled with return activated sludge of the aeration basin unit of the petroleum refinery wastewater treatment plant. Temperature, COD, mixed liquor volatile suspended solids (MLVSS), mixed liquor suspended solids (MLSS), and pH for return activated sludge were 34°C, 259; 240; 1,142 mg/L, and 7, respectively. The remaining volume was filled with wastewater passed through the dissolved air flotation unit of the petroleum wastewater treatment plant. To provide organics and nutrients required for the growth of

microorganisms, wastewater, and activated sludge were daily added to the aeration basin. This process had been done 25 times before we started pilot testing. That is, 25 cycles of treatment had been performed during the operational period. The temperature varied from 25°C to 30°C during the adaptation phase. pH generally affects the treatability of wastewater in biological wastewater treatment processes [35]. Because of this, pH was measured, and it was between 6.5 and 8.5. It was observed that after 7.5 h aeration in the aeration basin, the COD removal rate decreases because of the decline in the concentration of MLVSS. This decline can be attributed to the decrease in the food to microorganism ratio, and the death of microorganisms in the aeration basin, which normally increase COD concentration. Moreover, based on the inlet wastewater flow rate and the dimensions of the settling tank, settling time in the activated sludge system was 2.5 h. As a result, 10 h was chosen for the whole hybrid activated sludge system as the optimum hydraulic retention time. The settling tank and the sand filter column were added to the system after the adaptation phase. Raw wastewater was added to the feeding tank by a pump. Afterward, wastewater samples were collected to measure the influent wastewater characteristics. Raw wastewater with approximately 100 mL/min flow rate was discharged into the aeration basin. Finally, the treated wastewater from the settling tank passed through the sand filter column for tertiary treatment. In addition to the mass of microorganisms to aerobically contaminants removal, the attached growth biomass resulted in contaminates removal in our hybrid reactor. In other words, the suspended growth process and the attached growth process work together for contaminates removal in hybrid reactors [36]. Predation, mechanical trapping, natural death, and adsorption typically occur in sand filtration for contaminants removal. The particulate contaminants are physically removed by filtration on the surface of the sand filter's

Table 3  
Characteristics of rotating biological contactors used in this study

Parameter	Value
Number of disks	4
Diameter of disks (cm)	25
Thickness of disks (cm)	4.5
Surface material	PUF
Spacing between disks (cm)	5
Total surface area of disks (m <sup>2</sup> )	0.4
Porosity of disks (%)	85
Disk submergence (%)	100

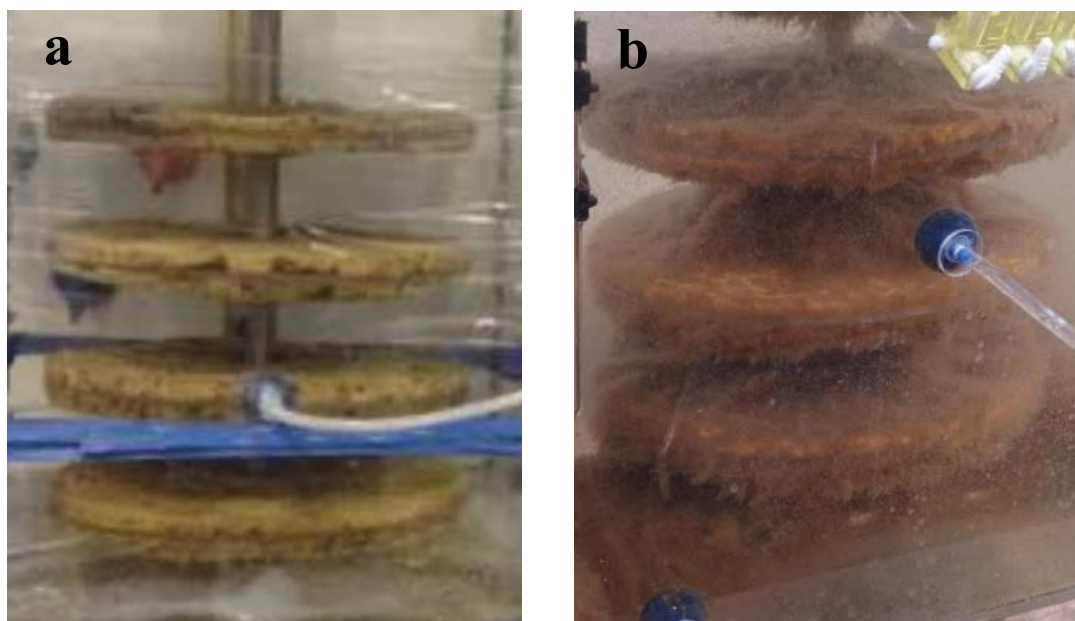


Fig. 2. Rotating biological contactors before biofilm formation (a) and after biofilm formation in the aeration basin (b).

bed. Dissolved contaminants are removed by biological or physical-chemical processes in sand filtration [37].

2.4. Analytical method

COD, BOD<sub>5</sub>, dissolved oxygen (DO), TSS, MLSS, MLVSS, ammonia (NH<sub>3</sub>), total dissolved solids (TDS), temperature, turbidity, pH, and oil were measured in this study. The pH and the temperature were measured by a digital pH meter. TDS was measured by AZ8371, and turbidity was measured by PC CECKIT Loviband. A spectrophotometer (Loviband laboratory spectrophotometer) was used to measure NH<sub>3</sub> and COD at the petroleum refinery wastewater treatment plant laboratory. TSS, MLVSS, MLSS, BOD<sub>5</sub>, DO, and oil were measured according to standard methods [38].

2.5. NN-based model development

ANNs are inspired by how the animals’ brain works [39]. Artificial neurons are connected with synapses, which can transmit signals to the next neuron, in neural network systems [40]. The NNs are typically composed of a lot of artificial neurons; thus, the connections among these neurons determine the network’s function [41]. In other words, the NNs attempt to project a relationship between inputs and outputs without any specific rule by assessing examples from the training data set, and this the most advantages of neural networks [42]. A layer neuron cannot effectively

detect the relationships between many inputs and outputs in the network; because of this, a multi-layer perceptron (MLP) is employed for building the NN models [43]. In this research, we used an MLP-NN with three layers, including an input layer, a hidden layer with ten neurons, and an output layer. The single-output MLP-NN, executed in this research with M neurons in the hidden layer, can be indicated by Eq. (1):

$$y(w, x) = \phi_{out} \left( \sum_{i=1}^M (W_{i,out} \times x_{ih}) + b_{out} \right) \tag{1}$$

where  $b_{out}$  is the bias of output neuron,  $\phi_{out}$  is the transfer function of the output layer,  $W_{i,out}$  is the weight between the  $i$ th neuron in the hidden layer and the output neuron, and  $x_{ih}$  is the output of each neuron in the hidden layer, and this is determined by Eq. (2):

$$x_{ih} = \phi_h \left( \sum_{i=1}^N (W_{i,n} \times x_i) + b_n \right) \tag{2}$$

where  $b_n$  is the bias of  $n$ th neuron in the hidden layer,  $x_i$  is the  $i$ th input,  $W_{i,n}$  is the weight between the  $n$ th neuron and the  $i$ th input in the hidden layer,  $N$  is the number of inputs, and  $\phi_h$  is the transfer function of the hidden layer.

Fig. 3 shows the architecture of the MLP-NN used in the study for the prediction of effluent wastewater characteristics. Nine parameters, including influent COD,

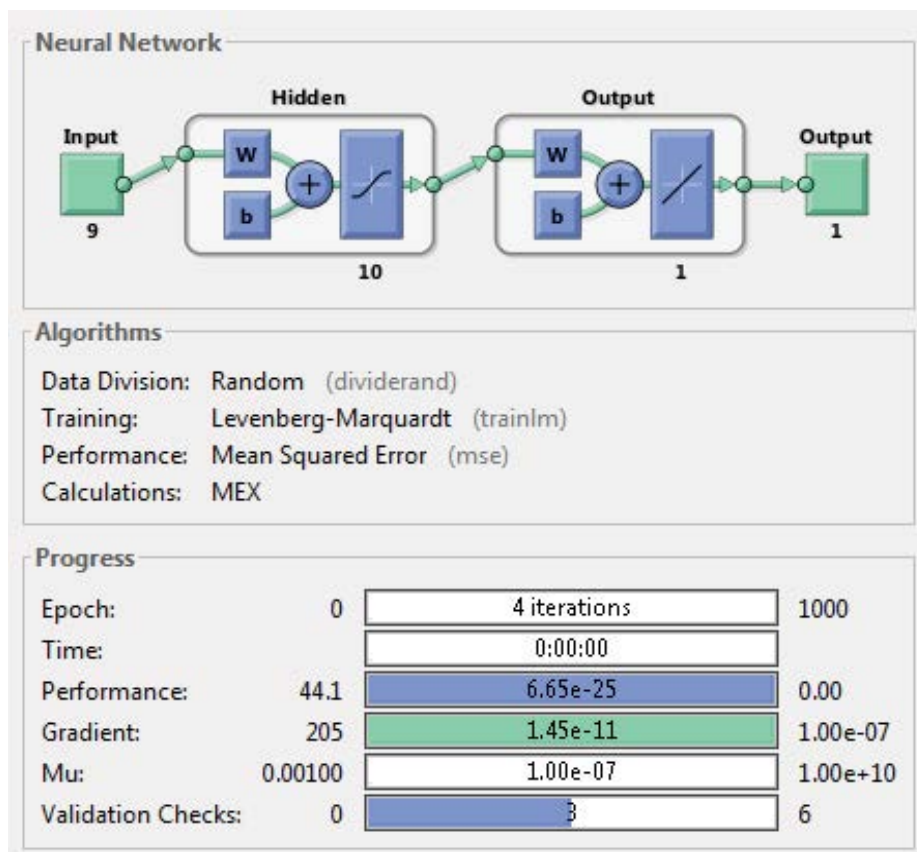


Fig. 3. Architecture of MLP-NN model for the prediction of effluent COD, TSS, oil, NH<sub>3</sub>, and turbidity.



BOD<sub>5</sub>, TDS, TSS, oil, NH<sub>3</sub>, turbidity, pH, and DO, were used as inputs of the MLP-NN to predict effluent COD, TSS, oil, NH<sub>3</sub>, and turbidity. The characteristics of input and output variables in the NN modeling process are given in Table 4. Via a random data division, the data set was divided into three sets, 80% for training, 10% for testing, and 10% for validation of the MLP-NN model. Indeed, ten data points were used in the MLP-NN model, and the model allocated eight data points for training, one data point for testing, and one data point for validation. In this research, the Levenberg–Marquardt algorithm (which is a robust algorithm) were used for training the MLP-NN model. The performance of the MLP-NN model in predicting effluent COD, TSS, oil, NH<sub>3</sub>, and turbidity was measured using mean squared error (MSE) and correlation coefficient (R).

### 3. Results and discussion

#### 3.1. COD removal efficiency

Fig. 4 shows the changes of COD concentration in the petroleum refinery wastewater and also the removal efficiencies after the activated sludge system with immersed RBCs and the sand filter column. The average COD concentration was about 234 mg/L in the influent of the raw wastewater, which decreased to 14.4 mg/L in the effluent (lower than the standard limit of 60 mg/L by U.S. EPA) [44]. In Fig. 4, the standard deviation for raw wastewater, effluent after the hybrid activated sludge system, and effluent after the sand filter column are 57.82, 9.57, and 5.69, respectively. The average COD removal efficiency after the activated sludge system with immersed rotating biodisks was 87.6%, and the removal efficiency increased to 94% after the sand filter column. The results of this study state that the activated sludge system with immersed rotating biodisks in the biological reactor, and the sand filter column is effective in terms of COD removal. The microorganisms in our hybrid system (attached/suspended growth), have a higher ability to remove organic carbon than an activated sludge system with the single suspended growth process [45]. Besides, our integrated system performed better, in terms of COD removal, than an aerated baffled reactor, which was coupled with an aerated biological filter [46]. In a study,

Tong et al. [47] used a conventional activated sludge process coupled with an immobilized biological filter [47]. Our integrated system displayed higher COD removal efficiency as compared with the removal efficiency of around 64% in the treatment of heavy oil wastewater in that system. Also, the COD removal in our integrated system is higher than a hybrid oil refinery wastewater treatment system, which was composed of a moving bed biofilm reactor and a slow-rate sand filter [48]. Shokrollahzadeh et al. [49] employed an activated sludge system for treating petrochemical wastewater; the COD removal efficiency in that system was lower than the COD removal in our integrated system.

#### 3.2. TSS removal efficiency

Fig. 5 shows the changes of TSS concentration and TSS removal efficiencies after the activated sludge system with immersed RBCs and the sand filter column. The settling tank and the sand filter column are the two prime steps of TSS removal in the integrated wastewater treatment system. The average TSS concentration in the influent of the raw wastewater was about 43.8 mg/L, which decreased to 31.3 mg/L after the settling tank and then decreased to 4.4 mg/L in the effluent. The results show TSS removal efficiency of 90% for the integrated wastewater treatment system. In Fig. 5, the standard deviation for raw wastewater, effluent after the hybrid activated sludge system,

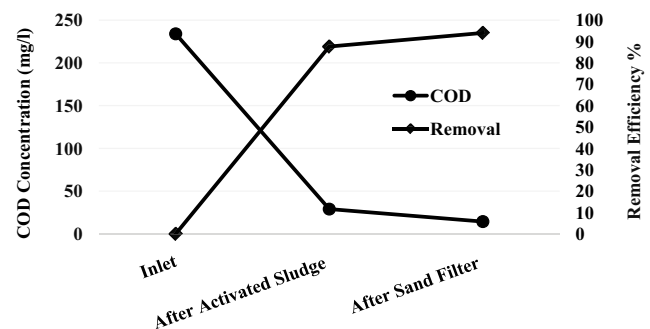


Fig. 4. COD concentration and removal efficiencies in the integrated system.

Table 4  
Characteristics of the measured variables in the NN modelling process

Input variable	Value	Output variable	Value
Influent concentration		Effluent concentration	
COD (mg/L)	140–320	COD (mg/L)	7–25
BOD <sub>5</sub> (mg/L)	35–75	BOD <sub>5</sub> (mg/L)	2.6–14
TDS (mg/L)	1,173–2,000	TDS (mg/L)	801–1,230
TSS (mg/L)	35–60	TSS (mg/L)	3–7
Oil (mg/L)	32–57	Oil (mg/L)	1–10
NH <sub>3</sub> (mg/L)	3.1–4.6	NH <sub>3</sub> (mg/L)	0.07–0.43
Turbidity (NTU)	18.2–28.3	Turbidity (NTU)	0.3–3.5
pH	7.4–8	pH	7–7.9
DO (mg/L)	0.4–1.2	DO (mg/L)	2.5–3.8

and effluent after the sand filter column are 7.23, 8.46, and 1.23, respectively. According to the results, the integrated system is efficient for TSS removal in petroleum refinery wastewater. Our integrated system showed a higher TSS removal efficiency in comparison to conventional activated sludge systems. As an example, 90% TSS removal in our integrated system is higher than the maximum 71% TSS removal in an extended aeration activated sludge system for petroleum refinery wastewater treatment used by Gasim et al. [50]. In a study, Xie et al. [51] used an aerated biological filter process for the treatment of slightly polluted wastewater in an oil refinery. Ninety percent of TSS removal in our integrated system is higher than 83% TSS removal in that system. In another study, Ahmed et al. [52] used a system, which consisted of three different configurations of sequencing batch reactors; the maximum TSS removal was 65%, which is lower than the average TSS removal in our integrated system. Comparing our results with Perez et al. [53], who used an anaerobic thermophilic fluidized bed for treating cutting-oil wastewater, our integrated system is more effective than that system in terms of TSS removal.

### 3.3. Oil removal efficiency

The average oil concentration in the influent of the raw wastewater was about 46 mg/L, which decreased to the average concentration of 5.7 mg/L in the effluent. The oil removal efficiency was approximately 88% at the end of the integrated petroleum refinery wastewater treatment system. The standard deviation for raw wastewater and effluent after the sand filter column are 7.51 and 2.68, respectively. Based on the results, the integrated system is effective in oil removal from petroleum refinery wastewater. In a study, Otadi et al. [54] used an oil refinery wastewater treatment system. That system was composed of a dissolved air flotation system, an activated sludge system, and a clarifier. The oil removal efficiency of our integrated system is higher than that system. In another study, Sekman et al. [55] utilized electrocoagulation to treat oily wastewater. Oil removal in that system is lower than oil removal in our integrated system. Also, oil removal in our integrated system is higher than oil removal in a system, which was employed by Dumore and Mukhopadhyay [56]. Comparing the results of our integrated system with the findings of Wang et al. [57],

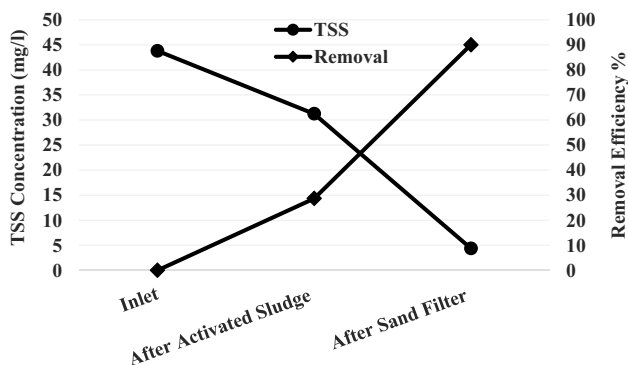


Fig. 5. TSS concentration and removal efficiencies in the integrated system.

who used an up-flow anaerobic sludge bed (UASB) reactor to treat heavy oil refinery wastewater and achieved the oil removal efficiency of up to 72%, our system is more efficient.

### 3.4. $\text{NH}_3$ removal efficiency

The changes of  $\text{NH}_3$  concentration and  $\text{NH}_3$  removal efficiencies after various steps in the integrated system are shown in Fig. 6. The average  $\text{NH}_3$  concentration in the influent of the raw wastewater was about 4.02 mg/L, which decreased to the average concentration of 0.26 mg/L in the effluent. The average ammonia removal efficiency after the activated sludge system with immersed RBCs was about 84.3%, and the removal efficiency increased to about 93% after the sand filter column. In Fig. 6, the standard deviation for raw wastewater, effluent after the hybrid activated sludge system, and effluent after the sand filter column are 0.53, 0.43, and 0.12, respectively. According to the results of this study, the integrated system with immersed RBCs in the aeration basin, and the sand filter column is efficient in terms of  $\text{NH}_3$  removal. In a study, Cao and Zhao [58] used a moving bed biofilm reactor (MBBR) to treat petrochemical wastewater. The 93%  $\text{NH}_3$  removal efficiency of our integrated system is higher than the approximately 80%  $\text{NH}_3$  removal efficiency of that system. In another study, Mirbagheri et al. [59] employed an activated sludge contact stabilization process for treating petroleum refinery wastewater;  $\text{NH}_3$  removal efficiency in our integrated system is higher than that system. Hamoda and Al-Haddad [60] evaluated the performance of a fixed-film reactor to treat petroleum refinery wastewater.  $\text{NH}_3$  removal in that wastewater system was lower than our integrated system. Also, Zhidong et al. [61] applied a submerged membrane bioreactor to treat oil refinery wastewater.  $\text{NH}_3$  removal in that study was approximately analogous to our system, but they used membranes, and they confronted the membrane fouling problem, which is a severe obstacle in these kinds of treatment systems [62].

### 3.5. Turbidity removal efficiency

The changes of turbidity concentration and turbidity removal efficiencies after various steps in the integrated system are shown in Fig. 7. The integrated system showed high performance in terms of turbidity removal, chiefly due to the settling tank and the sand filter column. The

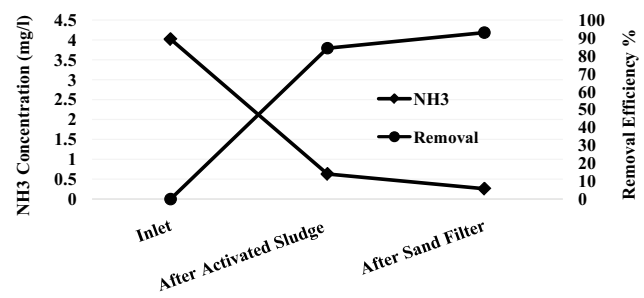


Fig. 6.  $\text{NH}_3$  concentration and removal efficiencies in the integrated system.

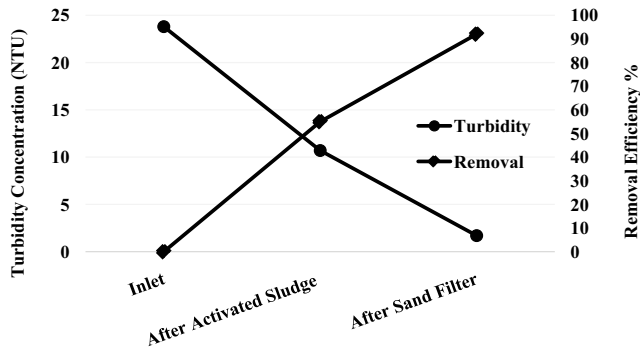


Fig. 7. Turbidity concentration and removal efficiencies in the integrated system.

average turbidity concentration of the influent of the raw wastewater was 23.8 NTU, which decreased to 10.7 and 1.7 NTU after the settling tank and the sand filter column, respectively. In Fig. 7, the standard deviation for raw wastewater, effluent after the hybrid activated sludge system, and effluent after the sand filter column are 3.90, 2.36, and 1.09, respectively. The turbidity removal efficiency

was approximately 92% at the end of the integrated petroleum refinery wastewater treatment system. The results of this study indicated that the integrated system is efficient for the removal of turbidity in wastewater with higher efficiencies than other kinds of integrated wastewater treatment systems [63]. Additionally, the average turbidity removal in our integrated system is higher than the average turbidity removal of a batch electrochemical reactor, which was used by Körbahti and Artut [64] to treat bilge water. In a study, Veliöğlu et al. [65] used an activated sludge system to treat olive oil-bearing wastewater. Turbidity removal in that system was lower than our integrated system. In another study, Pendashteh et al. [66] employed a sequencing batch reactor to treat produced water. The average turbidity concentration of the effluent in that wastewater treatment system was higher than the average turbidity concentration of the effluent in our integrated system.

### 3.6. NN-based prediction of effluent characteristics

In this research, several network architectures with neurons at the hidden layer were tested to predict COD, TSS, oil,  $\text{NH}_3$ , and turbidity. To maintain the network as

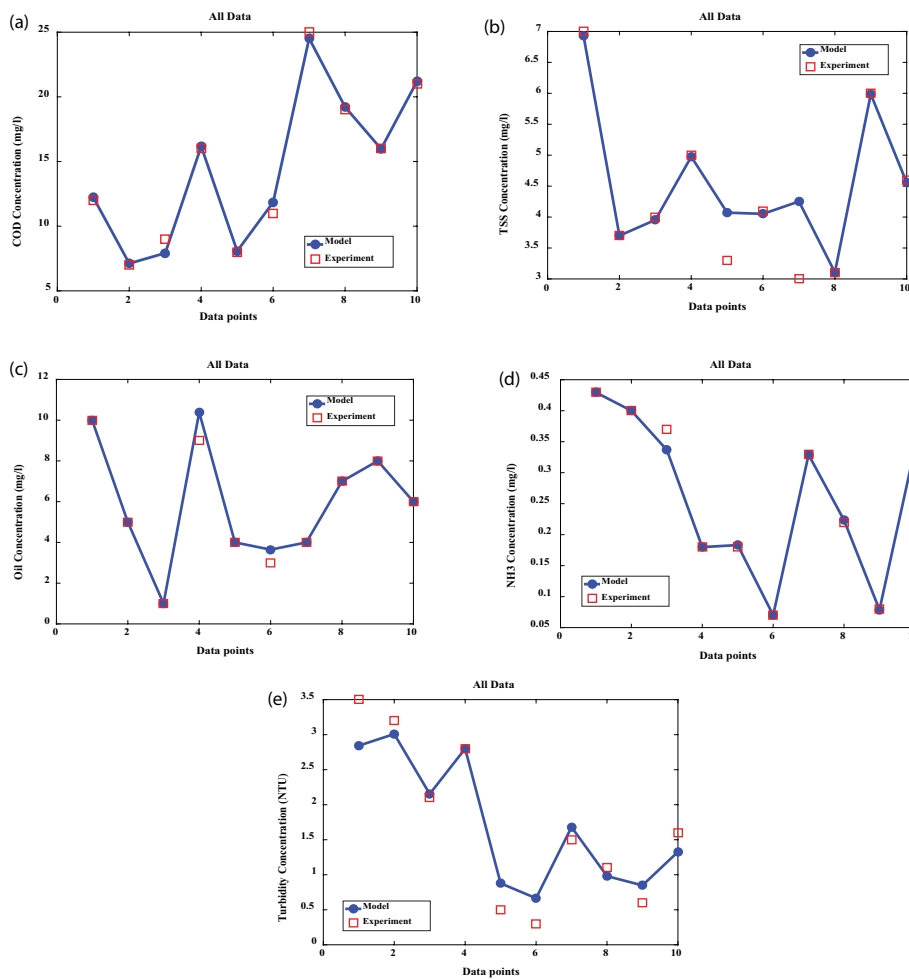


Fig. 8. Prediction of (a) effluent COD, (b) effluent TSS, (c) effluent oil, (d) effluent  $\text{NH}_3$ , and (e) effluent turbidity using MLP-NN model.



simple as possible after a lot of preliminary experiments for each output, the three-layer MLP-NN was selected. Although employing more hidden neurons in the neural networks may improve the performance of the network, using too many neurons may result in over-fitting, which typically undermines the generalization capacity of the model [67]. Accordingly, the MLP-NN with three layers and ten neurons in the hidden layer resulted in higher accuracies for most of the tested architectures and the effluent characteristics. Optimal architecture is crucial for training the algorithm with suitable speed and short simulation time for determined network performance [68]. In this study, the training procedure of the MLP-NN model

was successful for the prediction of effluent COD, TSS, oil,  $\text{NH}_3$ , and turbidity. Fig. 8 shows the results of the prediction for the six effluent characteristics using the MLP-NN algorithm. According to the results of the different data, there is a perfect match between predicted values and experimental values for the effluent COD, TSS, oil,  $\text{NH}_3$ , and turbidity. The results also confirm the high generalization capability of the MLP-NN algorithm, and this has been reported in some other studies [69,70].

The regression lines for the MLP-NN model predicting effluent COD, TSS, oil,  $\text{NH}_3$ , and turbidity based on all data sets are shown in Fig. 9. In this study, the results confirm the high correlation of experimental values with predicted

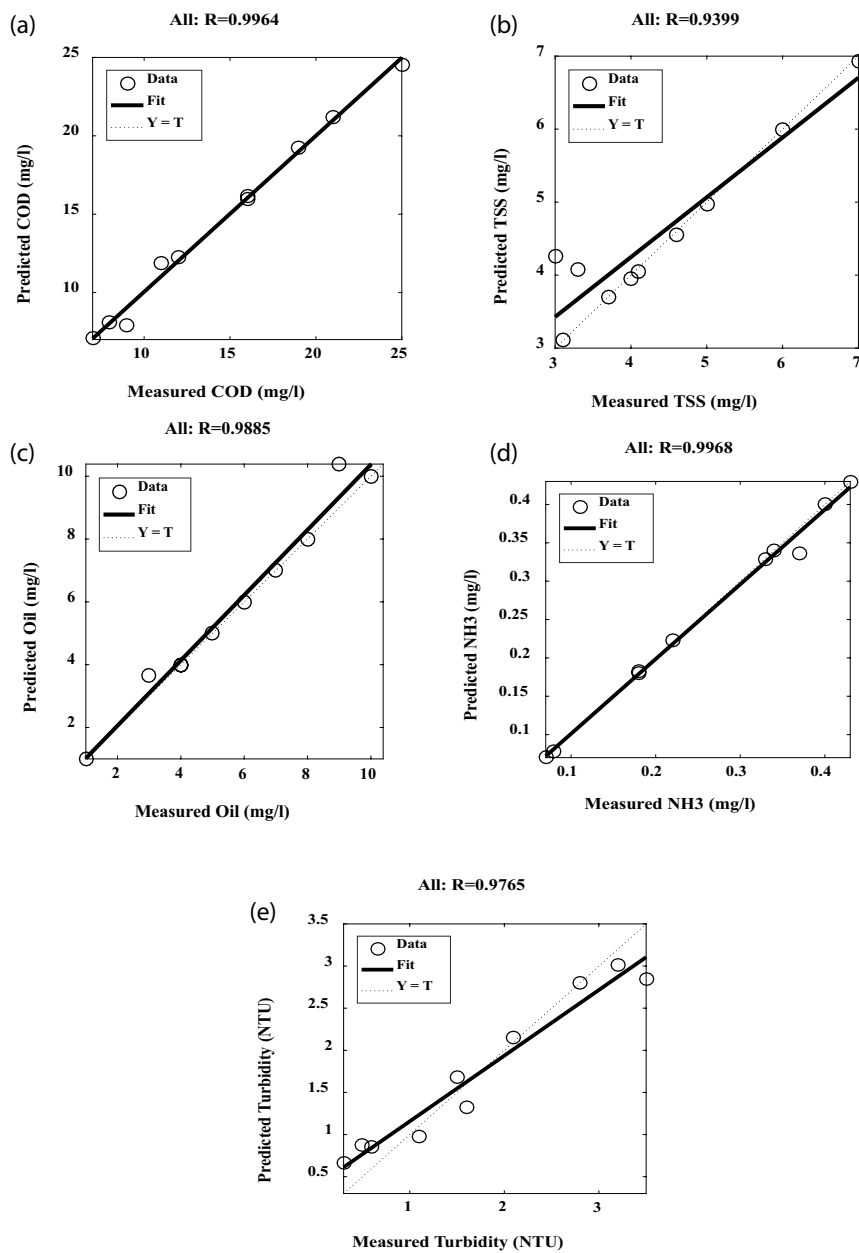


Fig. 9. Regression plots for the MLP-NN models predicting (a) effluent COD, (b) effluent TSS, (c) effluent oil, (d) effluent  $\text{NH}_3$ , and (e) effluent turbidity.

Table 5  
R and MSE values for measured parameters

Parameter	R	MSE
COD	0.9964	0.231
TSS	0.9399	0.217
Oil	0.9885	0.234
NH <sub>3</sub>	0.9968	0.0001
Turbidity	0.9765	0.093

values. Table 5 presents R and MSE values for each of the measured parameters. The results of our modeling for the prediction of effluent characteristics using the MLP-NN model display higher accuracies in comparison to some previously developed models [71,72]. In this study, the optimal architecture of the MLP-NN model was discovered to be acceptable since the error based on all data sets was satisfactory for the effluent COD, TSS, oil, NH<sub>3</sub>, and turbidity.

#### 4. Conclusion

This study examined the novel application of an integrated system for the treatment of petroleum refinery wastewater. The integrated system was composed of an activated sludge system, which was coupled with immersed vertical RBCs, and a sand filter column at the end of the treatment process to improve the performance of the system. In order to make a tradeoff between attached growth and suspended growth, the aeration basin of the activated sludge system was filled with four fully immersed RBCs. With the HRT of ten hours, COD, TSS, oil, NH<sub>3</sub>, and turbidity removal efficiencies were 94%, 90%, 88%, 93%, and 92%, respectively. In our integrated system, the removal efficiencies were higher than as compared to conventional activated sludge systems or biofilm reactors. In other words, the integrated system obtained a successful result in petroleum refinery wastewater treatment. The application of a sand filter column as a post-treatment step after the biological wastewater treatment is a promising technology for wastewater reclamation and reuse in countries, which are suffering from the water crisis. Additionally, the findings showed that using the MLPNN model had high prediction accuracies, and high correlation coefficients (R) between the measured and predicted output variables were achieved. Therefore, the MLPNN model is suggested for designing and estimating the performance of integrated wastewater treatment systems since the MLPNN model could successfully predict the performance of our integrated system.

#### Acknowledgments

The authors are grateful to Tehran oil refinery for their logistical assistance during this work.

#### References

- [1] H. Wake, Oil refineries: a review of their ecological impacts on the aquatic environment, *Estuarine Coastal Shelf Sci.*, 62 (2005) 131–140.
- [2] P. Falàs, P. Longrée, J. La Cour Jansen, H. Siegrist, J. Hollender, A. Joss, Micropollutant removal by attached and suspended growth in a hybrid biofilm-activated sludge process, *Water Res.*, 47 (2013) 4498–4506.
- [3] M. Christensson, T. Welander, Treatment of municipal wastewater in a hybrid process using a new suspended carrier with large surface area, *Water Sci. Technol.*, 49 (2004) 207–214.
- [4] K.V. Gernaey, M.C. Van Loosdrecht, M. Henze, M. Lind, S.B. Jørgensen, Activated sludge wastewater treatment plant modelling and simulation: state of the art, *Environ. Modell. Softw.*, 19 (2004) 763–783.
- [5] A. Pala, E. Tokat, Color removal from cotton textile industry wastewater in an activated sludge system with various additives, *Water Res.*, 36 (2002) 2920–2925.
- [6] I. Martínez-Alcalá, J.M. Guillén-Navarro, C. Fernández-López, Pharmaceutical biological degradation, sorption and mass balance determination in a conventional activated-sludge wastewater treatment plant from Murcia, Spain, *Chem. Eng. J.*, 316 (2017) 332–340.
- [7] F. Jung, M.C. Cammarota, D.M.G. Freire, Impact of enzymatic pre-hydrolysis on batch activated sludge systems dealing with oily wastewaters, *Biotechnol. Lett.*, 24 (2002) 1797–1802.
- [8] G.T. Tellez, N. Nirmalakhandan, J.L. Gardea-Torresdey, Performance evaluation of an activated sludge system for removing petroleum hydrocarbons from oilfield produced water, *Adv. Environ. Res.*, 6 (2002) 455–470.
- [9] E. Raper, A. Soares, J. Chen, A. Sutcliffe, E. Aries, D. Anderson, T. Stephenson Enhancing the removal of hazardous pollutants from coke-making wastewater by dosing activated carbon to a pilot-scale activated sludge process, *J. Chem. Technol. Biotechnol.*, 92 (2017) 2325–2333.
- [10] M.A. Cardete, J. Mata-Álvarez, J. Dosta, R. Nieto-Sánchez, Sludge settling enhancement in a pilot scale activated sludge process treating petrochemical wastewater by implementing aerobic or anoxic selectors, *J. Environ. Chem. Eng.*, 5 (2017) 3472–3482.
- [11] E. Loupasaki, E. Diamadopoulos, Attached growth systems for wastewater treatment in small and rural communities: a review, *J. Chem. Technol.*, 88 (2013) 190–204.
- [12] J. Chung, W. Bae, Y.W. Lee, B.E. Rittmann, Shortcut biological nitrogen removal in hybrid biofilm/suspended growth reactors, *Process Biochem.*, 42 (2007) 320–328.
- [13] Y. Zaoyan, S. Ke, S. Guangliang, Y. Fan, D. Jinshan, M. Huanian, Anaerobic-aerobic treatment of a dye wastewater by combination of RBC with activated sludge, *Water Sci. Technol.*, 26 (1992) 2093–2096.
- [14] S.J. You, C.L. Hsu, S.H. Chuang, C.F. Ouyang, Nitrification efficiency and nitrifying bacteria abundance in combined AS-RBC and A<sub>2</sub>O systems, *Water Res.*, 37 (2003) 2281–2290.
- [15] D. Di Trapani, M. Christensson, M. Torregrossa, G. Viviani, H. Ødegaard, Performance of a hybrid activated sludge/biofilm process for wastewater treatment in a cold climate region: influence of operating conditions, *Biochem. Eng. J.*, 77 (2013) 214–219.
- [16] Y.K. Park, C.H. Lee, Dyeing wastewater treatment by activated sludge process with a polyurethane fluidized bed biofilm, *Water Sci. Technol.*, 34 (1996) 193–200.
- [17] S. Aslan, B. Alyüz, Z. Bozkurt, M. Bakaoglu, Characterization and biological treatability of edible oil wastewaters, *Pol. J. Environ. Stud.*, 18 (2009) 533–538.
- [18] F. Gebara, Activated sludge biofilm wastewater treatment system, *Water Res.*, 33 (1999) 230–238.
- [19] J.L. Su, C.F. Ouyang, Nutrient removal using a combined process with activated sludge and fixed biofilm, *Water Sci. Technol.*, 34 (1996) 477–486.
- [20] K. Tang, G.T.H. Ooi, E. Toressi, K.M.S. Kaarsholm, A. Hambly, K. Sundmark, S. Lindholm, C. Sund, C. Kragelund, M. Christensson, K. Bester, H.R. Andersen, Municipal wastewater treatment targeting pharmaceuticals by a pilot-scale hybrid attached biofilm and activated sludge system (Hybas™), *Chemosphere*, 259 (2020), 127397, <https://doi.org/10.1016/j.chemosphere.2020.127397>.
- [21] K. Hassan, O. Hamdy, M. Helmy, H. Mostafa, Enhancing treated wastewater effluent characteristics using hybrid biofilm/

- activated sludge process—a case study, *Water Sci. Technol.*, 81 (2020) 217–227.
- [22] H.T. Dang, C.V. Dinh, K.M. Nguyen, N.T. Tran, T.T. Pham, R.M. Narbaitz, Loofah sponges as bio-carriers in a pilot-scale integrated fixed-film activated sludge system for municipal wastewater treatment, *Sustainability*, 12 (2020), 4758–4773.
- [23] M. Bagheri, K. Al-jabery, D. Wunsch, J.G. Burken, Examining plant uptake and translocation of emerging contaminants using machine learning: implications to food security, *Sci. Total Environ.*, 698 (2020), 133999–134011.
- [24] L. Rossi, M. Bagheri, W. Zhang, Z. Chen, J.G. Burken, X. Ma, Using artificial neural network to investigate physiological changes and cerium oxide nanoparticles and cadmium uptake by *Brassica napus* plants, *Environ. Pollut.*, 246 (2019) 381–389.
- [25] M.B. Fard, S.A. Mirbagheri, A. Pendashteh, J. Alavi, Biological treatment of slaughterhouse wastewater: kinetic modeling and prediction of effluent, *J. Environ. Health Sci.*, 17 (2019) 731–741.
- [26] M. Bagheri, K. Al-jabery, D.C. Wunsch, J.G. Burken, A deeper look at plant uptake of environmental contaminants using intelligent approaches, *Sci. Total Environ.*, 651 (2019) 561–569.
- [27] M. Bagheri, A. Akbari, S.A. Mirbagheri, Advanced control of membrane fouling in filtration systems using artificial intelligence and machine learning techniques: a critical review, *Process Saf. Environ.*, 123 (2019) 229–252.
- [28] H.A. Mokhtari, M. Bagheri, S.A. Mirbagheri, A. Akbari, Performance evaluation and modelling of an integrated municipal wastewater treatment system using neural networks, *Water Environ. J.*, (2020) (in Press), <https://doi.org/10.1111/wej.12565>.
- [29] F. Shokri Dariyan, A. Eslami, E. Aghayani, M. Pourakbar, A. Oghazyan, Comparison of artificial neural network and multi-kinetic models to predict optimum retention time for dairy wastewater treatment in the integrated fixed-film activated sludge, *Int. J. Environ. Anal. Chem.*, (2020), <https://doi.org/10.1080/03067319.2020.1785442>.
- [30] A. Noroozi, M. Farhadian, A. Solaimanyazar, Kinetic coefficients for the domestic wastewater treatment using hybrid activated sludge process, *Desal. Water Treat.*, 57 (2016) 4439–4446.
- [31] N. Delgrange, C. Cabassud, M. Cabassud, L. Durand-Bourlier, J.M. Laine, Neural networks for prediction of ultrafiltration transmembrane pressure—application to drinking water production, *J. Membr. Sci.*, 150 (1998) 111–123.
- [32] Y. Yang, K. Tsukahara, S. Sawayama, Performance and methanogenic community of rotating disk reactor packed with polyurethane during thermophilic anaerobic digestion, *Mater. Sci. Eng.*, 27 (2007) 767–772.
- [33] Y. Liu, Y.M. Lin, S.F. Yang, J.H. Tay, A balanced model for biofilms developed at different growth and detachment forces, *Process Biochem.*, 38 (2003) 1761–1765.
- [34] A. Gjaltema, L. Tjihuis, M. Van Loosdrecht, J.J. Heijnen, Detachment of biomass from suspended nongrowing spherical biofilms in airlift reactors, *Biotechnol. Bioeng.*, 46 (1995) 258–269.
- [35] P.S. Kodukula, T. Prakasam, A.C. Anthonisen, Role of pH in Biological Wastewater Treatment Processes, M. Bazin, Ed., *Physiological Models in Microbiology*, CRC Press, Boca Raton, FL, 2018, pp. 113–135.
- [36] W. Jianlong, S. Hanchang, Q. Yi, Wastewater treatment in a hybrid biological reactor (HBR): effect of organic loading rates, *Process Biochem.*, 36 (2000) 297–303.
- [37] S.A. Mirbagheri, S. Malekmohamadi, M. Ehteshami, Designing activated carbon and zeolite amended biosand filters: optimization using response surface methodology, *Desal. Water Treat.*, 93 (2017) 48–60.
- [38] American Public Health Association and American Water Works Association, *Standard Methods for the Examination of Water and Wastewater*, American Public Health Association, 1989.
- [39] S.S. Haykin, *Neural Networks and Learning Machines/Simon Haykin*, Prentice Hall, New York, NY, 2009.
- [40] G. Onkal-Engin, I. Demir, S.N. Engin, Determination of the relationship between sewage odour and BOD by neural networks, *Environ. Modell. Softw.*, 20 (2005) 843–850.
- [41] M. Kubat, *Artificial Neural Networks*, M. Kubat, Ed., *An Introduction to Machine Learning*, Springer, Cham, Switzerland, 2015, pp. 91–111.
- [42] Z.H. Zhou, J. Wu, W. Tang, Ensembling neural networks: many could be better than all, *Artif. Intell.*, 137 (2002) 239–263.
- [43] R. Soleimani, N.A. Shoushtari, B. Mirza, A. Salahi, Experimental investigation, modeling and optimization of membrane separation using artificial neural network and multi-objective optimization using genetic algorithm, *Chem. Eng. Res. Des.*, 91 (2013) 883–903.
- [44] U.S. Environmental Protection Agency, *National Recommended Water Quality Criteria-Correction*, EPA 822-Z-99-001, 1999.
- [45] F. Ma, J.B. Guo, L.J. Zhao, C.C. Chang, D. Cui, Application of bioaugmentation to improve the activated sludge system into the contact oxidation system treating petrochemical wastewater, *Bioresour. Technol.*, 100 (2009) 597–602.
- [46] S. Delin, W. Jianlong, L. Kaiwen, Z. Ding, Kinetic performance of oil-field produced water treatment by biological aerated filter, *Chin. J. Chem. Eng.*, 15 (2007) 591–594.
- [47] K. Tong, Y. Zhang, G. Liu, Z. Ye, P. Chu, Treatment of heavy oil wastewater by a conventional activated sludge process coupled with an immobilized biological filter, *Int. Biodeterior. Biodegrad.*, 84 (2013) 65–71.
- [48] I.N. Dias, A.C. Cerqueira, G.L. Sant’Anna, M. Dezotti, Oil refinery wastewater treatment in biofilm reactor followed by sand filtration aiming water reuse, *J. Water Reuse Desal.*, 2 (2012) 84–91.
- [49] S. Shokrollahzadeh, F. Azizmohseni, F. Golmohammad, H. Shokouhi, F. Khademhaghighat, Biodegradation potential and bacterial diversity of a petrochemical wastewater treatment plant in Iran, *Bioresour. Technol.*, 99 (2008) 6127–6133.
- [50] H. Gasim, A.R.M.M.A. Megat, R.M.K. Shamsul, Treatment of petroleum refinery wastewater using extended aeration activated sludge, *Int. J. Eng. Res. Afr.*, 13 (2015) 1–7.
- [51] W. Xie, L. Zhong, J. Chen, Treatment of slightly polluted wastewater in an oil refinery using a biological aerated filter process, *Wuhan Univ. J. Nat. Sci.*, 12 (2007) 1094–1098.
- [52] G.H. Ahmed, S.R.M. Kutty, M.H. Isa, Petroleum refinery effluent biodegradation in sequencing batch reactor, *Int. J. Appl. Sci.*, 1 (2011) 179–183.
- [53] M. Perez, R. Rodriguez-Cano, L. Romero, D. Sales, Performance of anaerobic thermophilic fluidized bed in the treatment of cutting-oil wastewater, *Bioresour. Technol.*, 98 (2007) 3456–3463.
- [54] N. Otadi, A. Hassani, A. Javid, F. Khiabani, Oily compounds removal in wastewater treatment system of pars oil refinery to improve its efficiency in a lab scale pilot, *J. Water Chem. Technol.*, 32 (2010) 370–377.
- [55] E. Sekman, S. Top, E. Uslu, G. Varank, M. Bilgili, Treatment of oily wastewater from port waste reception facilities by electrocoagulation, *Int. J. Environ. Res.*, 5 (2011) 1079–1086.
- [56] N.S. Dumore, M. Mukhopadhyay, Removal of oil and grease using immobilized triacylglycerin lipase, *Int. Biodeterior. Biodegrad.*, 68 (2012) 65–70.
- [57] Y. Wang, Q. Wang, M. Li, Y. Yang, W. He, G. Yan, S. Guo, An alternative anaerobic treatment process for treatment of heavy oil refinery wastewater containing polar organics, *Biochem. Eng. J.*, 105 (2016) 44–51.
- [58] C.Y. Cao, Y.H. Zhao, The comparison of MBBR and ASP for treatment on petrochemical wastewater, *Pet. Sci. Technol.*, 30 (2012) 1461–1467.
- [59] S.A. Mirbagheri, M. Ebrahimi, M. Mohammadi, Optimization method for the treatment of Tehran petroleum refinery wastewater using activated sludge contact stabilization process, *Desal. Water Treat.*, 52 (2014) 156–163.
- [60] M. Hamoda, A. Al-Haddad, Treatment of petroleum refinery effluents in a fixed-film reactor, *Water Sci. Technol.*, 20 (1988) 131–140.
- [61] L. Zhidong, L. Na, Z. Honglin, L. Dan, Study of an A/O submerged membrane bioreactor for oil refinery wastewater treatment, *Pet. Sci. Technol.*, 27 (2009) 1274–1285.
- [62] M. Bagheri, S.A. Mirbagheri, Critical review of fouling mitigation strategies in membrane bioreactors treating water and wastewater, *Bioresour. Technol.*, 258 (2018) 318–334.

- [63] T.P. Moisés, B.H. Patricia, C. Barrera-Díaz, R.M. Gabriela, R. Natividad-Rangel, Treatment of industrial effluents by a continuous system: electrocoagulation-activated sludge, *Bioresour. Technol.*, 101 (2010) 7761–7766.
- [64] B.K. Körbahti, K. Artut, Electrochemical oil/water demulsification and purification of bilge water using Pt/Ir electrodes, *Desalination*, 258 (2010) 219–228.
- [65] S.G. Velioglu, K. Curi, S.R. Çamlilar, Activated sludge treatability of olive oil-bearing wastewater, *Water Res.*, 26 (1992) 1415–1420.
- [66] A. Pendashteh, A. Fakhru'l-Razi, T. Chuah, A.D. Radiah, S. Madaeni, Z. Zurina, Biological treatment of produced water in a sequencing batch reactor by a consortium of isolated halophilic microorganisms, *Environ. Technol.*, 31 (2010) 1229–1239.
- [67] A. Giwa, S. Daer, I. Ahmed, P. Marpu, S. Hasan, Experimental investigation and artificial neural networks ANNs modeling of electrically-enhanced membrane bioreactor for wastewater treatment, *J. Water Process Eng.*, 11 (2016) 88–97.
- [68] D. Aguado, J. Ribes, T. Montoya, J. Ferrer, A. Seco, A methodology for sequencing batch reactor identification with artificial neural networks: a case study, *Comput. Chem. Eng.*, 33 (2009) 465–472.
- [69] S. Mirbagheri, M. Bagheri, M. Ehteshami, Z. Bagheri, M. Pourasghar, Modeling of mixed liquor volatile suspended solids and performance evaluation for a sequencing batch reactor, *J. Urban Environ. Eng.*, 9 (2015) 54–65.
- [70] M. Bagheri, S.A. Mirbagheri, M. Ehteshami, Z. Bagheri, A.M. Kamarkhani, Analysis of variables affecting mixed liquor volatile suspended solids and prediction of effluent quality parameters in a real wastewater treatment plant, *Desal. Water Treat.*, 57 (2016) 21377–21390.
- [71] A.R. Pendashteh, A. Fakhru'l-Razi, N. Chaibakhsh, L.C. Abdullah, S.S. Madaeni, Z.Z. Abidin, Modeling of membrane bioreactor treating hypersaline oily wastewater by artificial neural network, *J. Hazard. Mater.*, 192 (2011) 568–575.
- [72] H. Gong, R. Pishgar, J. Tay, Artificial neural network modelling for organic and total nitrogen removal of aerobic granulation under steady-state condition, *Environ. Technol.*, 40 (2019) 3124–3139.