Modeling the Fate of Phenol in Moving Bed Biofilm Reactor Sewage Treatment Plant

Haider M. Zwain, Mohammadtaghi Vakili, Ahmed M. Faris and Irvan Dahlan

Abstract Phenol is one of a major pollutant existing in the wastewaters from several industrial activities. Biological removal of the phenolic compounds at full scale treatment plant is very complicated processes. This study investigates the fate of phenol biodegradation in moving bed biofilm reactor sewage treatment plant (MBBR-STP) was investigated. A computer-based mechanistic model, TOXCHEM V4.1, and as well as sensitivity analysis were used to predicts the fate of phenol biodegradation in MBBR-STP. The results showed that MBBR-STP has successfully achieved removal efficiency of 90%, 87%, 100%, 89%, 48%, 90%, and 96% for COD and BOD_5, PO_4, H_2S, oil and grease, SO_4, NH_4, TSS, and phenol, respectively. However, a high nitrate (NO_3) concentration of 60 mg/L was produced, indicating the occurrence of nitrification process. The phenol fate analysis demonstrated that 0.3, 0.5, 6, 93.2% went for volatilization, sorption, untreated and biodegradation, respectively. Nevertheless, the sensitivity analysis indicated that increasing the influent flow rate from 200 to 1000 m^3/day has reduced biodegradability of phenol from 96 to 82%, respectively, whereas increasing MLSS

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concentration 500–6000 mg/L incorporated resilient sorption and degradation processes by 0.2–66% and 93–34%, respectively, and MBBR media fill fraction of 88% achieved about 100% phenol removal. The results conclude that MBBR-STP is efficient in removing phenol, and the TOXCHEM V4.1 has successfully predict the fate of phenol.

Keywords Modeling · Fate · Phenol · Biodegradation · Moving bed biofilm reactor (MBBR) · Sewage treatment plant

1 Introduction

Organic pollution of surface water has become one of the most emerging environmental problems worldwide. This is due to the discharge of huge amounts of organic compounds from numerous sources, derived from agricultural runoff, industrial effluents, and domestic sewage, into the water bodies. Phenol and its derivatives are among the most toxic organic pollutants for humans and aquatic life and they are difficult to treat. The presence of phenolic components is increasing in water resources due to its extensive applications [2].

In domestic sewage, phenol is a compound that is in various household chemicals such as slimicides, antiseptics, and disinfectants. Phenol is also found in other household products such as perfumes soaps, varnish removers, paints, toys, and lacquers. In addition, it's also found in pharmaceutical or medical products including body ointments, lotions, mouthwashes, and sore throat treatment oral anesthetics. Phenol is an aromatic organic compound consisted of hydroxyl group tied to a hydrocarbon compound of benzene ring. Phenol is denser than water and sinks to the bottom. It's slightly soluble in water and continues to form toxic solutions even at low concentration [5].

Due to its adverse health effects, phenol is one of the 129 top toxic chemicals enlisted by the United States Environmental Protection Agency (USEPA) as pollutants of priority concern. The World Health Organization (WHO) has set a water purity standard of 1 mg/L as the maximum permissible phenol concentration in drinking water [10]. Meanwhile, phenol concentration in the range of 5–25 mg/L is toxic for both humans and aquatic life [3]. As an aromatic compound, phenol is resistant to natural degradation and persist in the environment. It is capable of long-range transportation and bioaccumulation in animal and human tissue [2].

Therefore, phenol must be removed from wastewater before discharge into water bodies. Various physical, chemical, and biological methods have been developed and applied for the treatment of phenol. However, the application of physico-chemical treatment methods is quite expensive and not completely effective. Furthermore, these methods do not degrade phenols but rather transfer it to another compounds, resulting in the formation of hazardous toxic secondary byproducts [3]. On the other hand, the biological treatment of phenols is considered a favorable alternative because of the opportunity of complete degradation to nontoxic products and lower operation expenses. There are several microorganisms that can tolerate phenol and consume it as a carbon and energy source. In particular, aerobic process is more effective due to the presence of large number of microorganisms including bacteria, fungi, and algae that grow faster and commonly transform organic compounds to inorganic [2].

Attached growth reactors were found more resistant to high concentrations of phenol, which results in higher removal efficiency more than suspended growth reactors. Moving Bed Biofilm Reactors (MBBR) is one of the most efficient attached growth aerobic process that has several advantages such as no clogging issues, better biofilm thickness control and lower pressure drop [1]. Therefore, several wastewater treatments plants (WWTPs) are using MBBR. However, none of them were reported for the treatment of phenol at full scale, except for some laboratory studies [1, 7, 8].

Furthermore, very limited data on the removal mechanisms of phenol at full scale sewage treatment plant is available. There are quite many models that have been applied to predict the behavior of emerging contaminants of sewer networks and wastewater treatment plants, which require empirical derivations of mass transfer coefficients (kL and kG). These include BASTE, CINCI, WATER9, TOXCHEM, and Gostelow. From these models, the most widely applied models are WATER9, Gostelow and TOXCHEM [11]. Therefore, this study aims to understand the behavior of phenol biodegradation at full scale MBBR sewage treatment plant (MBBR-STP) located in Karbala Governorate, Iraq. TOXCHEM V4.1 model was used to predict the fate and removal mechanisms of phenol in the full scale MBBR-STP.

2 Materials and Methods

2.1 MBBR-STP

A full-scale sewage treatment plant employing MBBR system used in the present study is shown in Fig. 1. The MBBR-STP is located in Karbala Governorate, about 110 km south of Baghdad, Iraq. It was designed to serve a residential area of 4000 capita, with an estimated discharge flow rate of 800 m^3/day. It is consisted of grit chamber, equalization tank with a surface area of 50 m^3, two MBBR tanks with 80 m^3 each, lamella settling tank of 50 m^3; along with tertiary treatment process consisted of sand filter and disinfection unit using sodium hypochlorite (NaClO). The packing materials used in the MBBR-STP are made of high-density polyethylene (HDPE), having a surface area of 400 m^2/m^3, 15 mm in height and 20 mm in diameter, and constituted 35% of the aeration tank volume. The system had a mixed liquor suspended solids (MLSS) of 1000 mg/L.

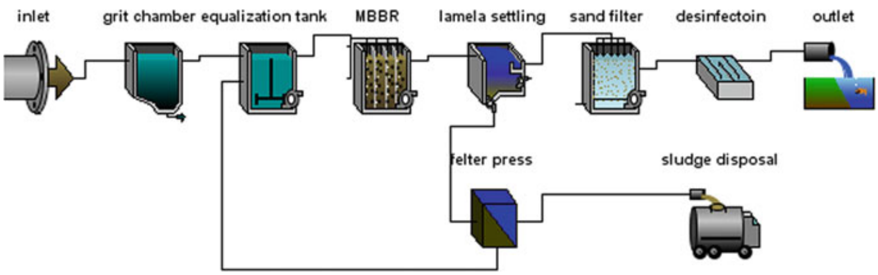

Fig. 1 Schematic diagram of sewage treatment plant employing MBBR system in Karbala, Iraq

2.2 Sampling and MBBR-STP Operation

To evaluate the performance of the MBBR-STP, 12 samples were taken on a monthly basis from inlet and outlet, and then analyzed for pH, COD, BOD_5, PO_4, H_2O, O, SO_4, NO_3, NH_4, TSS and phenol. The phenol was measured using Perkinelmer clarus 500 GC-MS equipped with Clarus 600T equipped with head-space (Perkin Elmer, Connecticut, USA). The average results of the operating performance of the MBBR-STP are shown in Table 1. The effluent results have been compared according to the Iraqi effluent standard [4] for the use of secondary treated effluents in agricultural irrigation [4].

2.3 TOXCHEM Model Setup

Phenol was selected as the model micro-organic compound due to it is increasingly detected in wastewater, adverse health effects, resistant to natural degradation, and

Table 1 Operating performance of MBBR sewage treatment plant

Parameter	Inlet concentration (mg/L)	Outlet concentration (mg/L)	Limits of effluent (mg/L)
pH*	7.2	7.5	6–9
COD	450	45	100
BOD_5	400	40	40
PO_4	8	1	3
H_2S	45	ND	3
Oil and grease	90	10	10
SO_4	200	104	400
NO_3	0	60	50
NH_4	5	0.5	10
TSS	250	25	60
Phenol	1.6	0.06	0.05

*Unitless

hard to be treated at biological sewage treatment plants. TOXCHEM is notable mass balance-based model operated to predict the behavior of compounds in WWTP's under either steady state or dynamic conditions.

In this study, TOXCHEM V4.1 was used to predict the fate of phenol at MBBR-STP. TOXCHEM V4.1 model simulates removal rates by a combination of mass transfer, biodegradation, and sorption, but not all of these processes would happen for each compound. In this study, the removal of phenol by biodegradation and volatilization and sorption in the equalizer, MBBR and secondary clarifier were of interest.

Due to the combination of treatment mechanisms in MBBR system, attached and suspended growth biodegradations were applied in the model. Biodegradation of phenol in small concentration during suspended growth processes was expressed as:

$$r_b = k_b \left(\frac{C}{1 + \frac{C}{K_s}} \right) XV \tag{1}$$

Furthermore, the biodegradation kinetics of phenol as a trace contaminant during biofilm processes was expressed as:

$$rb = kb \times f \times CV \tag{2}$$

where r_b is the biodegradation rate (mg/h), k_b is the first order biodegradation rate coefficient (L/mg VSS/h), X is the biomass concentration (mg/L), X_f is the biofilm density (mg/L), C is the contaminant concentration (mg/L), Ks is the half saturation constant (mg/L), and V is the vessel volume (L).

Pollutants can move from the liquidity state to the dead mass and also to the remaining suspended solid materials by sorption. Sorption of phenol onto the sludge was represented by a linear isotherm in low contaminant concentrations, expressed as:

$$q = Kp \times C \tag{3}$$

where q is the concentration of contaminant in solid phase (μg/g), and K_p is the sorption partition coefficient (L/g).

For the mass transfer mechanisms, volatilization of phenol from the surfaces of equalizer, MBBR and secondary clarifier was expressed as:

$$KL = \left(\frac{1}{kL} + \frac{1}{kGH} \right)^{-1} \tag{4}$$

where *KL* is the overall mass transfer coefficient, *H* is the Henry's law coefficient, k_L is the liquid phase mass transfer coefficient (m/h), and k_G is the gas phase mass transfer coefficient (m/h).

2.4 Sensitivity Analysis

Sensitivity analysis is very important indicator to know the influence of different parameters on the output of the model and this analysis process is necessary for the calibration of the model. In this study, a sensitivity analysis was performed to understand the fate of phenol using the most affecting parameters on the treatment process in MBBR systems which are influent flow rate, MLSS, and MBBR media fill fraction. Different flow rate (200–1000 m^3/day), MLSS concentration (500–1000 mg/L), and MBBR media fill fraction (18–88%) were applied.

3 Results and Discussion

3.1 The MBBR-STP Performance

The performance of the MBBR-STP at a residential area in Karbala, Iraq during half capacity operation is shown in Table 1. The MBBR-STP had performed very well to treat most of the parameters in accordance to the Iraqi standard. Removal efficiency of 90%, 87%, 89%, 48%, 90%, and 96% were achieved for both of COD and BOD_5, PO_4, oil and grease, SO_4, NH_4, TSS, and phenol, respectively.

H_2S was not detected at the effluent due to its emission to the atmosphere during the treatment process. The results also showed an increase in the nitrate concentration in the effluent due to the oxidation of ammonium (NH_4) and low organic content in the sewage, which is very common for aerobic wastewater treatment process [12]. The increased NO_3 was found to be associated with a decreased NH_4, indicating the occurrence of nitrification process and organic compounds are not sufficient as electron donors for denitrification. Interestingly, oil and grease have been significantly removed at ambient temperature. This confirms the outstanding ability of MBBR-STP systems in removing oil compounds, due to the attached growth process bacteria (i.e. *Stenotrophomonas rhizophila*, *Sphingobacterium* sp., *Pseudomonas libanensis*, *Pseudomonas aeruginosa*, and *Pseudomonas poae*) that are more efficient to degrade and break large molecules of oil and grease by secretion of the lipases and esterases enzymes [6]. In general, the treatment process had a stable neutral pH level due to the adequate amount of carbonate in the effluent, even if the nitrification process consumed carbonate.

3.2 The Fate of Phenol

The fate of phenol at the MBBR-STP is shown in Fig. 2. The results showed that 93% of the phenol is removed by degradation at the MBBR process, 6% of phenol was not treated and discharged with the effluent, 0.5% was by sorption at the biomass and solids surfaces in the system, and 0.3% was by emission to the air. This indicated that most of the phenol was successfully degraded at the MBBR-STP. This could be due to the low phenol concentration that didn't inhabit degradation process and the ability of MBBR system to tolerate toxic contaminants. This is similar to several studies who confirmed the effectiveness of MBBR systems in treating phenol compounds even at high concentrations [1, 13, 14]. In a laboratory scale MBBR treating synthetic wastewater, Zhou et al. [14] reported that up to 90% of phenol was degraded. In addition to nitrification process in the system, phenol degradation process suggested an interspecific cooperation removal mechanism. While Zheng et al. [13] indicated that *Pseudomonadales*, *Burkholderiales* and *Bdellovibrionales* were predominate bacteria in MBBR system degrading phenol and ammonia.

Furthermore, phenol volatilization to air at the MBBR-STP is shown in Fig. 3. It was observed that phenol emission was safe at all MBBR-STP units according to the hotspots rage, and most of the emission (85%) occurred at the equalizer and gradually decreased towards the end of the treatment plant. This indicates that phenol is rapidly volatized once it's entered the sewage treatment plant, however, air/odor treatment facilities are not required in MBBR system treating sewages.

3.3 Sensitivity Analysis

The biodegradation of phenols is affected by several factors such as substrate inhibition effect, biomass concentration and their formation process, microbial community and their metabolic potential, and nutrient concentration. In this research, sensitivity analysis was performed at different influent flow rate, MBBR MLSS concentration, and MBBR media fill fraction. Figure 4 shows the sensitivity of

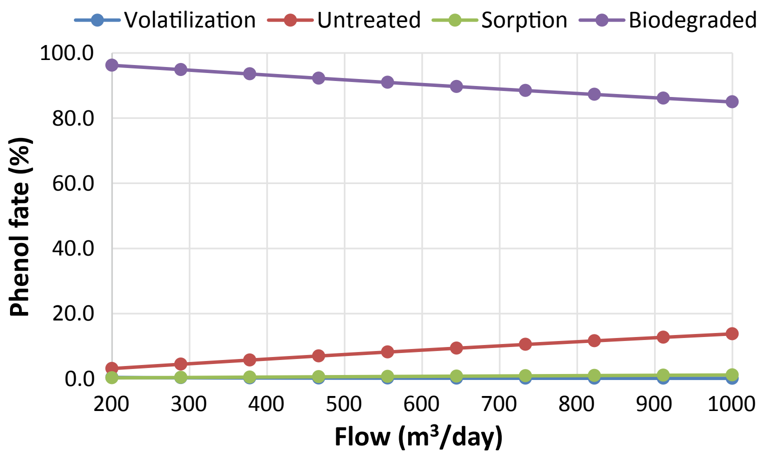

Fig. 2 The fate of phenol at MBBR-STP

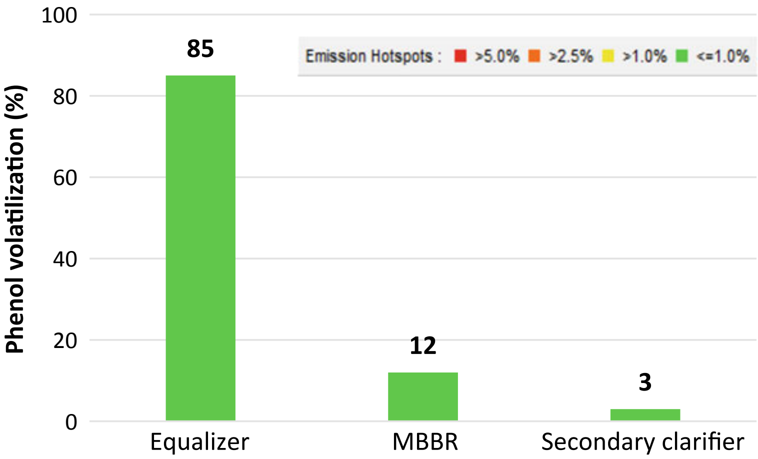

Fig. 3 The evaluation of phenol volatilization to air at MBBR-STP

different influent flow rate on the fate of phenol at MBBR-STP. The flow rate was varied from 200 to 1000 m^3/day, while actual operational flow rate was 400 m^3/day. The results showed a reduction in phenol removal when the flow rate was increased leading to decrease in biodegradability rate up to 82%. Reduction in phenol removal could be due to increased phenol concentration in the system. Compared to volatilization and sorption, biodegradation was more sensitive towards variation in flow rate, and some of the phenol was left untreated. Similarly, Zheng et al. [13] implied that high phenols loading suppressed the activity of phenolic degraders, resulting in decreased phenol utilization rate (PUR). However, phenol removal remains high at maximum flow rate of 1000 m^3/day due to the ability of MBBR systems to rapidly form biomass that can absorb load fluctuation [1].

Figure 5 depicted the effect of MLSS concentration variation on the biodegradation of phenols in MBBR-STP. The actual operational MLSS concentration was 1000 mg/L that contributed to 93% phenol degradation. The sensitivity analysis was modeled at MLSS ranged from 500 to 6000 mg/L. From the results, it was observed that low MLSS concentration did not affect the fate of phenol and its removal remains high as 94%. On the other hand, increasing the MLSS concentration beyond 3000 mg/L has changed the trends. At MLSS concentration of 5500–6000 mg/L, phenol biodegradation decreased to 34% and phenol sorption was increased up to 66%. It was found that sorption and biodegradation were more sensitive towards variation in MLSS concentration with no phenol left untreated. This reveals that higher MLSS concentration will offer larger surface area for phenol sorption instead of biomass for degradation process, resulting in sludge containing phenol compounds. Increasing sludge in the system may cause bioactivity decay, leading to severe foaming, and indicating distinct impact resistance to phenols. The accumulated organic pollutants in the sludge implying considerable decomposed biomass and overload pressure, which is induced by phenolic toxicity [13].

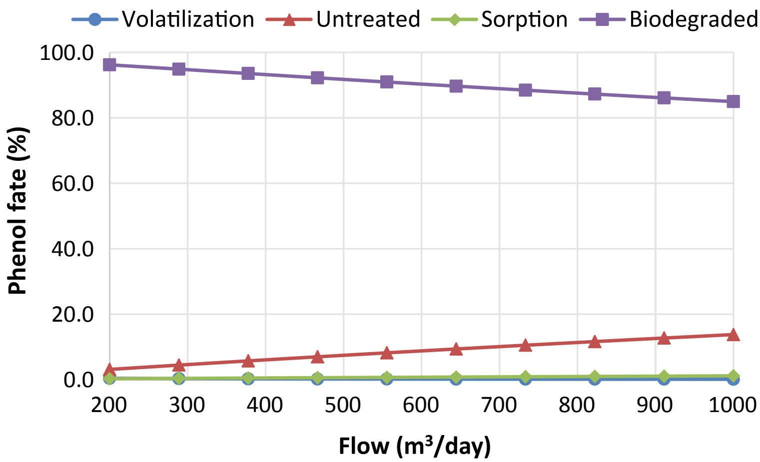

Fig. 4 Sensitivity analysis of phenol fate at different influent flow rate at MBBR-STP

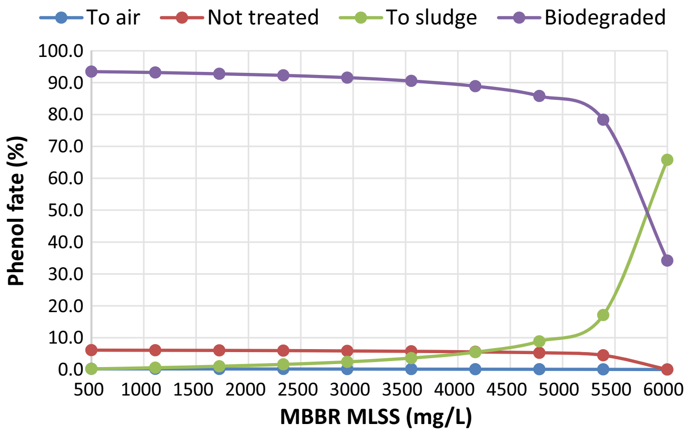

Fig. 5 Sensitivity analysis of phenol fate at different MLSS concentration at MBBR-STP

At different MBBR media fill fraction, the phenol fate at MBBR-STP is shown in Fig. 6. The percentage of MBBR media fill fraction was varied at 18–88% of the total MBBR unit volume. The existing operational MBBR media fill fraction was 35%, which resulted in 93% of phenol degradation. The increase in MBBR media fill fraction demonstrated increase in phenol degradation, leading to phenol removal up to 100%. This reveal that phenol degradation was more sensitive towards variation in of MBBR media fill fraction. High MBBR media fill fraction amplified the moving media containing a thin biofilm, leading to enhanced biomass activity in the reactor. This will improve the contact between substrate and biomass, and

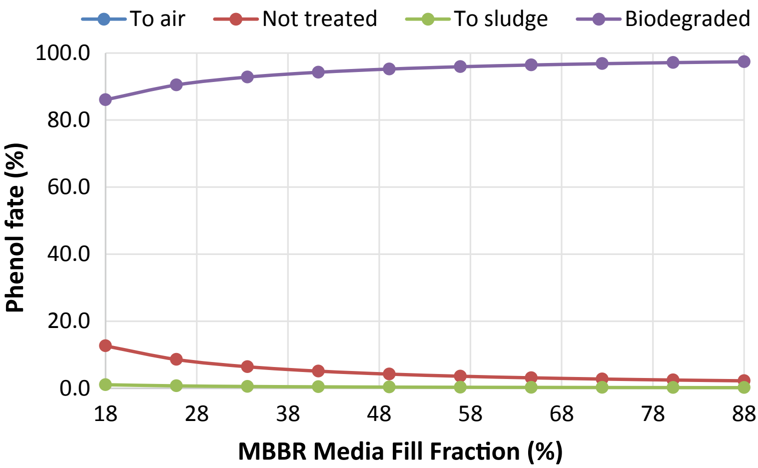

Fig. 6 Sensitivity analysis of phenol fate at different MBBR media fill fraction at MBBR-STP

transfer rate of oxygen and substrate, thus, enhances degradation rate of phenol [9]. Increasing media fill ratio offers differences in biofilm structure and composition. As the media fill ratio increased, the minimum air flux for carrier fluidization increased. Subsequently, the shear stresses on biofilm formed became higher and the biofilm became smoother with higher density [7].

4 Conclusion

The full scale MBBR-STP treating of domestic sewage has performed very well for the removal of organic contaminants, phenol and ammonia, except for the nitrate. The TOXCHEM V4.1 has successfully predicted the fate of phenol at the equalizer, MBBR and secondary clarifier. Most of the phenol removal took a place in the MBBR unit, mostly by degradation up to 93%, and volatilization to air and sorption were limited. The sensitivity analysis indicated that increase in influent flow rate has decreased the phenol biodegradability rate to 82% with 13% remains untreated, while increase in MBBR MLSS concentration has amplified phenol sorption capacity from up to 66% and increment in MBBR media fill fraction has enhanced biodegradability of phenol up to 100%.

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