Effect of hydraulic retention time and influent substrate concentration on nutrient removal and membrane fouling in an anoxic/anaerobic-aerobic MBR

Zubair Ahmed [a*]

[a] Department of Civil Engineering, King Abdulaziz University, Jeddah, Kingdom of Saudi Arabia
Telephone number: +966-2-640-2000 (ext. 68239);
Fax number: +966-2-695-2179
Email: himatali@gmail.com

ABSTRACT

In this study, effects of organic loading rates on biological nutrient removal and in an MBR based nutrient removal system were investigated. Furthermore, trans-membrane pressure at different applied conditions was monitored to observe impact on membrane fouling Three different levels of COD_{inf} and three HRTs (4, 5 & 8 hrs) were applied. An improved performance with 74 % & 89% of TN & TP removal respectively was observed when the SAM was fed with high COD_{inf}. When the MBR based BNR process was operated at relatively higher influent COD and at short HRT, efficient TN and TP removal efficiencies were observed at stable TMP variation throughout the test. The membrane fouling was accelerated at short HRT but suppressed when concentration of COD_{inf} was increased. The optimum specific organic loading rate was found to be 0.3 kg $\text{COD}\cdot\text{kg}^{-1}$ $\text{MLVSS}\cdot\text{day}$ for the MBR based BNR system.

KEYWORDS: Hydraulic retention time, organic loading rate, nitrogen, phosphorus, membrane bioreactor, membrane fouling.

INTRODUCTION

Excessive addition of nitrogen and phosphorus to surface water is a well known contributor to eutrophication of aquatic environments. One of the major sources of these nutrients is the discharge from wastewater treatment plants. Therefore, removal of nitrogen and phosphorus is becoming a strict legislative requirement for such discharge (1). The membrane bioreactor (MBR) based nutrient removing process has many advantages over conventional processes e.g. small footprint and reactor requirements, high effluent quality, good disinfection capability, higher volumetric loading and less sludge production (2). The membrane technology, with its versatile separation capability, is making an impact in a number of treatment areas including enhanced biological nutrient removal (BNR) processes due to possibility of keeping wide range of denitrifiers and phosphate accumulating organisms in membrane based systems (3). Ahn et al.(4, 5) developed an MBR based nutrient-removing system and demonstrated its better performance in comparison to modified Ludzack-Ettinger type MBR for simultaneous nitrogen and phosphorus removing in domestic wastewater.

In countries such as Korea, municipal wastewater usually contains low levels of soluble organic carbon (COD approximately 100 $\text{mg}\bullet\text{L}^{-1}$) and relatively high levels of nutrients (4 mg phosphorus L^{-1} and 25 mg nitrogen L^{-1})(6). The limited availability of organic substrate is a concern in the design and operation of BNR systems. Enhanced BNR processes depend on the nature and concentration of carbonaceous organic compounds undergoing degradation (7). A $\text{COD}/\text{NO}_x\text{-N}$ ratio in the range of 5-7 has been suggested for efficient denitrification in conventional denitrification systems (8). Particularly, the success of activated sludge plants for enhanced biological phosphorus removal (EBPR) processes largely depends on the composition of the wastewater (9). The availability of organic compounds during the anaerobic phase is important for the storage of polyhydroxybutyrate (PHB) by polyphosphate accumulating organisms (PAOs). During the subsequent aerobic phase, PHB is required for growth of the PAO's and for P-uptake (10, 11). The process doesn't proceed in cases of limited organic substrate. Therefore, wastewater treatment plants designed for EBPR often encounter problems due to low organic loading even for short periods (9). In denitrification systems, such as the SAM, where internal recycling is utilized to create anoxic conditions (5), substrate concentration becomes very important for efficient BNR. The organic loading rate can be increased to achieve good BNR by decreasing the hydraulic retention time (HRT), which is possible in MBR based systems due to presence of membranes. In their study, Song et al. found that the nutrient removal efficiency increased at short HRT (higher flux) along with increases in membrane fouling (12). This inherent problem of enhanced membrane fouling at short HRT can be handle by supplying additional carbon sources in the influent. In this way, nutrient removal in anoxic/anaerobic MBRs can be enhanced at higher flux without frequently need of membrane cleaning when strength of wastewater become temporarily low e.g. during raining season. However, optimum operating conditions for operating flux and requirement of additional carbon should be investigated..

The objective of this study was to investigate the effects of different HRTs and influent substrate concentrations on nitrogen and phosphorus removal and membrane fouling behavior in the MBR utilized for nutrient removal from wastewater.

1. MATERIALS AND METHODS

Laboratory-scale MBR

Two sequential anoxic/anaerobic-aerobic membrane bioreactor were set up, each composed of a sequential anoxic/anaerobic reactor (SAAR), which provided alternating anoxic/anaerobic conditions, and an aerobic reactor (AR) in which a flat-sheet micro-filter (MF) membrane module was immersed (Figure 1). The membrane module (Kubota, Japan) had an effective filtration area of 0.1 m^2 and a nominal pore size of 0.25 μm. The membrane module was fully immersed and placed vertically in the system. The airlift was installed underneath the membrane module to provide dissolved oxygen in the reactor as well as to control membrane fouling by hydraulic shear force and agitation. For permeate generation, outside-in type filtration was induced by a suction pump. The influent was continuously fed into the sequential reactor and the effluent was continuously generated through the submerged membrane in the aerobic reactor. The SAM process applies intermittent internal recycling of the mixed liquor directly from the aerobic reactor to the sequential reactor. The recycling rate (Q_r) from the aerobic reactor to the sequential reactor was maintained at 2.5 times the influent flow (Q_{in}). The internal recycling was continued for 3 h and paused for 1 h. During the recycling period, anoxic conditions were created as the result of the nitrification, which took place in the aerobic reactor, whereas pausing the internal recycling created anaerobic conditions in the sequential reactor. The working volume of the aerobic reactor (AR) and sequential anoxic/anaerobic reactor were 6 L and 4 L respectively. The MBRs were inoculated using sludge taken from a sludge line returning to the aeration tank of a wastewater treatment plant (Gwachon, Republic of Korea). The sludge was allowed to settle for two hours, supernatant was discarded and the SAM reactors were filled with settled sludge (10 liters of sludge for each reactor).

Experimental Details

The operating conditions adopted in this study are given in Table 1. The operational sludge retention time (SRT) was calculated considering the amount of biomass present in both the aerobic reactor and anoxic/anaerobic reactors. The desired SRT was maintained by wasting the excess sludge daily from the aeration reactor of the SAMs. The HRT was set considering the combined volume of anaerobic/anoxic reactor and aerobic reactor as given by the following relation:

$$HRT_{total} = \frac{V_{SAAR} + V_{AR}}{Q_{in}}$$

, where VSAAR and VAR are volumes of anoxic/anaerobic reactors, respectively ,whereas Q_{in} stands for influent flow rate. The MBRs were fed with synthetic wastewater with nutrient. Acetate and glucose were used as organic substrate (in ratio of 7:3). The current study was composed of three phases. The applied concentrations of chemical oxygen demand (COD), total nitrogen (TN) and total phosphorous (TP) in different phases of the experiment are given in Table 2. In phase-I, low and moderate strength synthetic water (in terms of COD) was introduced to the SAMs, which were operated at longer HRT. In phase-II, the HRT was decreased to 4 hrs aiming to increase the organic loading. In phase III, a high ratio of COD/TP was applied (HRT of 5 hr). The influents were named, according to concentration of COD in influent, as low COD_{inf}: 109~109.8 mg•L^{-1}, moderate COD_{inf}: 216.2~227.5 mg•L^{-1},and high COD_{inf}: 331 mg•L^{-1}. The influent COD, TN, and TP concentrations (low and moderate COD_{inf}) were similar in phase I & II but levels of COD and TP were increased in phase III.

A typical concentration profile of nitrate, nitrite and phosphate (track study) was made for a single anoxic/anaerobic cycle of the sequential anoxic/anaerobic reactor (duration of 4 hrs) after steady state conditions were reached in all three phases. Steady state condition was assumed after operation period exceeded twice of applied SRT. Samples from sequential anoxic/anaerobic reactor were collected every 15 minutes during the anaerobic period and every 30 minutes during the anoxic period. Samples were centrifuged, filtered with 0.45μm filter paper and immediately analyzed for nitrate, nitrite and ortho-P. The dissolved oxygen (DO) concentrations and pH values were monitored throughout the study. The DO values were 4.1-4.3 mg•L^{-1} in the aerobic reactor and 0.15-0.18 mg•L^{-1} in the sequential anoxic/anaerobic reactor, and pH values were in the range 7.69~7.91.

Analytical methods

Nitrite, nitrate and ortho-P were determined for filtered samples using an ion-chromatography system (IC DX-120, Dionex, USA) equipped with anion-exchange column Ionpac AS12A. Total organic carbon (TOC) was measured using a TOC analyzer (TOC-VCPN-6000, Shimadzu, Japan). Total nitrogen (TN), total phosphorus (TP) and COD were measured using Hach Digestion Vials (Hach, USA) and a Beckman spectrophotometer (DU 520, Germany). Ammonia nitrogen was measured using Hach method 8038. Dissolved oxygen concentration was measured using a DO meter (YSI, Model 58, USA); pH was determined using a glass electrode pH meter (Orion, Model 525A, USA). Measurement of mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) followed the standard method (13).

2. Results and Discussion

Treatment efficiencies at long HRT

The COD removal efficiencies were above 97% where both low and moderate COD_{inf} were introduced in the SAMs (phase I & II). The major part of influent COD was consumed during the anoxic/anaerobic period, as indicated by low effluent COD concentration (below 5 mg·L^{-1}) and efficient nitrification processes in the aerobic period. The MBR fed with low COD_{inf} (HRT= 8 hrs) did not demonstrate nitrogen removal and just behaved like a nitrification reactor with almost all the nitrogen converted into nitrate (99.9% NH_3-N conversion), and no nitrite detected in the effluent. The dominance of nitrification processes was probably due to the low COD/TN ratio (4.8) in the MBR fed with low COD_{inf} as compared to the case of moderate COD_{inf} (COD/TN= 10), and about 1.5 times higher specific TN loading rate in the MBR fed with low COD_{inf} (0.024 kg TN· kg^{-1} MLVSS·day^{-1}) than that in the MBR fed with moderate COD_{inf} (0.017 kg TN·kg^{-1} MLVSS·day^{-1}). The TN and TP removal efficiencies were slightly higher in the case when the MBR was fed with moderate COD_{inf} (Table 3).

Treatment efficiencies at Short HRT

The treatment efficiencies achieved at reduced HRT (4 hrs) in the MBRs fed with low COD_{inf} and moderate COD_{inf} are summarized in

Table 3. It is evident that the TN and TP removal efficiencies were enhanced at short HRT due to the slight increase in organic loading rates in the MBRs compared with those of the long HRT MBRs. However, the extent of organic loading rate was not high enough to enhance biological nitrogen and phosphorus removal (volumetric organic loading rate less than 0.6 kg $COD \cdot m^{-3} \cdot day^{-1}$. In a previous study, enhanced nutrient removal was observed when volumetric organic loading rate was 3.95 kg $COD \cdot m^{-3} \cdot day^{-1}$, corresponding to average influent concentration of 304 mg $COD \cdot L^{-1}$ (14). It seems that phosphorus release and uptake became less significant at low COD/TP. Lee et al. found that a higher COD/TP ratio was necessary for luxury uptake of phosphorus (15). Enhanced biological nutrients removal was observed by Zhang et al. in a MBR based system at COD/TN/TP ratio of 28.5/5.1/1 (16). The phosphorus release during the anaerobic period may have been influenced by nitrate in recycled sludge due to the increased sludge recycling rate at short HRT.

COD/TP ratio

In the MBR operated at short HRT (4 hrs), low treatment efficiencies were observed. Therefore, the concentration of influent COD was increased and the concentration of TP was reduced (COD/TP = 103). In addition, the HRT was set to 5 hrs in the phase III. The MBR was operated at these conditions until steady state conditions were reached. The TP and TN removal efficiencies were increased up to 89.2% and 74.16% respectively (Table 3). It can be concluded that increased organic loading and higher COD/TN and COD/TP ratios positively influence performance of the MBR.

Track studies

Typical concentration profiles of nitrate, nitrite and phosphate for a single anoxic/anaerobic cycle in the sequential anoxic/anaerobic reactor are shown in Figure 2. In the case of MBR fed with low COD_{inf}, nitrate accumulated during the anoxic period and disappeared in the anaerobic period [Figure 2(a)]. On the other hand, nitrite concentration increased during the anaerobic period. This suggests that nitrate converted to nitrite during the anaerobic period, however, conversion of nitrite to nitrogen gas was not completed and it oxidized back to nitrate in the aeration reactor; so a low nitrogen removal was observed (Table 3). In the case of the MBR fed with moderate COD_{inf}, nitrate did not accumulate in the sequential anoxic anaerobic reactor but the concentration of nitrite increased during the anoxic period and the nitrite disappeared in the anaerobic period [Figure 2(b)]. This suggests that the nitrate introduced into the sequential anoxic/anaerobic reactor from the aerobic reactor, via recycling, reduced to nitrite and finally to nitrogen gas. However, the extent of denitrification was not sufficient and total nitrogen removal efficiency was around 31%. The concentrations profile of PO_4-P in the sequential anoxic/anaerobic reactors [Figure 2(a), 2(b)] shows that EBPR activities were not occurring and almost no TP removal was observed (Table 3).

A track study was also conducted in the sequential anoxic/anaerobic reactor of both the MBRs after reaching steady state conditions at short HRT (4 hrs). The concentration profiles of NO_3-N, NO_2-N and PO_4-P at different time intervals are shown in Figure 2(c) and 2(d). The improvement of TN and TP removal at short HRT (4 hrs) is reflected in the denitrification and phosphorus release processes in the sequential anoxic/anaerobic reactors. In case of the MBR fed with low COD_{inf} at short HRT, relatively higher concentration of NO_3-N accumulated during anoxic period as compared to the MBR operated at long HRT (8 hrs). Similarly, low concentrations of NO_2-N were detected during anoxic/anaerobic period. This trends show that nitrogen removal efficiency was improved at reduced HRT, though at low extent (TN removal efficiency 19%). Furthermore, no accumulation of NO_3-N both during anoxic and anaerobic periods was observed in sequential anoxic/anaerobic reactor of the MBR fed with moderate COD_{inf} [HRT 4 hrs, Figure 2(d)], whereas low NO_2-N concentrations were found only in anoxic period. It suggests that nitrate conversion ($NO_3 \rightarrow NO_2 \rightarrow N_2$) was occurring at relatively much higher extent than other three cases. A higher TN removal rate in case of MBR fed with moderate COD_{inf} (57.6%) was observed, which was almost two times higher than that of the MBR operated at long HRT (8 hrs). The PO_4-P concentration profile demonstrated some EBPR activity (PO_4-P release in anaerobic period), indicated the recovery of active PAOs in the sludge (TP removal 17.1%). The PO_4-P concentration declined during the anoxic period (when internal recycling was continued), so it seems that phosphorus uptake has occurred. In fact, phosphorus uptake was not taking place during the anoxic period but concentrations of PO_4-P were affected by combined dilution effect of internal recycling and influent in the sequential anoxic/anaerobic reactor.

Figure 3 depicts the concentration profile of PO_4-P in sequential anoxic/anaerobic reactor of the MBR fed with high COD_{inf}. The concentrations of NO_3-N, NO_2-N were below detection limit in the sequential anoxic/anaerobic reactor of the MBR fed with high COD_{inf} both in the anoxic and the anaerobic periods which indicates efficient denitrification. Similarly, the extent of PO_4-P release in anaerobic conditions was much higher than those observed in the track study of phase-I and II of the MBR operation. In brief, the track studies reveals improvements in TN and TP removal efficiencies in the MBR when organic loading rate was increased either via reduction in HRT or by increase in influent COD concentration

Specific organic loading

The TN and TP removal efficiencies achieved in the three phases of the experiment are shown as a function of specific loading rate in Figure 4. The specific organic loading rates were calculated on the basis of variations in influent COD concentration or reduction in HRT. The increase in TN removal efficiency was linear as the specific organic loading rate increased. However, the TP removal efficiency improved sharply when applied specific loading rate was raised up to 0.3 kg COD $kg^{-1}\cdot$ MLVSS$\cdot$ day^{-1}. This sharp increase in TP removal can be understood by focusing on the fact that the COD/N and COD/TP ratios (15.2 and 103.2 respectively) were comparatively higher than the MBRs fed with low and moderate COD_{inf} (ranges 4.8~10 and 18.7~40.4 respectively).

Variation of TMP at different HRTs.

The treatment efficiencies were increased in the MBR at short HRTs due to increased organic loading rates. However, application of short HRTs brings an inherent problem: increased flux consequently increases membrane fouling (17). In order to observe membrane fouling behavior at different HRTs or organic loading rates, variations of trans-membrane pressure (TMP) were monitored (Figure 5). An abrupt increase in TMP indicated that fouling was enhanced and subsequently physical cleaning was performed by scrubbing the bio-cake from the surface of membrane. Chemical cleaning was not performed during the entire experiment. The MBRs which operated at long HRT (flux of 12.5 $L\cdot m^{-2}\cdot hr^{-1}$) had a longer time between cleanings than the MBR operated at short HRT (flux of 25 $L\cdot m^{-2}\cdot hr^{-1}$) [Figure 5(a) and 5(b)]. However, the TMP variations in the MBR operated at HRT of 5 hrs (high COD_{inf}) reveals that stable operation of the MBR was prolonged due to less frequent cleaning [Figure 5(c)]. In other words, although the MBR was operated at short HRT (5 hrs) the higher loading rate was one of the reasons for suppression of membrane fouling. This trend of stable TMP or less frequent fouling at increased organic loading rate observed in this study is not in accordance with previous findings. For example, Trussell et al. demonstrated increases in fouling rate with increases in specific loading rate (17). Ahmed et al. also showed decreases in the value of specific cake resistance or reduced fouling with decreases in specific loading rate (18). It is known that biomass at low specific organic loading predominately undergoes endogenous respiration and cell lysis occurs (19). This would cause the accumulation of soluble microbial products (SMP) in bulk solution, which is considered as one of the major causes of membrane fouling (19, 20). It may be that the cell lysis cell lysis was decreased when specific organic loading rate was increased to 0.3 kg COD$\cdot$ kg^{-1} MLVSS $\cdot day^{-1}$, therefore, less frequent membrane fouling was observed.

3. Conclusions

Different organic loading rates were applied in an MBR based BNR system (MBR) by varying influent concentration, termed as low, moderate, and high COD_{inf}, and by changing hydraulic retention time (HRT). The treatment efficiencies, in terms of COD, total nitrogen (TN), and total phosphorus (TP), were measured. Furthermore, trans-membrane pressure at different applied conditions was monitored to observe impact on membrane fouling. In addition, microbial community structure was studied using the quinine profile method. The results demonstrated that increases in organic loading rate had a positive impact on BNR process. The BNR process may face problems when influent is low strength domestic wastewater. The organic loading rate can be increased by decreasing HRT in MBR based BNR systems, however, increased flux resulted in increased membrane fouling. When the MBR based BNR process was operated at relatively higher influent COD and at short HRT, efficient TN and TP removal efficiencies were observed at stable TMP variation throughout the test. It means that a suitable specific organic loading rate is necessary for stable operation of MBR based BNR system. It further indicates that an external carbon source will be required in case of treatment of low strength domestic wastewater if an efficient BNR is desired, particularly during raining period when strength of wastewater becomes low. A specific organic loading rate above 0.3 kg $COD \cdot kg^{-1}$ $MLVSS \cdot day$ would be required for enhanced BNR in the MBR based BNR system used in this study.

Acknowledgement

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Table 1. Operating conditions of lab-scale MBR used in this study.

Operating factors	Phase		
	I	II	III
Influent Flow , L· day^{-1}	30	60	48
Flux, constant flux mode, (L· m^{-2}· hr^{-1})	12.5	25	20
HRT (hours)	8	4	5
SRT (days)	60		
Internal Recycling rate	2.5 × of influent flow		

Table 2 Influent characteristics used in this study.

Parameters, mg•L^{-1}	Phases				
	I		II		III
	Low	Moderate	Low	moderate	High
COD*	109.8 ±7.3	227.5 ±20.4	109 ±7.2	216.2 ±10.7	331.6 ±17.5
TN	22.7 ±1.5	22.73 ±0.4	23.1 ±1.8	22.5 ±1.3	21.8 ±0.5
TP	5.7 ±0.5	5.64 ±0.5	5.9 ±0.3	5.7 ±0.5	3.2 ±0.3
NH_3-N	24.4 ±0.3	22.61 ±1.2	21.1 ±1.1	22. 3 ±2.3	21.3 ±1.9

Average ±standard deviation, * *Acetate:Glucose = 7:3*

Table 3 Treatment efficiencies achieved in different phases of the experiment.

Parameter	Phases (removal, %)				
	I		II		III
	Low COD_{inf}	Moderate COD_{inf}	Low COD_{inf}	Moderate COD_{inf}	High COD_{inf}
COD	97.5 ±2.7	99.9 ±0.05	99.9 ±0.04	99.9 ±0.04	99.85 ±0.2
TN	0.8 ±6.2	31.7 ±2.3	17.2 ±2.4	57.6 ±3.1	74.16 ±4.9
TP	5.5 ±o.6	13.1 ±0.7	7.0 ±1.3	17.2 ±2.1	89.2 ±4.5
NH_3-N	99.9 ±0.05	99.9 ±0.05	97.5 ±1.29	98.9 ±0.52	99.89 ± 0.1

Average ±standard deviation

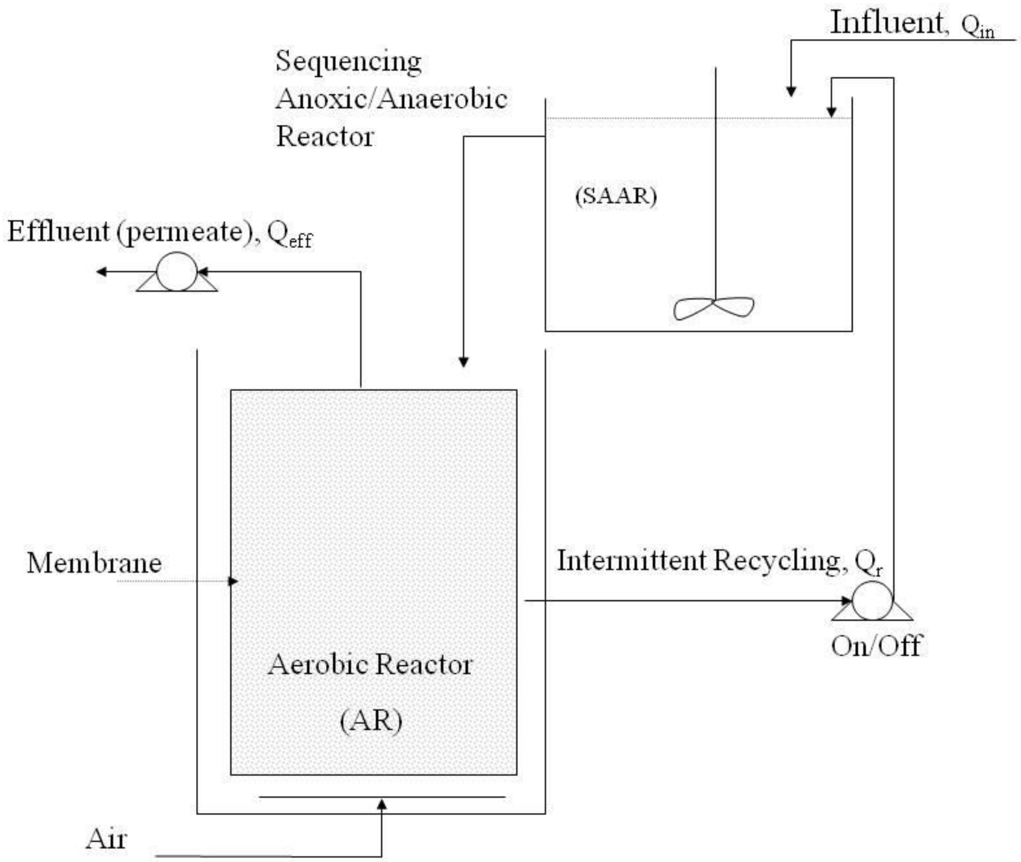


Figure 1. System configuration used in this study.

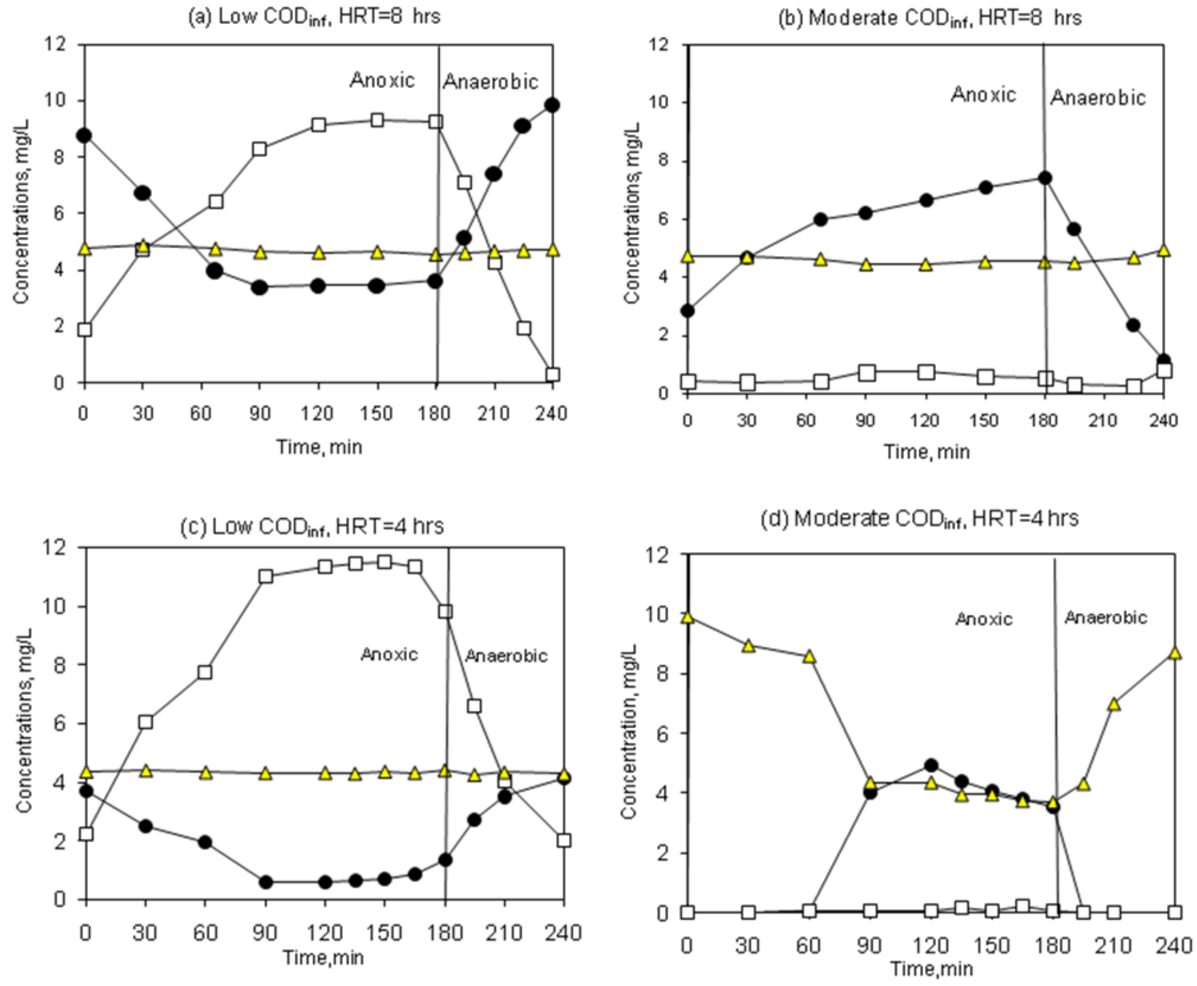


Figure 2. Concentration profiles of NO_3-N, PO_4-P, and NO_2-N in sequential anoxic/anaerobic reactor of the MBRs operated at low and moderate COD concentrations. (a) low COD_{inf} (HRT=8 hrs), (b) moderate COD_{inf} (HRT=8hrs), (c) low COD_{inf} (HRT=4 hrs), (d) moderate COD_{inf} (HRT=4hrs)

NO_2-N —●—, NO_3-N —□— , PO_4-P —△—

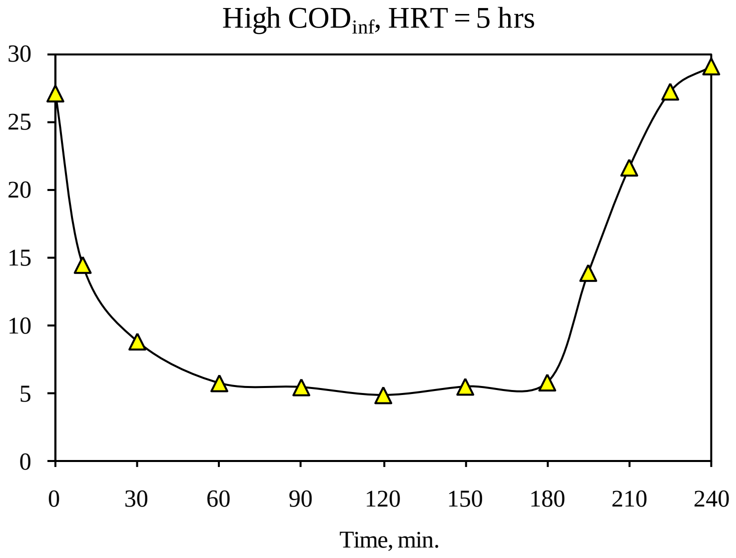


Figure 3. Concentration profile of PO_4-P in sequential anoxic/anaerobic reactor of the MBR fed with high COD_{inf} (HRT= 5 hr).

PO_4-P —△—

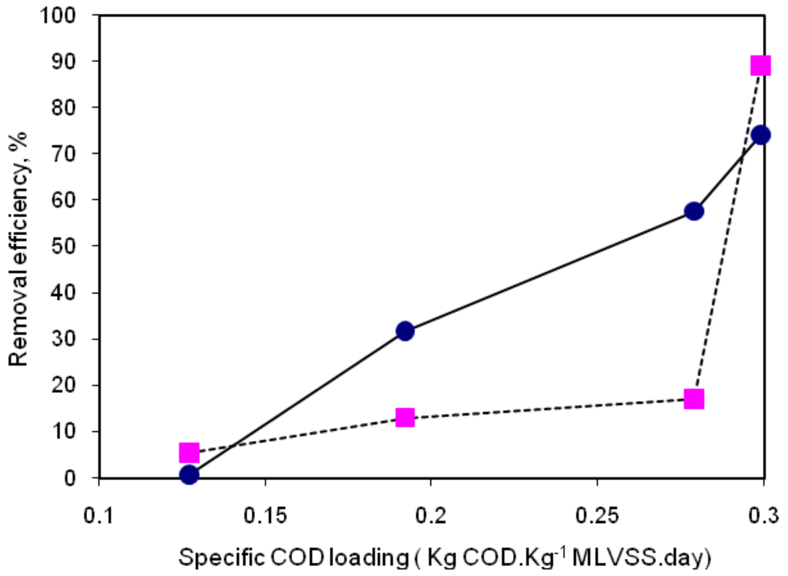
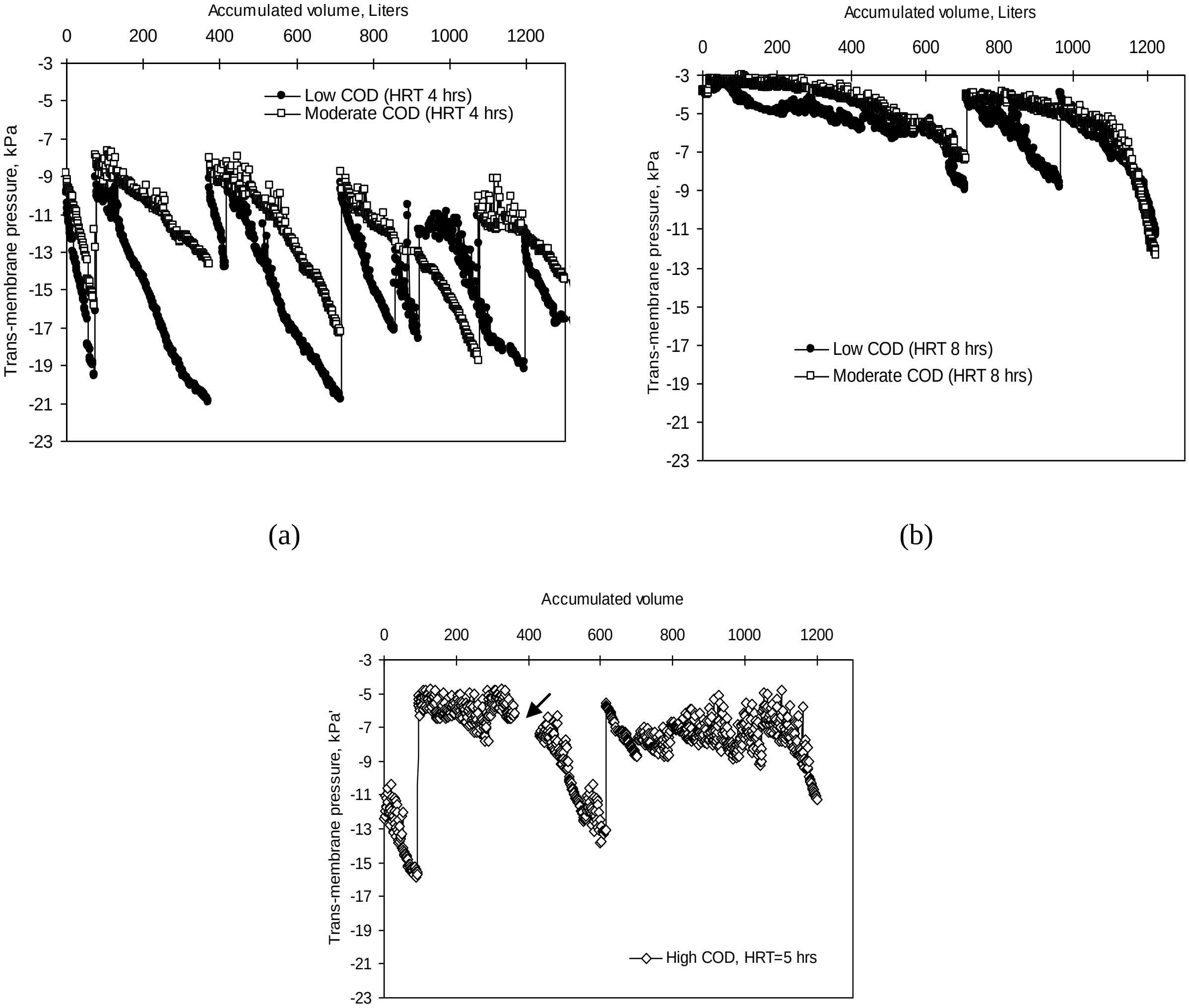


Figure 4. TN and TP removal efficiencies at different specific organic loadings.

TP ········■········, TN —●—



(a) (b)

(c)

Figure 5. Trans-membrane pressure in MBRs fed with different influent COD and operated different HRTs (a) low & moderate COD_{inf} (HRT 8 hrs), (b) low & moderate COD_{inf} (HRT 4 hrs), (c) High COD_{inf} (HRT 5 hrs) , ↙ Data acquisition system was not functional.