Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2015.1044912

57 (2016) 11300–11308
May

Taylor & Francis
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Nitrobenzene removal from micro-polluted water resource by a submerged MBR and the importance of activated sludge

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Received 17 July 2014; Accepted 15 April 2015

ABSTRACT

A submerged membrane bioreactor (MBR) employed for nitrobenzene-contain water resource treatment was operated for investigation of system effectiveness and for specification of nitrobenzene (NB) removal pathway. Two sludge retention times (2 and 25 d) that regulated biomass content in MBR were adopted to evaluate the importance of activated sludge for NB removal through biodegradation way. From a long-term examination, both of organic biodegradation by activated sludge and rejection by fresh and/or fouled membrane were the major ways to remove organic matters including NB. However, activated sludge played a rather significant role in NB removal. A lengthening solid retention time form 2 to 25 d in MBR resulted in a significantly improvement of removal efficiency in dissolved organic matters, ammonia-N and NB, due mainly to the continuous accumulation of biomass within bulk. Especially, the NB removal efficiency displayed an apparently positive correlation with the biomass content. After sludge acclimation process, the MBR was able to remove NB by a removal rate at 1.266 mg/(gMLSS h), equivalent to the NB removal efficiency up to 80%. In addition, by Fluorescence excitation-emission matrix observation, it was found that biomass growth and accumulation could also impact the effective removal of organic fractions, as that there were obvious selectivity of organic removal by biodegradation of activated sludge. The experimental results obtained herein are very useful to recognize the refractory organics removal pathway in MBR, and to understand the importance of activated sludge.

Keywords: Activated sludge; Micro-polluted water resource; Nitrobenzene (NB); Submerged MBR

1. Introduction

Due to the fast development of industrialization and urbanization, industrial wastewater that is irregularly discharged to water body without effective treatment, as well as domestic sewage, lead to severe surface water resource contamination. Especially, in developing countries, lack of stringent legislation and limitation of environmental capacity cause frequent water resource pollution accidents. Nitrobenzene (NB) is a toxic and suspected carcinogenic compound [1] that was widely used in industrial production for

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chemical syntheses of aniline, dyes, drugs, pesticides, synthetic rubber [2,3], which was found in surface water in some water body of China [4,5]. For instance, in the Songhua river pollution accident, as high as 600 μg/L NB concentration was found in the water body that take a role as water resource for Harbin city, and more than 200 μg/L NB was expected in a large section of the river within a quite long period due to possible sediment desorption [6]. In view of the resistance to chemical or biological degradation, it is very difficult to reduce NB in conventional drinking water treatment [7]. Hence, various new techniques were developed recent years, including chemical treatment [8], bioaugmentation [9], adsorption [10] and membrane-based processes [11], in order to enhance NB removal efficiency from water resource.

Membrane bioreactor (MBR) has been proved to possess stably excellent organic substances and ammonia removal capability from sewage treatment and drinking water resource, due mainly to its high membrane intrinsic rejection rate to refractory organics, and long solid retention time (SRT) for microorganisms growth and accumulation by membrane retention [12]. From last two decades, MBR begins to be employed for micro-polluted water resource treatment to guarantee drinking water quality that was evidenced by stably good results from the several experimental reports [13,14]. Some researchers observed that a single activated sludge process with sufficient biomass had efficient for biological removal of NB [15], bringing about a high expectation for MBR to stably treat micro-polluted water resource loaded with NB. Moreover, MBR system could also employ adsorption by activated sludge and biodegradation by biomass, both of which take some responsible for NB removal, while the possibility of NB desorption from solid was completely avoided.

In contrast to the MBR treating wastewater biologically that contained a high organic substances concentration, whose treatment performance could be easily obtained due mainly to the high biomass content [16], the MBR employed for drinking water treatment has to confront to insufficient biomass situation resulted from a limited organic matters available. In order to improve MBR capability in organic removal during water resource treatment, many efforts were tried by researchers, including to develop a hybrid processes combined activated sludge process [17,18] with coagulant [19], advanced oxidation [20] and activated carbon, resulting in an increasing of operational cost. These studies mainly focused on the enhanced treatment performance, however, eventual capability and the importance of activated sludge in MBR tank were always underestimated to some extent. Different from conventional activated sludge process, MBR retained suspended solids by membrane rejection could also affect sludge properties and system loading rate, which may give rise to an expectation for its biodegradation enhancement for refractory organics removal [21,22]. For instances, Chen et al. [16] observed an excellent Bisphenol-A (BPA) removal effectiveness and confirmed that biodegradation was the dominant way for BPA removal process in MBR system. Fallah et al. [23] found that a long-term operation of submerged MBR was capable to remove styrene at a high efficiency above 99% mainly through biodegradation, which had a greater removal rate compared with the conventional ASP did. Thus, the activated sludge content is a key operational parameter for MBR technology [24] that was worthy for comprehensively investigation. In this study, different SRT in MBR were regulated to evaluate the importance of activated sludge, and the relationship of biomass and organics, especially NB removal efficiencies in MBR system, was examined. The aims of the experimental study was to improve the contaminants removal efficiency of MBR treating micro-polluted water resource by an easy way, and to indicate the importance role of activated sludge for organic matter removal.

2. Materials and methods

2.1. Experimental set-up and design

Two laboratory-scale submerged plexiglas MBR reactors, marked as R1 and R2, respectively, were employed to investigate micro-polluted water resource treatment. R1 and R2 had identical working volume of 4.4 L, and the same aeration intensity for DO supply and continuously membrane cleaning. Plat-sheet polyvinylidenefluoride ultra-filtration membranes (an average pore size of 0.08 μm, working area of 0.5 m^2, Peier, China) were immersed to form submerged MBR, which was operated in a constant flux mode and the trans-membrane pressure (TMP) was monitored with a manometer in mmHg. Suction pumps (BT1002J, Longer, China) were used to withdraw the effluent of the MBRs through the membranes at a filtration-to-idle cleaning ration of 8 min: 2 min.

Two operational runs, named as Phase I and II, were designed for evaluation of importance of activated sludge in system treatment by varying SRT in R1 and R2, and for investigation of the impact of dosed NB onto sludge properties, respectively. As shown in Table 1, during the Phase I, R1 was set as an incomplete mixing mode where mixed sludge was discharged regularly to maintain a short SRT of around 2 d, whilst R2 was operated under a complete mixing

Table 1
Experimental conditions and comparison of treatment performance in Phase I

		R1	R2
Operations parameters	Duration (d)	60	40
	SRT (d)	2	20
	HRT (h)	2	2
	MLSS (mg/L)[a]	50	496
Particle size of sludge[b]	D_{10} (μm)	22	22
	D_{50} (μm)	44	104
	D_{90} (μm)	249	592
Biomass *n* mol P	In mixed liquor/mL	6.8	12.3
	On membrane/cm^2	8.1	27.6
Influent	Ammonia-N (mg/L)	1.29 ± 0.25	0.97 ± 0.18
	DOC (mg/L)	3.37 ± 0.48	4.38 ± 0.85
	UV_{254} (cm^{-1})	0.08 ± 0.03	0.1 ± 0.03
Effluent	Ammonia-N (mg/L)	1.11 ± 0.39	0.51 ± 0.21
	DOC (mg/L)	2.56 ± 0.68	2.05 ± 0.25
	UV_{254} (cm^{-1})	0.04 ± 0.02	0.05 ± 0.03
Removal	Ammonia-N (%)	14	47.4
	DOC (%)	24	53.2
	UV_{254} (%)	51.3	50

[a] Based on the value of stable operation *n* = 15.

[b] For R1 and R2, *n* = 15 and 12.

mode to keep a long SRT up to 25 d without any sludge discharge except sludge sampling. Apart from different SRT, the other operational conditions, such as flux and HRT, were kept identically at 18 L/h-m^2 and 2 h, respectively. In the second phase, both of R1 and R2 were operated under a complete mixing mode with identical operational conditions except that there was not NB added into R1.

Surface water collected from a local reservoir (Xili Reservoir, Shenzhen, China) was diluted with tap water at a ratio of 1:2 to simulate micro-polluted water resource. NB (Aladdin, purity 98%) was selected as the target synthetic organic contaminant to simulate the actual situation (containing 200 μg/L NB) reported in previous research [6]. The chemical reagent NB was first dissolved into the simulated raw water by stirring for more than 24 h at 25℃ before use. In order to obtain raw water with stable concentration of ammonia, reagent NH_4Cl was also dissolved in the raw water.

2.2. NB elimination capacity of activated sludge test

The maximum NB elimination capability ($q_{\max}$) of the sludge sample was estimated by a batch-test carried out in a 2 L glass cylinder reactor with an air diffuser fixed at the bottom to provide DO following the procedure described in the previous report [25]. Target sludge sample was poured into the reactor, and concentrated NB solution (300 mg/L) was continuously injected into the reactor at a slow flow rate about 0.5 mL/min. At the same time, NB concentration in the reactor was determined intermittently for its degradation rate calculation. The mass balance equation of NB in the reactor could be described by Eq. (1), while the volume of miscible liquids in the reactor was assumed constant.

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \frac{S_\mathrm{i} \times Q}{V} - qX \qquad (1)$$

where S = NB concentration in the reactor, mg/L, t = hydraulic retention time, d, S_i = NB concentration in influent, mg/L, Q = influent flowrate, L/d, V = mixed liquor volume in reactor, L, q = specific substrate removal rate, d^{-1}, X = biomass concentration in batch reactor, mg/L.

Upon the steady-state of MBR, centrifuged mixed sludge solid from MBR was re-suspended with 0.05% NaCl solution to form a mixed solution. During the substrate utilization estimation test, 2 L raw water resource mixed with pre-dissolved NB was prepared in two reactors, which was with and without the sludge sample, respectively. Since the beginning of the test, 50 mL liquor was sampled and its NB concentration was measured for every 30 min. Afterwards, cumulative NB removal rate could be calculated by plotting NB concentration with reaction time. A blank trial was conducted side-by-side to examine any mistakes encountered. Accordingly, Eq. (1) was converted to Eq. (2), where $v = \mathrm{d}S/\mathrm{d}t$, $v_\mathrm{i} = S\mathrm{i} \times Q/V$.

$$q_{\max} = \frac{v_\mathrm{i} - v}{X} \tag{2}$$

2.3. Analytical methods

The treatment performances of the MBRs were evaluated in terms of dissolved natural organic (DOC and UV_{254}), ammonia-N, and NB removal efficiencies. DOC was measured by TOC analyzer (TOC-L CPN, Shimadzu, Japan) using the high-temperature combustion method. UV absorbance at 254 nm (UV_{254}) was determined by a spectrometer (UV2600, Shimadzu, Japan). Ammonia-N was also determined by the spectrometer. NB was determined following US EPA Method 502.2 by gas chromatography (7890A, Agilent, USA). The particle size of suspension of MBR was determined by a particle size analyze (Malven Laser Mastersizer 2000). The biomass content was estimated using lipid-P determining [26], and the method was modified according to the phosphor lipid analysis methods [27,28]. In order to analyze the organic fractions and removal, a spectrofluorometer was employed. Fluorescence excitation-emission matrix of a water sample was developed by scanning it over an excitation range of 220–460 nm by 10 nm increments and an emission range of 220–600 nm by 2 nm increments using spectrofluorometer [29].

3. Results and discussion

3.1. Comparison of MBR treatment performance

During the operations of MBRs in Phase I, the removal efficiencies of ammonia-N, dissolve organics (DOC), UV_{254} and NB were measured and shown in Table 1. As for R1 with a feeding containing ammonia-N concentration of 1.29 ± 0.25 mg/L, its ammonia removal efficiency averaged about 14.0%, which was much lower than those in R2 that eliminated ammonia-N from 0.97 ± 0.18 to 0.51 ± 0.21 mg/L on average. There was an apparent maturation process of nitrifying bacteria in R2, reflected by an increasing of ammonia-N removal efficiency from 20% in the beginning to more than 70% in the stationary phase after 40 d operation. The same phenomenon was also observed in organic matters DOC removal profile in R1 and R2, whose DOC removal efficiency was 24.0% and 53.2%, respectively. Under a rather long SRT, R2 played comparable performances on ammonia and organics removal with experimental results by other researches [30], while R1 with a SRT as short as 2 d displayed rather poor treatment performance that may related with its limited activated sludge biomass. The divergence also indicated the important of long SRT for ammonia removal in MBR which had a great effect on the sludge characteristics and mass. Notwithstanding, both of R1 and R2 had comparable UV_{254} removal efficiency that averaged at 51.3 and 50.0%, respectively. Due mainly to that suspended pollutants, particular and colloidal organics, and DOC components had large molecular weight preferentially rejected by ultrafiltration membrane, the UV_{254} contained or formed accompanying with microbial metabolism could be effectively retained in the MBR then subjected to be degraded, which was not significantly affected by biomass content in MBR bulk.

Fluorescence chromatogram results showed that there was great difference in organic substances components in two MBRs as a result of SRT variation. There were five fractional components in DOC detected and specified by spectrofluorophotometer: Thereinto, Tryptophane, SBM, Fulvic and Humic (Fig. 1). In feeding water, SBM was the major organic fractional component took a proportion about 35.5%, followed by 30.2% humic, 17.5% Tryptophane, 12.6% Tyrosine and 5.2% Fulvic. However, after sufficient biomass degradation in R2, Tryptophane, Tyrosine and SBM were found to be readily biodegraded. It was noted that MBR could remove Tryptophane, Tyrosine and SBM by 65.6, 76.8 and 78.2% through biodegradation way, while fulvic and humic would not be degraded and then passed through membrane. The results agreed well with the previous research of Sun and Li [31]. Moreover, it was indicated that, MBR system, comprised of membrane interception and biomass degradation, seemed as one selective removal process that dependent on the components and

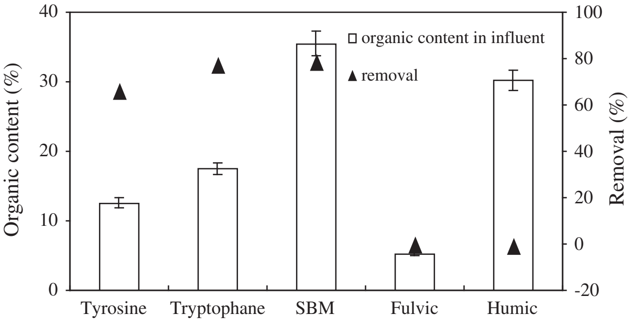

Fig. 1. Organic fractional components and removal.

properties of feeding organic substances, especially for the situation of refractory organic substances removal.

3.2. Importance of activated sludge and its capability estimation

The biomass content in MBR took important role in organics removal, e.g. NB. As illustrated in Fig. 2(a), R1 performed relatively poor NB reduction whose removal efficiency average at 47.4%, which was rather stable with the experimental progress. By comparison, NB removal performance enhanced significantly in R2, where the NB concentration in effluent decreased from 100 μg/L to less than 30 μg/L on day 12. Afterwards, stable NB removal efficiency above 80% was observed throughout the experimental phase (Fig. 2(b)). The results concurred well with the reports of Van den Broeck et al. [32] and Li et al. [33], the former found that there was importance of long SRT in MBR system to organic matters removal and the latter confirmed effective NB biodegradation ability of natural microorganisms.

It was reasonable to divide MBR into two processes for organic matters removal: (i) biodegradation by sludge biomass, and (ii) rejection by membrane filtration [30]. By using Eqs. (3) and (4) the NB removed in R1 and R2 through these two ways could be quantificated, where R_{as} and R_{fil} were defined as follows:

$$R_{as} = \frac{C_{in} - C_{mix}}{C_{in}} \tag{3}$$

$$R_{fil} = \frac{C_{mix} - C_{eff}}{C_{mix}} \tag{4}$$

where C_{in}, C_{mix} and C_{eff} were the NB concentration in MBR influent, mixed liquor and effluent, respectively. In addition, R_{as} was defined as the NB reduction efficiency by activated sludge that includes bioadsorption and biodegradation in MBR, while R_{fil} was the NB removal efficiency of membrane filtration. As illustrated by Kawasaki et al. [34] the initial MLSS in MBR system affected the organics removal performance . In this study, NB removal in MBR showed good relativity with the variation of MLSS. The relationships between R_{as} and MLSS in R1 and R2 were illustrated in Fig. 3. With slow accumulation of biomass content from 10 to 50 mg/L after 20 d operation in R1, NB removal rate by activated sludge slightly increased from 20 to 50%, and its mean value was 35.6% that mainly attributed to activated sludge adsorption and direct discharge under a short SRT (Fig. 3(a)). In contrast, the biomass content enriched from 10 to 500 mg/L, resulting to a continuous improvement of NB removal from 20 to 80% after 15 d (Fig. 3(b)). Except for the small NB removal efficiency in R2, a close correlation between biomass content with the abundance of NB removed was observed in R2, implying the important role of activated sludge onto NB removal, other than membrane filtration.

Similar with the reports on MBR for water and wastewater treatment, membrane fouling caused by cake layer formation on the membrane surface occurred in both of R1 and R2 [32]. As for the water resource treatment, the sludge cake layer would take a positive role in organic pollutants retention. As showed in Fig. 4, during 60 d operation, both of R1 and R2 subjected to severe fouling for six times. R_{fil} was observed to closely depend with TMP evolution, i.e. a higher TMP resulted in a better membrane NB rejection efficiency that could be improved from 10 to

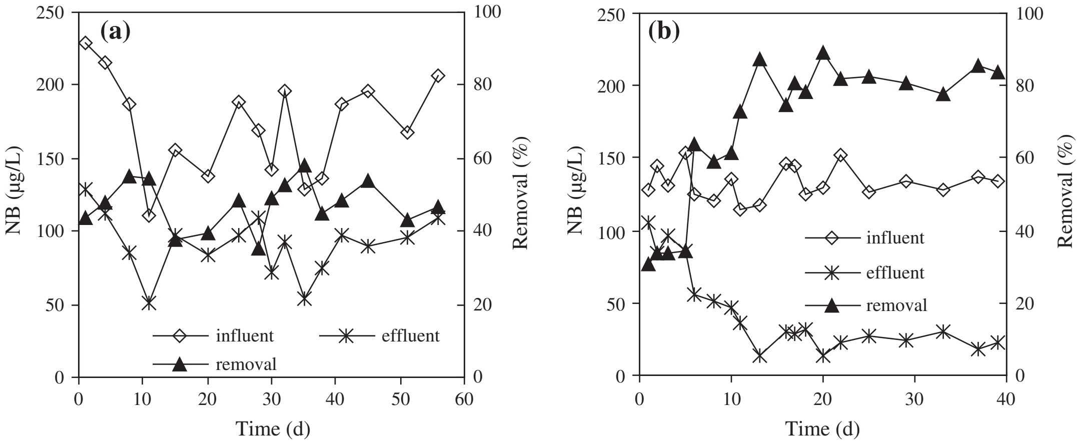

Fig. 2. NB removal of R1 (a) and R2 (b).

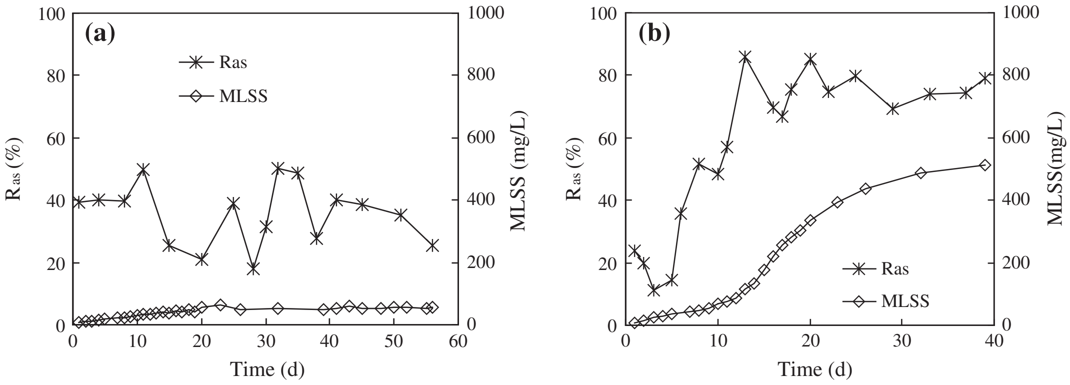

Fig. 3. Relationships between MLSS and R_{as} in R1 (a) and R2 (b).

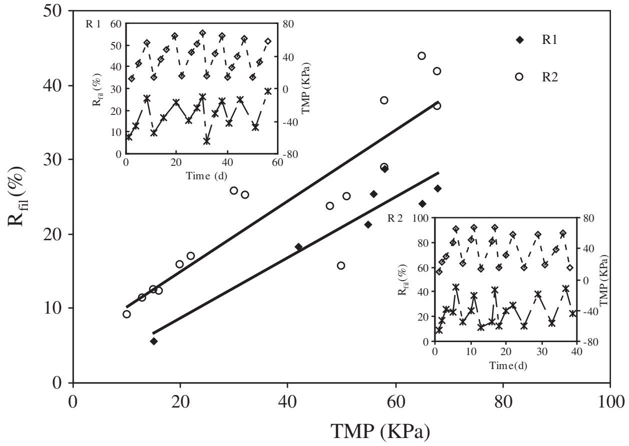

Fig. 4. Relationship between TMP and $R_{fil.}$

30%. The similar trend and correlation could also be found in R2. These phenomena confirmed the enhancement of NB retention efficiency by sludge layer revealed by Tian et al. [35]. Notwithstanding, a high biomass content in R2 apparently caused a better rejection rate improvement compared with those in R1, which was well illustrated in Fig. 4. A small amount of biomass and co-existed organic matters in R1 preferentially to induce pore clogging and gel-layer fouling, and the cake layer biomass was more than 8.1 nmol P/mL, whilst a high biomass content in R2 would form cake layer on the membrane surface above 27.6 nmol P/mL that may favor the NB removal enhancement (Table 1). On the other hand, the sludge particle size of R2 was much larger than those in R1. The particle size D_{50} and D_{90} in R1 was 44 and 249 μm, respectively, which was much smaller than that in R2 of 104 and 590 μm. A larger sludge particle was more susceptible to solid–liquid separation, and much easily to be attached onto membrane surface, in contrast to those small sludge particles were suspended well within bulk.

The concentration of activated sludge was important for NB removal in MBR system, and its biodegradation capability was significant for the system operation. As reported by Ling et al. [36], there was microbial acclimation process in activated sludge process for refractory organic removal, after which the activated sludge became more effective in organic-pollutants elimination. For NB, which is known as a refractory organic difficult to biodegrade, effective biodegradation process was also revealed. According to the previous work, there were two main pathways to biodegrade NB. By the first way, NB was easily converted to aniline, which could be further degraded to low molecular weight organic containing amino group [37], and by the other way, ntrosobenzene was a product of NB deoxidization catalyzed by nitroreductase, whose biodegradation products contained nitrite [38]. In a study of NB biodegradation, microbial communities in water and sediments along the Songhua River after a NB pollution event showed high rates where ntrosobenzene and aniline were both detected [33].

Herein, the NB removal capabilities by MBR activated sludge before and after acclimatization were compared. Fig. 5 showed that sludge without NB acclimatization had poor NB elimination capability, reflected by a rather low q_e of 0.07 mgNB/(gMLSS h) that was recognized as NB adsorption capability. However, the sludge after one month acclimatization could reduce NB by a rate of 1.266 mg/(gMLSS h) which mainly attribute to biodegradation process. Thus, the activated sludge in MBR would adapt to the contaminants and influent properties, and then played an important role in the treatment of micro-polluted water resource. Furthermore, the NB elimination performance of MBR could be calculated by Eq. (5).

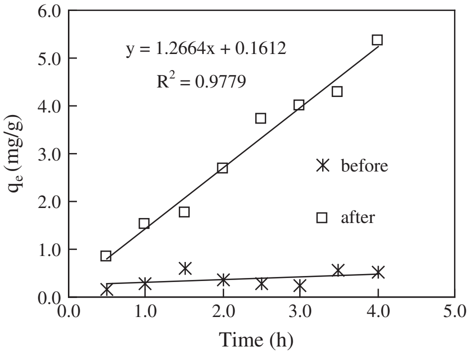

Fig. 5. NB elimination capability of sludge before and after acclimatization.

$$C_{\mathrm{eli}} = q_{\mathrm{e}} \times \mathrm{MLSS} \times \mathrm{HRT} \times 1,000 \quad (5)$$

where C_{eli} was NB elimination μg/L; q_{e} was maximum NB elimination capability of sludge, 1.266 mg/g in this test. For the real applications, sludge acclimation and concentration are eventually essential factors in the treatment performance of low-degradation rate refractory organics in drinking water treatment, and a long time acclimation and a high MLSS would be effective to shorten MBR treatment time that was of great economical efficiency.

3.3. *Effect of operational conditions onto MLSS and its influence to system treatment performance*

In the MBR operation, membrane flux, HRT and biomass content had effects on organic matters removal. In order to figure out the dominant parameters that eventually impact the MBR performance, both of R1 and R2 were continuously operated up to 100 d under various operational conditions. Fig. 6 showed the DOC and NB removals at three different HRTs (0.5, 1.0, and 2.0 h) and a wide range of MLSS (0.5–1.1 g/L) with different membrane flux from 18.3 to 27.5 L/(m^2 h). A short HRT inevitably led to a high volumetric loading rate for MBRs, and hence resulted in a high biomass content, which agree well with the finding of Li et al. [33]. It was observed that when HRT decreased from 2.0 to 1.0 h, MBR biomass content in R1 and R2 increased from 0.54 to 0.73 g/L and from 0.52 to 0.70 g/L, respectively, which improved the NB removal rate in R2 from 80.8 to 83.7%. Further decreased HRT to 0.5 h could also increase biomass content and improve NB removal efficiency. By using Eq. (3), the maximum NB removal capability of R2 was 1,282, 2,333 and 1,176 μg/L during HRT of 2.0, 1.0 and 0.5 h, respectively, which were greater than the NB content in raw water. Thus, for organics removal in MBR, the influence of NB in raw water onto MBR system is also worthy to study because of that there were potential biotoxicity accumulation to microorganisms after long-term experiment. In Phase II, R1 and R2 were operated under the same conditions except that no NB dosing in raw water of R1. R1 and R2 performed comparable DOC removal efficiency, as showed in Table 2, and both of R1 and R2 had similar particle size of sludge with same D10 and D90, which also had similar SOUR profiles, in spite of that R1 had a small amount of biomass in bulk solution and on membrane surface.

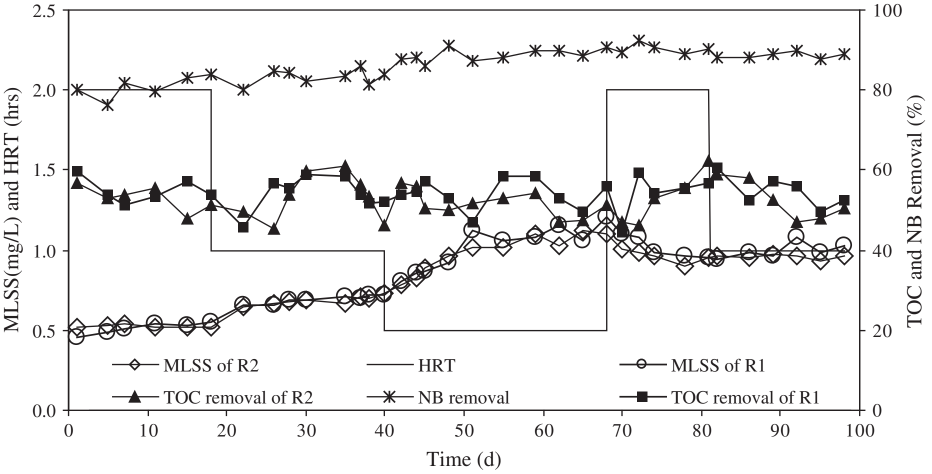

Fig. 6. Performances comparison of R1 and R2.

Table 2
Experimental conditions and comparison of sludge properties in Phase II

			R1	R2
Operations parameters	Duration (d)		100	100
	SRT (d)		60	60
	HRT (h)		–	–
Particle size of sludge[a]	D_{10} (μm)		26	26
	D_{50} (μm)		176	148
	D_{90} (μm)		704	704
Activated sludge properties[b]	SOUR mgO_2/(gMLSS min)		0.08	0.08
	Biomass *n* mol P	in mixed liquor/mL	18.6	16.4
		on membrane/cm^2	45.2	32.6

[a] Based on the value of stable operation $n = 33$

[b] Based on the value of stable operation $n = 3$.

4. Conclusions

The experimental results on the MBR treatment performance on organic pollutants removal from micro-polluted water resource showed that the activated sludge biomass is important to system effectiveness. MBR with a long SRT, and high sludge content could effectively remove ammonia-N, DOC and NB at an average efficiency of 70, 50 and 90%, respectively. NB was eliminated mainly by activated sludge biodegradation and membrane interception that dependent significantly with cake layer formation. After sludge acclimation process, the MBR was able to remove NB by a removal rate at 1.266 mg/(gMLSS h). The degradable organic matters removal performance by MBR has to relate with the organic property and abundance of activated sludge. The organic fractional components comparison showed that there was selectivity of organic removal by biodegradation of activated sludge, as that amino acid and SBM could be effectively eliminated, while humic and fulvic were uneasily removed from MBR.

Acknowledgements

This research was supported by grants No. 51408149 from National Natural Science Foundation of China, grant No. KQCX20120802095942112 from the Shenzhen Peacock Technique Funding Project, grants 2012ZX07313001-008 from water pollutant control and treatment, and grants S2012040006350 from Chinese Guangdong Natural Science Foundation Funding.

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