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Performance determination of a novel dynamic membrane reactor for slightly contaminated surface water treatment

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ABSTRACT

A novel lab-scale bio-diatomite dynamic membrane reactor (BDDMR) using a module made of stainless steel mesh with an equivalent aperture of 80 μm was proposed to treat the slightly contaminated surface water. This work presents complete information on the impurities removal performance and mechanisms involved in this process. The BDDMR was very effective at reducing particle number and removing turbidity, chemical oxygen demand (COD_{Mn}), dissolved organic carbon, UV absorbance at 254 nm (UV_{254}), NH_3-N and trihalomethane formation potential with a hydraulic retention time of 4 h. The filtration resistance of each of the selected fluxes was investigated, which indicated that the resistance of high flux increased faster than that of lower flux. Three individual effects responsible for removing pollutants, such as microbial biodegradation, bio-diatomite adsorption and bio-diatomite dynamic membrane were determined. Furthermore, dissolved organic matter fractionation and molecular weight distribution were applied to reveal the pollutant removal mechanisms of the BDDMR.

Keywords: Bio-diatomite dynamic membrane reactor; Dissolved organic matters; Drinking water treatment; Membrane fouling; Slightly contaminated surface water

1. Introduction

In developing countries, organic matter and ammonia (NH_3-N) pollution of surface water is caused by a part of domestic and industrial wastewater discharged into natural water bodies without sufficient treatment. Water was defined as slightly contaminated surface water (SCSW). In China, it has been generally assumed that organics (around 10 mg L^{-1}) and ammonia were the major pollutants in SCSW [1,2]. Recent years, membrane bioreactors (MBR) have dramatically attracted a considerable amount of attention in the field of drinking water treatment, wastewater treatment and reclamation because they offer a host of technical advantages over conventional wastewater treatment plants, such as a superior effluent quality and a smaller footprint [3,4]. Nevertheless, membrane cost, energy consumption and membrane fouling and its mitigation are considered as major obstacles to

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more widespread applications of MBRs [5]. As a novel water treatment technology, the dynamic membrane method can be a promising approach to resolve these problems.

In 1965, dynamic membranes were first reported by workers at the Oak Ridge Laboratories engaged in desalination research [6]. Dynamic membranes can be formed on a support mesh by filtering a solution containing either inorganic or organic materials through a porous support [7]. These membranes are also called secondary membranes or formed-in-place membranes to more precisely elucidate the concept of membrane forming [8,9]. There are two types of dynamic membranes; these are pre-coated and self-forming. The precoated membrane is generated by passing a solution of one or more specific components over the surface of a porous support. A self-forming membrane is a dynamic membrane in which the membrane forming materials are the same as those to be separated [10,11]. Therefore, the difference between the two types is that the mixed liquor can be filtered with or without the substance to form a dynamic membrane. Dynamic membrane is superior compared with a conventional membrane reactor because of the low cost of the membrane module, easy backwash and low energy consumption.

Additionally, it is crucial to select a suitable formation material of dynamic membrane. To date, diatomite [12,13], MnO_2 [14,15], Kaolin clay [16], activated sludge [17,18], powder activated carbon [19–21], and so on, have been adopted to be formation materials of dynamic membrane. It is worth noting that diatomite is a silicon primarily composed of amorphous SiO_2 and a small amount of Al_2O_3, Fe_2O_3, Na_2O and CaO [22]. Diatomaceous earth, otherwise known as diatomite, has a unique combination of stable physical and chemical characteristics, such as high porosity, good hydrophilicity, bulk volume, high purity, large specific surface area, high adsorption capacity and good adsorptive properties. With these properties, diatomite can be applied as a filtration medium in a number of industrial processes [23,24]. It has been approved as a food-grade material by the US Food and Drug Administration (FDA).

Diatomite particles that are used in biological reactors as carriers for microorganisms, on which microbial colonies can form zoogloeas as through microbial capsules and surface mucus, are called bio-diatomite [25–27]. Yang et al. [28] examined the effect of diatomite addition on membrane fouling and process performance in an anoxic/oxic submerged MBR and showed that diatomite addition is a reliable and effective approach in terms of both membrane fouling mitigation and pollutants removal improvement. Tsai et al. [29] studied the feasibility of using spent and treated diatomite as an adsorbent for the removal of herbicide paraquat. Other investigators conducted some applications of dynamically formed membranes in the microfiltration of tap water and activated sludge processes using diatomite [30]. Recently, bio-diatomite combined with a dynamic membrane, which forms a bio-diatomite dynamic membrane reactor (BDDMR), has been developed as a newly emerged water and wastewater treatment technology that can effectively remove contaminants [31].

In the present study, a bench-scale BDDMR was developed to treat SCSW. In an integrated diatomite adsorption/dynamic membrane system, unit processes, such as the adsorption–biodegradation of organic matter, liquid–solid separation and sludge accumulation and withdrawal, can be completed in a single reactor. The objective of this study was to evaluate the pollutant removal efficiency in the BDDMR, and the mechanisms of organic pollutant removal were also investigated.

2. Materials and methods

2.1. Experimental set-up

A bench-scale BDDMR with an effective volume of 9.6 L and a water depth of 23.5 cm was developed to investigate pollutant removal from SCSW. A module made of stainless steel mesh with an equivalent aperture of 80 μm was used as the dynamic membrane support mesh and immersed inside the BDDMR. The module has a total surface area of $0.048\ m^2$ (20 × 12 cm). The schematic diagram of the experimental set-up is shown in Fig. 1.

2.2. Influent water

Raw water used in the experiment was collected from a lake in campus, Tongji University. During the experiment stage, the pH and temperature of the raw water were 7.24–8.56 and 18.2–29.6°C, respectively. Chemical oxygen demand (COD_{Mn}), UV absorbance at 254 nm (UV_{254}) and dissolved organic carbon (DOC, pre-filtered through a 0.45 μm membrane) are employed to indicate the concentration of organic compounds in water. The major physicochemical characteristics of the raw water during the experiment could be found in Table 1.

2.3. Experimental methods

Continuous aeration (around $50\ L\ h^{-1}$) was provided through an air diffuser at the bottom of the reactor to generate strong turbulence to control the

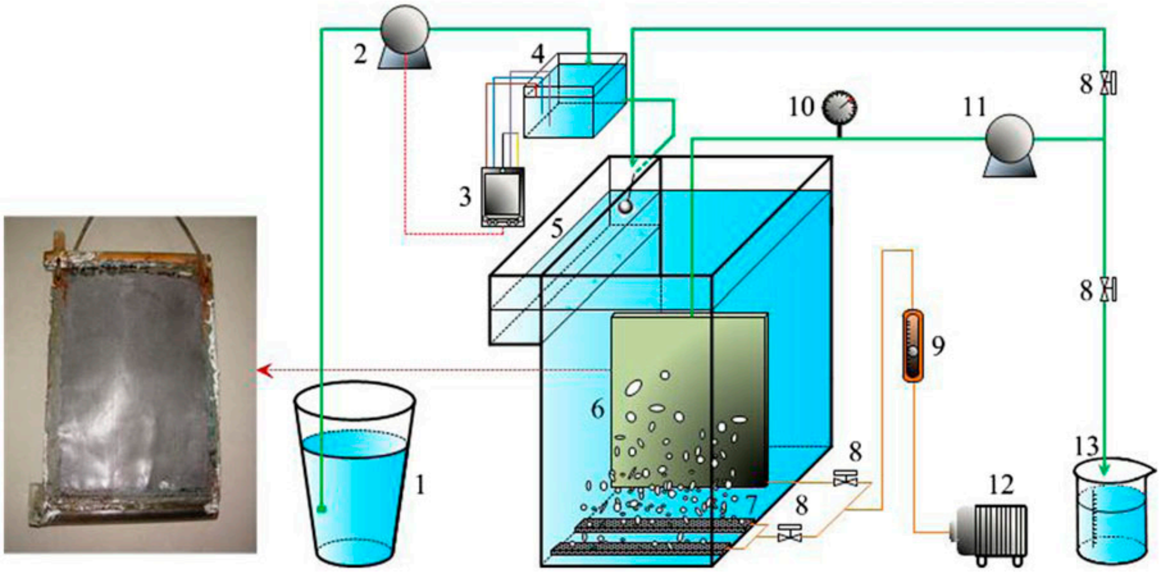

Fig. 1. Flow diagram of the bench-scale BDDMR: (1) influent tank; (2) feed pump; (3) water level controller; (4) high level tank; (5) constant level tank; (6) dynamic membrane support module; (7) air diffuser; (8) solenoid valves; (9) air flow meter; (10) manometer; (11) suction pump; (12) air supplier and (13) effluent tank.

Table 1
Removal efficiencies of the BDDMR process

Items	Influent	Effluent	Removal (%)
Turbidity (NTU)	3.46 ± 0.83	0.28 ± 0.13	91.6 ± 3.7
COD_{Mn} (mg L^{-1})	6.55 ± 0.76	2.72 ± 0.24	58.2 ± 4.4
DOC (mg L^{-1})	7.59 ± 0.89	3.82 ± 0.45	49.3 ± 6.66
UV_{254} (cm^{-1})	0.09 ± 0.004	0.049 ± 0.002	45.8 ± 2.85
NH_3-N (mg L^{-1})	0.23 ± 0.03	0.015 ± 0.006	93.7 ± 2.26
$CHCl_3$ (μg L^{-1})	143.8 ± 25.4	55.7 ± 14.5	60.9 ± 8.8
$CHCl_2Br$ (μg L^{-1})	81.2 ± 13.9	39.7 ± 9.4	50.8 ± 11.0
$CHClBr_2$ (μg L^{-1})	17.5 ± 5.1	8.3 ± 1.39	49.2 ± 14.3
$CHBr_3$ (μg L^{-1})	0	0	0
THMFP (μg L^{-1})	242.0 ± 25.5	103.7 ± 13.0	56.9 ± 6.0

Notes: Values are given in average ± standard derivation. For turbidity COD_{Mn}, DOC, UV_{254} and NH_3-N, the measurement number $n = 36$; for THMFP, $n = 13$.

growth of the dynamic membrane and to provide dissolved oxygen (DO, 7.36–8.62 mg L^{-1} in the BDDMR) for microorganisms in the bioreactor. A constant level tank was provided to ensure a constant reactor volume. The effluent was drawn directly from the membrane module by a suction pump. A high-precision vacuum pressure gauge was installed between the module and the suction pump to monitor the transmembrane pressure (TMP).

Table 1 shows that the SCSW was dystrophic for microbial growth due to the lack of nitrogen (i.e., 0.23 ± 0.005 mg L^{-1} NH_3-N) and organic carbon (6.55 ± 0.12 mg L^{-1} COD_{Mn}). Thus, to provide the nutrients for microbial growth, glucose was externally added to the influent to increase COD_{Mn} to around 10 mg L^{-1} in the first 5 d of the system startup period. Since the NH_3-N concentration in raw water was below 1.0 mg L^{-1}, ammonium sulfate was added to maintain 1.0–3.0 mg L^{-1} NH_3-N in the influent. Then, the system was continuously operated by feeding the raw water and withdrawing the effluent. After 30 d of cultivation, the bio-diatomite was matured.

In the precoating stage, the dynamic membrane needed to be pre-coated. The effluent was drawn from the membrane module and fed back to the feeding point (constant level tank) through the precoating valve. It could be regarded as a completion of precoating on the module after the effluent was qualified. When precoating was completed, the solenoid valve on the effluent pipe was simultaneously opened while the one on the precoating pipe was turned off. During the experimental period, diatomite was added to the reactor, and 2% of the mixed liquor volume in the reactor was discharged daily, corresponding to a solids retention time (SRT) of 50 d. The mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentrations in the BDDMR were maintained at around 10,000 and 2,500 mg L^{-1}, respectively. During the stationary operation, dynamic membrane flux was set at 50 L (m^{-2} h^{-1}) with a hydraulic retention time (HRT) of 4.0 h. As the operation proceeded, 45 kPa TMP was reached, indicating the completion of dynamic membrane filtration. Therefore, an on-line air supplier backwash was used to remove the dynamic membrane from the support mesh.

To investigate three individual mechanisms that might contribute to the removal of pollutants in the BDDMR (microbial biodegradation, bio-diatomite adsorption, and bio-diatomite dynamic membrane (BDDM) rejection), diatomite adsorption was processed alone in a fully mixed reactor with a HRT of 4.0 h and a MLSS of 10,000 mg L^{-1}. The pre-coated dynamic membrane (support module attaching cake layer) was carefully taken out of the bench-scale bioreactor and submerged into another tank that was filled with the same raw water.

2.4. Analytical techniques

2.4.1. Routine monitoring parameters

Routine water quality items included turbidity, UV_{254}, NH_3-N and COD_{Mn}, in the influent, effluent and mixed liquor. Influent, effluent and mixed liquor were simultaneously sampled each time. Turbidity was monitored by a turbidity meter (Model 2100P, Hach, USA). COD_{Mn}, MLSS and MLVSS were analyzed following the Chinese State Environmental Protection Agency (SEPA) Standard Methods [32]. NH_3-N concentrations were measured spectrometrically (Pocket Colorimeter II, Hach, USA) using corresponding reagent kits. UV_{254} was determined using a spectrometer (Model DR5000, Hach, USA) in a quartz cell with an optical path length of 1 cm. Total organic carbon (TOC) and DOC was measured using a TOC analyzer (TOC-VCPH, Shimadzu, Japan). The supernatant of the mixed liquor was obtained by centrifuging at 5,400 rpm for 10 min followed by filtration through a 0.45 μm membrane. DO concentrations in the reactor were recorded with a DO electrometer (Model LDO HQ30d, Hach, USA). Particle counting was conducted using a particle counting system (MODEL Versa Count, IBR, USA) with a detection range of 2–25 μm.

2.4.2. Molecular weight distribution and isolation of dissolved organic matter

Molecular weight distribution (MWD) was analyzed with a gel permeation chromatography (GPC) method on a high performance liquid chromatography (LC-10ADVP, Shimadzu, Japan) system coupled with an SPD-20A UV detector and a TSK-GEL G3000PWXL column (7.8 mm × 300 mm). Anhydrous sodium sulfate (0.05 M) was used as the isocratic mobile phase. The separated compounds were detected by UV absorbance at 254 nm. The MWD pattern was derived by calibration with polystyrene sulfonate molecular weight standards of 14, 7.5, 4.3, 1.4, 0.7, 0.5 and 0.21 kDa. Dissolved organic matter (DOM) was fractionated based on the distribution pattern of the affinity of functional groups in organics substances for water [33]. Amberlite XAD-8, XAD-4 and IRA-958 were used to separate the DOM into strong hydrophobic fractions (SHPs), weak hydrophobic fractions (WHPs), neutral hydrophilic fractions (NHPs), and charged hydrophilic fractions (CHPs). All DOM fractions were quantified according to DOC analysis. A flow chart of the preparative scheme of the DOC fractionation is shown in Fig. 2.

2.4.3. Trihalomethanes formation potential

The trihalomethanes formation potential (THMFP) tests were performed using the methods developed by Chu et al. [34] using free chlorine, except that the incubation time after chlorine dosing was extended from 3 to 7 d. Water samples were chlorinated in sealed 50 mL amber glass bottles at 24°C in the dark. Solution pH was buffered at pH 7 with phosphate salts (0.3-M NaH_2PO_4, and 0.2-M, Na_2HPO_4) obtained from Sigma-Aldrich. The disinfectant dosages for FP tests were calculated by Eq. (1). Free chlorine condition was achieved by addition of sufficient chlorine to break out any ammonia.

$$\mathrm{Cl_2\ dosage\ (mg\ L^{-1})} = 3 \times \mathrm{DOC\ (mg\ L^{-1})} + 7.6 \times \mathrm{NH_3\ (mg\ L^{-1})} + 10\ \mathrm{(mg\ L^{-1})} \tag{1}$$

After 7 d of incubation, prior to analysis, $Na_2S_2O_3$ at twice as the initial normality of chlorine added was

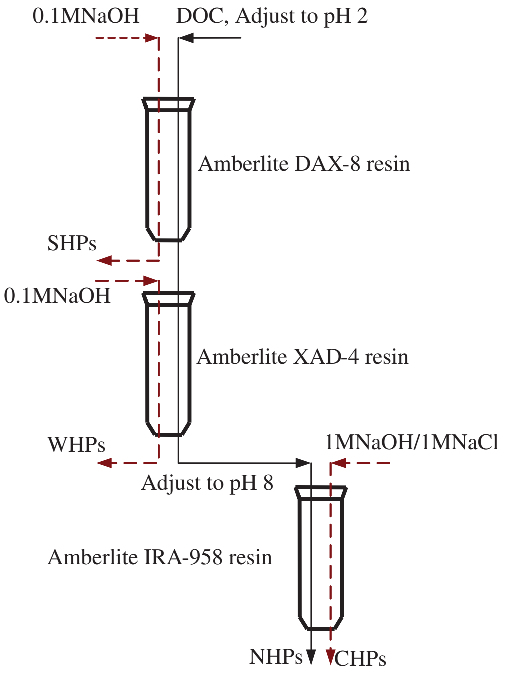

Fig. 2. Analytical procedure for preparative DOC fractionation.

added to quench any residual disinfectant [35]. Subsequently, the solution pH was adjusted to 4.5–5.0 with glacial acetic acid for THMs analysis. THMs were analyzed using a gas chromatograph (Shimadzu-QP2010, Japan) with an electron capture detector (ECD), based on U.S. Environmental Protection Agency (USEPA) method 551.1.

3. Results and discussion

3.1. *Pollutant removal efficiency of the BDDMR*

3.1.1. *Turbidity and particle removal*

As shown in Fig. 3(a), the influent turbidity was not very high fluctuating from 1.7 to 4.59 NTU. Due to bio-flocculation and membrane separation, turbidity decreased from 3.46 ± 0.13 NTU in the raw water to 0.28 ± 0.13 NTU in the effluent, with a removal efficiency of greater than 91.6 ± 3.7%. The effluent turbidity was always less than 1.0 NTU during the filtration period complying with the Chinese National Standards for Drinking Water Quality (CNSDWQ, GB5749-2006). Thus, the BDDMR displayed remarkable solid–liquid separation.

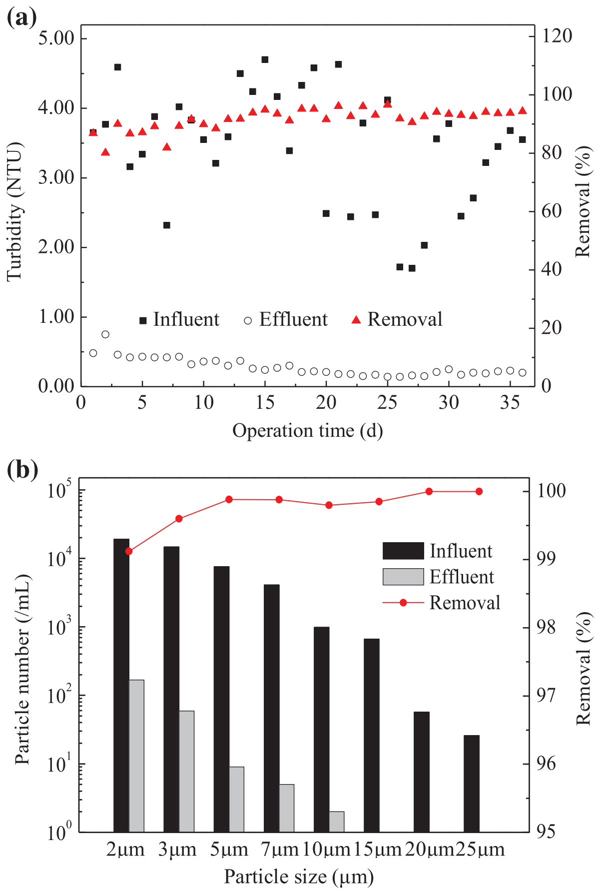

Fig. 3. Removals of turbidity (a) and particle number (b) by the BDDMR.

A particle counting technique was applied to determine the physical characteristics of the particles in the water. The particles were counted in a size range of 2–25 μm. Fig. 3(b) shows the reduction of particles in the influent and the effluent by the BDDMR. It could be seen that the total number of particles, which was 47,284 per mL in the influent, was eliminated to 244 per mL with a removal efficiency of 99.48%. Most particles in the influent ranged from 2 to 10 μm, and these particles accounted for 98.42% of the total particle number. Thus, the BDDMR demonstrated high liquor–solid separation capacity probably attributable to the mechanical screening and the surface adsorption of the cake layer. It was reasonable to infer that some large particles were retained within the pores of cake layer after the formation of the cake layer, which might result in pore blockage and partial compression of the cake. As a result, fine particles were rejected for the reduction of the permeation space. Therefore, it could be postulated that pore blockage and cake compression shed light on the high removal of turbidity and particles.

3.1.2. Organic matter removal

COD_{Mn} was used as the surrogate parameter to represent the content of organic matter; this measure is widely used in the field of drinking water treatment. COD_{Mn} in water might be classified into particulate fraction and dissolved organic fraction. Particulate organic matter (POM) could be easily reduced even through the conventional treatment process including coagulation, sedimentation, filtration and disinfection. By contrast, the conventional water treatment operations are not designed for the effective removal of DOM. During the stationary experiments, the influent had an average COD_{Mn} of 6.55 ± 0.76 mg L^{-1} ranging from 5.48 to 9.35 mg L^{-1}. As illustrated in Fig. 4(a), COD_{Mn} was decreased to 2.72 ± 0.24 mg L^{-1} in the effluent through BDDMR treatment, with a removal efficiency of 58.2 ± 4.4%.

As described above, DOM is one of the major concerns (i.e. the predominant THM precursor) in drinking water purification due to its difficulty to be removed. It should be noted that UV_{254} used as an indirect surrogate parameter representing aromatic substances with unsaturated bonds is highly effective, especially with the current financial situations of many developing countries [36]. The BDDMR influent had average DOC and UV_{254} concentrations of 7.59 ± 0.89 mg L^{-1} and 0.09 ± 0.004 cm^{-1}, respectively. DOC and UV_{254} removal efficiencies by the BDDMR system were shown in Fig. 4(b) and (c), which shows that DOC and UV_{254} were reduced by 49.3 ± 6.66 and 45.8 ± 2.85% on average, respectively. It was reasonable to infer that the residual organic in the effluent was likely the inert organic content that was to a large extent non-biodegradation. Compared to other water purification technologies, such as traditional coagulation–sedimentation–sand filtration, ultrafiltration and microfiltration membrane separation, the removal efficiencies of COD_{Mn} and UV_{254} were much higher [37–41], and they were even higher than those (COD_{Mn}: 40.05%, UV_{254}: 29.43%) in a MBR for drinking water treatment [1,42]. The experimental results suggest that the BDDMR process can be both technically and economically feasible for use in drinking water treatment, not only for organic degradation, but also for the possible replacement of the conventional treatment process.

3.1.3. NH_3-N removal

Ammonia concentration was monitored throughout the present study. It could be seen from Fig. 5(a) that the average NH_3-N concentration of the influent was relatively low (0.23 ± 0.005 mg L^{-1}), and the effluent NH_3-N concentration was 0.015 ± 0.006 mg L^{-1} with an average removal efficiency of 93.7 ± 2.26%.

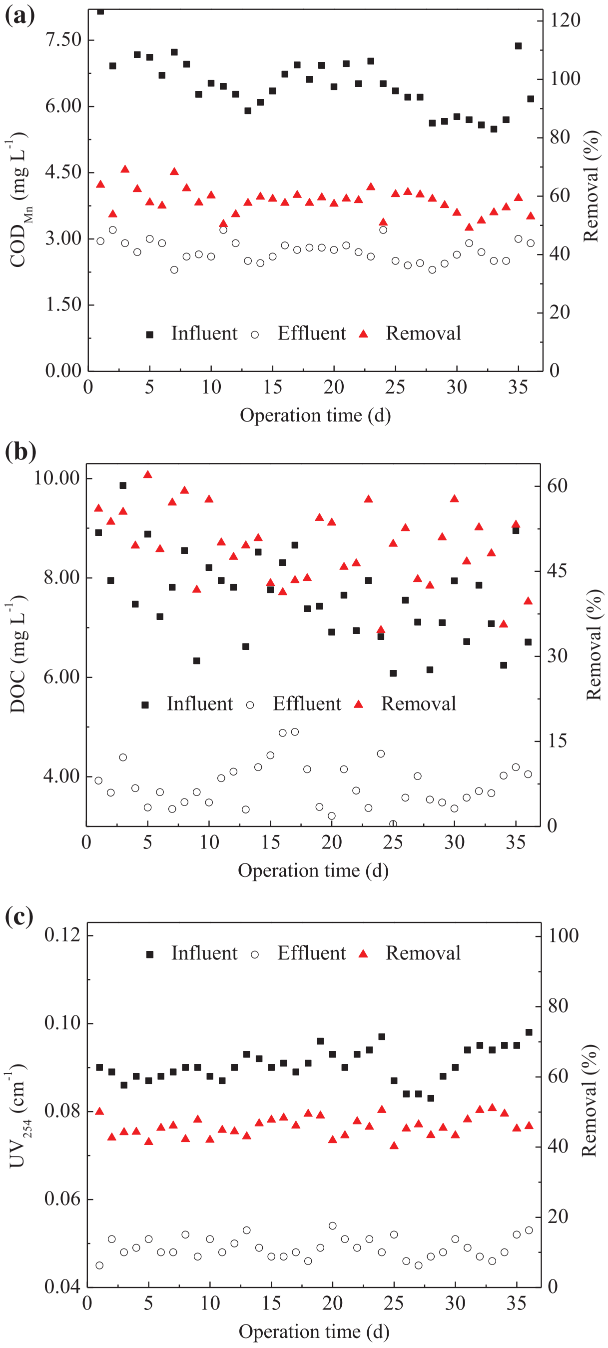

Fig. 4. Removals of (a) COD_{Mn}; (b) DOC and (c) UV_{254}.

In addition, the NH_3-N removal efficiencies by the diatomite adsorption alone and the BDDM alone were also examined. There are only 6.1 and 7.8% of the influent NH_3-N were removed by diatomite adsorption alone and BDDM, respectively (Fig. 5(b)). Some

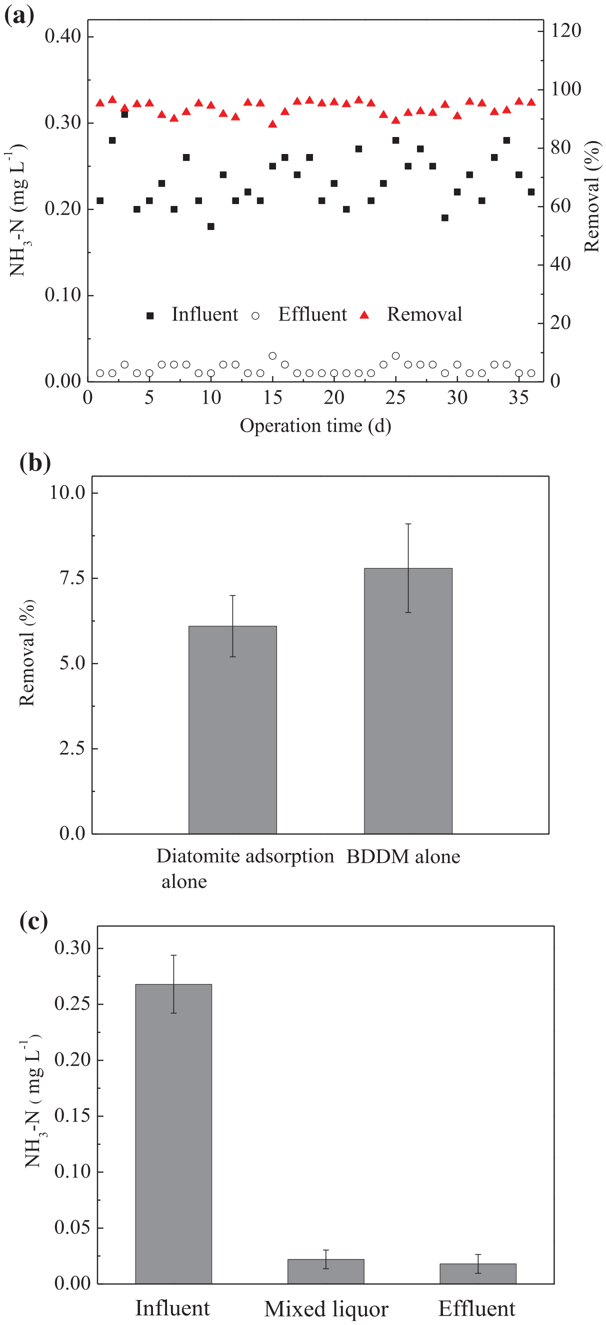

Fig. 5. (a) Removal of NH_3-N by the BDDMR; (b) removal of NH_3-N by BDDM alone and bio-diatomite adsorption alone; and (c) NH_3-N in the waters from the BDDMR. Data of (b) and (c) represent the average of two samples, each measured three times.

investigators also found that the microfiltration or ultrafiltration membrane was less effective for NH_3-N removal by retention capacity [43,44]. To illustrate the removal mechanism of ammonia in the BDDMR, the ammonia concentrations in influent, mixed liquor and effluent were measured. As shown in Fig. 5(c), the influent had the average NH_3-N concentration of 0.268 ± 0.026 mg L^{-1}. 0.022 ± 0.008 mg L^{-1} was detected in the mixed liquor of the BDDMR. This concentration was essentially at the same level of the 0.018 ± 0.008 mg L^{-1} in the effluent, while much lower than that in the influent. Therefore, it is reasonable to infer that high NH_3-N removal efficiency in the BDDMR was reached through better biological nitrification performance, which could be attributed to the long SRT (50 d) provided by the BDDMR for the population of nitrifying bacteria.

3.1.4. Relationship between THMFP and DOM

The THMs included chloroform ($CHCl_3$), bromodichloromethane ($CHCl_2Br$), and dibromochloromethane ($CHClBr_2$) and tribromomethane ($CHBr_3$). As illustrated in Fig. 6, $CHCl_3$, $CHCl_2Br$, $CHClBr_2$, and $CHBr_3$ levels were 143.8 ± 25.4, 81.7 ± 13.9, 17.5 ± 5.1 and 0 μg L^{-1}, respectively, in the influent and 55.7 ± 14.5, 39.7 ± 9.4, 8.3 ± 1.4 and 0 μg L^{-1}, respectively, in the effluent. These measurements corresponded to removal efficiencies of 60.9 ± 8.8, 50.8 ± 11.0, and 49.4 ± 14.3%, for $CHCl_3$, $CHCl_2Br$, and $CHClBr_2$, respectively. Bromoform ($CHBr_3$) was not detected in either the influent or the effluent. The average removal efficiency of the THMs was 56.9 ± 6.0%. All of the THMs concentrations met the requirements of the CNSDWQ (GB5749-2006).

It has been considered that THM was closely related to the DOM [45–48]. In the present study, the feeding water was surface water slightly polluted by sewage discharge. Therefore, organic matter form sewage in the raw water was supposed to be the main THM precursor [49,50]. As described in Section 3.1.2, DOC was reduced by 49.3 ± 6.66% after the BDDMR

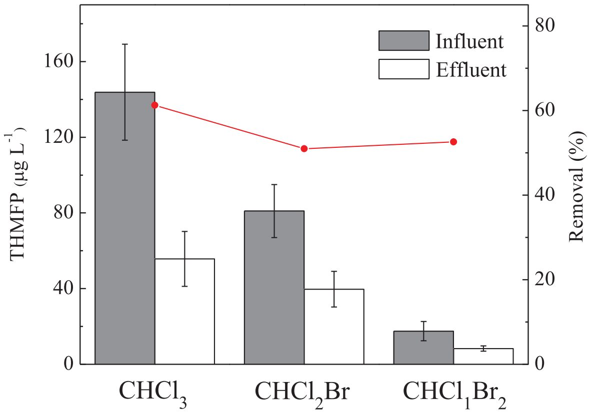

Fig. 6. Removal of THMFP by the BDDMR.

treatment. It is worth noting that the DOC removal has been the main cause of the 56.9 ± 6.0% THM removal. In other words, the unit carbon THMFP was eliminated from 31.9 to 27.1 μg TH mg^{-1} DOC (by 15%). Additionally, UV_{254} as an indirect surrogate parameter representing aromatic substances with unsaturated bonds was applied to indicate source water THMFP. In this study, UV_{254} was reduced by 45.8 ± 2.85% on average causing THMFP reduction. Furthermore, it has been suggested that DOM with low molecular weight (<3 kDa) had a dominant effect on THMFP during the disinfection process by chlorine in purifying raw source water [51]. The fractions of low molecular weight are more aromatic having a higher content of carboxyl groups and more reactive to form THMFP in the disinfection process [52]. The MWD of the organic in the raw water ranged from 3,000 to 200 Da below (Fig. 9). This observation is consistent with the finding of THMFP formation aforementioned.

3.2. *TMP and filtration resistance under different design flux*

The most important disadvantage of dynamic membrane is declination of permeate flux due to membrane fouling. In the present research, the variations of the TMP as a function of operation time at different fluxes were monitored to evaluate dynamic membrane fouling. 45 kPa was used to represent a termination of a period operation and to analyze quantitatively.

After the precoating stage, the effluent flux was set at 30, 50, 80 and 120 L ($\text{m}^{-2}\,\text{h}^{-1}$). Fig. 7(a) shows that the periodic durations of the four selected fluxes 30, 50, 80 and 120 L ($\text{m}^{-2}\,\text{h}^{-1}$) were 33, 22, 12, and 7 d, respectively. It could be seen that TMP was invariant of each selected flux in the early filtration stage. During the final stage of the operation period, TMP rose to 45 kPa for each of the selected fluxes. The slopes of the TMP curves were different. As expected, the greater the flux, the steeper the curve of the TMP. It was presented that the dynamic membrane filtration cycles approach its completion when 45 kPa was reached. Thus far the BDDMR needed to be backwashed.

It was obvious that TMP of the BDDMR rose due to the increasing of filtration resistance. The filtration resistance is generally described by Darcy′ law (Eq. (2)):

$$R = \frac{\Delta P}{\mu J} \tag{2}$$

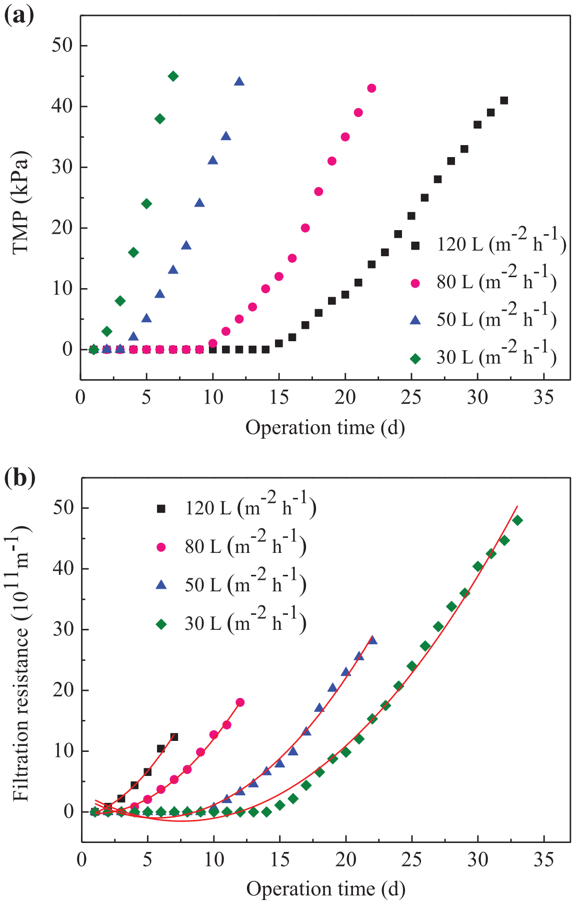

Fig. 7. TMP (a) and filtration resistance (b) as a function of operation time at different fluxes.

where ΔP is the TMP, μ the solution viscosity, R is the filtration resistance, J is the filtrate flux.

Fig. 7(b) shows that the filtration resistance of each of the selected fluxes increased with the operation time. During the early stage of all the fluxes, the time maintaining invariant resistance ($R = 0\ \text{m}^{-1}$) at 30, 50, 80 and 120 L ($\text{m}^{-2}\,\text{h}^{-1}$) were 14, 9, 3 and 1 d, respectively. The resistance increased slowly thereafter. It could be seen that the operation time for the pressure to reach 45 kPa was substantially prolonged when the flux was decreased. Obviously, the resistance of high flux increased faster than that of lower flux. Thus, it should be noted that, in the present bench-scale experiment, the thickness of cake layer was apt to grow up to 2 cm due to the weak cross-flow effect and the high concentration of bio-diatomite in the mixed liquor. It can be called thickness-increase resistance [13]. In addition, operation pressure caused cake layer compression due to restructuring of bio-diatomite particles, resulting in an increased resistance of dynamic membrane which is called compaction resistance. Therefore, TMP of the BDDMR rose mainly due to the increasing of the filtration resistance, resulting from cake layer thickness-increase and cake layer compression. Moreover, the blocking of pores by rejected solutes and adsorption of some substances (which may be microorganisms and extra-cellular polymer substance) on the pores were also the important contributors to the operation resistance. However, the effect of various factors on dynamic membrane fouling is a significant research frontier that still merits further study.

3.3. Mechanisms of BDDMR for the organics removal

The removal efficiencies of organics can be ascribed to bio-diatomite adsorption, microbial biodegradation and BDDM rejection, especially the microbial biodegradation function. Some studies have been conducted to elucidate the effect of various factors on organics' removal.

3.3.1. Identification of individual effects on organics removal

To determine which effect mainly accounted for the organics removal, the dynamic membrane operated by 24 h was carefully taken out from the BDDMR and then submerged into another tank that was filled with the same raw water but contained little MLSS. The same flux (50 L (m^{-2} h^{-1}) and identical automatic control system were applied. As shown in Fig. 8, the cake layer alone reduced influent COD_{Mn}, UV_{254} and TOC by 7.8, 9.5 and 9.2%, respectively, which indicate that cake layer alone was much less effective in eliminating the organics. Additionally, diatomite static adsorption was undertaken to examine the removal efficiency of pollutants through diatomite adsorption alone. Three hours of static experiments with the same raw water and diatomite concentration were conducted. There are only 13.2, 4.8 and 16.4% of COD_{Mn}, UV_{254} and TOC, respectively, were removed through static adsorption. It was reasonable to infer that the additional purification of pollutants was ascribed to microbial degradation and diatomite adsorption in the BDDMR, which corresponds to the findings of previous study [26]. Therefore, diatomite addition probably had a more significant contribution to the synergetic effect of adsorption and the biodegradation of microorganisms, which provided a carrier material for attaching microorganisms in the system.

3.3.2. Fractionation and MW distributions of DOM

Based on the distribution pattern of the affinity of functional groups in organic substances for water, DOM was separated into four fractions: SHPs, WHPs, NHPs and CHPs. The removal efficiency of various DOM by the BDDMR was demonstrated in Fig. 9(a). As discussed above, the organics was supposed to be removed by bio-diatomite adsorption, microbial biodegradation and BDDM rejection. It could be seen that SHPs and WHPs concentrations in the mixed liquor were much lower than the values of the influent due to microbial degradation together with the bio-diatomite adsorption. These two fractions corresponded to high removal efficiencies of approximately 45.8 and 46.2%. On the other hand, NHPs and CHPs, with removal efficiencies of 10.5 and 12.8%, respectively, were much lower than the removal of SHPs and WHPs. It has been investigated that the cake layer alone achieved little removal effect (Section 3.3.1). The rejection efficiencies of SHPs, WHPs, NHPs and CHPs by the cake layer alone were only 9.5, 6.4, 6.8 and 3.7%, respectively. These results shed light on that the BDDMR was able to effectively eliminate SHPs and WHPs, but little NHPs and CHPs were reduced. Therefore, the conclusion could be drawn that bio-diatomite adsorption and bio-degradation were the main functions of the system and cake layer filtration alone was not very effective in rejecting various DOM fractions. This observation is consistent with the results described in Section 3.3.1.

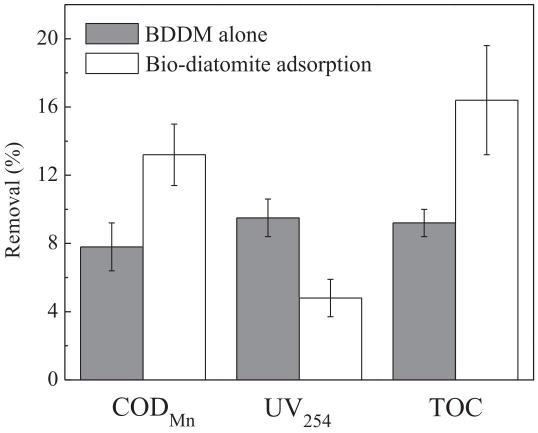

Fig. 8. Removals of COD_{Mn}, UV_{254} and TOC by BDDM alone and bio-diatomite adsorption alone. Data represent the average of two samples, each measured three times.

The GPC method was applied to determine the apparent MWD of DOM in the raw water, mixed liquor and the effluent of the BDDMR. Fig. 9(b), shows that organic matter with MWD ranging from 2,813 to 242 Da was predominate in the raw water, and five peaks in the MW (2,813, 2,285, 1,588, 950 and 242) were detected. It could be observed that the absorbance intensity of the DOM in the raw water was

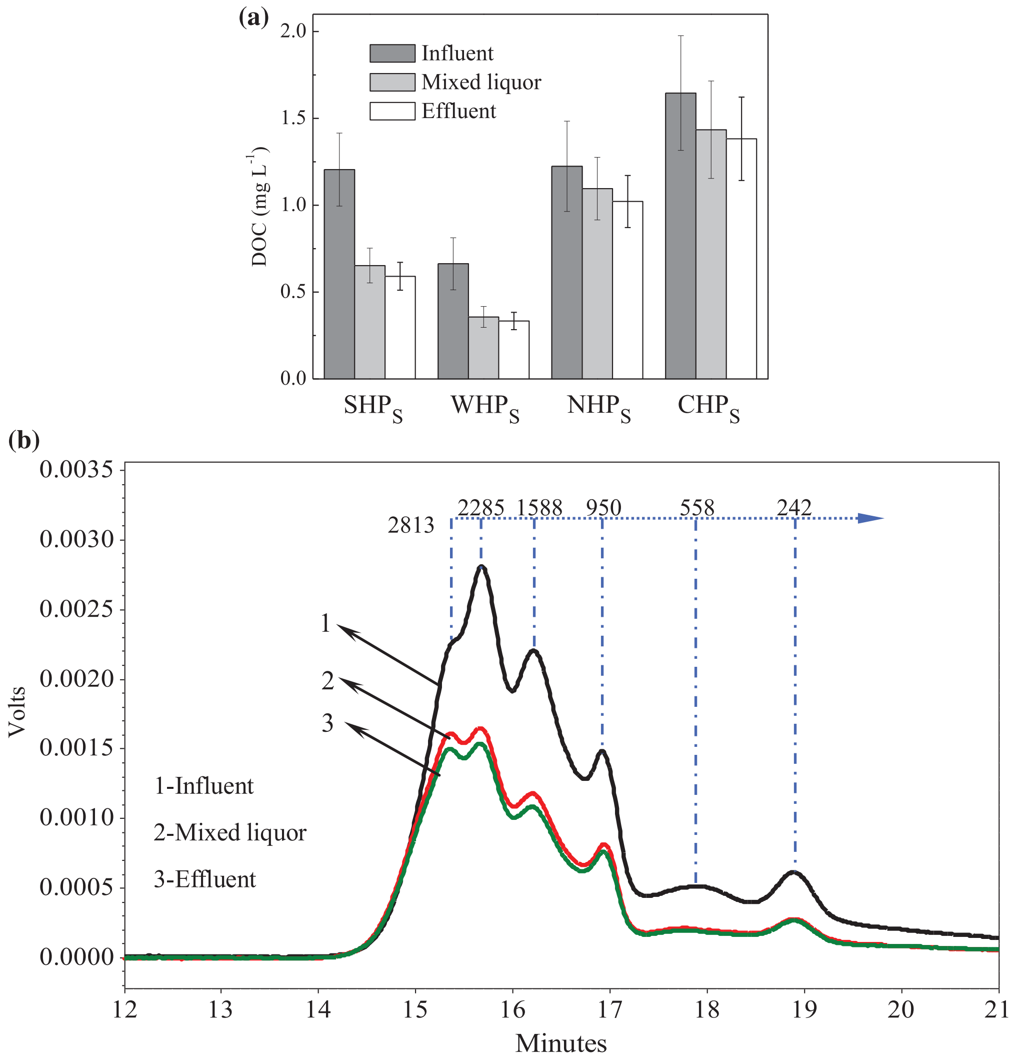

Fig. 9. Fractionation (a) and MW distribution (b) of DOM in influent, effluent and mixed liquor. Bars represent the average of two isolated samples, each measured once.

much higher than the absorbance intensity of the DOM in the mixed liquor, which indicated that the organic matter was removed strongly by approximately 40% after bio-diatomite adsorption and microbial biodegradation. By comparing the spectra of the mixed liquor and the effluent, we found that the dynamic membrane alone could partially remove the MWD 950–2,800 Da peak cluster. Therefore, it is reasonable to infer that the augmented separation function for the MWD 950–2,800 Da peak cluster was provided by gel layer formed on the surface of the module [53]. Although the organic matter with a MWD less than 950 Da peak cluster could be decontaminated by bio-diatomite adsorption and microbial degradation, the organics was able to penetrate the cake layer without reduction.

4. Conclusions

In this study, a bench-scale BDDMR was proposed to decontaminate the SCSW. The experimental results illustrates that BDDMR is highly effective for impurity removal with an HRT of 4.0 h, an MLSS of 10 g/L, and a constant flux of 50 L (m^{-2} h^{-1}) and may be one of the sustainable technologies for drinking water treatment. The following conclusions can be drawn:

(1) An excellent solid–liquor separation function was achieved during the treatment process. Turbidity and particle number were eliminated by 91.6 and 99.48%, respectively.
(2) The BDDMR reduced COD_{Mn}, UV_{254} and DOC by 58.2 ± 4.4%, 45.8 ± 2.85% and 49.3

± 6.66%, respectively. NH_3-N was decreased by 93.7 ± 2.26 and the main way was the biological nitrification. THMFP reduction (by 56.9 ± 6.0%) was also examined.

(3) Cake layer alone reduced influent COD_{Mn}, UV_{254} and TOC by 7.8, 9.5 and 9.2%, respectively. Only 13.2, 4.8 and 16.4% of COD_{Mn}, UV_{254} and TOC, respectively, were reached through bio-diatomite static adsorption.

(4) The dynamic membrane alone could partially remove the DOM in the MWD range of 950–2,800 Da peak cluster. DOM fractionation indicated that SHPs, WHPs, NHPs, and CHPs were removed due to microbial degradation and bio-diatomite adsorption with the efficiencies of 45.8, 46.2, 10.5 and 12.8%, respectively; while 9.5, 6.4, 6.8 and 3.7%, respectively, were obtained through cake layer rejection.

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Abbreviation

BDDM — bio-diatomite dynamic membrane
BDDMR — bio-diatomite dynamic membrane reactor
CHPs — charged hydrophilic fractions
COD_{Mn} — chemical oxygen demand
DO — dissolved oxygen
DOC — dissolved organic carbon
DOM — dissolved organic matter
GPC — gel permeation chromatography
HRT — hydraulic retention time
MBR — membrane bioreactors
MLSS — mixed liquid suspended solids
MLVSS — mixed liquid volatile suspended solids
MWD — molecular weight distribution
NHPs — neutral hydrophilic fractions
POM — particulate organic matter
SCSW — slightly contaminated surface water
SHPs — hydrophobic fractions
SRT — solids retention time
THMFP — trihalomethane formation potential
TMP — trans-membrane pressure
TOC — total organic carbon
UV_{254} — UV absorbance at 254 nm
WHPs — weak hydrophobic fractions

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