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Ultraviolet membrane bioreactor for enhancing the removal of organic matter in micro-polluted water

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ABSTRACT

This research investigated the organic matter removal performance using an ultraviolet (UV) treatment membrane bioreactor (UV–MBR) in micro-polluted surface water. Compared with a conventional MBR process, the UV–MBR achieved higher removal efficiencies in terms of the reduction of COD_{Mn} and UV_{254}, which were reduced by 58 and 63%, respectively. The organic matter removal rate increased significantly arising from biodegradation mechanisms. Furthermore, the ammonia nitrogen and nitrite nitrogen were removed almost completely and not affected by the subsequent UV treatment. Through the UV treatment, a fraction of refractory organics was transformed into biodegradable organics available to the microbial consortium in the reactor. UV treatment of the raw water leads to an increase of BDOC content from 0.22 to 0.88 $mg\,L^{-1}$ after 5 d inoculation. According to the analysis of the characteristics of hydrophobic/hydrophilic compounds, the relative molecular weight distribution and the fluorescence excitation–emission matrix of the organics, it can be indicated that the organics were transformed into biodegradable organics because part of molecules were split into simple structures during the UV treating process. Furthermore, the organics removed by the UV–MBR were hydrophobic fractions.

Keywords: UV treatment; MBR process; Organic matter; Micro-polluted water

1. Introduction

Organic matter removal in raw water is one of the major concerns in drinking water treatment. Currently, the principle process to remove organics in China is still a conventional treatment by flocculation and sedimentation. However, the performance of such treating processes for organic removal is unsatisfactory, especially for purifying the micro-polluted water, since dissolved organic matter (DOM) in water is heterogeneous and of low concentration and its structure changes spatially and temporally [1,2]. The micro-polluted water contains low molecular weight components which tend to be more hydrophilic and are considered difficult to remove by coagulation [2]. The principle components, DOM may affect the water quality in the subsequent processes during the conventional treatment. For example, the production of disinfection by-products (DBPs) may be higher during

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chlorination in a subsequent disinfection process [3] and particles can stimulate heterotrophic bacterial regrowth in drinking water distribution systems [4].

In order to improve the organic matter removal efficiency in the micro-polluted water and to decrease DBPs generation, membrane filtration technology associated with biological processes has been employed in drinking water treatment [5,6]. The membrane bioreactor (MBR) process has been introduced to drinking water treatment because of its high reduction of organic matter and ammonia nitrogen in water treatment. Furthermore, in recent years, the MBR process in drinking water treatment is often applied with powered activated carbon (PAC), which could be the carrier of micro-organisms in the bioreactor and can provide protection for them from shear forces [7] and it has been reported that high pollutant reduction can be achieved using PAC-MBR process as an effective way of treating surface water [8,9].

Ultraviolet (UV) treatment is considered as an alternative disinfection method to chlorination, since no DBPs are generated and UV can inactivate pathogenic micro-organisms effectively at low UV doses [10,11]. It has been reported that UV treatment can change the structure of organics and lead to decrease in the formation potentials of trihalomethanes and haloacetic acids when the UV doses were in high level [11].

Although the MBR system exhibits good contaminants reduction in most cases, the organic matter reduction is not favorable in the micro-polluted water. It was well known that micro-organism activity is the key factor in influencing the efficiency of the MBR. Nevertheless, the organic matter available to bacteria is insufficient in the micro-polluted water and part of the organics is refractory substances. Micro-organisms attached to the PAC particles have insufficient biodegradable substrates, which can lead to poor biodegradation of pollutants. Advanced oxidation is widely used in treating refractory organics by breaking up the structure of complex organic matter and changing its characteristics. UV treatment can increase the assimilable organic carbon (AOC) and biodegradable organic matter concentration when applied in treating the raw water [12,13]. In previous researches, the UV treatment may result in biological instability in the drinking water distribution system, thereby increasing the AOC level; this is a significant drawback which limits the popular utilization of UV-oxidation.

However, in this research, UV treatment was used as a pre-treatment of the MBR system. It was applied at the beginning of the process since much more biodegradable organic carbon (BDOC) was expected to be produced. The drawback of UV in disinfection process may bring advantages in the UV–MBR treatment. Compared to the MBR, AOC can continue to be effective after pre-treatment even when the organic matter level was low in the raw water [14,15], therefore, micro-organisms in the pre-treatment of MBR can metabolize better, and the contaminant removal rate would also be enhanced. In view of this, the organics removal efficiency was investigated to compare the UV–MBR process and MBR system. For clear understanding, the contaminant removal mechanism in the combined treatment, the hydrophobic and hydrophilic performance, the relative molecular mass distribution and the fluorescence excitation–emission matrix (EEMs) of water samples were analyzed to investigate the organic matter characteristics variance during the process.

2. Materials and methods

2.1. Raw water

The raw water used in the study was from the Sanhaowu Lake in Tongji University (Shanghai, China). It was a typical micro-polluted raw water in winter when the experiment was launched from October 2012 to February 2013. Table 1 presents the raw water quality parameters.

2.2. Experimental setup

2.2.1. The MBR system

A bench-scale MBR system was operated under the controlled laboratory conditions; the schematic diagram is shown in Fig. 1. The experimental setup consisted of 2 L bioreactor and an immersed hollow fiber PVC module (Liters Ltc., Hainan, China). Each membrane tube had an internal diameter of 0.6 mm with 0.01 μm membrane pore size, and the total area was $0.10\,m^2$. The apparatus was operated in continuous mode at a flux of $0.67\,L\,h^{-1}$ controlled by a peristaltic pump. The suction pump was controlled by a timer for 8 min working followed by a 2 min time-out in each cycle. The hydraulic retention time (HRT) was set at 3 h. Air was injected into the base of the module via an aeration pump to scour the membrane fibers, provide enough O_2 and prevent membrane fouling [16]. The ratio of gas and water amount was maintained at 20:1. At the beginning of the operation, $1\,g\,L^{-1}$ PAC was added into the reactor and 0.5 L activated sludge (Quyang Sewage Plant, Shanghai) was inoculated as the initial mix liquid suspended solid (MLSS) ($0.20\,g\,L^{-1}$). In order to prevent the PAC and activated sludge from precipitating on the bottom of bioreactor, a magnetic stirring was applied during the experiment.

Table 1
Sanhaowu Lake water quality characteristics

Parameters	Range	Mean
COD_{Mn} (mg L^{-1})	3.02–3.87	3.55
UV_{254} (cm^{-1})	0.058–0.074	0.065
Ammonia nitrogen (mg L^{-1})	0.31–1.70	0.85
Nitrite nitrogen (mg L^{-1})	0.035–0.098	0.067
Nitrate nitrogen (mg L^{-1})	1.312–1.761	1.519

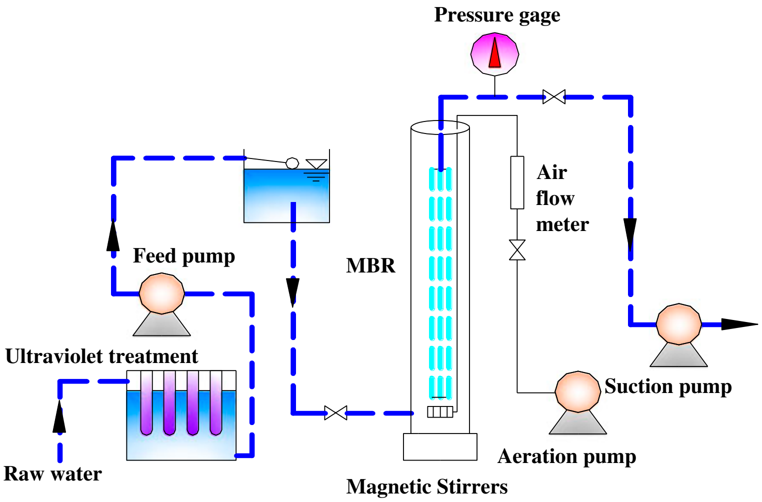

Fig. 1. Schematic diagram of the UV–MBR system.

At the beginning of the installation, four UV lamps (254 nm wavelength, 11 W, Philips, Netherlands) were installed with quartz sleeves (15 cm in length and 2 cm in diameter) in a water tank (25 cm in length, 25 cm in width, and 35 in depth) which was enclosed during the experiment. Since the UV system switch was independent of the MBR system, it could be controlled easily at different stages during the experiment. The HRT in the water tank was set at 3 h.

2.2.2. Operating procedures

Before the research, the MBR system had been operated for more than 80 d and the PAC adsorption capacity had reached saturation, which means, the organic matter parameters of the effluent were stable. It can be inferred that the variation of the contaminants removal had no relation to the PAC adsorption.

As the contaminants removal performance is almost dependent on the biological metabolism, it can be deduced that the microbial activity which may influence the treatment result in the MBR was stable.

The experimental data in this research were separated in two stages. In order to compare the treatment efficiency with the UV–MBR process, first 17 d' data was chosen from the long-term stable operating of the latter part of MBR operating. In the experimental days, the MBR system was operated by feeding raw water directly. In the second stage, from the 18th day to 25th day, the UV lamps were turned on and the UV–MBR treatment came into operation.

2.3. Analysis methods

Before measuring the organic parameters, the water samples were filtered through a 0.45 μm membrane. The permanganate index (COD_{Mn}) and ultraviolet adsorption (UV_{254}) were measured according to standard methods as well as ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen (HACH DR5000, USA) [17].

2.3.1. BDOC analysis

BDOC is defined as the fraction of DOC which can be used and removed by heterotrophic microorganisms. In this study, BDOC was measured according to the batch procedure by Khan et al. [18]

and Ratpukdi et al. [19]. Samples were filtered through a 0.45 μm membrane, the first 100–200 mL filtrates were discharged for avoiding contamination in the filter before being collected into a 500 mL vial and inoculated with 2 mL of MLSS from the bioreactor. Then the incubation was conducted in the dark at 20°C for 5 d. After incubation, the sample was again filtered through a 0.45 μm membrane before measurement. The deionized water incubated with MLSS was used as the blank sample. The BDOC value was calculated as the measured TOC (Sievers 900, German) difference before and after the incubation.

2.3.2. High-performance size exclusion chromatography

High-performance size exclusion chromatography (HPSEC) (e2695, Waters, USA) followed by a TOC detector was employed to determine the relative molecular weight distribution of organics in the raw water, the effluent of the UV treatment and the effluent of the UV–MBR system.

2.3.3. Hydrophobic/hydrophilic fraction analysis for organics

Determination of the hydrophobic/hydrophilic fraction was based on the experimental method by Lee et al. [20]. Three fractions were separated from raw water by two resins: XAD-4 and XAD-8 (Amberlite, USA) which were pre-cleaned by submersing in methanol (chromatographically pure) for 24 h. Then, the resins were immersed in ultrapure water and decanted, and repeated the process for three times. After that, the resins were placed in a column (0.6 cm in diameter, 2 cm in length). Before use, the resins were cleaned again by passing ultrapure water through the columns until the trace amount of TOC present in the effluent was less than 0.1 mg L^{-1}, and the detected UV_{254} was less than 0.001 cm^{-1}. A water sample (40 mL) passed through XAD-8 and XAD-4 columns was adjusted to pH 2 using concentrated HCl solution (37%). The adsorbate attached to the XAD-8 and XAD-4 resins were hydrophobic and slightly hydrophobic fractions, which were then eluted by passing a NaOH solution (0.1 M) through the column, respectively. The final effluent after the two resins was the hydrophilic fraction. In order to maximize the fraction recovery, a very low flow rate was set at 1.5–2 mL min^{-1} by means of a pump. The separated samples were analyzed by TOC and UV_{254}, respectively.

2.3.4. Fluorescence characteristics analysis for organics

The fluorescence characteristics of the organic matter were measured using a fluorescence spectrophotometer (CARY Eclipse Varian, Canada). The excitation spectral range was from 200 to 450 nm in 10 nm intervals, and the emission scans were performed from 250 to 600 nm in 2 nm steps. The EEM was obtained based on the excitation and emission spectra which revealed a picture of the organic matter fluorescence characteristics.

3. Results and discussion

3.1. Effect of UV treatment on pollutant removal

3.1.1. Organic matter removal

During the first 17 d, the MBR system performed stably. The average effluent COD_{Mn} concentration through the MBR system was 2.22–2.91 mg L^{-1} and its removal rate was around 26%, as shown in Fig. 2(a). Meanwhile, the UV_{254} removal rate was less than 15% (Fig. 2(b)). It was found that the MBR system alone could not obtain desirable effect for treating micro-polluted water. Tian et al. [21] also found that the reductions of COD_{Mn} and UV_{254} were limited by using the MBR.

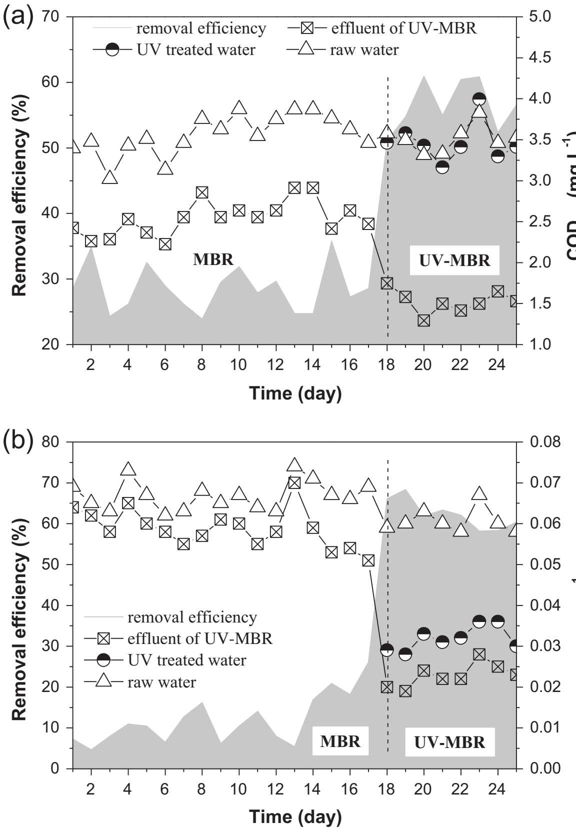

Fig. 2. Organic matter removal efficiency: (a) COD_{Mn} and (b) UV_{254}.

During the second stage of the experiment (from 18th day to 25th day), the organic matter removal rate was investigated using the UV–MBR combined system. The results showed that more than 50% COD_{Mn} and 60% UV_{254} removal rates were achieved, as shown in Fig. 2. Compared with the first stage, the organic matter removal performance was enhanced greatly. Treguer et al. [22] demonstrated that ozone treatment can break up organics with large molecular weight to that with small molecular weight, which led to an obvious increase in biodegradability. Other researchers also reported that high UV dose led to the change of organic structures [23]. The principle factor influencing the organic matter removal efficiency is the biodegradable action in the MBR system. Usually, heterotrophic bacteria maintain a stable metabolism feeding on biodegradable carbon. Nevertheless, most of the organic matter present in the raw water is non-biodegradable [21]. Because of insufficient nutrients for the micro-organisms, the organic matter assimilated by heterotrophic bacteria was inadequate and the removal performance was affected. However, in the UV–MBR system, the organics biodegradability increased through the UV treatment, which provided more nutrients for the heterotrophic bacteria in the subsequent MBR system.

In Fig. 2(a), the COD_{Mn} concentration in the UV treatment was close to that in the raw water; however, the UV_{254} decreased markedly. The UV_{254} response to aromatic hydrocarbons [22,24] and unsaturated hydrocarbons consisted of aldehyde, ketone, and conjugated diene. The latter consisted of small molecules which were the decomposition products by micro-organisms with low biodegradability. From Fig. 2(b), it can be assumed that the UV treatment can effectively change the organic matter structures and transform them into simple components available to micro-organism.

3.1.2. Ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen removal

Through the biological treatment, the ammonia nitrogen removal efficiency was much better than that of organic matter in the MBR. As shown in Fig. 3, the ammonia nitrogen concentration fluctuated from 0.3 to 1.7 mg L^{-1} in the influent, whereas it was lower than 0.2 mg L^{-1} in the effluent both in the presence and absence of UV treatment during the MBR system. The nitrite nitrogen removal performance was similar to that of ammonia nitrogen, which showed that nearly complete nitrification occurred in the UV–MBR, as presented in Fig. 3(b). After 17th day, the nitrite nitrogen amount after UV treatment nearly doubled its original concentration in the raw water; however, the high nitrite nitrogen content did not influence the treating performance, since it was still as low as 0.005 mg L^{-1} in the effluent of the UV–MBR system. In other words, the UV treatment did not affect nitrification. Fig. 3(c) shows the variation of nitrate nitrogen concentration. The nitrate nitrogen level decreased a little after the UV treatment, leading to the nitrite nitrogen increase which was much more than that in the raw water. It can be inferred that the UV treatment

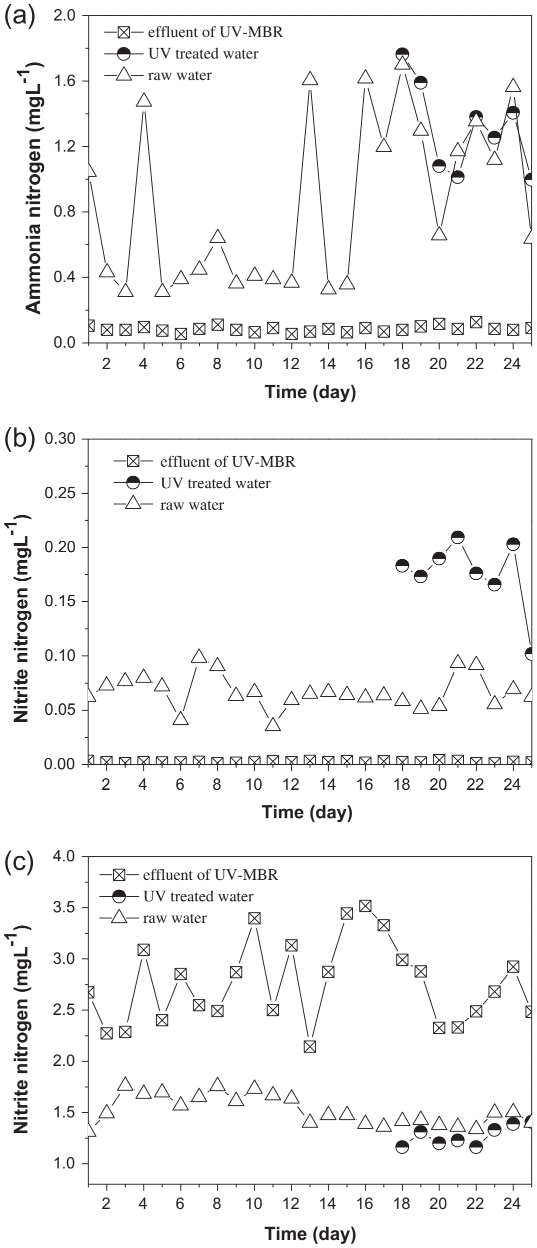

Fig. 3. Nitrogen removal efficiency, (a) ammonia nitrogen, (b) nitrite nitrogen, and (c) nitrate nitrogen.

can induce nitrate nitrogen to transform into nitrite nitrogen.

3.2. The characteristics of organic matter

3.2.1. Effect of UV treatment on BDOC and relative molecular weight distribution of organics

To illustrate the organic matter removal mechanism during the UV–MBR treatment, the relative molecular weight distribution was measured by HPSEC. Two peaks were observed in the relative molecular weight distribution pattern of the raw water, as shown in Fig. 4. The first peak in the chromatogram responds to humic substances with a molecular weight of 2,000–3,000 Da, and the second peak responds to simple organics with small-size molecules (molecular weight below 1,000 Da) [25]. Compared with the first peak, the second one had higher intensity, which implies that small-size molecules are the main substances in micro-polluted water.

After the UV treatment, the humic substances concentration decreased from 0.014 to 0.009 mg L^{-1}, which may be attributed to that high UV dose can induce organics to transform into lower molecular fractions [19,23,26]. It was inferred that the UV treatment was very effective in changing higher molecular weight characteristics such as aromatic hydrocarbon and humic substances [27]. From the relative molecular weight distribution pattern of organics in the UV–MBR effluent, the intensity associated with simple organics had a significant reduction which arose from the biodegradation by heterotrophic bacteria in the activated sludge. However, the first peak increased a little based on the effluent analysis. It was supposed that microbial metabolic products with molecular weight around 2,000 Da were generated and part of them leaked from the membrane. Nevertheless, the total organic matter notably decreased in the raw water. It was more feasible for micro-organisms to take in small-size organics, as the organic molecular weight distribution patterns of water samples are shown.

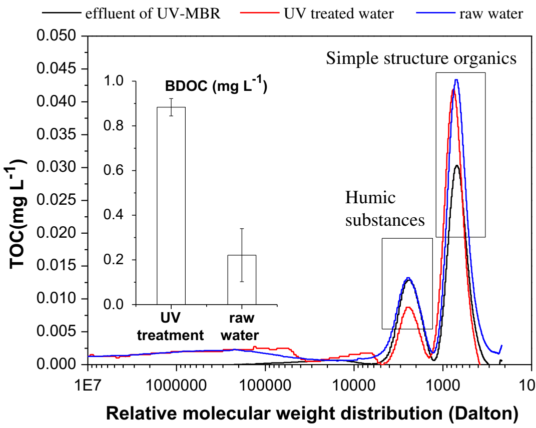

Fig. 4. HPSEC–TOC chromatograms and BDOC of raw water, water samples by UV treatment and UV–MBR.

As seen in Fig. 4, the BDOC value increased significantly with the UV treatment. Compared with that in the raw water, the BDOC content rose from 0.22 to 0.88 mg L^{-1} and BDOC/DOC ratio increased from 6.3 to 26.7%. This indicated that the UV treatment can change the organic matter structure in the raw water to that easily assimilated by micro-organisms. In addition, the high BDOC content can promote the bacterial regrowth potential generation as well. In other words, part of refractory contaminants was transformed into biodegradable matter and the extra generated nutrients can help the microbes thrive.

3.2.2. Hydrophobic/hydrophilic fraction

Fig. 5 shows the hydrophobic/hydrophilic fraction distribution in the raw water, water sample by UV

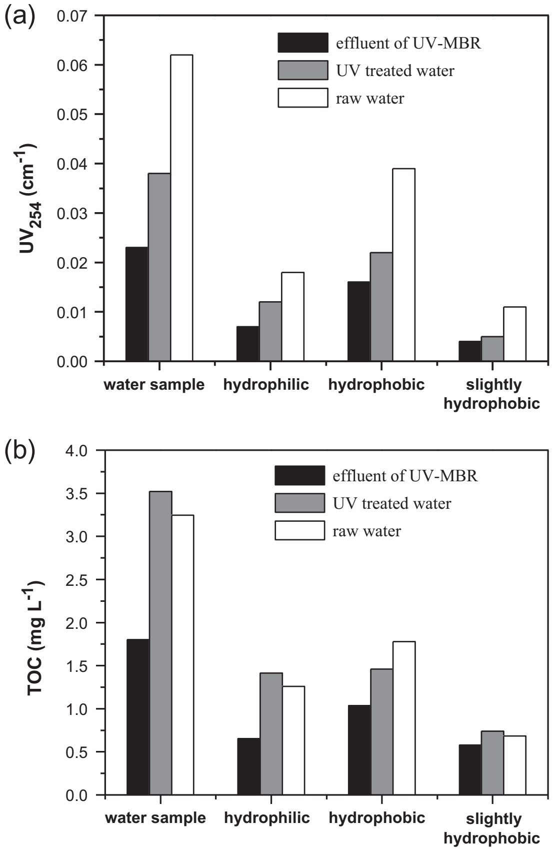

Fig. 5. Distribution of raw water, water samples by UV treatment and UV–MBR: (a) characterized by UV_{254} and (b) characterized by TOC.

and the UV–MBR system treatment. As shown in Fig. 5(b), the organics in the raw water were composed of 48% hydrophobic, 34% hydrophilic, and 18% slightly hydrophobic fractions. The organic distribution of raw water characterized by UV_{254} had the same pattern as that by TOC, that is, the hydrophobic fraction was the main component in the influent (almost 60%).

The variation trends of UV_{254} and TOC volume were significantly different through the UV treatment. According to Fig. 5(a), the hydrophilic fraction decreased from 0.019 to 0.014 cm^{-1}, while it increased a little bit based on the TOC in Fig. 5(b). Compared with the hydrophilic fraction, similar trends were found in the terms of the other two fractions as shown in the profile analysis of the UV_{254} and TOC. As aromatic hydrocarbon had a high response to UV treatment, this kind of organic matter was transformed into organics with simple structures and its intensity of UV_{254} response decreased. This is why the TOC profile is opposite to that of the UV_{254}.

As shown in Fig. 5(a), the UV_{254} in the effluent through the UV–MBR system had a sharp decrease and the hydrophilic and hydrophobic fractions were the major removal components with reductions of 48 and 42%, respectively. It has been reported that the hydrophilic fraction in natural raw water consists of polysaccharide and protein, and other small molecular-weight organics [28]. Substances such as polysaccharide and protein can be changed by UV treatment, or removed by membranes [20]. In Fig. 5(b), the hydrophilic fraction increased a little bit after the UV treatment. It was inferred that some complex organics were changed to small molecular-weight hydrophilic fractions with high biodegradability, which resulted in higher removal efficiency of this fraction by the MBR. The UV_{254} and TOC level of hydrophobic fraction both decreased by the UV treatment. For slightly hydrophobic fractions, the UV_{254} decreased through UV pre-treatment but it did not drop down further in the subsequent MBR system. Compared with the UV_{254}, the TOC of slightly hydrophobic fractions changed a little bit during the process. However, the removal performance of slightly hydrophobic fraction was poor through both the UV treatment and subsequent biodegradation.

3.2.3. The fluorescence characteristics

In natural water, humic substances and biological metabolites are the main components which have responses to fluorescence analysis [29]. Chen has reported that four regions in the EEM could be marked. Regions 1 (EM/EX: 280–380/200–250) and region 4 (EM/EX: 280–380/250–340) are associated with aromatic protein and soluble microbial products, respectively. The other two are associated with humic

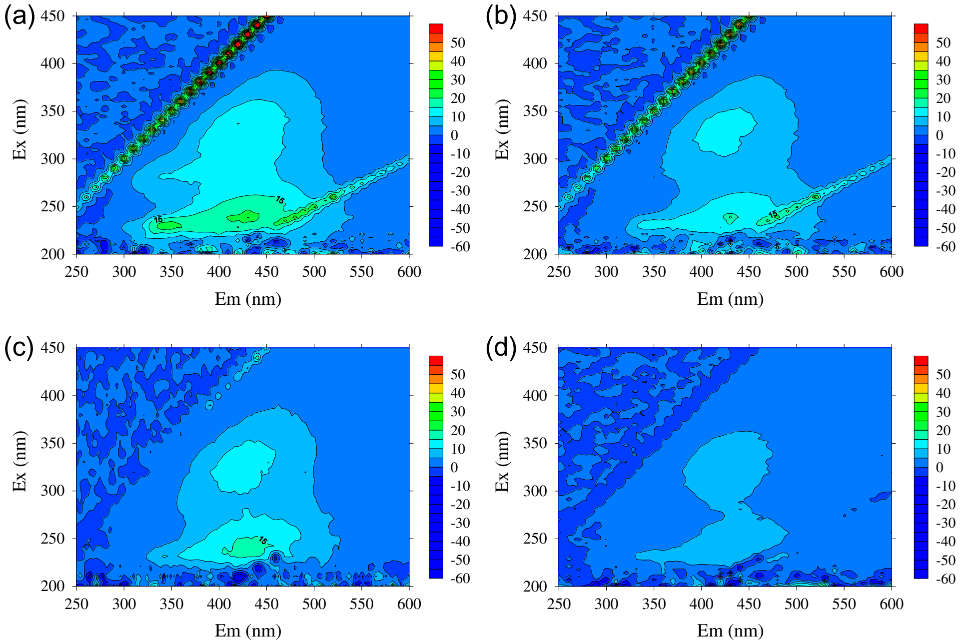

Fig. 6. EEMs fluorescence spectra of (a) raw water, (b) water samples by UV treatment, (c) water samples by MBR, and (d) water samples by UV–MBR.

substances (EM/EX: 380–480/250–400) and fulvic-like substances (EM/EX: 380–480/200–250) [30].

In order to better understand the fluorescence characteristics variation in water samples during the process, the EEMs of the raw water and UV–MBR effluent are shown in Fig. 6. Three regions can be observed in the raw water in Fig. 6(a): aromatic protein, humic-like and fulvic-like regions. These components were investigated by Elliott et al. [31] as well and the humic-like and fulvic-like intensity was higher. It was found that the intensity of the three regions decreased compared with that in the raw water. The protein peak had a significant reduction, which was attributed to the retention of large molecules by the membrane (Fig. 6(c)). As shown in Fig. 6(d), the peak areas of the three regions were lower than those in Fig. 6(a) and (c). This showed that the pollutant removal performance of the UV–MBR was superior to that of the MBR only.

To illustrate the organics removal mechanism during the UV–MBR treatment, the water sample fluorescence characteristics after the UV treatment were analyzed as well (Fig. 6(b)). Although the three regions' fluorescence intensity decreased, the organics were not removed from the raw water. The lower fluorescence intensity was attributed to the variation of the organic structure [32]. Through the UV treatment, part of humic substances was changed to simple structure organics with low response to fluorescence which provided more nutrients for micro-organisms in subsequent treatment [23].

4. Conclusions

The COD_{Mn} and UV_{254} removal efficiencies were enhanced significantly by the UV–MBR and nearly most of ammonia nitrogen and nitrite nitrogen were removed because of superior biodegradation. UV treatment leads to the increase of BDOC content. It was indicated that UV can change the organic matter characteristics according to the HPSEC and fluorescence spectra analysis. The refractory organics were split into simple structures which can be easily biodegraded by micro-organism. From the hydrophobic and hydrophilic fraction distribution, the hydrophilic fraction increased after the UV treatment and it is the principle substrate for heterotrophic bacteria.

Acknowledgments

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