AQUA
Water Infrastructure, Ecosystems and Society
Corrected Proof
Check for updates

AQUA — Water Infrastructure, Ecosystems and Society Vol 00 No 0, 1 doi: 10.2166/aqua.2023.191

A pilot study on the dual membrane process of ultrafiltration and nanofiltration to treat algae-laden raw water compared to O_3-BAC

Xingqi Ding[a], Zhenxun Yu[a,*], Huaqiang Chu[b], Dong Wang[c], Kunqiao Liu[d], Buyun Wang[a], Caihuan Tong[e] and Bingzhi Dong[b]

[a] School of Civil and Environmental Engineering, Ningbo University, Ningbo 315211, China
[b] State Key Laboratory of Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Tongji University, Shanghai 200092, China
[c] Ningbo Tianhe Aquatic Ecosystems Engineering Co. Ltd, Ningbo 315012, China
[d] CCDI (Suzhou) Exploration & Design Consultant Co. Ltd, Suzhou 215127, China
[e] Centre Testing International (Ningbo) Co. Ltd, Ningbo 315048, China
*Corresponding author. E-mail: yuzhenxun@nbu.edu.cn

ABSTRACT

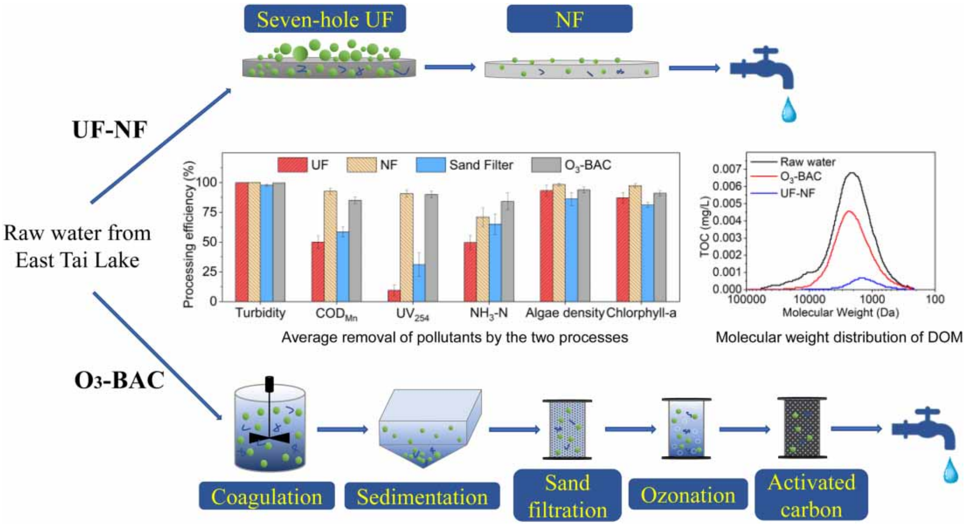
In this study, a dual membrane process that combines seven-hole ultrafiltration and nanofiltration was designed and compared to O_3-biological activated carbon (O_3-BAC) for high-quality drinking water production. The pollutant removal, membrane fouling, long-term operational characteristics, and technical economy were systematically investigated using raw water from Tai Lake, which has a high algae content. The results elucidate that the dual membrane process has superb decontamination. This method has much better removal of turbidity, COD_{Mn}, UV_{254}, and algae than O_3-BAC. Its removal of ammonia nitrogen and fluorescent substances is slightly lower than that of O_3-BAC, but the effluent still satisfies the drinking water standard. The dual membrane process is also much more capable of dealing with high algae-laden raw water. Compared to O_3-BAC, the cost of the dual membrane process is 46.4% higher per ton without consuming chemicals, so it is more environmentally friendly. In summary, the dual membrane process offers a promising and effective technology to treat high algae-laden water with the advantages of high stability, reliable effluent, and zero emissions.

Key words: high algae-laden water, membrane fouling, pilot study, technoeconomic analysis, the dual membrane process

HIGHLIGHTS

- A pilot study on the dual membrane process of ultrafiltration and nanofiltration was conducted compared to O_3-biological activated carbon (O_3-BAC).
- The dual membrane process can remove algae effectively without breaking them.
- Providing a reference for large water plants to produce high-quality drinking water.
- The dual membrane process has high economic and technical feasibility.

GRAPHICAL ABSTRACT



1. INTRODUCTION

The frequent outbreaks of algal blooms due to eutrophication in drinking water resources are a grave threat to the water supply (Maeng *et al.* 2021; Xu *et al.* 2021). Treating high algae-laden water is a problem in drinking water treatment in China. Raw water with an algal density of 1 million algae per litre or more is defined as high algae-laden water (Chen *et al.* 2009). During algal blooming, the increase in algae cells may penetrate the filter tank into the water supply network. It will increase assimilable organic matter, promote microbial growth, and reduce the microbiological safety of drinking water (Zhu *et al.* 2022). In addition, algae can release dissolved organic matter (DOM) during their metabolism. This process will produce disinfection by-products with potential carcinogenic effects during chlorine disinfection (Fang *et al.* 2010; Xiao *et al.* 2020; Du *et al.* 2022; Liu *et al.* 2022; Pandian *et al.* 2022). As a source of freshwater in China, East Tai Lake has received widespread attention. Its pH is stable, but the turbidity, organic matter, ammonia nitrogen, and algae vary with the seasons. To be specific, the pH of the raw water in Lake Taihu was relatively stable in the range of 7.76–8.2. On the contrary, the difference between the lowest and highest turbidity is nearly 100 NTU, COD_{Mn} fluctuates in the range of 3.47–4.74 mg/L and the concentration of ammonia nitrogen varies from 0.0645 to 0.2382 mg/L. The number of algae is low in winter (96.32×10^4 per litre) and increases rapidly in summer, reaching a maximum of 573.44×10^4 per litre. The average density of algae in Taihu Lake can reach 241.83×10^4 per litre. The density of algae in reservoirs as a source of drinking water is generally 1×10^4 per litre. The density of algae in Taihu Lake water is 200 times higher, which satisfies the characteristics of high algae-laden water.

Treating high algae-laden water is a great challenge for large water supply plants. In general, traditional treatment methods for high algae-laden water can be divided into physical, chemical, and biological methods (Su *et al.* 2016; Pompei *et al.* 2022; Ren *et al.* 2022). Physical methods are simple and environmentally friendly but inefficient. Chemical methods are cheap and convenient, but they may cause secondary pollution and cause microorganisms to generate drug resistance. The advantages of biological methods are environmental friendliness and minimal sludge production, but the disadvantages are long-running periods. In addition, pre-oxidation-enhanced coagulation is widely used in drinking water treatment. The main principle is to deactivate and remove algae cells by coagulation and sedimentation (Lin *et al.* 2021), but cell deactivation is difficult to control. Traditional pre-oxidation or ultrasound and ultraviolet treatment may damage algal cells, so this method has certain limits. The ozone-biological activated carbon process (O_3-BAC) has been applied widely in drinking water treatment. It can remove a proportion of the hydrophobic organic matter. But it may produce ozone by-products that have an impact on drinking water. Therefore, there is still a need to develop a better drinking water treatment (Liu *et al.* 2021; Sun & Chu 2021).

As an emerging water treatment technology, membranes are receiving increasing attention. The dual membrane process is used in desalination and decontamination (Touati *et al.* 2018; Ağtaş *et al.* 2020; Kamp *et al.* 2021) due to its high efficiency, good stability, small footprint, and easy management (Wang *et al.* 2020). The first step in the dual membrane process is usually microfiltration or ultrafiltration (UF), which is used primarily to remove larger particulate matter (Sangrola *et al.*

2020). Then, nanofiltration (NF) or reverse osmosis (RO) is used as a second step to effectively remove various contaminants (Guo *et al.* 2019), including DOM (Ersan *et al.* 2016) and heavy metals (Zhang *et al.* 2016). The dual membrane process has been applied extensively in desalination and decontamination, but it is rarely, if ever, designed as the core part of drinking water treatment, and there is minimal research related to the dual membrane process. It is noteworthy that RO basically removes minerals that are good for human health. So, this study uses a combination of UF and NF membranes to remove algae, to ensure substances that are beneficial to people are not completely removed from the water. Theoretically, the membrane removes pollutants mainly relying on size exclusion, and it can completely remove algae cells without breaking them (Cheng *et al.* 2021). Therefore, the membrane is suitable for treating high algae-laden water. Moreover, UF–NF could not only remove the target contaminants but also retain the minerals beneficial to human health in drinking water. Thus, UF–NF is considered an effective method to prepare high-quality drinking water. Nevertheless, compared with O_3-BAC, the application experience of a full-scale dual membrane process has not been well established. As a result, a pilot study on the dual membrane process is an important research subject that merits further investigation of the dual membrane process efficiency and membrane fouling in treating high algae-laden water.

This study explores the pollutant removal capacity and membrane fouling of the dual membrane process consisting of UF and NF to treat high algae-laden water. A pilot-scale experiment was built at the No. 1 Water Treatment Plant in Wujiang District, Suzhou, China. To better explain the feasibility of the dual membrane process, an O_3-BAC (the drinking water treatment used at the plant) was set up for comparison. The feasibility of the dual membrane process for treating high algae-laden water was explained in terms of pollutant removal efficiency, stability, and technical economy. Furthermore, this pilot study aims to provide a reference for large water plants to produce high-quality drinking water.

2. MATERIALS AND METHODS

2.1. Raw water

A pilot-scale experiment was set up at the first Water Treatment Plant in Wujiang District, Suzhou, China. The raw water was taken from East Tai Lake, which is the main source of drinking water in Suzhou, Jiangsu Province. The features of the raw water quality are summarized in Table 1.

2.2. Pilot experimental setup

UF–NF and O_3-BAC were simultaneously carried out in water plants. The membranes in the dual membrane process are UF and NF. Ultrafiltration (INGE, Germany) is a seven-hole UF membrane with an average molecular-weight cut-off (MWCO) of 150,000 Da. The hydrophilic UF membrane is made of polyethersulfone (PES). The UF that contains seven membranes with a total effective filtration area of 6.5 m^2 adopts internal dead-end filtration with a constant flow, which is shown in Figure 1. The hydrophilic nanofiltration membrane (Toray, Japan) with an average MWCO of 200 Da is made of polyamide (PA). The spiral-wound NF membrane that contains two NF membranes with a total effective filtration area of 8 m^2 adopts cross-flow filtration with a constant flow.

O_3-BAC mainly includes coagulation, sedimentation, sand filtration, ozonation, and activated carbon. Poly aluminium chloride (PAC) was added to a mechanical agitation tank with an effective area of 0.3 m^3. Sedimentation adopts a sloping pipe settling tank with an effective area of 2 m^3. The sand filter column is 200 mm in diameter and 2 m in height and uses quartz sand filler. The diameter of the ozone contact column is 200 mm, and the height is 2.5 m. The dosage of ozone is 1.5 mg/L. The diameter of the activated carbon column is 400 mm, and the height is 2.5 m. The height of the filler is 1.5 m.

Table 1 | Characteristics of the raw water

Water quality indicator	Values	Water quality indicator	Values
pH	7.38–8.88	Algae density, million/L	0.85–3.75
Turbidity, NTU	15.1–412	Chlorophyll-a, μg/L	1.77–7.07
Conductivity, μS/cm	172.39–262.44	COD_{Mn}, mg/L	2.25–5.75
TOC, mg/L	2.22–3.59	NH_3-N, mg/L	0.07–0.38
UV_{254}, cm^{-1}	0.051–0.087	TDS, mg/L	360–543

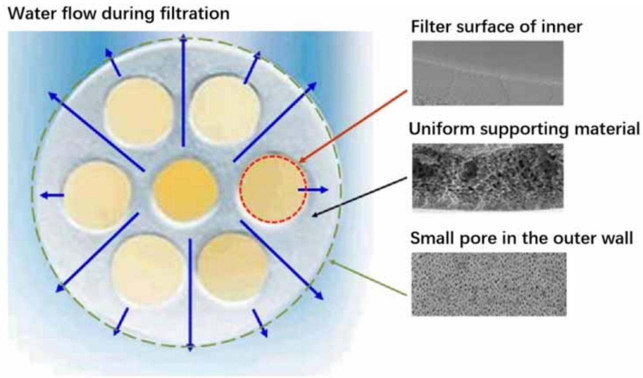


Figure 1 | Electron microscopy of the UF membrane.

2.3. Operating conditions

Figure 2 shows the flow chart of the dual membrane process of UF–NF and the O_3-BAC. The water from East Tai Lake was filtered by a 200-μm disc filter to remove floating matter and was sent to the feed tank. Then, the effluent was pumped into the UF with a flux of 325 L/h. The UF with a total effective filtration area of 6.5 m^2 used an internal dead-end filtration with 50-L/(m^2·h) flux. The automatic backwash was periodically performed every 30 min for 1 min with the effluent of UF and air. When the trans-membrane pressure (TMP) reached 1,500 Mbar or every 30 h, chemically enhanced backwash (CEB)

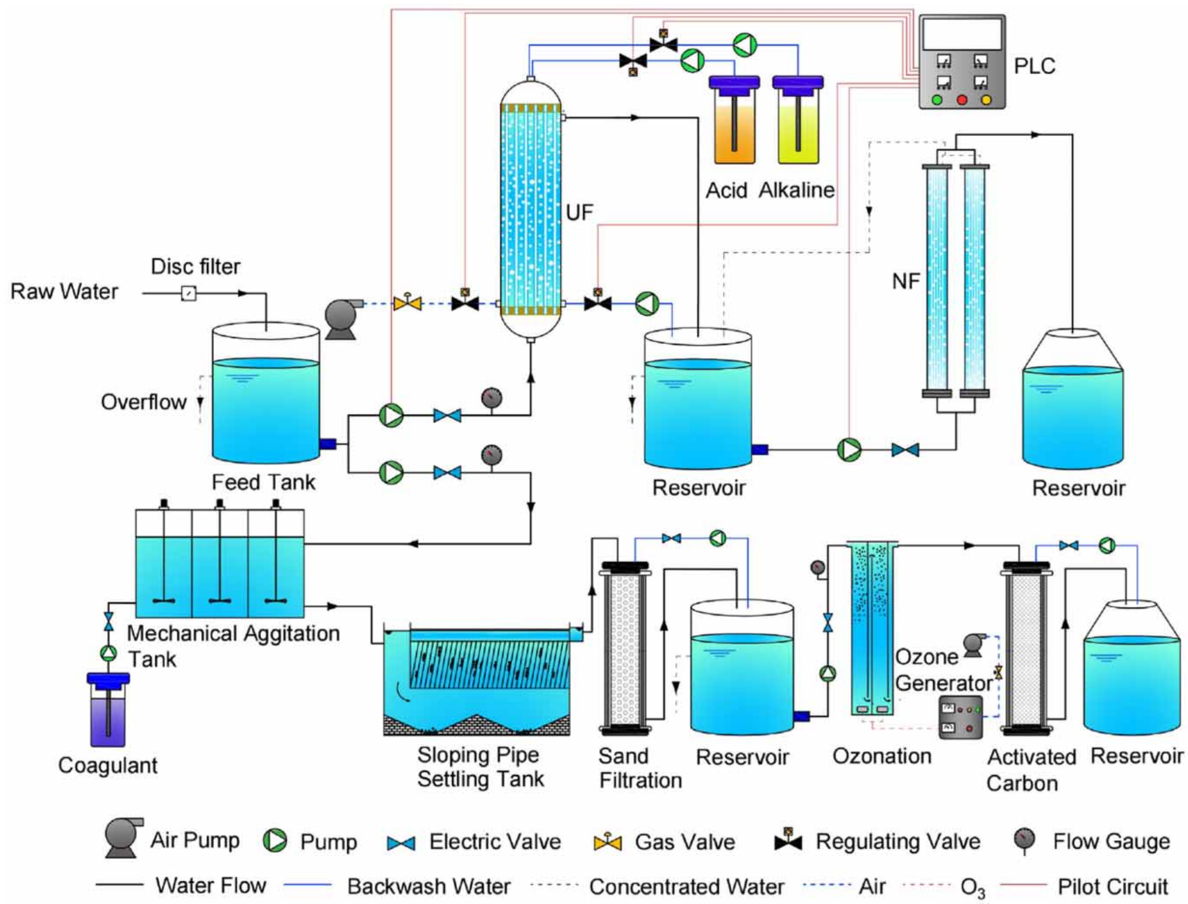


Figure 2 | Schematic diagram of UF–NF and O_3-BAC.

was automatically performed. CEB contains acid pickling and caustic scrubbing. NaOH and NaClO were added to remove organic matter, and HNO_3 was added to remove inorganic matter. Then, the effluent of UF was pumped into the NF. The NF was controlled by a programmable logic controller (PLC) for complete automation. NF was used to mitigate membrane fouling by manually dosing 100 mL of NaClO (recorded as active chlorine) at a concentration of 200 ppm per week.

For O_3-BAC, the raw water from East Tai Lake was sent to a feed tank. Then, the effluent was pumped into a mechanical agitation tank. The coagulant was added to the mechanical agitation tank using a peristaltic pump with a dosing rate of 20 mg/L. The water subsequently flowed to the sloping pipe settling tank with an effective area of 2 m^3. After 1.5 h of precipitation, the supernatant was sent to a sand filtration column with a flux of 0.002 m/s. Then, the effluent was pumped into an ozone contact column with a reaction time of 15 min. Finally, the water entered the activated carbon column with a flux of 0.002 m/s. The contact time in the empty bed of the activated carbon column is 12 min. The backwash of the sand filtration and activated carbon was periodically performed with the effluent of the reservoir.

2.4. Analytical methods

An online conductivity detector (CR 300) was used to measure the total dissolved solids (TDS) and conductivity. Turbidity was measured with a portable turbidity meter (HACH 2100P, USA), and pH was measured with a pH meter (PHS-25, Shanghai). Chemical oxygen demand (COD_{Mn}) and ammonia nitrogen (NH_3-N) were determined by the methods of the National Environmental Protection Standard of the People's Republic of China (GB 11892-89 and HJ 535-2009). Three-dimensional excitation-emission matrix (3D-EEM) spectra were investigated on a 3D fluorescence spectrophotometer (VARIAN Cary Eclipse, USA). The UV_{254} absorption spectra were obtained by a DR 6000 UV spectrophotometer (Hash, USA). Total organic carbon (TOC) was measured using a TOC analyser (TOC-VCPH, Japan), and algae were determined by a water quality analyser (YSIEXO2, USA). The molecular-weight (MW) distribution was determined using a high-performance liquid chromatography (Waters 2695, USA) system with a detector (Sievers 900).

3. RESULTS AND DISCUSSION

3.1. Trans-membrane pressure

TMP is an important indication to examine the stability of the membrane and characterize membrane fouling. Figure 3(a) shows that the TMP was proportional to filtration time. After chemical cleaning, the TMP was restored, but it slightly increased during filtration. This is because of the blockage of membrane pore, formation of the cake layer, and rising temperature (Yang *et al.* 2022). Figure 3(a) and Figure 4 demonstrate that increase in temperature and flux led to an increase in the TMP (Al-Amri *et al.* 2010). The UF membrane still upheld stability, which indicates that effective chemical cleaning significantly reduces membrane fouling and ensures the stability of subsequent NF (Cai & Liu 2016; Tang *et al.* 2019).

The flux of the membrane is an essential factor that affects the treatment effectiveness, stability, energy consumption, and membrane fouling. The constant permeate fluxes of 45 and 60 $L \cdot m^{-2} \cdot h^{-1}$ (LMH) were, respectively, employed to assess the operation under different fluxes. Figure 3(b) shows that the TMP of NF displayed a linear increasing trend with extending

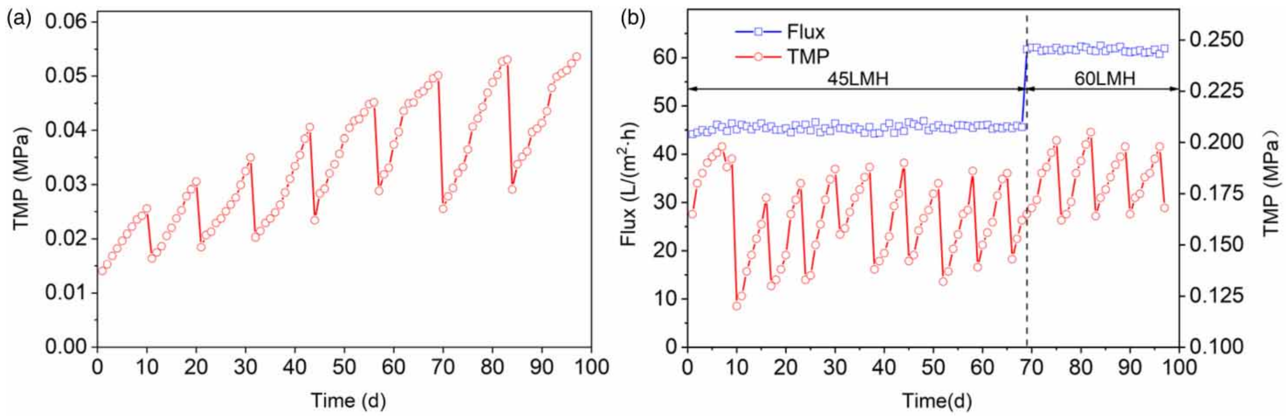


Figure 3 | The changes in membrane pressure difference between (a) UF and (b) NF.

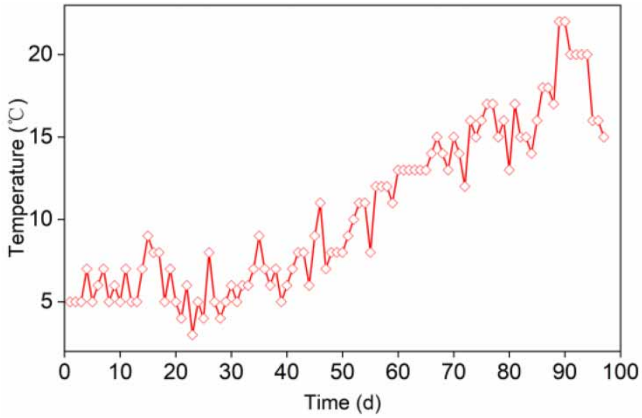


Figure 4 | The changes in temperature in the pilot experiment.

filtration. Besides, the TMP of NF changes from 0.13–0.198 to 0.162–0.201 MPa when the flux increases from 45 to 60 LMH. This is attributed to the fact that NF is more susceptible to contamination at a higher permeate flux. To avoid microbial exposure, a fungicide (HS-301 RO membrane specific fungicide containing DBNPA (2,2-dibromo-3-nitroso-propionamide)) was applied by shock dosing. DBNPA is a broad-spectrum industrial fungicide that rapidly penetrates the cell membranes of microorganisms and causes cell death by suspending normal redox in the cells (Da-Silva-Correa *et al.* 2022). The fungicide was added at 100 mg/L every 7 days for 30 min. To prevent the effects of residual fungicide and its decomposition products, the effluent of NF was completely discharged during sterilization (Liu *et al.* 2021). The pressure significantly decreases due to the fungicide because the fungicide can inhibit the growth of microorganisms (Yu *et al.* 2012; Wang *et al.* 2018a, 2018b). As a result, UF as a pretreatment can maintain the long-term stability of NF.

3.2. Improvements in pollutant removal

3.2.1. Turbidity removal

Turbidity is a parameter that reflects suspended matter in water. Moreover, germs, viruses, and other toxic matter tend to attach to suspended matter (Medema & Schijven 2001). Therefore, it is important to maintain the turbidity of water within certain limits. The removal of turbidity by the dual membrane process and O_3-BAC is shown in Figure 5. The average effluent of UF and NF was 0.067 ± 0.020 NTU and 0.035 ± 0.011 NTU, respectively. The removal of turbidity by UF and NF was stable at 99.9 ± 0.05% and 99.94 ± 0.03%, respectively. The removal of turbidity by sand filtration and O_3-BAC was 97.72 ± 0.88% and 99.65 ± 0.12%, respectively, with an average effluent of 1.432 ± 0.72 NTU and 0.22 ± 0.10 NTU,

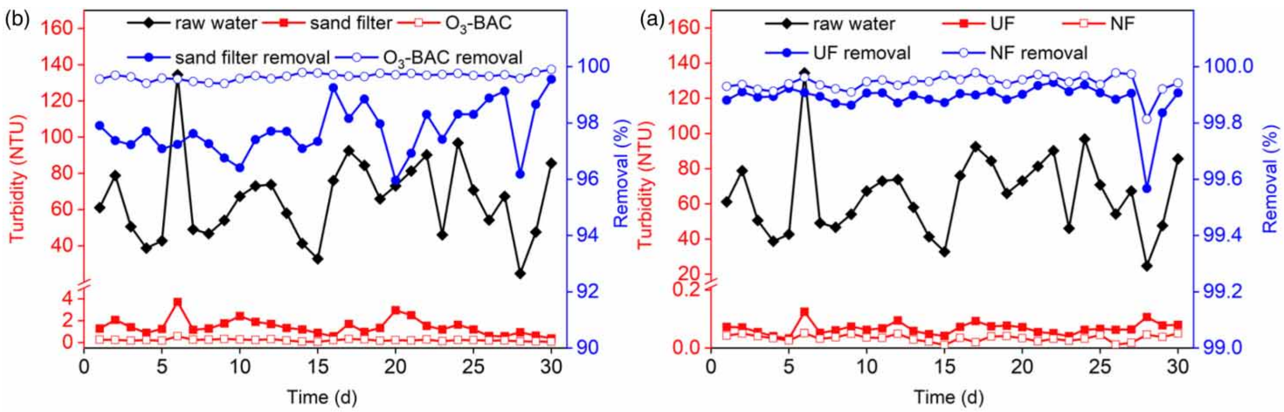


Figure 5 | The removal effect of turbidity by (a) the dual membrane process and (b) O_3-BAC.

respectively. The dual membrane process has a slightly better effect than the O_3-BAC. The effluent of the dual membrane process is stable.

The removal of turbidity by the dual membrane process is less affected by raw water and performing conditions, and the removal is extremely high. In contrast, the removal of turbidity by O_3-BAC is largely influenced by raw water. Therefore, the dual membrane process is much more effective than O_3-BAC in treating high-turbidity water. In addition, most of the suspended matter is removed by UF mainly because the size of the suspended matter is larger than the pores of UF. UF retention supplies effluent quality, guarantees biological safety, and ensures subsequent stable operation.

3.2.2. Organic matter removal

The COD_{Mn} and UV_{254} are parameters that reflect the total organic matter. Organic matter is always divided into particulate and dissolved matter. The most important factor affecting drinking water treatment is DOM. Figure 6 describes the removal of COD_{Mn} by the dual membrane process and O_3-BAC. The average effluent of COD_{Mn} from the UF was 2.02 ± 0.13 mg/L with an average removal of 50.07 ± 5.26%. The effluent from NF was 0.29 ± 0.10 mg/L with removal of 92.78 ± 2.55%. The effluent from the sand filtration was 1.68 ± 0.19 mg/L with removal of 58.56 ± 4.23%. The effluent from O_3-BAC was 0.6 ± 0.13 mg/L with a removal of 85.19 ± 2.77%. Then, as shown in Figure 7, the average effluent UV_{254} from the UF was 0.0511 ± 0.004 cm^{-1} with an average removal of 9.55 ± 4.61%. The effluent from NF was 0.0052 ± 0.002 cm^{-1} with removal of 90.82 ± 3.06%. The effluent from the sand filtration was 0.039 ± 0.006 cm^{-1} with removal of 31.01 ± 10.01%. The effluent from O_3-BAC was 0.0056 ± 0.002 cm^{-1} with removal of 90.07 ± 2.99%. The quality of raw water greatly impacts the effluent of O_3-BAC, while the effluent of the dual membrane process is relatively stable.

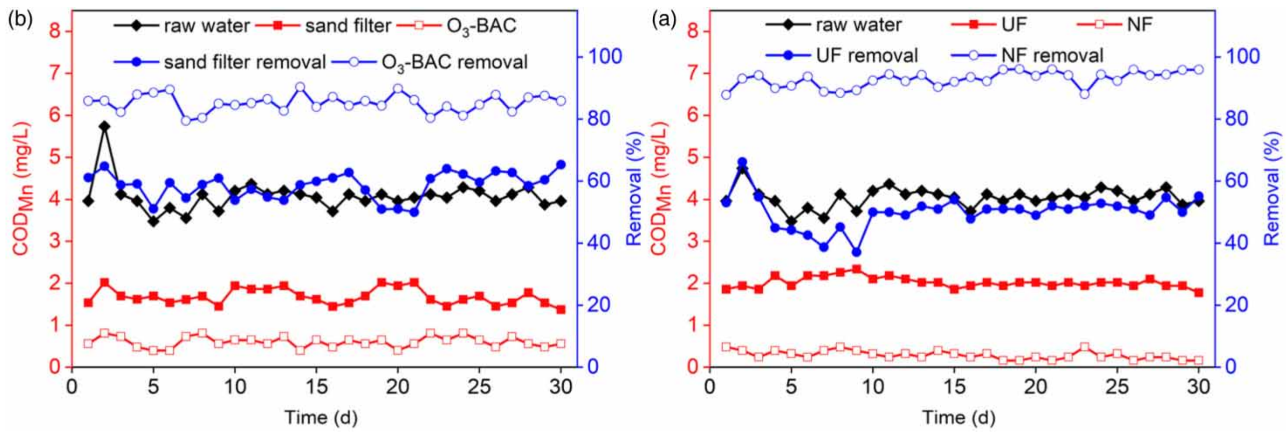


Figure 6 | The removal effect of COD_{Mn} by (a) the dual membrane process and (b) O_3-BAC.

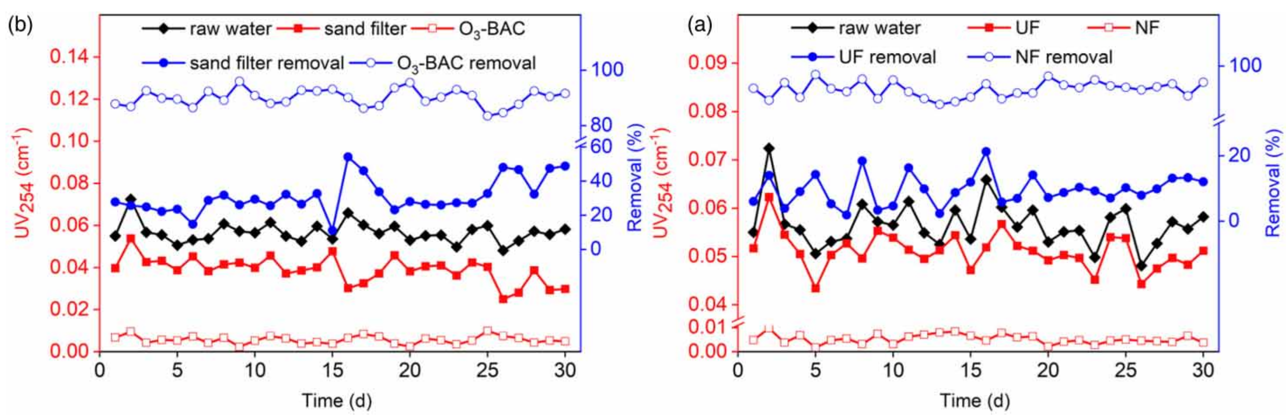


Figure 7 | The removal effect of UV_{254} by (a) the dual membrane process and (b) O_3-BAC.

These results show that O_3-BAC can effectively remove organic matter mainly due to a biofilm attached to the surface of the sand filter after a long period (Guo *et al.* 2018). Rejecting DOMs by the UF membranes remains difficult, since the particle of dissolved organics is much smaller than the pores of the UF membrane (Guo *et al.* 2018). The NF membrane achieves high retention of DOM. For rejection, NF can retain all organic matter with an MW of 200–1,000 Da. Moreover, ion adsorption and charged groups of the NF membrane can fundamentally affect the removal of DOM (Castaño Osorio *et al.* 2022). Humic substances and other hydrophobic organic matter in water are usually negatively charged, while the NF membrane is also negatively charged under neutral pH conditions. Therefore, NF can seize negative ions or negatively charged organic matter (Verliefde *et al.* 2008). In addition, the NF in the experiment is a hydrophilic membrane that plays a critical role in the removal of hydrophobic substances (Nghiem *et al.* 2005). As a result, the NF membrane directly compensates for the UF membrane, which ensures that the system is effective in removing organic matter.

3.2.3. Ammonia nitrogen removal

As shown in Figure 8, the removal of ammonia nitrogen by UF and sand filtration was 49.67 ± 6.01% and 65.06 ± 8.65%, respectively. The removal of ammonia nitrogen by the dual membrane process and O_3-BAC was 71.00 ± 7.88% and 84.21 ± 7.06%, respectively. The effect of O_3-BAC was slightly better than that of the dual membrane process, mainly because the biodegradation of sand filtration has an obvious removal capacity for ammonia nitrogen (Guo *et al.* 2018). Theoretically, UF has no removal ability of ammonia nitrogen because ammonia nitrogen is much smaller than the pore size of UF (Wang *et al.* 2018a, 2018b; Maeng *et al.* 2021). However, in this study, UF was effective in the removal of ammonia nitrogen, probably due to the adsorption of microorganisms (Guo *et al.* 2018). Biofilms gradually developed on the UF membrane surface because of the accumulation of microorganisms. Therefore, O_3-BAC is more effective than the dual membrane process, but the effluent from O_3-BAC is influenced by raw water, whereas the dual membrane process is stable.

3.2.4. Algae removal

In this experiment, algal density and chlorophyll-a were used as parameters to characterize algae. As shown in Figures 9 and 10, the average removal of algal density by UF and sand filtration was 93.25 ± 4.56% and 86.19 ± 5.28%, respectively. The average removal by the dual membrane process and O_3-BAC was 98.31 ± 1.24% and 93.89 ± 2.62%, respectively. The average removal of chlorophyll-a by UF and sand filtration was 87.24 ± 4.41% and 81.17 ± 2.07%, respectively. The average removal by the dual membrane process and O_3-BAC was 97.34 ± 1.72% and 91.09 ± 2.21%, respectively. Thus, the removal effects of the two processes on algal density and chlorophyll-a were excellent, and the dual membrane process had a slightly better effect than O_3-BAC. Clearly, the two parameters are correlated, since chlorophyll-a is generally found in chloroplasts within algal cells.

As shown in the figure in the chart, O_3-BAC is influenced by raw water, whereas the dual membrane process is stable. In O_3-BAC, stirring paddles and oxidation prefer to disintegrate algal cells (Zamyadi *et al.* 2013), releasing phycotoxins and odour during algal blooming. The secretion from algae can adhere to the surface of filter media and undermine the adsorption capacity and biological effect (Yu *et al.* 2017), which affects the security of water quality. Additionally, the coagulation and

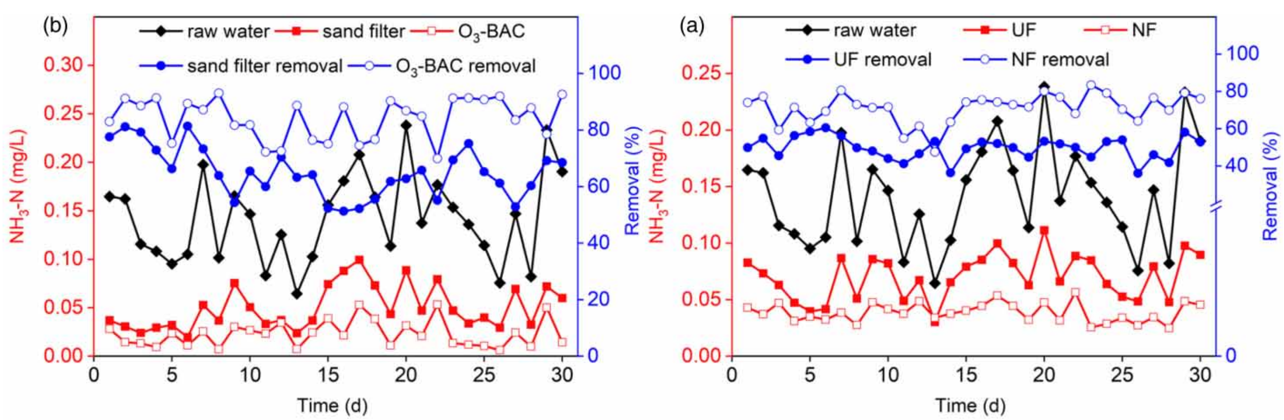


Figure 8 | The removal effect of ammonia nitrogen by (a) the dual membrane process and (b) O_3-BAC.

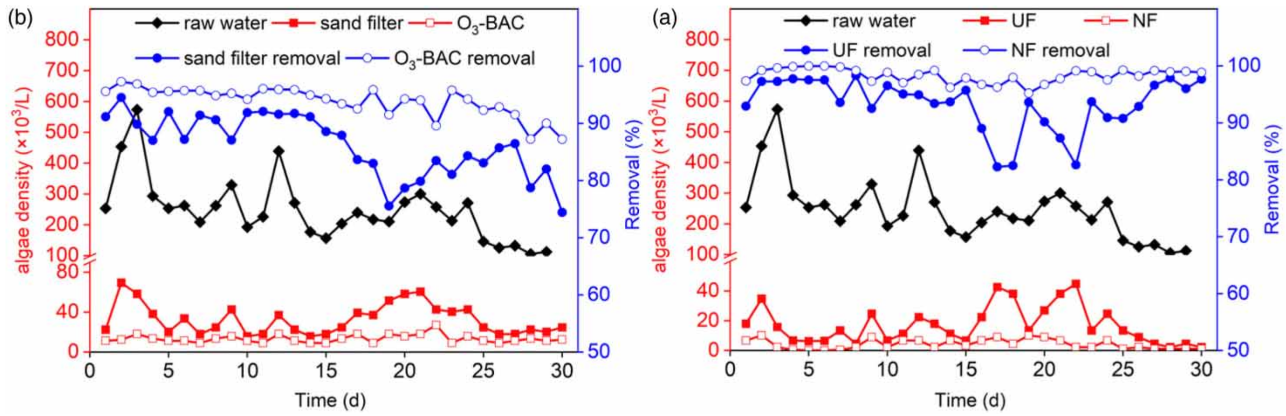


Figure 9 | The removal effect of algal density by (a) the dual membrane process and (b) O_3-BAC.

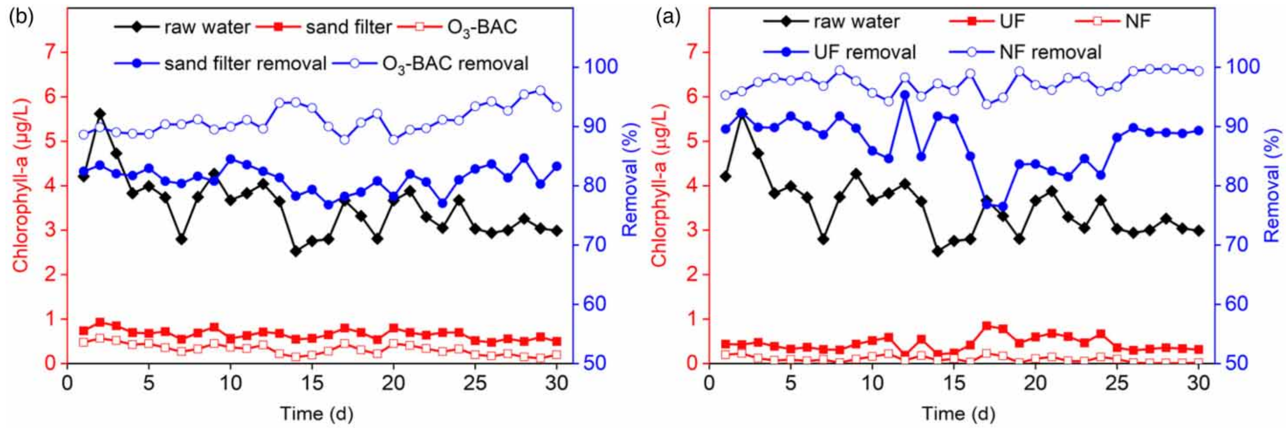


Figure 10 | The removal effect of chlorophyll-a by (a) the dual membrane process and (b) O_3-BAC.

sedimentation in the O_3-BAC are ineffective in removing algae, so most algae are retained and subsequently enter the sand filter (Henderson *et al.* 2008). The algae can adhere to the surface of the sand filter and decrease the removal effect. The flux cannot be restored with periodic backwashing and even increase the frequency of backwashing, which will increase the long-term operational burden. The reason why the removal rate of chlorophyll-a in O_3-BAC has been rising steadily is due to microbial action and appropriate temperature. Biofilms gradually developed due to the accumulation of microorganisms. Besides, the range of optimum temperature for microbial growth is 16–30 °C. During the experiment, the temperature rose steadily and gradually reached a temperature suitable for microbial growth, which was beneficial to microbial action.

In contrast, most algae are removed by the UF membrane, and the removal rate is up to 90%. The diameter of algae is larger than the pore diameter of the UF membrane (Chiou *et al.* 2010; Cheng *et al.* 2018), so it can be completely rejected by the UF membrane. It is noteworthy that the removal effect of UF in the graph suddenly decreased. This may be because UF is not stable enough as a pretreatment. But it also could reduce the burden of the NF membrane and does not affect the effluent quality of the dual membrane process.

3.3. Mechanism of two processes for pollutant removal

3.3.1. Analysis of 3D excitation–emission matrix fluorescence spectra

A three-dimensional excitation-emission matrix (3D-EEM) was utilized in this experiment to provide information about chemical components of DOM in the water (Liu *et al.* 2011). Five fluorescence regions were classified using fluorescence regional integration (FRI) methods and were represented by I, II, III, IV, and V (Chen *et al.* 2003), including tryptophan-like proteins (Ex: 220–250 nm, Em: 280–330 nm); tyrosine-like proteins (Ex: 220–250 nm, Em: 330–380 nm); fulvic acid-like substances (Ex: 220–250 nm, Em: 380–500 nm); soluble microbial product substances (Ex: 250–280 nm, Em: 280–380 nm); and humic acid-like substances (Ex: 250–400 nm, Em: 380–500 nm), respectively.

As shown in Figure 11(a), four obvious fluorescent peaks are observed in the spectrum of raw water. Peak B (Ex = 230 nm, Em = 340 nm) and peak T (Ex = 280 nm, Em = 310 nm) respond strongly, and they represent protein and microbial product substances, respectively. Peak A and peak C are attributed to fulvic acid-like and humic acid-like substances, and they respond slightly. The organic matter of Tai Lake is mainly composed of protein and microbial product substances. The removal of the dual membrane process for regions I–V was 82.15, 81.88, 75.73, 78.49, and 78.66%, respectively, with an average removal rate of 79.38%. The removal of O_3-BAC for regions I–V was 85.46, 89.04, 88.00, 86.72, and 91.13%, respectively, with an average removal rate of 88.07%. Thus, the dual membrane process is slightly inferior to O_3-BAC for the removal of fluorescent substances.

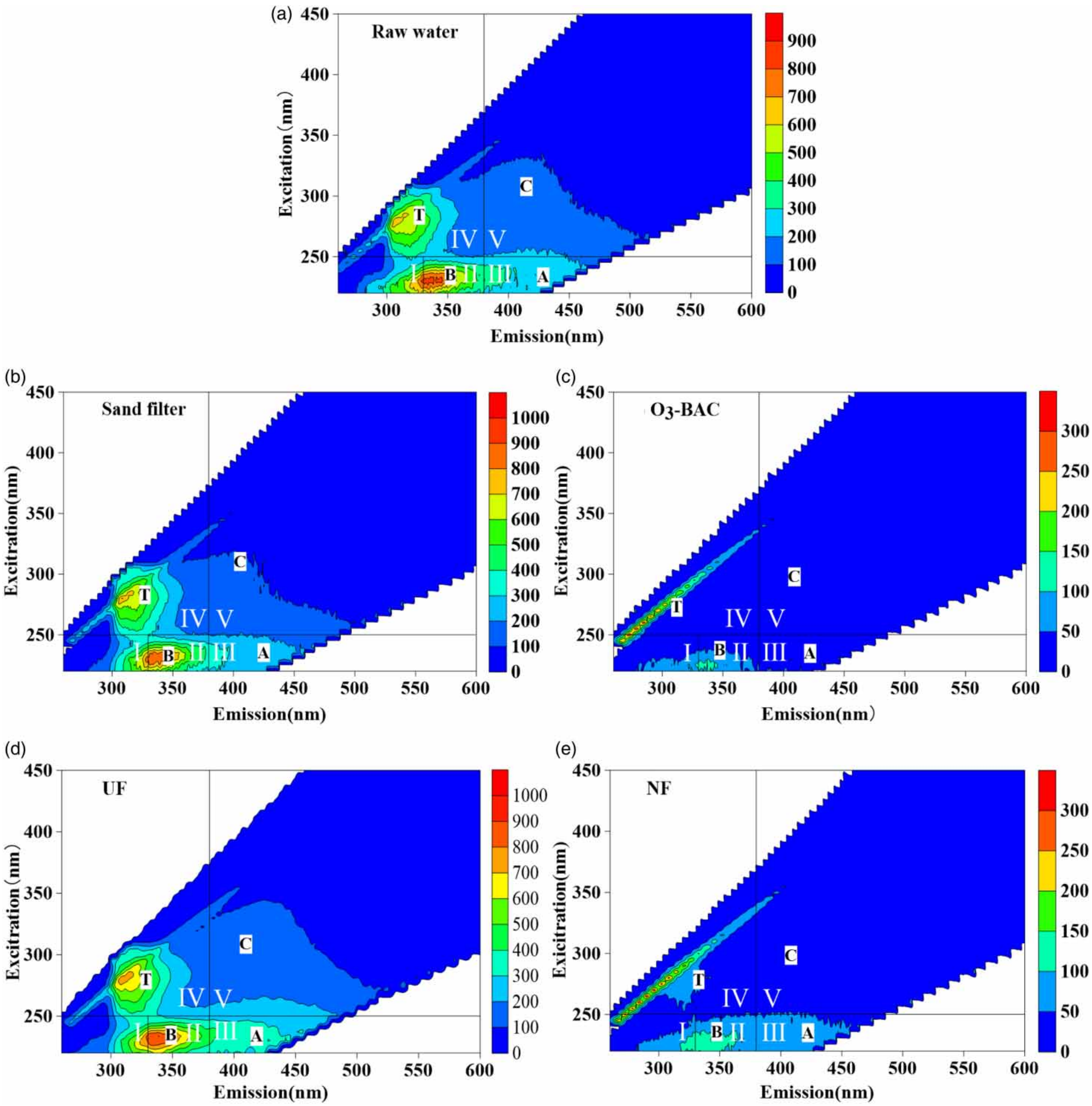


Figure 11 | 3D-EEM spectra of water samples, including (a) raw water, and effluent from (b) sand filter, (c) O_3-BAC, (d) UF, and (e) NF.

Figure 12 shows that sand filtration and UF are less effective at removing fluorescent substances, but O_3-BAC and NF are more effective. O_3-BAC greatly affects the removal of fulvic and humic acid-like substances. Ozone can oxidize organic matter such as humic acid (Rivas *et al.* 2009). As a strong oxidant, it can effectively destroy the structure of organic pollutants and degrade hydrophobic aromatic humic substances (Chen *et al.* 2017). A high-MW protein substance is mainly removed by the UF membrane, while most low-MW pollutants will pass through the membrane unhindered. The NF membrane can remove an amount of low-MW organic matter relying on size exclusion, adsorption of surface charge, and hydrophilicity (Li *et al.* 2021). However, a portion of the DOM will still pass through the membrane unhindered.

3.3.2. Characterization of the MW

The chromatograms of MW are shown in Figure 13. Raw water includes a large proportion of medium-MW organic matter. The results indicate that high-MW (MW > 10 kDa) and low-MW (MW < 1 kDa) organic matter are effectively removed by the dual membrane process and O_3-BAC, and the difference between the two processes was not obvious. In regard to medium MW (1 kDa < MW < 10 kDa), the response of effluent from O_3-BAC to TOC is strong, while the response to UV is mild. However, the responses of effluent from the dual membrane process to TOC and UV_{254} are both mild. This result suggests that the effort of the dual membrane process is slightly better than that of O_3-BAC.

According to the above analysis results, O_3-BAC plays a critical role in the removal of medium- and high-MW organics mainly because of the adsorption of sand filtration and the degradation of biofilm attached to the surface of sand filtration (Guo *et al.* 2018). Additionally, ozone can oxidize medium-MW organics to low-MW organics and subsequently remove them by adsorption of activated carbon (Wei *et al.* 2021). The dual membrane process is effective in removing almost all

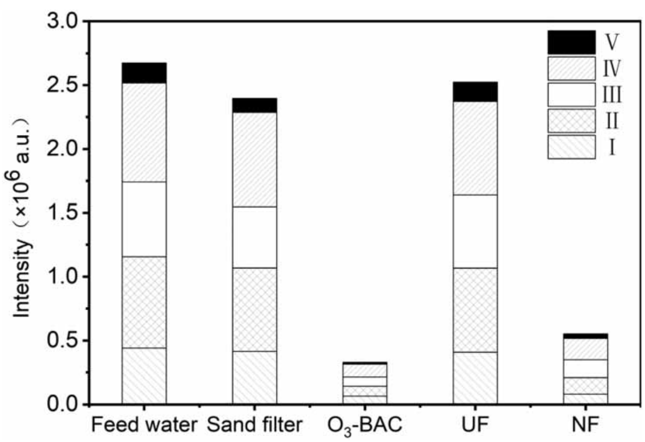


Figure 12 | Fluorescence distribution of 3D-EEM spectra in the water samples under different treatments.

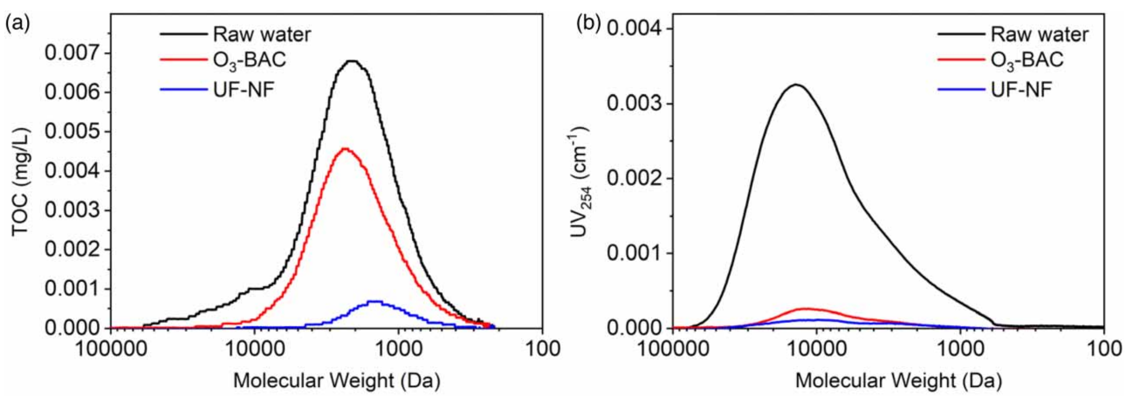


Figure 13 | MW distribution of DOM in (a) the dual membrane process and (b) O_3-BAC.

organics. The medium- and high-MW organics are almost completely removed by the size exclusion of the UF. NF can remove low- and medium-MW organics, which can be attributed to its size exclusion and adsorption (Teixeira & Sousa 2013). Surface water contains both hydrophilic and hydrophobic substances. Figure 13 shows that the dual membrane process is slightly more effective in removing UV_{254} than TOC because the UF and NF membranes in this test are hydrophilic, so they play less role into removing hydrophilic substances from the water, which is consistent with the research findings of the previous study (Nghiem *et al.* 2005). A component of hydrophilic substances is also responsible for hydrophilic polysaccharides and neutral hydrophilic matter secreted by algal cells (Gray *et al.* 2007). In summary, the dual membrane process can effectively remove organic matter, which ensures water safety.

3.3.3. Removal efficiency of various processes

Figure 14 shows the average removal efficiency of various processes. The dual membrane process is slightly more effective than O_3-BAC in pollutant removal, and the dual membrane process is equally stable. In addition, the removal efficiency of the UF membrane, as a pretreatment, is identical to that of sand filtration in O_3-BAC. Although the removal of organic matter is not good, the following effluent satisfies the standard. From the results, this pilot-scale experiment provides a technical reference for large drinking water treatment plants using the dual membrane process. However, another major reason for the limited application of the dual membrane process is its high cost. Therefore, based on a pilot-scale experiment, this study is a projection of the average annual cost of a large drinking water treatment plant using the dual membrane process. It will be further demonstrated whether the process is necessary on a large scale.

3.4. Technoeconomic analysis

A detailed analysis of the cost for the dual membrane process and O_3-BAC is shown in Table 2. It is well known that the water quality, water quantity, and scale of the treatment system greatly affect the cost. Thus, the economic analysis is performed based on the plant capacity in this study, i.e., 12,000 m^3/day. The unit costs ($/$m^3$) for both processes are estimated for the comparison.

O_3-BAC includes the building of coagulation, sedimentation, sand filtration, ozonation, and activated carbon with a total area of 360.08 m^2. The cost of construction, facilities, and materials is $898,524, the cost of depreciation is $34,150 per year, and the annual operating cost is $324,920. Operating costs are mainly the purchase of quartz sand, activated carbon and electricity, which can be saved by changing the material. According to the calculation, we find that the treatment cost is $0.082/$m^3$, and the payback period is 7 months with an internal rate of return of 178.34%. At present, as the most popular water treatment process, low cost is its major advantage.

The dual membrane process mainly includes the structure of UF and NF with a total area of 200 m^2. The cost of construction and facilities is $1,223,538, the cost of depreciation is $38,739 per year, and the annual operating cost is $525,766. Operating costs are mainly for the purchase of UF membranes, NF membranes, electricity, chemicals and bactericidal additives. Accounting for the calculation, we find that the treatment cost is $0.13/$m^3$, and the payback period is 11 months with an internal rate of return of 111.44%. As an emerging water purification technology, NF has drawbacks such as high cost and

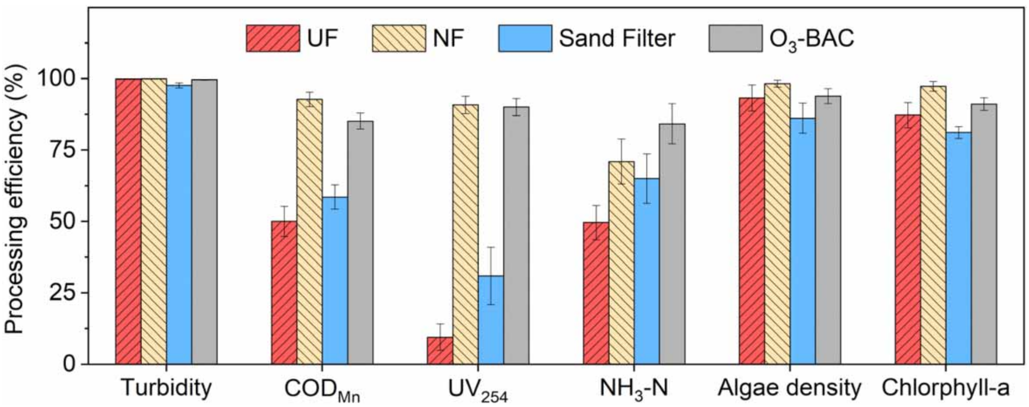


Figure 14 | Average removal of pollutants by the two processes.

Table 2 | Comparison of the cost estimated for the dual membrane process and O_3-BAC

Items	O_3-BAC	UF–NF
Occupied area, m^2	360.08	200
Construction cost, $	508,243	306,454
Facility and material, $	390,281	917,084
Service life, years	25	30
Depreciation rate	3.8%	3.2%
Depreciation cost, $	34,150	38,739
Operating cost (per year), $	324,920	525,766
Annual cost (per year), $	359,070	564,504
Unit cost, $/$m^3$	0.082	0.13
Payback period, months	7	11
Internal rate of return	178.34%	111.44%

low recovery rate, but the shorter process and smaller footprint create more economic benefits. When the cost of membranes decreases in the future, people may incline to think in terms of footprint. Therefore, the dual membrane process is considered an effective method to prepare high-quality drinking water.

4. CONCLUSION

In this paper, a dual membrane process of UF–NF was set up to purify algae-laden water from Tai Lake. The pilot-scale experiment was conducted in a water treatment plant on the southeastern shore of Tai Lake. Compared to O_3-BAC, the test systematically evaluates the removal efficiency, membrane contamination, long-term characteristics, and technical economy of the dual membrane process. The results show that the dual membrane process is effective in removing turbidity, ammonia nitrogen, algae, and organic matter. The dual membrane process is stable, even with high concentrations of fouling, and the effluent satisfies the need for high quality. During operation, the dual membrane process controls biological fouling and reduces the rate of membrane fouling through the application of biocides. UF as pretreatment effectively reduces fouling and prolongs the lifetime of the NF membrane. In addition, O_3-BAC consumes many chemicals and is less effective in removing contaminants, while the dual membrane process is environmentally friendly and highly efficient. In summary, the dual membrane process has high economic and technical feasibility in potable water treatment in the future.

ACKNOWLEDGEMENT

This work was supported by the National Natural Science Foundation of China (Grant No. 51922078) and the Open Project of the State Key Laboratory of Pollution Control and Resource Reuse Foundation (Grant No. PCRRF14011). The authors acknowledge helpful conversations regarding the interpretation of these data with Dr Juncheng Han and we thank Qianqian Xu for providing access to instrumentation.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

REFERENCES

Ağtaş, M., Yılmaz, Ö., Dilaver, M., Alp, K. & Koyuncu, O. 2020 Hot water recovery and reuse in textile sector with pilot scale ceramic ultrafiltration/nanofiltration membrane system. *Journal of Cleaner Production* **256**, 120359.

Al-Amri, A., Salim, M. R. & Aris, A. 2010 The effect of different temperatures and fluxes on the performance of membrane bioreactor treating synthetic-municipal wastewater. *Desalination* **259** (1), 111–119.

Cai, W. & Liu, Y. 2016 Enhanced membrane biofouling potential by on-line chemical cleaning in membrane bioreactor. *Journal of Membrane Science* **511**, 84–91.

Castaño Osorio, S., Biesheuvel, P. M., Dykstra, J. E. & Virga, E. 2022 Nanofiltration of complex mixtures: the effect of the adsorption of divalent ions on membrane retention. *Desalination* **527**, 115552.

Chen, W., Westerhoff, P., Leenheer, J. A. & Booksh, K. 2003 Fluorescence excitation – emission matrix regional integration to quantify spectra for dissolved organic matter. *Environmental Science & Technology* **37** (24), 5701–5710.

Chen, M. Z., Mo, J. N., Lu, W. & Li, F. 2009 Discussion on the high algae content water treatment. *Water & Wastewater Engineering* **45** (07), 28–32.

Chen, Z., Li, M., Wen, Q. & Ren, N. 2017 Evolution of molecular weight and fluorescence of effluent organic matter (EfOM) during oxidation processes revealed by advanced spectrographic and chromatographic tools. *Water Research* **124**, 566–575.

Cheng, X., Wu, D., Liang, H., Zhu, X., Tang, X., Gan, Z., Xing, J., Luo, X. & Li, G. 2018 Effect of sulfate radical-based oxidation pretreatments for mitigating ceramic UF membrane fouling caused by algal extracellular organic matter. *Water Research* **145**, 39–49.

Cheng, X., Lian, J., Ren, Z., Hou, C., Jin, Y., Zhang, L., Zhu, X., Luo, C., Wu, D. & Liang, H. 2021 Coupling sodium percarbonate (SPC) oxidation and coagulation for membrane fouling mitigation in algae-laden water treatment. *Water Research (Oxford)* **204**, 117622.

Chiou, Y., Hsieh, M. & Yeh, H. 2010 Effect of algal extracellular polymer substances on UF membrane fouling. *Desalination* **250** (2), 648–652.

Da-Silva-Correa, L. H., Smith, H., Thibodeau, M. C., Welsh, B. & Buckley, H. L. 2022 The application of non-oxidizing biocides to prevent biofouling in reverse osmosis polyamide membrane systems: a review. *Aqua (London, England)* **71** (2), 261–292.

Du, J., Shi, X., Wang, Y., Tang, A., Zhang, Z., Fu, M., Sun, W. & Yuan, B. 2022 Effects of chlorination on the nitrosamines formation from two algae species in drinking water source-M. aeruginosa and C. meneghiniana. *Chemosphere (Oxford)* **287**, 132093.

Ersan, M. S., Ladner, D. A. & Karanfil, T. 2016 The control of N-nitrosodimethylamine, Halonitromethane, and Trihalomethane precursors by Nanofiltration. *Water Res* **105**, 274–281.

Fang, J., Yang, X., Ma, J., Shang, C. & Zhao, Q. 2010 Characterization of algal organic matter and formation of DBPs from chlor(am)ination. *Water Research* **44** (20), 5897–5906.

Gray, S. R., Ritchie, C. B., Tran, T. & Bolto, B. A. 2007 Effect of NOM characteristics and membrane type on microfiltration performance. *Water Res* **41** (17), 3833–3841.

Guo, Y., Bai, L., Tang, X., Huang, Q., Xie, B., Wang, T., Wang, J., Li, G. & Liang, H. 2018 Coupling continuous sand filtration to ultrafiltration for drinking water treatment: improved performance and membrane fouling control. *Journal of Membrane Science* **567**, 18–27.

Guo, H., Peng, L. E., Yao, Z., Yang, Z., Ma, X. & Tang, C. Y. 2019 Non-polyamide based nanofiltration membranes using green metal-organic coordination complexes: implications for the removal of trace organic contaminants. *Environ Sci Technol* **53** (5), 2688–2694.

Henderson, R., Parsons, S. A. & Jefferson, B. 2008 The impact of algal properties and pre-oxidation on solid-liquid separation of algae. *Water Res* **42** (8–9), 1827–1845.

Kamp, J., Emonds, S., Borowec, J., Restrepo Toro, M. A. & Wessling, M. 2021 On the organic solvent free preparation of ultrafiltration and nanofiltration membranes using polyelectrolyte complexation in an all aqueous phase inversion process. *Journal of Membrane Science* **618**, 118632.

Li, G., Gao, Y., Song, W., Xu, F., Wang, Y., Sun, S. & Jia, R. 2021 Parameter optimization and performance analysis of nanofiltration membrane in treatment of compound-contaminated high-hardness water. *Aqua (London, England)* **70** (8), 1145–1158.

Lin, J., Nugrayanti, M. S., Ika, A. R. & Karangan, A. 2021 Removal of Microcystis Aeruginosa by oxidation-assisted coagulation: effect of algogenic organic matter fraction changes on algae destabilization with Al hydrates. *Journal of Water Process Engineering* **42**, 102142.

Liu, T., Chen, Z., Yu, W. & You, S. 2011 Characterization of organic membrane foulants in a submerged membrane bioreactor with pre-ozonation using three-dimensional excitation-emission matrix fluorescence spectroscopy. *Water Research* **45** (5), 2111–2121.

Liu, M., Wang, S., Wang, T., Duan, M., Su, Y., Han, H., Lin, X. & Li, Z. 2021 Application of microfiltration-nanofiltration combined technology for drinking water advanced treatment in a large-scale engineering project. *AQUA: Water Infrastructure, Ecosystems and Society* **70** (4), 619–636.

Liu, W., Yang, K., Qu, F. & Liu, B. 2022 A moderate activated sulfite pre-oxidation on ultrafiltration treatment of algae-laden water: fouling mitigation, organic rejection, cell integrity and cake layer property. *Separation and Purification Technology* **282**, 120102.

Maeng, M., Shahi, N. K. & Dockko, S. 2021 Enhanced flotation technology using low-density microhollow beads to remove algae from a drinking water source. *Journal of Water Process Engineering* **42**, 102131.

Medema, G. J. & Schijven, J. F. 2001 Modelling the sewage discharge and dispersion of Cryptosporidium and Giardia in surface water. *Water Res* **35** (18), 4307–4316.

Nghiem, L. D., Schäfer, A. I. & Elimelech, M. 2005 Nanofiltration of hormone mimicking trace organic contaminants. *Separation Science and Technology* **40** (13), 2633–2649.

Pandian, A. M. K., Rajamehala, M., Singh, M. V. P., Sarojini, G. & Rajamohan, N. 2022 Potential risks and approaches to reduce the toxicity of disinfection by-product – A review. *The Science of the Total Environment* **822**, 153323–153323.

Pompei, C., Campos, L. C., Vieira, E. M. & Tucci, A. 2022 The impact of micropollutants on native algae and cyanobacteria communities in ecological filters during drinking water treatment. *Sci Total Environ* **822**, 153401.

Ren, B., Weitzel, K. A., Duan, X., Nadagouda, M. N. & Dionysiou, D. D. 2022 A comprehensive review on algae removal and control by coagulation-based processes: mechanism, material, and application. *Separation and Purification Technology* **293**, 121106.

Rivas, J., Gimeno, O. & Beltrán, F. 2009 Wastewater recycling: application of ozone based treatments to secondary effluents. *Chemosphere* **74** (6), 854–859.

Sangrola, S., Kumar, A., Nivedhitha, S., Chatterjee, J., Subbiah, S. & Narayanasamy, S. 2020 Optimization of backwash parameters for hollow fiber membrane filters used for water purification. *Journal of Water Supply : Research and Technology – AQUA* **69** (6), 523.

Su, J. F., Ma, M., Wei, L., Ma, F., Lu, J. S. & Shao, S. C. 2016 Algicidal and denitrification characterization of Acinetobacter sp. J25 against Microcystis aeruginosa and microbial community in eutrophic landscape water. *Mar Pollut Bull* **107** (1), 233–239.

Sun, W. & Chu, W. 2021 Editorial: advanced treatment technologies for drinking water. *Aqua (London, England)* **70** (8), iii–iiv.

Tang, S., Li, J., Zhang, Z., Ren, B. & Zhang, X. 2019 Comparison of long-term ceramic membrane bioreactors without and with in-situ ozonation in wastewater treatment: membrane fouling, effluent quality and microbial community. *Science of The Total Environment* **652**, 788–799.

Teixeira, M. R. & Sousa, V. S. 2013 Fouling of nanofiltration membrane: effects of NOM molecular weight and microcystins. *Desalination* **315**, 149–155.

Touati, K., Gzara, L., Mahfoudhi, S., Bourezgui, S., Hafiane, A. & Elfil, H. 2018 Treatment of coastal well water using ultrafiltration-nanofiltration-reverse osmosis to produce isotonic solutions and drinking water: fouling behavior and energy efficiency. *Journal of Cleaner Production* **200**, 1053–1064.

Verliefde, A. R. D., Cornelissen, E. R., Heijman, S. G. J., Verberk, J. Q. J. C., Amy, G. L., Van der Bruggen, B. & van Dijk, J. C. 2008 The role of electrostatic interactions on the rejection of organic solutes in aqueous solutions with nanofiltration. *Journal of Membrane Science* **322** (1), 52–66.

Wang, Y. L., Xu, X., Jia, R., Liu, B. & Song, W. 2018a Experimental study on the removal of organic pollutants and NH3-N from surface water via an integrated copolymerization air flotation-carbon sand filtration process. *Journal of Water Supply: Research and Technology – AQUA* **67** (5), 506.

Wang, Y., Wang, Z., Wang, J. & Wang, S. 2018b Triple antifouling strategies for reverse osmosis membrane biofouling control. *Journal of Membrane Science* **549**, 495–506.

Wang, J., Tang, X., Xu, Y., Cheng, X., Li, G. & Liang, H. 2020 Hybrid UF/NF process treating secondary effluent of wastewater treatment plants for potable water reuse: adsorption vs. coagulation for removal improvements and membrane fouling alleviation. *Environmental Research* **188**, 109833–109833.

Wei, L., Wen, K., Lu, J. & Ma, J. 2021 Quantification of low molecular weight oxidation byproducts produced from real filtered water after catalytic ozonation with different pathways. *J Hazard Mater* **405**, 124674.

Xiao, R., Duan, Y. & Chu, W. 2020 The effectiveness of household water treatment and safe storage in improving drinking water quality: a disinfection by-product (DBP) perspective. *Journal of Water Supply : Research and Technology – AQUA* **69** (8), 785.

Xu, P., Chen, Y., Gui, B., Guo, X. & Zhang, J. 2021 Pilot study on the treatment of lake water with algae by ultrafiltration–ozone–biologically activated carbon. *Aqua (London, England)* **70** (8), 1192–1203.

Yang, W., Guo, Q., Duan, D., Wang, T., Liu, J., Du, X., Liu, Y. & Xia, S. 2022 Characteristics of flat-sheet ceramic ultrafiltration membranes for lake water treatment: a pilot study. *Separation and Purification Technology* **289**, 120677.

Yu, C., Wu, J., Contreras, A. E. & Li, Q. 2012 Control of nanofiltration membrane biofouling by Pseudomonas aeruginosa using d-tyrosine. *Journal of Membrane Science* **423**, 487–494.

Yu, W., Zhang, D. & Graham, N. J. D. 2017 Membrane fouling by extracellular polymeric substances after ozone pre-treatment: variation of nano-particles size. *Water Research* **120**, 146–155.

Zamyadi, A., Fan, Y., Daly, R. I. & Prevost, M. 2013 Chlorination of Microcystis aeruginosa: toxin release and oxidation, cellular chlorine demand and disinfection by-products formation. *Water Res* **47** (3), 1080–1090.

Zhang, Y., Zhang, S., Gao, J. & Chung, T. 2016 Layer-by-layer construction of graphene oxide (GO) framework composite membranes for highly efficient heavy metal removal. *Journal of Membrane Science* **515**, 230–237.

Zhu, T., Zhou, Z., Qu, F., Liu, B. & Van der Bruggen, B. 2022 Separation performance of ultrafiltration during the treatment of algae-laden water in the presence of an anionic surfactant. *Separation and Purification Technology* **281**, 119894.

First received 19 October 2022; accepted in revised form 3 February 2023. Available online 20 February 2023