

Application of forward osmosis in reusing the brackish concentrate produced in reverse osmosis plants with secondary treated wastewater as feed solution: a case study

W. D. Wang, M. Esparra, H. Liu and Y. F. Xie

ABSTRACT

This study evaluated the feasibility of forward osmosis (FO) in diluting and reusing the concentrate produced in a reverse osmosis (RO) plant in James City County, VA. Secondary treated wastewater (STW) was used as the feed solution. Findings indicated that pH had slight effects on the water flux of the FO membrane. As the concentration of total dissolved solids (TDS) in the concentrate was diluted from 12.5 to 1.0 g/L or the temperature in the STW decreased from 23 to 10 °C, the membrane flux decreased from 2.2 to 0.59 and 0.81 L/(m² h), respectively. The FO membrane showed a good performance in the rejection of organic pollutants, with only a small part of the protein-like substances and disinfection byproducts permeating to the diluted concentrate. During an 89-hour continuous operation, water flux decline due to membrane fouling was not observed. Controlling the TDS in the second-stage FO effluent at 1.5 g/L, approximately 8.3% of the pump energy input could be saved. The consumption of groundwater was reduced from 22.7×10^3 to 10.6×10^3 m³/d. FO was proved to be an effective method in both diluting the discharged concentrate and reducing the energy consumption of RO.

Key words | brackish water, drinking water, forward osmosis, secondary treated wastewater

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INTRODUCTION

With the increase in water demand, many utilities are turning to non-traditional water, such as brackish water, seawater, and even wastewater, as a possible water source in many countries and areas of the world (National Research Council (US) 2008). Accordingly, reverse osmosis (RO) is becoming more and more common as a technique to produce drinking water because of its high efficiency in pollutant removal (Herzberg *et al.* 2009). This is the case for James City County in Virginia, USA, where groundwater with a high salt content (brackish water) is the main water source. The raw water is desalinated by a RO system. One of the major issues that the actual process presents is disposal of the concentrate. Due to the environmental impacts of high salinity water, the discharge of desalination

concentrated solutions is highly controlled by the regulatory agencies. Strict regulations have been created over the last decade, making it more difficult to build a new facility or retrofit/expand existing ones.

In the RO process, hydraulic pressure is used to oppose and exceed the osmotic pressure of a saline aqueous feed solution. The applied pressure is the driving force for the mass transport through a semi-permeable membrane (Greenlee *et al.* 2009; Malaeb & Ayoub 2011). The membrane allows the passage of clean water, while the salt and other contaminants are held back (National Research Council (US) 2008). Accordingly, both the energy consumed in the RO unit and the salt content in the discharged concentrate are high (Stoughton & Lietzke 1965; Lattemann & Höpner

2008; Semiat 2008; Elimelech & Phillip 2011). Although great gains have been made over the last decade in the RO technology to increase its energy efficiency, energy costs still contribute as much as 75% of the operation cost of desalination plants (Semiat 2008). Innovations that help to reduce energy consumption and the amount of concentrate discharged will strengthen the suitability of RO in drinking water purification.

The amount of energy used in RO mainly depends on the salt concentration of the feed solution, and can range from 3.0 to 7.0 kWh/m³ for seawater desalination (National Research Council (US) 2008). For brackish water desalination, however, where the concentration of salt (1–10 g/L) is notably lower than that of the seawater (10–30 g/L), the energy consumption is approximately 0.5–3.0 kWh/m³ (National Research Council (US) 2008). For a feed solution with low salt content, a notable decrease in the osmotic pressure that must be overcome with applied hydraulic pressure can be achieved (Semiat 2008; Subramani *et al.* 2011). Accordingly, the RO process would require less energy to desalinate diluted brackish water than it does for the raw brackish water, which is the upgrading plan proposed by the authors for the upgrading of the existing RO system in James City County. The secondary treated wastewater (STW) was used as the dilution water.

Compared with direct dilution, forward osmosis (FO) can enable the use of impaired water to dilute the water entering the RO plant with high pollutant rejection efficiency (Cath *et al.* 2006, 2010). In the FO unit, a low salt concentration water source, acting as the feed solution, is separated by a selective FO membrane from water with a high salt concentration, which acts as the draw solution. The two solutions are placed on opposite sides of the membrane. Fresh water will move from the feed solution towards the draw solution, leaving contaminants retained in the membrane (McCutcheon & Elimelech 2007; Qin *et al.* 2010). Accordingly, FO is probably a competitive technique in diluting the influent of the RO system.

However, the quality of the STW differs from traditional water sources and has limited usages because of the presence of toxic organic materials (Carrara *et al.* 2008), which pose a proven or potentially high health risk to humans. Previous studies have mainly focused on the rejection performance of FO for a few parts of specific hazardous

materials, such as pharmaceuticals, personal care products, and flame-retardants (Hancock *et al.* 2011; Kong *et al.* 2014, 2015). The application of STW as an indirect source water in the hybrid FO/RO system and the organic rejection performance of FO have not yet been studied in depth. The objective of this work, therefore, is to investigate (i) the water flux variation law with the dilution of the concentrate and the variation in temperature and pH of the STW, (ii) the organic rejection performance of the FO membrane, and (iii) the feasibility of FO in diluting and reusing the concentrate produced in the RO unit using STW as the source water.

Existing RO desalination process in James City County, VA

James City County is a 144-square mile municipality located at the head end of the Virginia Peninsula, between the James and York Rivers in Virginia. The James City Service Authority operates the largest solely dependent groundwater based water system in the Commonwealth of Virginia. The majority of its groundwater supply system is derived from the Potomac and Chickahominy-Piney Point Aquifers. The water treatment facility consists of five wells drawing brackish groundwater from the Middle Potomac and Lower Potomac aquifers. To obtain potable water, RO is used to remove salts and other pollutants (Figure 1). The removed salts, also known as concentrate, is discharged into the James River. For every 18.9×10^3 m³/d of potable water produced, approximately 3.8×10^3 m³/d of concentrate is discharged.

Four RO skids are installed in the treatment facility for the desalination of the raw water extracted from the Lower Potomac Wells. The RO skids consist of a two-stage membrane system. The first stage consists of 20 vessels, and the second stage consists of 10 additional vessels. The vessels contain six membranes each. Groundwater is pumped into the first stage membranes of the RO skid. The permeate is discharged to the permeate line. The concentrate from the first stage RO becomes the feed water for the second stage RO. The permeate from the second stage combines with the permeate from the first stage. The concentrate from the second stage RO is piped to the concentrate line.

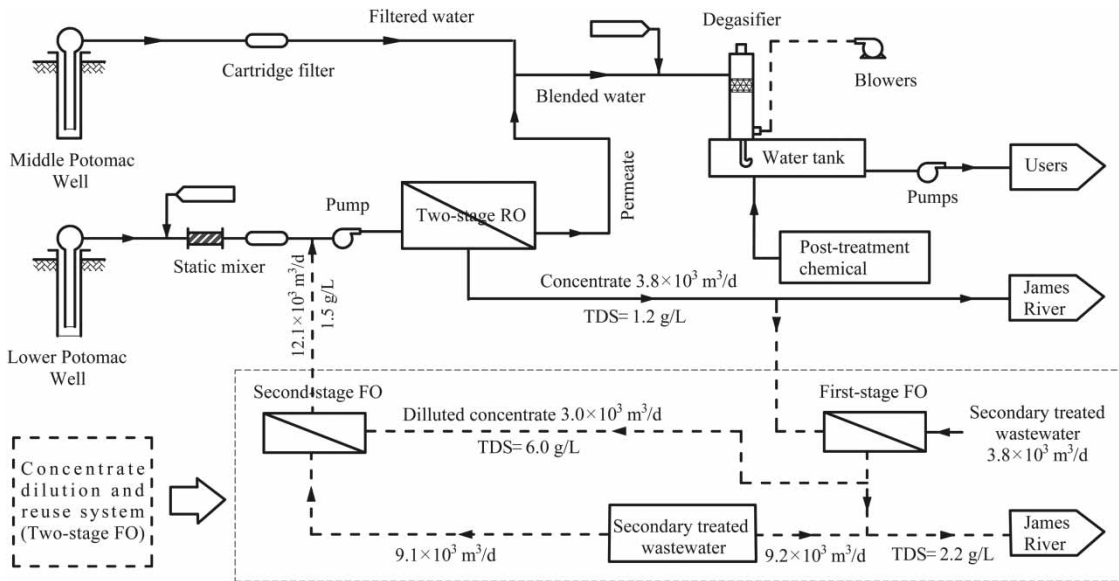


Figure 1 | Current drinking water treatment process and the proposed upgrading plan based on a two-stage FO system (dashed line) in James City County.

The raw water used in the drinking water plant is pumped from five wells. One of them supplies approximately $3.8 \times 10^3 \text{ m}^3/\text{d}$ groundwater with low salt concentration (Middle Potomac Well). The other four wells supply approximately $18.9 \times 10^3 \text{ m}^3/\text{d}$ groundwater with a higher salt concentration (Lower Potomac Well).

The water quality of both types of wells is presented in Table 1. Treatment with RO removes nearly all of the salts in the Lower Potomac Well water. To maintain a normal range of salt contents in the potable water, the RO effluent is blended with the groundwater pumped from the Middle Potomac Well as shown in Figure 1. The concentration of

Table 1 | Water quality of the well waters, concentrate, and the STW used in the experiment

Water quality parameters	Middle Potomac Well	Lower Potomac Well	Concentrate	STW
pH	7.9–8.2	7.6–7.8	8.3	7.5
Temperature (°C)	13	13	13	10–30
Conductivity (µS/cm)	1,500	4,500	25,430	9.1
TDS (mg/L)	1,000	2,300	12,500	280
Na ⁺ (mg/L)	417	1,100	6,127.4	–
Ca ²⁺ (mg/L)	6.1	20.3	105	18.0
Mg ²⁺ (mg/L)	1.5	4.2	25	14.5
Iron (mg/L)	0.01	0.02	0.1	0.05
SiO ₂ (mg/L)	41	29	164	–
Cl ⁻ (mg/L)	340	1,250	7,625	–
Alkalinity (mg/L)	320	340	1,910	52.0
Turbidity (NTU)	<1.0	<1.0	<1.0	5.2
THMs (µg/L)	ND	ND	ND	105.0
HAAs (µg/L)	ND	ND	ND	15.5
TOC (mg/L)	0.06	0.05	0.19	11.2

–, data not available; ND, under determination limit.

total dissolved solids (TDS) in the brackish water is approximately 2,500 mg/L, which increases to approximately 13,000 mg/L after being concentrated. To avoid the discharge of this highly concentrated brackish water, a two-stage FO is suggested for the upgrading of the RO system in James City County.

EXPERIMENTAL MATERIALS AND METHODS

Feed and draw solution of the FO system

The concentrate, which was used as the draw solution, was obtained from the RO system in James City County, VA. The STW, provided by a wastewater treatment plant in Middletown, PA, was used as the feed solution, as the plant is conveniently located near the Harrisburg campus where the experiments were conducted. This wastewater treatment plant consists of a solids grinder and grit remover, primary clarification, biological treatment with clarification, chlorine addition for disinfection, and de-chlorination prior to discharge. The quality of the concentrate and the STW are listed in Table 1.

FO cross flow setup

A bench-scale FO system including a membrane permeation unit, a water circuiting system, and a monitor and data recording system (Figure 2) was constructed and operated in the Environmental Engineering Laboratories at The Pennsylvania State University Harrisburg campus. The membrane permeation unit was made of acrylic plastic and had channel dimensions of 10.5 cm long, 5.0 cm wide, and 0.2 cm deep. The total effective membrane area was 52.5 cm². The FO membrane was provided by Hydration Technologies, Inc. (Albany, OR). The active layer made from cellulose triacetate (CTA) is supported by an embedded polyester screen to give the membrane asymmetry and additional mechanical stability (McCutcheon & Elimelech 2007). The recommended working pH and temperature of the FO membrane was in the ranges of 3.0–8.0 and 0–71 °C, respectively.

Feed and draw solutions were continuously circulated between the storage tanks and the membrane cell. The initial volumes of the feed and draw solutions were 350 mL and 1,000 mL, respectively. The cross-flow rate on each side of the membrane was the same, and was

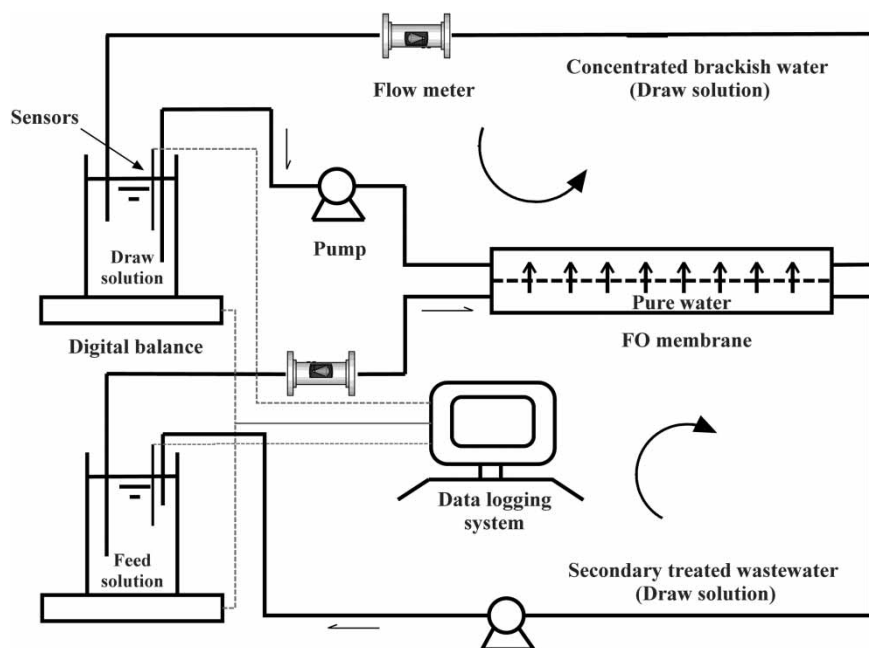


Figure 2 | FO setup used in the experiment.

controlled at 45 mL/min with a variable speed pump (Cole-Parmer, Vernon Hills, IL). Changes in the weight of the draw solution were recorded by a digital balance (TL2100, Mettler Toledo, Germany) to determine the membrane flux of the pure water. The active layer of the FO membrane was facing the feed solution throughout the experiment.

To examine the effect of pH on the flux of water, 0.1 mol/L NaOH was added to the feed solution to increase its pH from 7.5 to 8.5. The total running time was controlled at approximately 15 min. To determine the effects of the water temperature on the flux of the FO membrane, the temperature of the feed solution was adjusted between 3 and 30 °C in a constant-temperature incubator. To assess the effects of membrane fouling on water flux, the variation of the water flux of the FO membrane was monitored in 89 h of continuous running. The pH and temperature of the draw solution were maintained at 8.0 and 23 °C, respectively.

Water quality and membrane structure analysis methods

The trihalomethanes (THMs) and haloacetic acids (HAAs) were analyzed by gas chromatography with an electron capture detector (Agilent 7890A, Santa Clara, USA) after liquid-liquid extraction and methylation (for HAAs only) according to the standard method of EPA551.1 and EPA552.3, respectively. Standard solutions of four THMs, including CHCl_3 , bromodichlorobromomethane (CHCl_2Br), chlorodibromomethane (CHBr_2Cl), and bromoform (CHBr_3), and six HAAs, including monochloroacetic acid, monobromoacetic acid, dichloroacetic acid (DCAA), bromochloroacetic acid (BCAA), dibromoacetic acid, and trichloroacetic acid (TCAA), were purchased from Sigma-Aldrich (Germany). pH and temperature were monitored on-line using a Logger Pro 3.8.6 with corresponding sensors. Before measuring, all sensors were calibrated with standard solutions. TDS was measured according to standard method 2540c (APHA *et al.* 2005). The concentration of organic matters was measured by a total organic carbon (TOC) analyzer (TOC-VCPH, Shimadzu, Japan) and a spectrometer (T6, Puxi, China) at 254 nm.

Fluorescence measurements were conducted using a spectrofluorometer (FP-6500, Jasco, Japan) equipped with a 150 W xenon lamp at 23 °C. A 1.0 cm quartz cuvette

with four optical windows was used for the analyses. An emission scan was conducted from 250 to 600 nm at a wavelength step of 5 nm and excitation wavelengths from 220 to 450 nm at a 5 nm interval. The detector was set to high sensitivity, and the scanning speed was maintained at 2,000 nm/min. To observe the surface characteristics of the FO membrane, scanning electron microscopy (SEM) (JSM-6490LV, JEOL Ltd, Japan) was applied. The SEM samples were sputter-coated with gold before conducting scanning.

RESULTS AND DISCUSSION

Effects of TDS in the concentrate on the water flux of the FO membrane

The experiments were conducted for 15 min to avoid the effects of membrane fouling. As shown in Figure 3, the average water flux of the FO membrane was approximately 2.2 L/(m² h) when the TDS concentration in the concentrate (draw solution) was 12.5 g/L. This would benefit the application of FO in the dilution and reuse of the concentrate. Without membrane fouling, the water flux could be maintained above 2.0 L/(m²·h). Decreasing the concentration of TDS in the draw solution to 6.0 g/L, the water flux of the FO membrane decreased notably to 1.3 L/(m²·h).

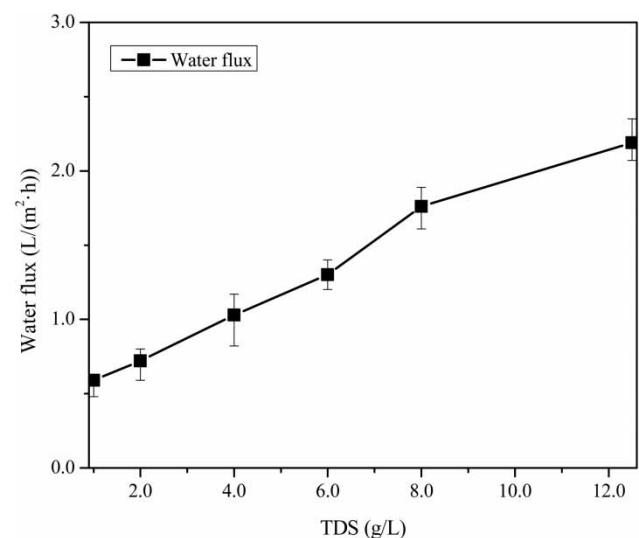


Figure 3 | Variation of water flux with the concentration of TDS in the draw solution.

Unlike the draw solutions with high TDS, for the draw solutions with TDS below 2.0 g/L, the variation of TDS showed fewer effects on the membrane flux (Figure 3). Decreasing the concentration of TDS in the draw solution further to 2.0 g/L and 1.0 g/L, the membrane fluxes were similar, at 0.72 L/(m²·h) and 0.59 L/(m²·h), respectively.

The water flux of the FO membrane is positively proportional to the content of TDS in the draw solution. To obtain a high water flux, a large difference in TDS between the feed and draw solution is desired. As described in Equation (1) (Cath *et al.* 2006), the greater the difference in osmotic potential, the faster water moves through the membrane. Meanwhile, the osmotic pressure is high for the raw concentrate and keeps decreasing with the permeation of pure water through the FO membrane. As the difference in osmotic pressure got lower, the water flux decreased with operation time:

$$J_w = A(\sigma\Delta\pi - \Delta P) \quad (1)$$

where J_w is the water flux, A is the water permeability constant of the membrane, σ is the reflection coefficient, and ΔP is the applied pressure (Cath *et al.* 2006). For FO, ΔP is zero, making the water flux directly proportional to the difference in osmotic pressure.

Effects of pH and temperature in the STW on the water flux of the FO membrane

The water flux of the FO membrane increased gradually as the solution pH increased. The highest value of approximately 2.45 L/(m²·h) was obtained at pH 8.5 (Figure 4(a)). The membrane used in this study was made from CTA with embedded polyester screen support. Under basic conditions, the number of deprotonated hydroxyl groups (with negative charge) in the membrane matrix increased notably, which probably forced adjacent polymers apart, thus increasing water permeability (Braghetta *et al.* 1997; You *et al.* 2012). However, because osmotic pressure is the driving force in FO, and this pressure was relatively stable under different pH conditions, the overall variation of membrane flux with pH was slight.

Compared with pH, temperature showed a more significant effect on the water flux through the FO membrane. As shown in Figure 4(b), the average water flux was

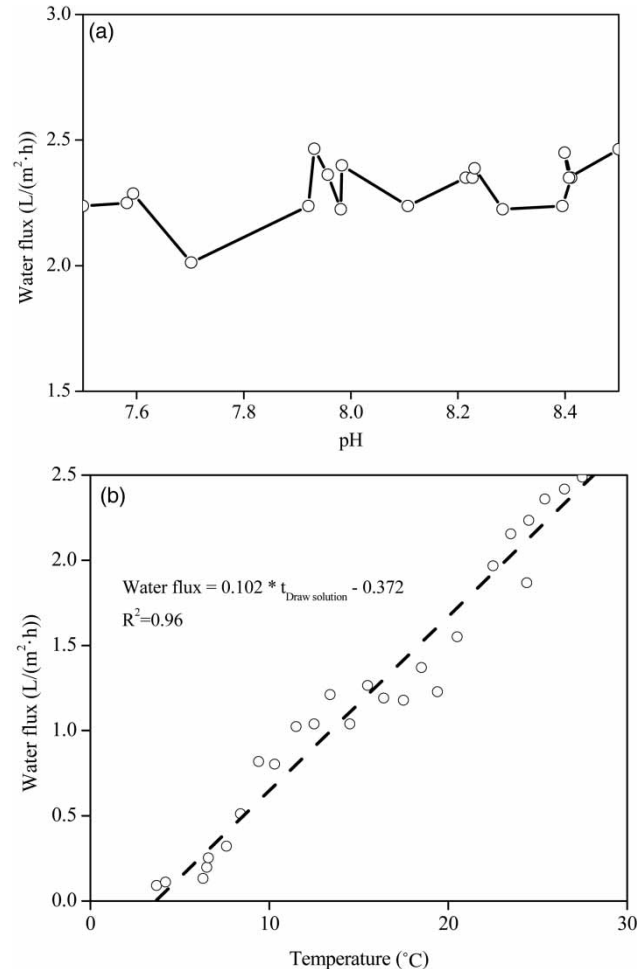


Figure 4 | Effects of pH (a) and temperature (b) in the STW on the water flux of the FO membrane.

approximately 2.75 L/(m²·h) when the temperature of the draw solution was 30 °C, corresponding to the STW temperature in summer, and decreased notably to approximately 0.81 L/(m²·h) at 10 °C, corresponding to the STW temperature in winter. A linear relationship was found between the membrane flux and the solution temperature. The slope and intercept of the best fit line were calculated to be 0.102 and -0.372 respectively, which was close to the values (0.064 and -0.227) obtained by Wang *et al.* (2014) using rainwater and cooling water as the feed and draw solutions, respectively.

However, based on the van't Hoff equation (You *et al.* 2012), the osmotic pressure of the feed solution decreases with the decrease of its temperature, which will increase the net osmotic pressure for driving the water in the FO

process. Besides net osmotic pressure, both the absolute and kinematic viscosities of the feed solution increased notably with the decrease of feed solution temperature (Phuntsho *et al.* 2012). Kim *et al.* (2015) determined that as the solution temperature decreased from 50 °C to 20 °C, osmotic coefficients and diffusivity decreased by 6.6% and 48.3% respectively, which increased the internal concentration polarization (ICP). The increased ICP and solution viscosity was probably the major reason that inhibited the permeation of pure water at low temperatures.

Organic rejection performance of the FO membrane

One of the challenges in using the STW as a potable water source is whether the FO membrane could effectively remove the organic matter existing in the STW. The amount of organic matter transported to the diluted concentrate was evaluated using both UV_{254} and TOC. The UV_{254}

of the draw solution increased slightly from 0.195 to 0.211, corresponding to an increase in TOC from 0.44 to 0.65 mg/L in 8 h of operation (Figure 5(a)). This indicated that the membrane used in this study provided a good organic matter rejection performance. The amount of organic pollutants entering the influent of the RO unit from the STW was few.

Besides UV_{254} and TOC, a 3D excitation–emission matrix fluorescence spectrum was used to investigate the type of organic matter that passed through the FO membrane. The organic matter contained in the STW (feed solution) was rich in humic acid-like substances (Figure 5(b)), associated with the peak of Ex/Em = 420–440/235–245 nm in the fluorescence spectrum (Henderson *et al.* 2009). Compared with the STW, the concentrate (draw solution) was low in organic pollutants, and no peak associated with organic matter was observed (Figure 5(c)). After 8 h of operation, only one peak (Ex/Em = 375/350 nm) contributed by protein-like

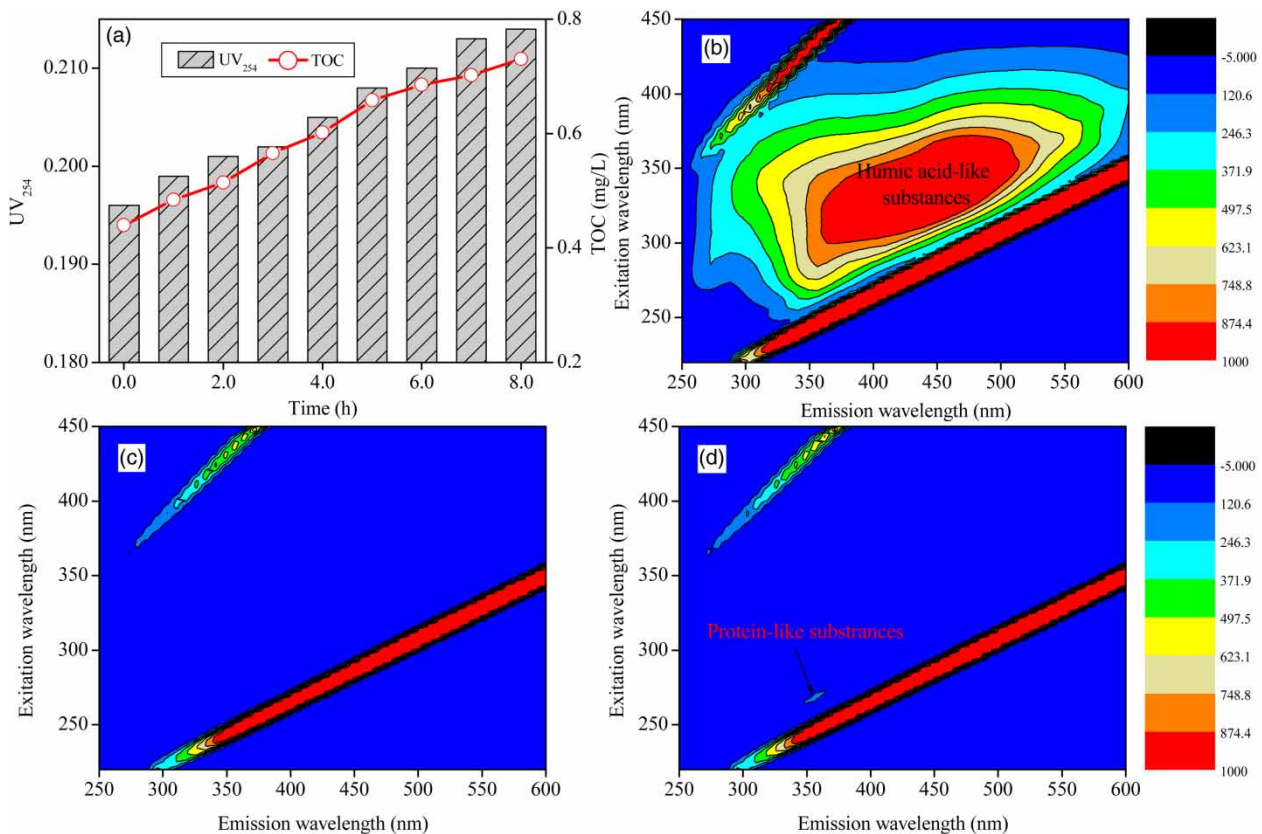


Figure 5 | Variations of UV_{254} and TOC in the draw solution (concentrate) with operation time (a), and the 3D excitation–emission matrix of the feed solution (STW) (b) and draw solution before (c) and after (d) 8 h of operation.

substances (Reynolds 2002; Liu *et al.* 2011) relating to the activity of bacteria was observed. This further proved that the FO membrane used in this study had a high capacity to remove organic matter (Figure 5(d)).

Besides the protein-like substances, part of the disinfection byproducts (DBPs) corresponding to the killing of bacteria might also permeate to the draw solution. Maintaining water temperature at approximately 23 °C, the system was run for 8 h at pH 6.5, 7.5, and 8.5, respectively. The concentration of THMs in the STW was 105 µg/L (Figure 6(a)), which was much higher than that of HAAs (15.5 µg/L). However, the total amounts of THMs and HAAs permeating through the FO membrane was similar under different pH conditions. The concentrations of both THMs and HAAs that permeated into the draw solution were in the range of 5.2–8.4 µg/L, indicating that the FO membrane was effective in DBP removal.

The removal rates for THMs were above 95%, higher than that of HAAs (50–60%) under the selected pH conditions (Figure 6(a)). The low rejection rate of HAAs was probably related to its low initial concentration in the STW used in our study (Xie 2003). These rejections are comparable to the values reported for NF membranes (56–100%) and RO membranes (86–94% for HAAs) (Agus & Sedlak 2010). We speculate that the mechanisms underlying the removal of THMs and HAAs might be size exclusion and charge repulsion, similar to the NF/RO membranes (Bellona *et al.* 2004; Kong *et al.* 2014). Unlike HAAs, all of the THMs were highly volatile organic materials (Lee *et al.* 2009). As the hydraulic retention time of the STW in the feed solution tank increased, part of the THMs might enter the air under the effect of volatilization. Further study showed that the concentration of THMs mainly existing in the forms of CHCl_2Br and CHCl_3 decreased notably from 101 to 6.7 µg/L after 4 days of retention (without stirring) in the feed solution tank (Figure 6(b)). Compared with THMs, the concentration of HAAs, mainly existing in the forms of DCAA, TCAA, and BCAA, was relatively stable (Figure 6(c)).

Feasibility analysis of FO in brackish water desalination system upgrading

Compared with RO, the water flux of FO was relatively small, especially for the draw solution with low salt content.

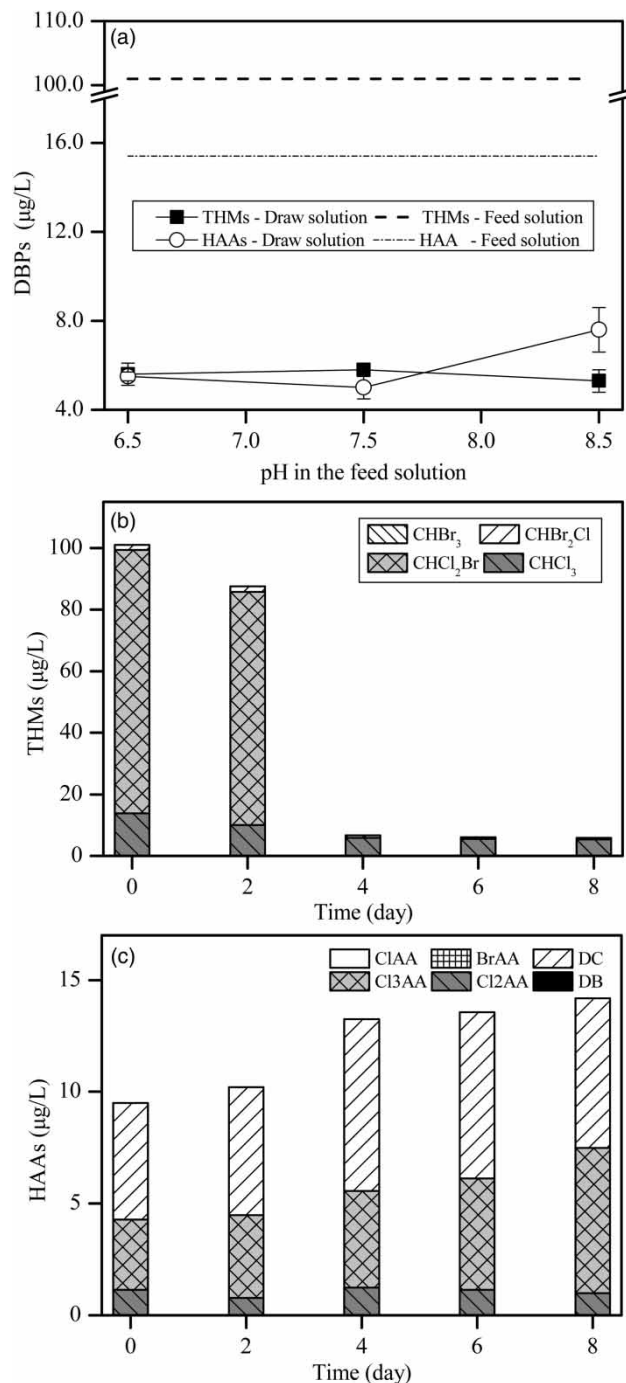


Figure 6 | Contents of DBPs permeating from the feed solution (STW) to the draw solution (concentrate) after running for 8 h (a), and the effects of hydraulic retention time on the residual concentrations of THMs (b) and HAAs (c) in the feed solution.

To guarantee the average water flux, a two-stage FO in which the TDS in the concentrate was diluted from 12.5 to 1.5 g/L in two series-connection draw solution tanks is

suggested for the upgrading of the existing RO system in James City County, as shown in Figure 1. STW was used as a feed solution. By controlling the retention time of the concentrate in the draw solution tank, the TDS in the first-stage FO effluent was maintained at 6.0 g/L, corresponding to an increase in the volume from 3.8×10^3 to 7.6×10^3 m³/d. Sixty per cent of the first-stage FO effluent was blended with 9.2×10^3 m³/d of the STW directly before it was discharged to the James River. In the second-stage FO, 40% of the first-stage FO effluent was further diluted four times. The effluent, with 1.5 g/L of TDS, was reused as the influent of the RO unit. The consumption of the groundwater could be reduced from 22.7×10^5 to 10.6×10^5 m³/d.

Considering the concentration of organic matter permeated into the diluted concentrate, part of which was reused as the influent for the RO, was limited based on accumulative values in 8 h, the effects of organic matter in the STW on the quality of the treated drinking water could be ignored. Before upgrading, the pump pressure needed in the RO unit was approximately 0.6 MPa. Based on the water quality shown in Table 1, NaCl and NaHCO₃ were assumed to be contributing to the osmotic pressure generated by the brackish water. Calculation results showed that after upgrading the osmotic pressure existing between the two sides of the RO membrane would decrease from 0.18 to 0.13 MPa so as the salt content in the influent decreased from 2.3 to

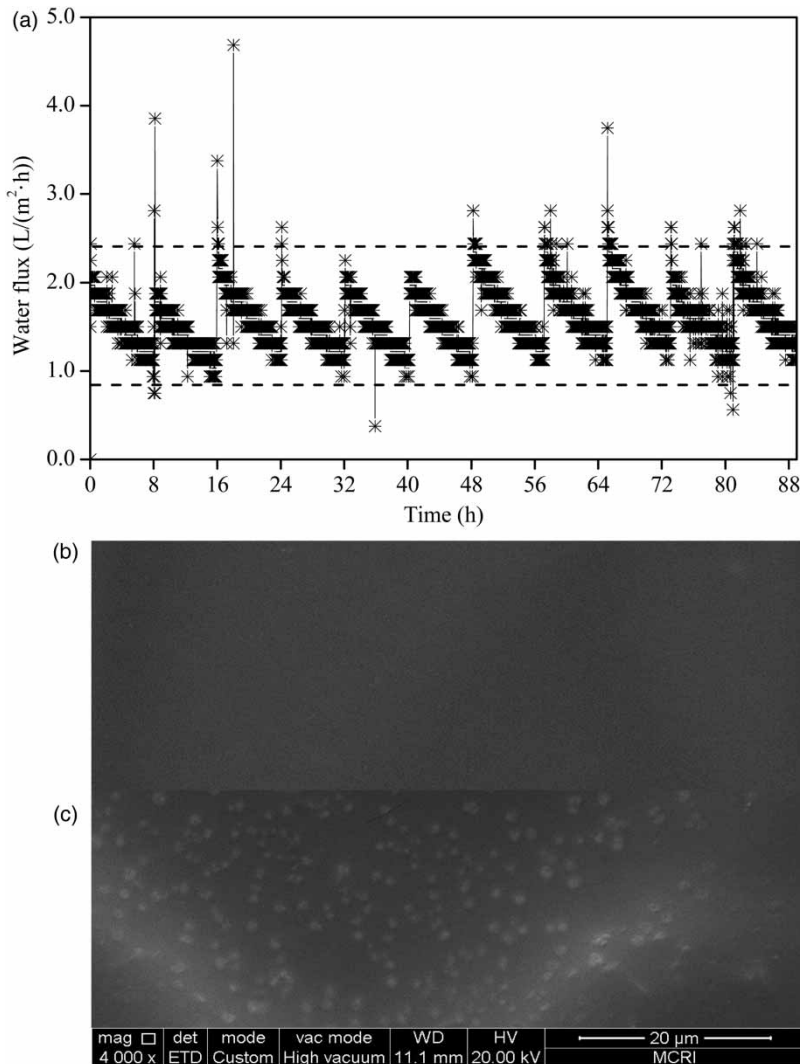


Figure 7 | Variation in water flux in an 89-hour operation (a), and the morphological characteristics of the FO membrane before (b) and after (c) running for 89 h.

1.78 g/L. Maintaining a water purification capacity of $22.7 \times 10^3 \text{ m}^3/\text{d}$, the reduced pump pressure was determined to be 0.05 MPa, so approximately 8.3% of the input energy of the pump station could be saved.

Besides, the fouling process of the FO membrane was slow. As shown in Figure 7(a), as the operation time increase, the water flux of the FO membrane decreased from 2.3 to approximately 1.0 L/(m²·h). This decrease was mainly caused by the dilution of the draw solution. Replacing the diluted concentrate (draw solution) with the concentrate taken from the pilot, the water flux of the FO membrane returned to 2.3 L/(m²·h) even after running for 88 h. The above phenomenon indicated that the fouling process of the FO membrane was slow or membrane fouling showed little effect on the water flux. From the morphological characteristics of the FO membrane (Figure 7(b) and 7(c)), the development of an obvious fouling layer, which mainly contributed to the water flux decline (Lee *et al.* 2010), was not observed on the surface of the active side. The low fouling rate of the FO membrane might be related to the low osmotic pressure of the feed solution, which limited the transport of foulants to the surface of the membrane (Tang *et al.* 2010).

CONCLUSIONS

Over the last decades, strict regulations have been established to protect water bodies by limiting the amount of TDS that are discharged into waterways. In this study, the implementation of FO was considered to treat and reuse the concentrate from the RO process. Solution pH showed a slight effect on the flux of both pure water and DBPs. With the decrease in water temperature, especially in winter, the water flux of the FO membrane decreased notably. Although part of the protein-like substances permeated into the draw solution, the FO membrane used in this study showed a good performance in the rejection of organic pollutants. More than 98% of TOC, 95% of THMs, and 50–60% of HAAs were removed from the feed solution. Considering that these removals were based on accumulative values in 8 h, the detrimental effect of using STW as source water on the quality of the produced drinking water could be ignored. Furthermore,

during an 89-hour operation, there was no observed flux decline due to membrane fouling. By controlling the effluent concentration of TDS in the second stage of FO at 1.5 g/L, the consumption of groundwater would decrease to $10.6 \times 10^3 \text{ m}^3/\text{d}$. Approximately 8.3% of the pump energy input was saved. Meanwhile, the environmental impact caused by the discharge of concentrate into the James River could be minimized.

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