

# Comparative analysis of reverse osmosis and nanofiltration for the removal of dissolved contaminants in water reuse applications

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## HIGHLIGHTS

- Bench- and engineering-scale systems were used to compare NF and RO membranes.
- High TOC rejection by NF90 and NF270, but lower rejection of inorganics with NF270
- Rejection of inorganics was similar between NF90 and RO except for monovalent ions.
- Rejection of TOCs was similar between NF90 and RO membranes.
- NF9 maintained high contaminants rejection in long-term engineering-scale operations.

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## ABSTRACT

The increasing demand for drinking water has led to the adoption of unconventional water sources, such as water reuse. Reverse osmosis (RO) and nanofiltration (NF) membranes are effective barriers against trace organic contaminants in potable water reuse applications. However, the use of RO is being challenged by NF, primarily due to NF's potential to achieve similar contaminant removal as RO but with higher productivity and lower energy requirements. This study compares NF and RO membranes in terms of contaminant removal and energy consumption for potable water reuse applications. RO (BW30XFR) and dense and loose NF (NF90 and NF270) membranes were tested in bench-scale systems, and RO (TW30) and NF (NF9) membrane elements were tested in an engineering scale system utilizing UF-filtered reclaimed wastewater. The highest solute passage was observed using NF270 membrane. There was no difference between NF90 and BW30XFR in terms of divalent ion passage, but NF90's total organic carbon and monovalent ion passages were higher. Both NF90 and BW30XFR highly rejected negatively charged trace organic contaminants (TOCs), though rejections were lower for neutral and positively charged compounds. Furthermore, all compounds were highly rejected in the engineering-scale system by NF9 and TW30. These results highlight the potential of dense NF membranes as an energy-efficient barrier for contaminant removal.

## 1. Introduction

Freshwater scarcity has become a global concern due to the rapidly growing population, which increasingly stresses the limited conventional water resources [1]. Potable water reuse is considered an attractive alternative to overcome the continuous decline in available water supplies [2]. The advanced purification of municipal wastewater for potable water reuse provides a safe and reliable water supply wherever wastewater is utilized [3]. Among different technologies, pressure-

driven membrane processes have proven their potential in advanced water purification. Membrane processes, including low-pressure technologies such as microfiltration (MF) [4,5] and ultrafiltration (UF) [6–8], as well as high-pressure processes such as nanofiltration (NF) [9,10] and reverse osmosis (RO) [11–13], are state-of-the-art for potable water reuse applications as they produce high-quality water [14]. The high rejection rates of membranes, specifically NF and RO, make them effective barriers to most contaminants in potable water reuse. While the NF membranes typically have pores in the range of 1 nm and a molecular

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weight cut-off between 200 Da (tight) and 500 Da (loose), RO membranes are considered nonporous; nevertheless, both types of membranes have emerged as standard processes in potable water reuse applications [15,16].

In potable water reuse applications, there has been a growing concern regarding the ubiquitous occurrence of trace organic contaminants (TOrcs), which are often poorly eliminated during conventional wastewater treatment [17–22]. The poor rejection of TOrcs can be attributed to their complex and diverse physicochemical properties, such as solubility, charge, hydrophobicity, polarity, and small molecular size [23–27]. Specifically, the molecular weight, partition coefficient ( $\log K_{ow}$ ), and charge of these compounds govern their fate in the aquatic environment. The rejection of TOrcs in potable water reuse applications can be achieved using different processes, such as adsorption [28–30], oxidation [31–33], and membrane separation [34–37]. However, due to the diverse properties of TOrcs, no single treatment process can assure the complete rejection of TOrcs, and a multiple barrier approach must be implemented [15,23]. The rejection of TOrcs is governed by several mechanisms, including size exclusion, oxidation, adsorption, and electrostatic interaction [38]. The full advanced treatment (FAT) is a sequential treatment process that includes integrated membrane systems and an advanced oxidation process (AOP). It has been widely implemented and has become the primary treatment choice for potable water reuse globally [15]. The FAT treatment approach consists of MF or UF followed by RO and then AOP, commonly UV/H<sub>2</sub>O<sub>2</sub>, to eliminate pathogens and remove persistent organic or inorganic compounds, ensuring the production of high-quality water that meets the required water quality standards.

In the FAT system, AOP eliminates neutrally charged and low molecular weight organics that might permeate through RO; nevertheless, FAT's high-quality water production is largely attributed to the RO's rejection of most solutes [14,15]. The specific energy consumption (SEC) of the FAT (the sum of the SEC for each unit process) is typically reported to be between 0.8 and 2.5 kWh/m<sup>3</sup> (MF/UF 0.1–0.3 kWh/m<sup>3</sup>, RO 0.6–1.5 kWh/m<sup>3</sup>, and UV/H<sub>2</sub>O<sub>2</sub> 0.07–1.0 kWh/m<sup>3</sup>) [39–41]. For example, the Orange County Water District (OCWD) Groundwater Replenishment System (GWRS) treatment scheme (MF-RO-UV/H<sub>2</sub>O<sub>2</sub>) reported an SEC of 1.12 kWh/m<sup>3</sup> [42]. Generally, the energy consumption of the RO process accounts for >50 % of the whole potable water reuse scheme [40]. NF could serve as an efficient alternative to RO. In contrast to RO, NF membranes have a relatively loose structure, allowing for more water permeation and producing low-saline concentrate, which results in lower energy consumption. In addition to the lower energy requirement, NF is effective in removing inorganic ions, such as divalent ions, as well as organics and viruses [43,44]. NF is commonly described as a water softening process due to its high rejection of divalent ions such as calcium and magnesium. Although a slightly higher permeate contaminant concentration could be observed when using loose NF membranes, which might require higher AOP dosing and thus increase energy consumption, it has been reported that the percent change in SEC as a result of increasing AOP dose to attenuate contaminants is minimal [39,45–47]. The energy consumption of UV/AOP at the OCWD GWRS was reported as 0.07 kWh/m<sup>3</sup> [48]. Furthermore, using UV/H<sub>2</sub>O<sub>2</sub> specifically, a minimum UV dose of 300 mJ/cm<sup>2</sup> (typically 900 mJ/cm<sup>2</sup>) is required to meet the OCWD pathogen log reduction objective for direct potable water reuse (6-log reduction) [49]. Moreover, when UV/AOP was utilized for the treatment of MF filtrate and RO permeate secondary effluent, the energy consumption was 0.93 and 0.62 kWh/m<sup>3</sup> respectively [50]. Accordingly, AOP energy consumption should be lower than MF and closer to RO when using NF membranes.

Despite the lower energy requirement of NF compared to RO, NF is rarely used in potable water reuse schemes. Most NF applications are limited to treating surface water, groundwater, as well as in the textile and food industry [51–58]. Nonetheless, a considerable number of works have reported the treatment of wastewater with NF membranes

[59–61]. Various studies have compared the rejection rates of RO and NF membranes in different water matrices, with NF demonstrating >90 % rejection of divalent ions and bulk organic matter [62–66]. In terms of TOrcs, studies found rejection to be similar between NF and RO membranes, while for some other compounds (especially neutral and low-molecular weight), rejection varied between the two types of membrane [67–71]. Although this suggests that RO may be a better option, it is important to balance the trade-off between solute rejection and energy consumption. Despite plenty of research comparing NF and RO membranes, most previous studies have been conducted using synthetic water spiked with targeted solute [67,72,73], which may not accurately reflect the conditions of natural water sources, as it does not account for the impact of other compounds that are present in the water, such as pathogens, salts, and organic matter. Additionally, while other works have used real water matrices with laboratory-scale filtration systems [66,68,69], a limited number of studies have addressed larger-scale testing, primarily short-term pilot-scale tests, and only reported low concentrations or non-detection of some TOrcs in the influent, leading to the presumption of complete rejection [62,63,74]. To comprehensively assess a membrane's capability, it is important to use real-world water conditions, such as municipal reclaimed water where substantial levels of TOrcs are present. There is a lack of long-term studies in continuously operated larger-scales, such as engineering-scale systems utilizing reclaimed municipal wastewater, assessing rejection efficiency, and addressing the energy requirements.

In this work, ultra-filtered treated reclaimed wastewater was used to study the transport of contaminants through NF and RO membranes for potable water reuse applications with a focus on assessing NF as a sustainable alternative to RO. This study compares the permeate water quality of NF and RO membranes to evaluate their water permeability and solute passage in bench-scale systems. Additionally, this study provides details on the organic compounds permeating through the membranes in terms of molecular weight, hydrophobicity, and TOrcs rejection. Subsequently, engineering-scale experiments employing NF were used to evaluate the rejection rates of contaminants and estimate the energy consumption.

## 2. Materials and methods

### 2.1. Membranes

Three different types of DuPont FILMTEC membranes (DuPont, Wilmington, DE, USA) were used for the bench-scale experiments, including two NF membranes (NF90 and NF270) and one RO membrane (BW30XFR). For the engineering-scale, RO membranes (DuPont FILMTEC TW30–4040) were operated for 48 months and then were replaced with NF membranes (Applied Membranes NF9 M-N4040A9) and operated for 14 months. Both TW30 and NF9 membranes are polyamide thin-film composite in spiral wound configuration with a 7.2 m<sup>2</sup> active membrane area.

### 2.2. Bench-scale permeation system

Illustrated in Fig. 1 is the bench-scale system, which consists of a feed tank, two flat sheet membrane cells (SEPA CF, Sterlitech, Auburn, WA, USA), electronic pressure controllers (Automation Direct, Cumming, GA, USA), proportional valves (Hass Manufacturing, Averill Park, NY, USA), pumps (Hydra-Cell, Wanner Engineering, Minneapolis, MN, USA), and scales for water flux data acquisition. The bench-scale experiments utilized the ultrafiltered effluent from the UF-RO engineering-scale system, which was delivered continuously to the bench-scale system feed tank. Permeation experiments were performed using two crossflow membrane cells (crossflow velocity of 0.11 m/s) with an active area of 0.014 m<sup>2</sup> at three permeate water fluxes of 35, 25, and 15 LMH without concentrate flow recirculation. For each water flux, once the desired water flux was acquired and maintained, the experiment was

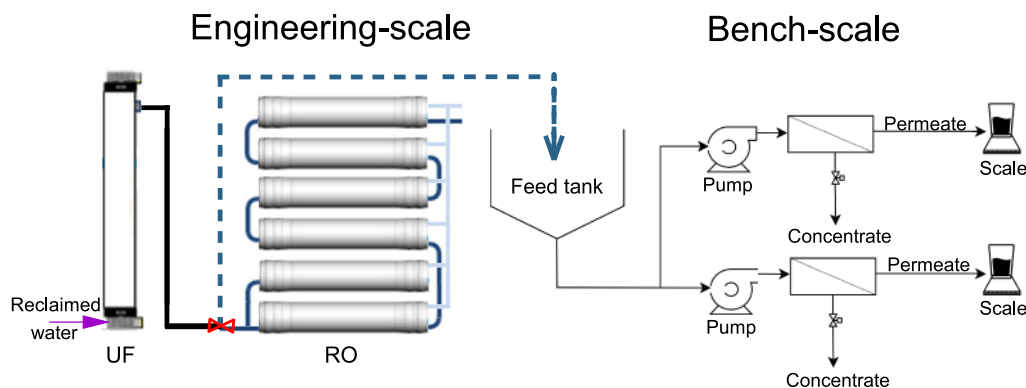


Fig. 1. Schematic of the experimental setup. The feed water from the engineering-scale RO system was collected in a bench-scale feed tank and then pumped through two bench-scale membrane cells, each containing different types of membranes that operated simultaneously.

performed for a duration of one hour. During the experiment, 50 mL samples were collected from both the feed and permeate water every 20 min for solute analysis. Prior to the experiments, all membranes were stored in deionized (DI) water and were compacted with DI water for 16 h using a water flux of 35 LMH.

### 2.3. Engineering-scale system

The engineering-scale system is a UF-RO train (Applied Membranes, Inc., Vista, CA, USA) that has been operating continuously, treating up to 75 m<sup>3</sup>/d of reclaimed wastewater with an influent flow rate of 3 m<sup>3</sup>/h (13 gpm) at a 62 % recovery rate, without recirculation, at the Water & Energy Sustainable Technology (WEST) Center, University of Arizona in Tucson, Arizona (USA). It comprises one UF membrane (DuPont IntegraFlux UXA-2680XP) and a two-stage RO skid. The first RO stage has four 4040 membrane elements housed in two stainless-steel pressure vessels in parallel, and the second stage has eight membrane elements housed in four stainless-steel pressure vessels in series. Details of the engineering-scale UF-RO schematic and regular cleaning have been published previously [13]. The engineering-scale system is fully automated and monitored; samples were collected and analyzed regularly.

### 2.4. Reclaimed wastewater

Tertiary treated wastewater effluent was pumped directly to the UF system from Pima County's Agua Nueva Water Reclamation Facility, and then the UF filtrate is passed to the RO/NF skid. The feed water quality is shown in Table 1.

### 2.5. Apparent permeability coefficients and solute passage

The apparent water permeability coefficient (A) was calculated using the solution-diffusion model equation [75]:

$$A = \frac{J_w}{(\Delta P - \Delta \pi)} \quad (1)$$

where  $J_w$  is the permeate water flux,  $\Delta P$  and  $\Delta \pi$  are the hydraulic and osmotic pressure differences across the membrane. The osmotic pressure was calculated using OLI Studio (OLI Systems Inc., Parsippany, NJ) with the composition of ultrafiltered water for bench-scale and considering the averaged ultrafiltered water for engineering-scale. The apparent solute permeability coefficient (B) was calculated from Fick's law:

$$B = \frac{J_s}{(C_f - C_p)} \quad (2)$$

where  $J_s$  is the solute flux,  $C_f$  and  $C_p$  are the solute concentrations in the feed and permeate water, respectively. It is worth noting that the term "apparent" is utilized for the water and solute permeability coefficients as they were calculated utilizing bulk feed concentrations instead of the rigorous interface concentrations that can be estimated considering concentration polarization.

The solute flux was calculated as:

$$J_s = \frac{Q_p * C_p}{A_m} \quad (3)$$

where  $Q_p$  is the permeate water flow rate and  $A_m$  is the membrane active area. The solute passage (SP) and rejection (R) were calculated as follows:

$$SP = 100 * \left( \frac{C_p}{C_f} \right) \quad (4)$$

$$R = 100 - SP \quad (5)$$

### 2.6. Analytical methods

#### 2.6.1. Inorganic analysis

An ion chromatography mass spectrometry system with a Dionex ICS-5000 Ion Pac, using 22 mM potassium hydroxide as the eluent, was used to measure anions concentrations (sulfate, chloride, and nitrate). An inductively coupled plasma mass spectrometry (ICP-MS) instrument (Agilent 7800x Series; Agilent, Santa Clara, CA) was used to measure cations concentrations (potassium, magnesium, calcium, and sodium). All samples were filtered using 0.45 μm glass fiber syringe filters (Tisch scientific, North Bend, OH). ICP-MS samples were acidified with 2 % (v/v) nitric acid (67–70 % HNO<sub>3</sub>, Fisher scientific, Fair Lawn, NJ).

#### 2.6.2. Organic analysis

A Total Organic Carbon (TOC) analyzer (TOC-L CSH, Shimadzu, Japan) was used to measure the TOC concentration. Samples were

Table 1  
Feed water quality.

Water quality parameter	Feed water
Turbidity (NTU)	0.1–0.2
pH	7.0–7.2
Conductivity (μS/cm)	1,100–1,300
TDS (mg/L)	700–850
TOC (mg/L)	5–7
Potassium (mg/L)	17–20
Magnesium (mg/L)	20–24
Calcium (mg/L)	90–100
Sodium (mg/L)	135–145
Sulfate (mg/L)	180–190
Chloride (mg/L)	140–150
Nitrate (mg/L)	5–10

acidified to pH 3 or lower by adding 0.1 mL of hydrochloric acid (24.5–26 % HCl, Sigma Aldrich, Burlington, MA).

Total fluorescence (TF) spectra were calculated from excitation-emission matrix (EEM) fluorescence data using a Duetta spectrofluorometer (Horiba Scientific, Japan). EEM fluorescence was scanned with excitation wavelengths from 250 to 450 nm, and emission wavelengths from 250 to 550 nm. EEM data were processed using MATLAB R2019b (MathWorks). Further details on the analytical methods can be found in a previous publication [13].

Apparent molecular weight was measured using size-exclusion chromatography (Agilent 1260 Infinity II high-performance liquid chromatography (HPLC), Santa Clara, CA) coupled with an organic carbon detector (OCD; Sievers M9 Portable TOC Analyzer, Suez Water Technologies and Solutions, Trevoze, PA), equipped with a custom-made column (250 × 20 mm) and Toyopearl HW-50S packing material (Tosoh Bioscience, Tokyo, Japan). Polyethylene oxide with molecular weights ranging from 194 to 100 k Da was used as molecular weight standards (Polymer Standards Service, Mainz, Germany).

A reverse-phase chromatography method using Agilent 1260 II HPLC with an in-line diode array detector (Agilent 1260 DAD) equipped with an Agilent Poroshell 120 EC-C18 column was used for organic hydrophobicity assessment. Peaks corresponding to a retention time ranging from 0 to 5 min were considered hydrophilic, and from 5 to 35 min were considered hydrophobic. Details of the analytical method have been previously published [76].

An ultra-HPLC (Agilent 1260) coupled with tandem mass spectrometer (MS/MS) (Agilent 6490) (Agilent Technologies, Palo Alto, CA) was used to quantify ten trace organic contaminants (TOCs). An Agilent ZORBAX Eclipse Plus reverse-phase C-18 RRHD column (2.1 × 50 mm, 1.8 μm particles) was used to separate analytes. The detailed procedure and information of the analytical method can be found in previous work [77,78]. A list of trace organics that were quantified with their physicochemical properties is presented in Table 2.

## 2.7. Specific energy consumption

The electricity usage (kWh) of the engineering-scale system was obtained from daily recorded data by a power meter. The actual specific energy consumption ( $SEC_a$ ) was calculated in kWh/m<sup>3</sup> as:

$$SEC_a = \frac{\text{Energy used}}{Q_p} \quad (6)$$

where  $Q_p$  is the permeate water flow rate. Additionally, the theoretical specific energy consumption ( $SEC_{th}$ ) was calculated as:

$$SEC_{th} = \frac{\sum (Q_f * \text{System pressure})}{\sum (Q_p)} \quad (7)$$

where  $Q_f$  is the feed flow rate and system pressure is the inlet pressure of the feed stream.

## 3. Results and discussion

### 3.1. Bench-scale experiments

#### 3.1.1. Apparent water permeability coefficient

Prior to engineering-scale testing, membranes were tested on bench-scale systems to evaluate their water permeability (Fig. 2a) and permeate conductivity (Fig. 2b), as conductivity is a direct and practical indicator of solute rejection by the membrane. As expected, using a membrane with a looser structure (NF270) resulted in higher apparent water permeability coefficient and permeate conductivity (~ 18 LMH/bar and ~ 750 μS/cm, respectively). In comparison, the dense NF (NF90) apparent water permeability coefficient and permeate conductivity were observed to be substantially lower (~ 10 LMH/bar and ~ 60 μS/cm, respectively), while the BW30XFR membrane results showed the lowest apparent water permeability coefficient and permeate conductivity (~ 6 LMH/bar and 40 μS/cm, respectively). The differences in conductivity observed between the NF270 and NF90 membranes could be attributed to differences in membrane structure, such as molecular weight cut-off (MWCO), surface roughness, and pore size. Interestingly, although the apparent water permeability coefficient of NF90 was higher than BW30XFR, similar permeate conductivity was observed for both membranes.

#### 3.1.2. Solute passage

Given the differences in permeate conductivity observed between NF90 and NF270 membranes, analyses were conducted to quantify the solute concentrations in the permeate water, including cations, anions, TOC, and total fluorescence, along with their corresponding passage rates (Fig. 3). Higher concentrations of contaminants were observed in the permeate using NF270 membrane compared to the NF90 membrane, as shown in Fig. 3a. While the passage of organics was similar between the two NF membranes, a substantial difference was observed in the passage of inorganic ions. For NF270, a low passage rate (<10 %) was observed for sulfate, whereas higher passage rates (~30 %) were observed for other divalent ions such as calcium and magnesium, with monovalent ions passing through more easily (Fig. 3b). Sulfate is negatively charged and has the highest molecular weight compared with

**Table 2**  
Analyzed trace organic contaminants.

Compound	MW (g/mol) <sup>a</sup>	LogK <sub>ow</sub> <sup>a</sup>	Charge <sup>b</sup>	Average feed water concentration (ng/L)	Limit of detection (ng/L)
Gemfibrozil	250.34	4.77	–	600	5
Ibuprofen	206.29	3.97	–	1,000	2
Sulfamethoxazole	253.28	0.89	–	3,000	5
Atenolol	266.34	0.43	+	220	2
Diltiazem	414.52	2.73	+	300	2
Diphenhydramine	255.36	3.65	+	40	1
Fluoxetine	309.33	3.82	+	850	1
Propranolol	259.35	2.58	+	30	2
Trimethoprim	290.32	1.28	+	90	1
Benzotriazole	119.13	1.3	o	12,000	5
Caffeine	194.19	–0.07	o	100	2
Carbamazepine	236.27	2.45	o	90	1
Hydrochlorothiazide	297.73	–0.58	o	8,000	5
Norgestrel	312.45	4.25	o	25	5
Sucralose	397.63	–0.47	o	35,000	1
Tris(chloropropyl) phosphate (TCPP)	327.56	3.36	o	700	20

+, positive; –, negative; and o, neutral.

<sup>a</sup> Data from [79].

<sup>b</sup> Data from [80].

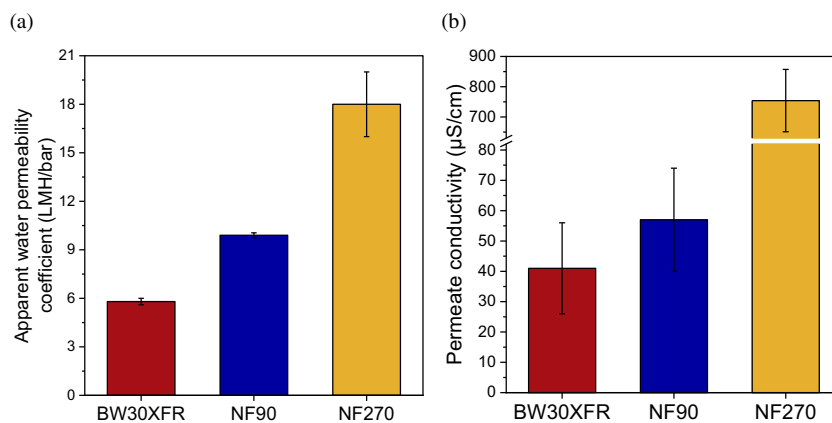


Fig. 2. Average values of a) apparent water permeability coefficient and b) permeate conductivity of BW30XFR, NF90, and NF270 membranes for all tested water flux conditions (35, 25, and 15 LMH). The error bars illustrate deviations from the average values determined from the varying water flux conditions. NF membranes have a more porous structure, leading to higher water permeability and permeate conductivity.

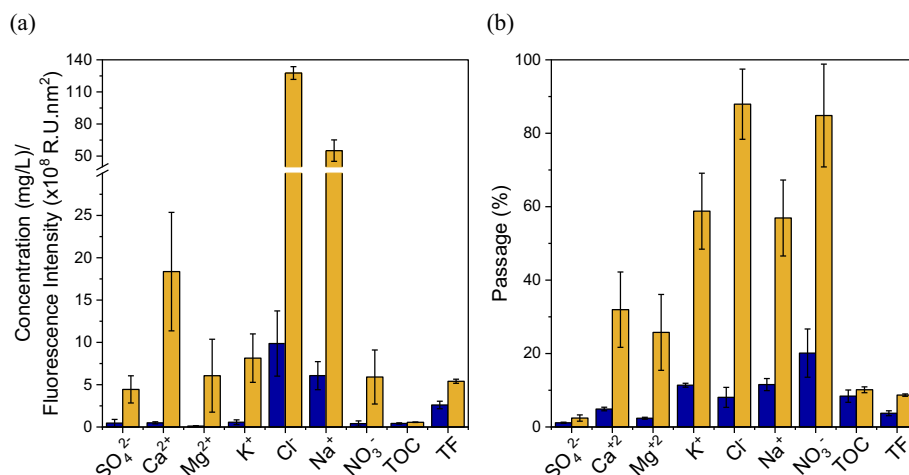


Fig. 3. NF90 (blue) and NF270 (yellow) membranes a) permeate concentration or fluorescence intensity (for TF) and b) passage for each solute. Organic passage was relatively similar between the two NF membranes, while inorganic passage was much higher for the NF270 membrane.

the other ions, while magnesium was rejected more than calcium due to the lowered smaller ionic radius [81]. The observed higher passage of monovalent ions is likely due to lower ionic radius compared with

divalent ions. In contrast, the NF90 membrane had a passage rate below 5 % for divalent ions and a slightly higher passage rate for monovalent ions. The higher solute passage observed in the NF270 membrane was

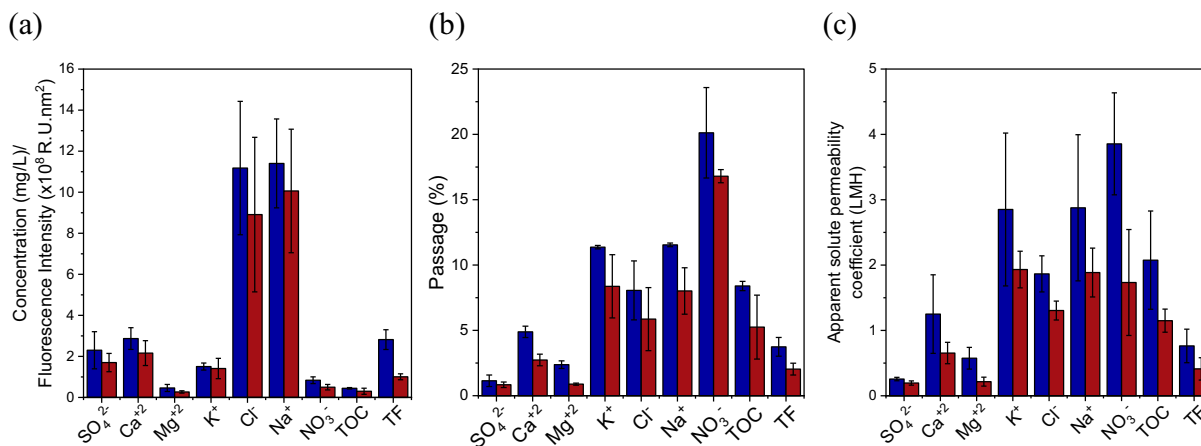


Fig. 4. NF90 (blue) and BW30XFR (red) membranes a) permeate concentration or fluorescence intensity (for TF), b) passage for each solute, and c) apparent solute permeability coefficient for each contaminant. The permeate concentration and passage of divalent ions were similar between NF90 and BW30XFR membranes, but higher passage of TOC and monovalent ions was observed for NF90.

attributed to its more porous structure compared to the NF90 membrane [14]. For organics, the NF270 membrane removed a large portion of TOC with a passage rate of approximately 10 %, yet the resulting permeate TOC concentration of 0.6 mg/L exceeded stringent regulatory limits, such as those set by the California Department of Public Health (CDPH) at 0.5 mg/L [82].

In comparing NF90 and BW30XFR, the concentrations of each solute in the permeate water, their corresponding passage rates, and apparent solute permeability coefficient are illustrated in Fig. 4. Interestingly, solute concentrations in the permeate water from NF90 and BW30XFR membranes were similar for most contaminants (Fig. 4a). For solute passage, divalent ions passage rates were similar between both membranes, while higher passage rates were observed for TOC and monovalent ions in NF90 membrane compared to BW30XFR (Fig. 4b). Divalent ions are more effectively rejected than monovalent ions in NF90 due to the membrane's selectivity and the physical characteristics of ions (ionic charge, density, ionic radius, and hydrated radius) [83]. Additionally, the solute apparent permeability coefficient was similar between the two membranes (Fig. 4c). Comparable divalent ion permeabilities were observed between NF90 and BW30XFR (approximately 0.2, 0.3, and 0.6 LMH for sulfate, magnesium, and calcium, respectively). In contrast, permeabilities for monovalent ions and organic solutes were considerably higher for NF90 compared to BW30XFR, with nitrate having the highest ion permeability coefficient. Nitrate's low ionic charge density and hydration energy facilitate faster transport and, consequently, lower charge repulsion interactions, resulting in higher passage [84].

### 3.1.3. Organic characterization assessment

To characterize the hydrophobicity of organic compounds, HPLC chromatograms of feed and permeate water from NF90 and BW30XFR membranes are presented in Fig. 5a. All detected peaks were observed at low retention times (below 5 min), which corresponds to the hydrophilic materials [76]. Most organic compounds detected in the feed water were relatively hydrophilic. Both NF90 and BW30XFR membranes effectively rejected the majority of hydrophilic compounds. Additionally, size-exclusion chromatography fingerprints indicated a similar apparent molecular weight (AMW) of organics detected in NF and RO permeate (Fig. 5b). Organics with apparent molecular weights below 50 kDa were detected in the feed water, with predominant apparent molecular

weights ranging between 10 and 1 kDa (Fig. 5b inset plot). In NF90 and BW30XFR permeate, the apparent molecular weight of organics was smaller than 1.7 kDa, with a higher response area for NF90, as shown in Fig. 5b. These findings highlight size exclusion as a primary mechanism for rejecting organic compounds in both NF90 and BW30XFR membranes.

### 3.1.4. Trace organic contaminants analysis

For the evaluation of NF90 and BW30XFR membranes in terms of

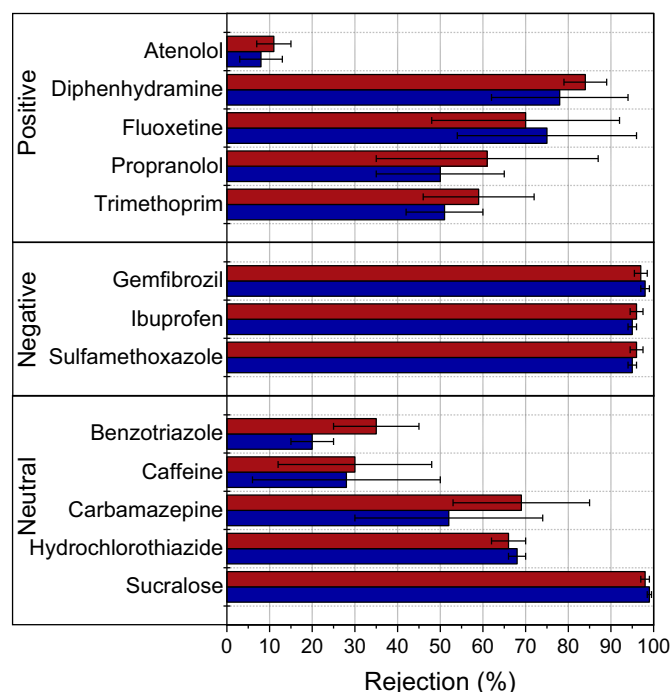


Fig. 6. Rejection of TOCs by NF90 (■) and BW30XFR (■) membranes in bench-scale experiments. TOCs are categorized into different groups based on their charge: neutral, negative charge, or positive charge. TCCP, diltiazem, and norgestrel were only detected in the feed water and not in the permeate.

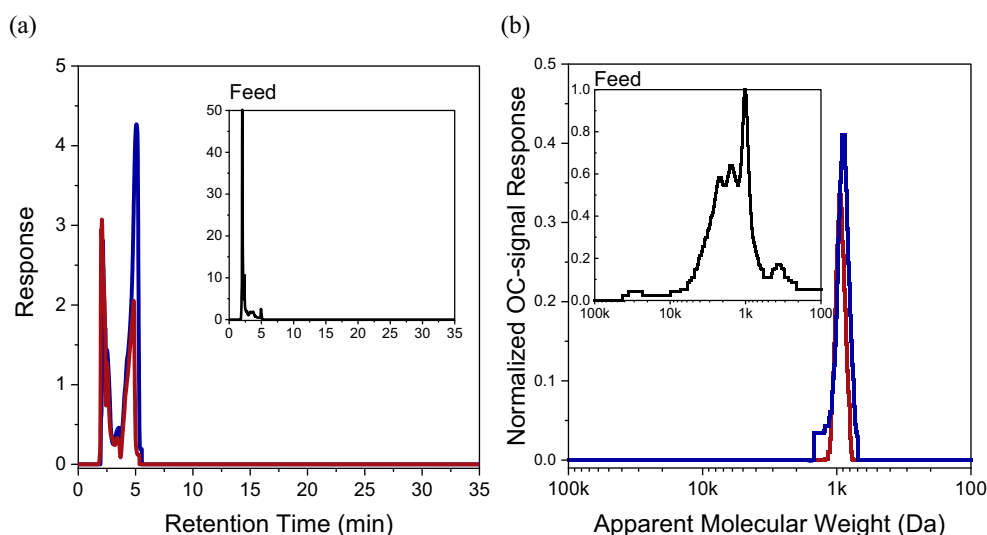


Fig. 5. a) HPLC chromatograms of NF90 (—) and BW30XFR (—) membranes permeate and feed water. Peaks detected within 5 min are considered hydrophilic, and >5 min are hydrophobic [76]. Interestingly, a similar peak response was observed for both membranes, indicating that organics in NF90 and BW30XFR permeates are predominantly hydrophilic. b) Normalized size-exclusion chromatograms of NF90 (—) and BW30XFR (—) membranes permeate and feed water (inset plot) as a function of apparent molecular weight. Both membranes had relatively similar chromatograms with apparent molecular weight of organics in the permeate below 1.7 kDa, with a higher response area for NF90. NF90 and BW30XFR permeate data were normalized based on feed water signal response.

trace organic compounds, Fig. 6 presents the rejection percentages of each compound by both membranes. Although concentration limits for TORCs in the aquatic environment have not been established, permeate concentrations varied from a few nanograms per liter to several micrograms per liter. Overall, the rejection of negatively charged compounds such as ibuprofen, sulfamethoxazole, and gemfibrozil was higher than that of neutral and positively charged TORCs. Although these negatively charged compounds have smaller molecular weights, and despite their hydrophilicity/hydrophobicity properties, the observed rejection was high (>96 %) by both membranes, which suggests that the primary rejection mechanism for negatively charged compounds is electrostatic exclusion. This phenomenon can be attributed to the repulsive forces between negatively charged solutes and the membrane surface, which hinder the solutes from engaging in hydrophobic interactions [85]. On the other hand, the rejection of positively charged compounds, including propranolol, fluoxetine, diphenhydramine, trimethoprim, and atenolol, varied from 10 % to 90 %. Rejection of propranolol, fluoxetine, and diphenhydramine, with  $\log K_{ow}$  of 2.58, 3.82, and 3.65, respectively, were higher than trimethoprim and atenolol, with respective  $\log K_{ow}$  of 1.25 and 0.43. Generally, more hydrophobic compounds tend to be better rejected than hydrophilic compounds. Moreover, negatively charged membranes can promote charge attraction with positively charged compounds, which could result in lower rejection rates for positively charged compounds. Electrostatic interactions between the negatively charged membrane and positively charged compounds might reduce their rejection efficiency due to limited electrostatic repulsion [86]. Furthermore, moderate rejection was observed for neutral compounds (below 65 %), such as hydrochlorothiazide and carbamazepine. However, the high rejection of sucralose, which has the highest molecular weight among the studied TORCs, was high (>95 %) despite its neutral and highly hydrophilic nature. In contrast, poor rejection was observed for caffeine and benzotriazole, which could be attributed to their lower molecular weights. Overall, variations in rejection among these compounds reflect a combination of polarity, charge, and size characteristics.

To examine the influence of polarity in rejection of TORCs, Fig. 7 illustrates the linear correlation between compound rejection and

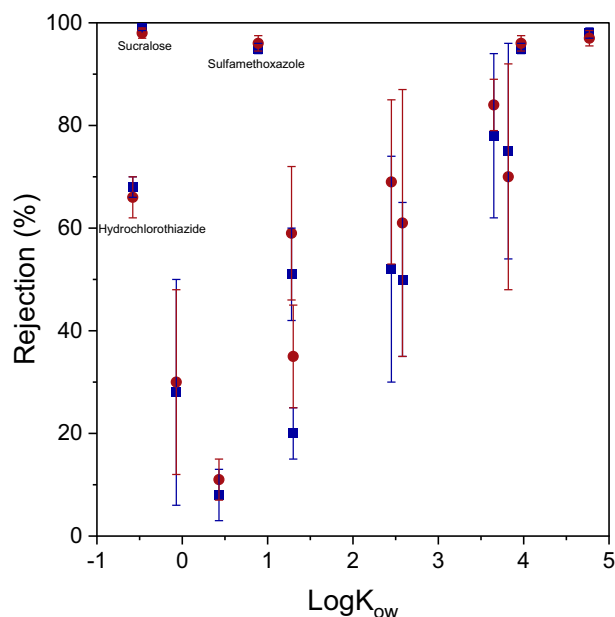


Fig. 7. Trace organics rejection rate by NF90 (■) and BW30XFR (●) membranes as a function of their partition coefficient ( $\log K_{ow}$ ). Excluding the high molecular weight and negatively charged compounds, there is a relatively linear correlations between the rejection of TORCs and their corresponding  $\log K_{ow}$  values.

$\log K_{ow}$  value, with the exception of sulfamethoxazole, sucralose, and hydrochlorothiazide, where the high rejection is likely due to charge and size exclusion. It is noteworthy that other compounds, including TCP, diltiazem, and norgestrel were only detected in the feed water and not in the permeate, indicating rejection rates of 97 %, 99 %, and 80 % respectively (based on detection limits of the analytical instrument). These compounds are hydrophobic with relatively high molecular weights, contributing to their non-detection in the permeate. Overall, the results demonstrate that NF90 membranes can provide high rejections of TORCs.

### 3.2. Engineering-scale experiments

#### 3.2.1. Apparent water permeability coefficient

Following bench-scale tests on organic and inorganic passage, RO (TW30) membranes were replaced with NF9 membranes in the engineering-scale system, which operated continuously for over a year. Fig. 8 illustrates the apparent water permeability coefficient (Fig. 8a) and apparent solute permeability coefficient based on conductivity (Fig. 8b) during operation. The apparent water permeability coefficient greatly decreased over time primarily due to membrane compaction (membrane stabilization) and irreversible fouling, leading to denser structure that hindered water flow. Additionally, the presence of organic and inorganic compounds led to their attachment to the membrane surface, further reducing its permeability. Highly selective NF membranes like NF90, characterized by rough surfaces, high rejection rates, and high apparent water permeability coefficients, typically has a severe initial reductions in apparent water permeability coefficient [64]. Despite the lower apparent water permeability coefficient, the membrane maintained its selectivity, as evidenced by the relatively low apparent solute (conductivity) permeability coefficient, similar to TW30. Furthermore, the apparent water permeability coefficient observed during engineering-scale experiments was roughly half of what was observed in bench-scale experiments. This discrepancy suggests that factors such as membrane compaction, concentration polarization, or differences in operating conditions could have influenced membrane water permeability.

#### 3.2.2. Solute passage

The average solute concentration in permeate water and the passage

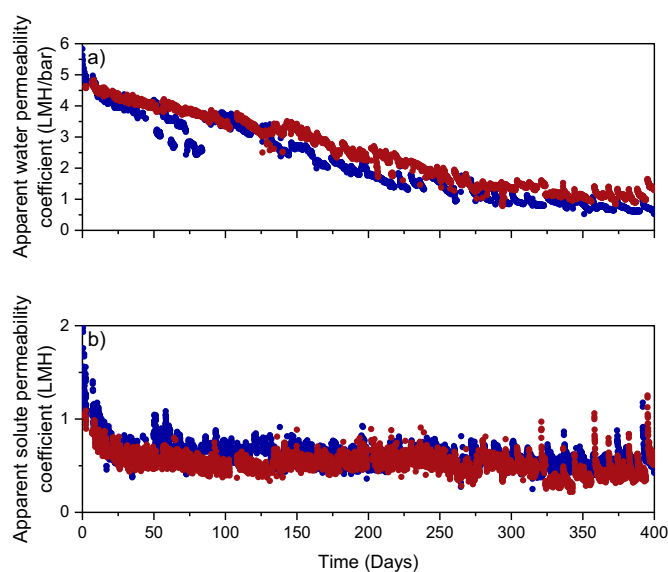


Fig. 8. Illustration of NF9 (●) and TW30 (●) membranes during operation displaying a) apparent water permeability coefficient (A), and apparent solute permeability coefficient based on conductivity (B) as a function of operating time. Each data point represents calculated daily average values.

rates of each solute during engineering-scale testing are shown in Fig. 9. The solute concentrations in the permeate water were lower or similar to those observed in bench-scale tests (Fig. 9a). Moreover, a slightly lower solute passage rates were observed in the engineering-scale experiments (Fig. 9b). Divalent ions were effectively rejected with passage rates below 2 % and maintained low passage levels throughout the operational period, while monovalent ion passage rates exceeded 5 %. Throughout long-term testing, all analyzed permeate samples maintained relatively constant TOC concentrations below 0.4 mg/L.

### 3.2.3. Trace organic contaminants analysis

The observed rejection of TORCs during engineering-scale testing was relatively higher compared to bench-scale experiments (Fig. 10). This difference can be attributed to factors such as membrane compaction, membrane fouling, and operation conditions, all contributing to improved rejection efficiency of TORCs. Moreover, membrane fouling and long-term operation may alter membrane surface properties, including hydrophobicity and charge, potentially affecting contaminant rejection compared to unfouled membranes. Various studies have suggested that membrane fouling and the coexistence of contaminants can enhance the rejection of TORCs [67,87]. However, it is important to note that adsorption may not be a sustainable long-term rejection mechanism, as membranes can become saturated, limiting their capacity for further adsorption [69]. Moreover, deposition of divalent cations (calcium and magnesium) on the membrane surface can neutralize membrane charge, weakening electrostatic attraction and repulsion forces between ionic TORCs and the negatively charged membrane [88]. These results demonstrate the potential of NF9 membrane as a viable alternative to RO membranes for potable water reuse applications with stringent TOC requirements.

### 3.2.4. Specific energy consumption

Fig. 11 illustrates the actual ( $SEC_a$ ) and theoretical ( $SEC_{th}$ ) specific energy consumption of NF9 and TW30 membranes as a function of operating time. The energy consumption increased over time primarily due to membrane fouling. Initially, within the first 150 days, the actual SEC for NF9 ( $\sim 0.7$  kWh/m<sup>3</sup>) was approximately half that of the TW30 membranes ( $\sim 1.6$  kWh/m<sup>3</sup>). However, NF9's  $SEC_a$  started to increase thereafter, eventually reaching similar energy consumption levels to TW30 membranes. Additionally, the theoretical SEC for NF9 was slightly lower than that of RO during the initial operation (first fifty days). However, as operation progressed, the  $SEC_{th}$  of NF9 gradually increased, ultimately exceeding TW30 energy consumption. These findings suggest that the NF9 membrane had severe fouling, leading to a progressive decrease in water permeability and an increase in energy

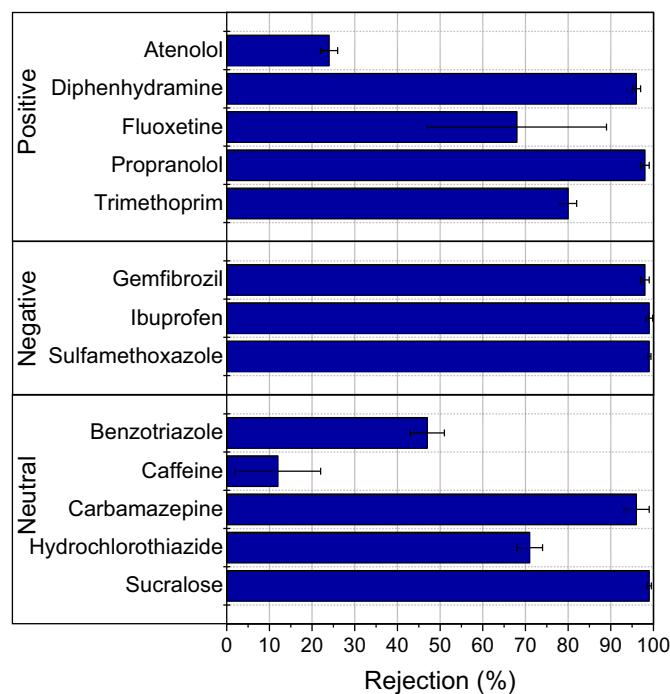


Fig. 10. Rejection of TORCs grouped based on their charge at NF9 engineering-scale membrane during one year of operation. Rejection of negatively charged TORCs was higher than neutral or positively charged compounds. Bars represent average values.

consumption. It is noteworthy that the regular cleaning procedures remained consistent for both NF9 and TW30 membranes. Accordingly, these results suggest the possibility of adapting cleaning protocols tailored to NF membranes to improve the efficiency of NF operations, mitigating challenges of rapid fouling while maintaining higher permeability. Bellona and coworkers [74] reported that NF90 had a large water flux decline during pilot testing due to fouling, resulting in minimal energy differences compared to TW30 membranes. These findings highlight the trade-offs in managing fouling, optimizing energy efficiency, and achieving effective contaminant rejection in NF membrane applications.

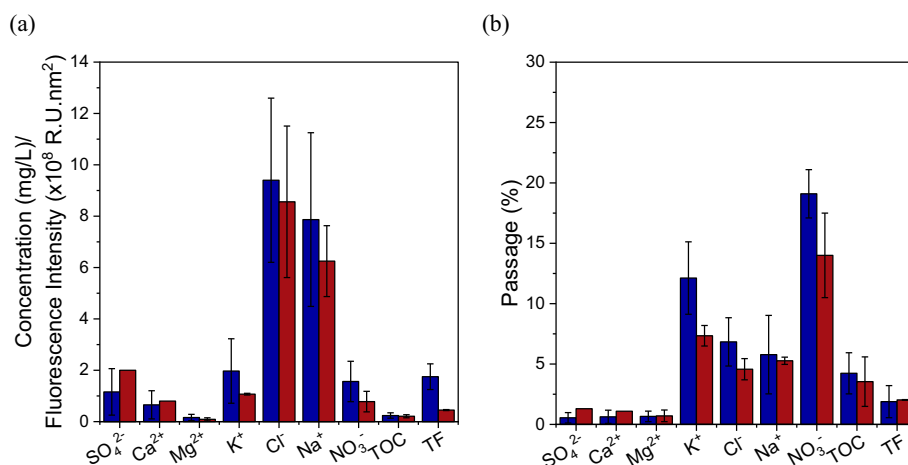


Fig. 9. NF9 (■) and TW30 (■) membranes a) permeate concentration or fluorescence intensity (for TF) and b) passage for each solute. Bars represent the calculated average values. Passage of both organics and divalent ions was lower than monovalent ions.



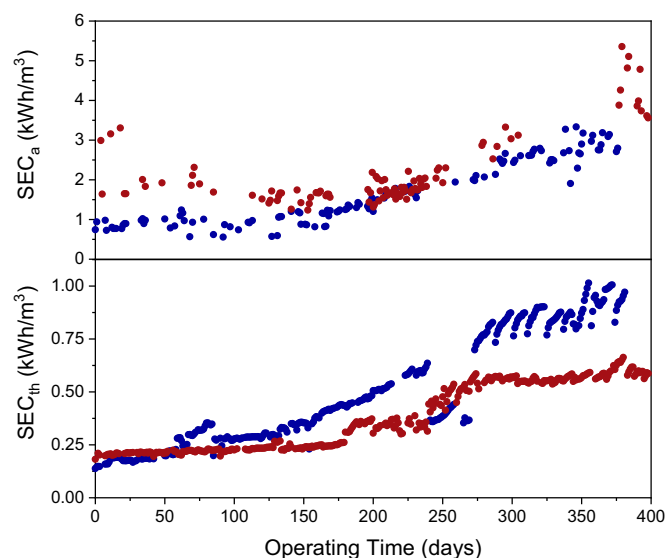


Fig. 11. Actual ( $SEC_a$ ) and theoretical ( $SEC_{th}$ ) specific energy consumption of NF9 (●) and TW30 (●) membranes in the engineering-scale system during continuous operation. The observed trend demonstrates an increase over time, despite disparities between actual and theoretical SEC values.

#### 4. Conclusion

While RO membranes are widely used and remain essential for water treatment, especially for highly saline waters, potable water reuse applications often involve treating reclaimed water with relatively low TDS. In these cases, there is an opportunity for NF to be employed. The higher operational costs associated with RO, due to its high-pressure operation, may not be justified by the additional treatment benefits it provides compared to NF. Nevertheless, despite the similar contaminant passage rates between dense NF and RO membranes, high water permeability decline and minimal energy savings due to high fouling propensity could limit the use of NF membranes as a substitute for RO. Effective fouling management will be essential for advancing the practical implementation and sustainability of NF in potable water reuse applications. Although NF could be a feasible alternative to RO in water reuse applications, it is important to consider the characteristics of the treated wastewater, such as high salinity, which could pose challenges for NF membranes. The membrane selection would be dependent on the specific application requirements. In this study, bench-scale experiments revealed high TOC passage rates for both NF90 and NF270 membranes; additionally, NF270 rejected divalent and monovalent ions poorly. The same passage rates were observed between the NF90 and BW30XFR for divalent ions, and slightly higher passage rates for monovalent ions. For organics, TOC passage results were slightly higher for NF90 compared to BW30XFR; however, both membranes had similar apparent molecular weight distributions of organics in the permeate. Moreover, no large differences were observed in the rejection of the studied TOCs between NF90 and BW30XFR membranes. Furthermore, when the NF9 membrane was tested in the engineering-scale system, it consistently achieved lower passage rates for all organic and inorganic compounds and maintained low contaminant passage rates during long-term operation. Additionally, NF9 specific energy consumption was lower during the initial stages of operation, and it progressively increased with continued operation, eventually reaching similar energy consumption levels as TW30 membranes. While challenges such as high fouling rates and limited energy savings need to be addressed, the increasing demand for sustainable and economically efficient processes, along with precise separation, highlights the potential of NF technology in advancing sustainable water practices.

#### CRediT authorship contribution statement

**Mohammed A. Alhussaini:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Bianca M. Souza-Chaves:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Varinia Felix:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Andrea Achilli:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

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