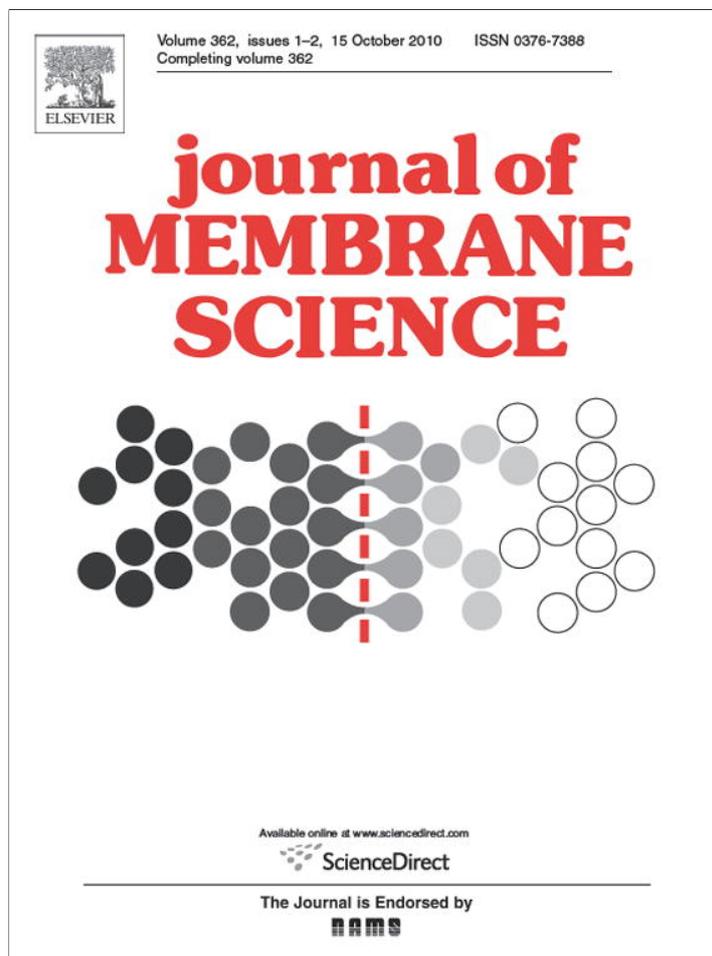


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A multi-barrier osmotic dilution process for simultaneous desalination and purification of impaired water

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ABSTRACT

Osmotically driven membrane processes use chemical potential difference between two aqueous solutions as a driving force for separation and concentration of feed streams or for recovery of energy. Forward osmosis was extensively studied in recent years and demonstrated that it can be successfully utilized in many applications of water treatment. In a new approach, the salinity difference between seawater and impaired water, such as reclaimed water, is used as a driving force for osmotic dilution of seawater before desalination. The osmotic dilution approach may provide at least four major benefits related to water and energy resources. These include lower energy desalination of seawater, multi-barrier protection of drinking water, reduction in reverse osmosis membrane fouling due to impurities in impaired water, and beneficial reuse of impaired water. The osmotic dilution process was investigated on both bench- and pilot-scales. Both secondary and tertiary treated effluents from a domestic wastewater treatment plant and impaired surface water were used as feed water to the process. Flux decline was minimal and under specific flow condition it was observed that membrane fouling was negligible. Additionally, the multiple membrane barriers provided high rejection of both organic and inorganic solutes and of trace organic chemicals.

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1. Introduction

Increasing water demands and diminishing water supplies due to over allocation and contamination are forcing many water authorities to explore unconventional methods of producing drinking water in higher quantity and at lower cost [1,2]. Two such options for augmenting water supplies are desalination of seawater or brackish water [3,4] or reuse of reclaimed water [5]. Among different treatment technologies, membrane desalination processes, such as reverse osmosis (RO) and nanofiltration (NF), are capable of rejecting most dissolved constituents, including trace organic chemicals [6,7]. Provided that adequate pretreatment is implemented, these processes can also produce high quality potable water from reclaimed water or from surface or ground water impacted by discharge from wastewater treatment plants [7,8].

Despite the prominence of RO and NF for desalination and treatment of impaired water, these technologies are not without limitations. High susceptibility to membrane fouling and scaling may require intensive chemical cleaning that can increase treatment cost and decrease membrane lifetime, while the use of scale inhibitors can add cost and complexity to system operation

[9]. Operating at elevated pressures increases both capital and operating costs compared to conventional water treatment [10].

In many cases RO is coupled with advanced oxidation processes (e.g., UV-AOP) to create multiple barriers to low molecular weight organic contaminants [5]; yet, these systems have high capital and O&M cost and they are energy intensive. Therefore, there is a growing need to design water treatment systems using alternative and simple processes that provide multiple barriers without substantially increasing chemical and energy demands.

1.1. Energy demand in water treatment processes

High energy demand has limited the use of desalination in many regions [11]. Current methods to reduce the energy required for desalination include incorporation of energy recovery devices (ERDs) or use of selective but more permeable membranes [12]. Yet, energy costs still contribute as much as 75% of the operating costs of desalination plants, or between 30 and 50% of the product water cost [10].

Dilution of saline water provides another method to reduce energy demand during RO desalination of highly saline water; dilution decreases the osmotic pressure that must be overcome to produce RO or NF permeate [13]. The relatively low salinity of most impaired waters makes them good candidates for use in diluting saline streams before desalination [5,14]; however, direct dilution

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may contaminate and alter the chemistry of the feed stream to the desalination process, accelerate membrane fouling, and subsequently may reduce product water quality. Therefore, adequate pretreatment of impaired water is necessary before desalination of diluted saline water; yet, many advanced pretreatment processes may be chemically or energy intensive [15].

In an alternative approach [16], the chemical potential of a saline stream can be used as a driving force for osmotic dilution in which water from an impaired stream spontaneously diffuses through a semipermeable membrane into the saline stream. This approach may provide several benefits related to water and energy resources; these include dilution of seawater before desalination that results in lower energy desalination, multi-barrier protection of drinking water, reduced RO membrane fouling due to impurities in impaired water, and an opportunity for safe and beneficial reuse of impaired water.

One of the main advantages of osmotically driven membrane processes (ODMP) (i.e., forward osmosis (FO) and osmotic dilution) is that they require limited external energy to perform separation of solvent and solutes because only very low hydraulic pressure is used to induce flow of the feed and draw solution on either side of the membrane [17]. Also, in osmotic dilution the draw solution can be a readily available renewable source (e.g., seawater, concentrated brackish water) that has relatively high osmotic pressure.

Furthermore, unlike FO, osmotic dilution does not require closed loop draw solution reconcentration; instead, the process can operate with both the feed and draw solution in a once through flow configuration [17]. This unique operating configuration provides the same benefits of FO (e.g., low energy operation and high resistance to irreversible fouling), while simultaneously eliminating energy costs associated with draw solution reconcentration and reducing operational complexities associated with bi-directional solute diffusion from and to the reconcentration process [18]. A seawater intake or a brackish water desalination plant can readily supply fresh draw solution; and because the draw solution is not recycled in the process, accumulation of sparingly soluble salts in the draw solution is not expected.

The magnitude of water flux (J_w) through the membrane during pressure driven and osmotically driven membrane processes depends on the difference in water chemical potential, and by extension the difference in osmotic pressure, across the semipermeable membrane [19]. Water flux is frequently expressed by the same general equation used for RO:

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

where A is the pure water permeability coefficient of the membrane, ΔP is the hydraulic pressure difference across the membrane (approximately zero in most ODMP), and $\Delta\pi$ is the difference in osmotic pressure across the membrane. Yet, lower water fluxes than estimated by Eq. (1) are observed during osmotic dilution of seawater with impaired water, and it is attributed to internal and external concentration polarization phenomena in and on the membrane. Past studies [20–22] have demonstrated that when accounting for internal concentration polarization (assuming negligible osmotic pressure in the feed and low salt permeability coefficient of the membrane), water flux can be expressed by:

$$J_w = A(\pi_D \exp(-J_w K)) \quad (2)$$

where π_D is the bulk draw solution osmotic pressure and $K(=t\tau/D\varepsilon)$ is the solute resistivity to diffusion within the porous support layer, and is a function of membrane thickness (t), support layer pore tortuosity (τ) and porosity (ε), and solute diffusivity (D) [21]. From Eq. (2) and the definition of K it is apparent that the osmotic dilution process is limited by the resistivity to diffusion of the draw solution solutes within the porous support layer. Because current ODMP membranes are not optimized, it is anticipated that future

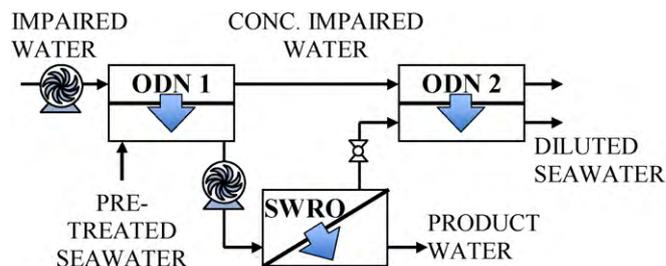


Fig. 1. Schematic drawing of a novel hybrid osmotic dilution (ODN)/SWRO process to reduce energy demand of SWRO, increase water recovery, improve beneficial use of impaired water, and mitigate stress to marine environments near outflow structures.

membranes with higher water permeability (A) would make the process even more practical. Despite the low water flux through current ODMP membranes, near future implementation of osmotic dilution to enhance seawater RO (SWRO) desalination is possible and economically viable.

1.2. Coupling osmotic dilution and SWRO

With the new approach, osmotic dilution may be employed to reduce the osmotic pressure of seawater prior to desalination by RO using a co-located source of impaired water; a secondary goal will be volume-minimizing treatment of the impaired water stream. An example of a dual stage osmotic dilution coupled with SWRO is illustrated in Fig. 1. Pretreated seawater is osmotically diluted by an impaired water stream. Pretreatment of the seawater is still required despite its osmotic dilution because contaminants in the seawater will ultimately contact the RO membrane. Before discharge, the concentrated SWRO brine can be osmotically diluted in the ODN2 by the concentrated impaired water, or it can be used in a pressure-retarded osmosis system to recover energy (not shown in Fig. 1) [23]. The concentrated impaired water can be used for beneficial purposes (e.g., further dewatering to produce high grade fertilizer [24]), or returned to a wastewater treatment plant for retreatment. Furthermore, the SWRO concentrate is diluted in the second osmotic dilution stage, reducing the environmental impacts associated with discharging concentrated brines back to the ocean.

1.3. Objectives

The main objective of this study was to investigate mass transport and performance of osmotic dilution in conjunction with SWRO desalination and wastewater reclamation. Specifically, the goals of the study were to perform a thorough investigation of membrane fouling tendency, to demonstrate long-term process performance, to investigate rejection of constituents of concern, such as inorganic compounds, nutrients, and trace organic chemicals, and to perform preliminary economic evaluation of the hybrid process. Although osmotic dilution is the topic of investigation in this study, experiments were conducted in FO mode (i.e., closed loop draw solution configuration) because continuous supply of seawater was not readily available at the locations where tests were conducted. Therefore, the term FO is used throughout this paper in accordance with this mode of operation. Nevertheless, the impaired water feed to the process was a side-stream from a water reclamation facility and the draw solution concentration was varied to simulate the conditions in different stages of a full-scale osmotic dilution process.

ODMP were tested in the past with highly impaired feed streams [24–29]. Results from these studies indicate that FO membranes are less susceptible to irreversible fouling compared to pressure-driven membrane processes because cake layer compaction due to

hydraulic pressure is negligible [24,27,28], and because FO membranes are highly hydrophilic [17]. However, these studies were mostly limited to bench-top investigations conducted over relatively short timeframes and with batches (rather than a continuous side-stream) of impaired water. In the current study both bench- and pilot-scale systems were used to investigate long-term performance of osmotic dilution using FO membranes with various impaired water feed sources.

2. Materials and methods

2.1. Membranes

Hydration Technology Innovations (Albany, OR) supplied FO membranes for this study. Membranes from a single production cast were used to minimize variability in results due to small differences in membrane characteristics between manufacturing batches. Flat sheet FO membranes were used in both bench- and pilot-scale experiments. SWRO membranes (SW30-2540, Dow Filmtec, Edina, MN) were chosen for the RO pilot system and were used in the pilot-scale RO-FO experiments. These membranes were chosen for their high salt rejection (99.4% NaCl rejection [30]) and high operating pressure that enable production of concentrated draw solution.

2.2. Bench-scale experiments

2.2.1. Bench-scale system design and operation

Baseline performance and the effect of feed water quality on water flux through the FO membrane were determined through a set of bench-scale experiments. A system was designed and constructed for this study with a membrane cell having an effective membrane surface area of 0.062 m² (four parallel channels 555 mm long, 29 mm wide, and 1.4 mm deep). A schematic drawing of the system is illustrated in Fig. 2. Feed and draw solution were recirculated from their respective tanks at 1.4 L/min through the FO membrane cell and back to the tanks using a rotary vane pump.

In-line sensors were used to measure solution conductivity, temperature, pH, and pressure at the inlets and outlets of the membrane cell. A supervisory control and data acquisition (SCADA) system was developed to record and control the system's operating conditions (UE9-Pro, LabJack Corporation, Lakewood, CO; and LabVIEW, National Instruments, Austin, TX). The concentration of the draw solution was maintained constant within ± 125 mg/L TDS of a set point value by intermittently dosing concentrated synthetic sea salt solution into the draw solution tank. Feed and draw solution temperatures were controlled by the SCADA system and were maintained at 19.0 ± 0.1 °C. Sodium hydroxide and hydrochloric acid (Fisher Scientific, Waltham, MA) were intermittently dosed to maintain the pH of the feed solution between 7.0 and 7.5.

The original concentration of feed water constituents was maintained constant during each experiment by replenishing permeating water with deionized water. This was accomplished by maintaining constant volume of solution in the feed tank using a float valve connected to a deionized water reservoir. The reservoir was positioned on an analytical balance that was also connected to the SCADA system, and water flux through the FO membrane was calculated by measuring the change in weight of the deionized water in the reservoir.

2.2.2. Bench-scale draw solution and feed solutions

Additional experiments were conducted to determine reverse diffusion rates of salts from the draw solution into the feed. Draw solution was prepared with synthetic seawater salt (Instant Ocean,

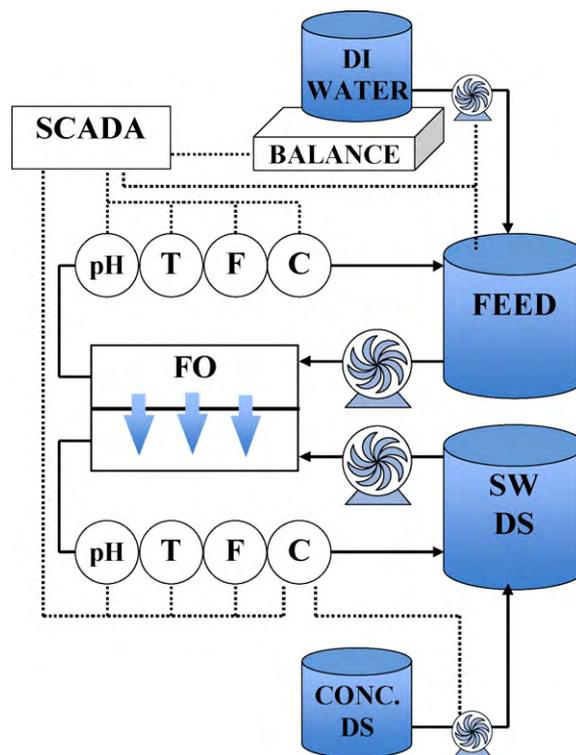


Fig. 2. Schematic drawing of the FO bench-scale system. Circles signify conductivity (C), pressure (P), flow rate (F), and pH probes installed in the draw and feed solutions loops.

Mentor, OH) dissolved in deionized water. Draw solution concentrations ranging from 5 to 35 g/L in increments of 5 g/L and of 70 g/L were used in these experiments. Prior to each experiment, both draw and feed solution loops were flushed with deionized water. Experiments were conducted for approximately 4 h.

Experiments were also conducted with three types of impaired feed water qualities, including secondary and tertiary treated effluents from the Denver water recycling plant (DWRP, Denver, CO) and effluent-impacted surface water from the South Platte River downstream of Denver, CO. Experiments were used to determine the effect of feed water quality on process performance, including water flux, membrane fouling, and solute rejection.

2.2.3. Measuring solute transport

Feed conductivity was continuously monitored and recorded during bench-scale tests. In ODMP the feed conductivity increases because draw solution solutes diffuse into the feed solution [18]. The feed conductivity increased at a constant rate during all experiments in this study. Specific reverse TDS flux was calculated by measuring the rate of feed conductivity increase (in mS-cm/m²-h), converting to mass flux (mg-TDS/m²-h), and dividing by the water flux through the FO membrane. This calculation provides a measurement of the mass of TDS that diffuses into the feed solution per liter of feed diffused into the draw solution (i.e., units of mg/L). During one experiment (35 g/L seawater draw solution with deionized water feed), samples for analysis of the feed were drawn at the beginning and end of the experiment. Analytical results were used to verify the specific reverse TDS flux derived from conductivity measurements and to provide further detail on the relative contribution of each draw solution solute. Ion analysis was conducted using a Dionex DC80 ion chromatography (IC) system (Dionex, Sunnyvale, CA) for anions, and Optima 3000 inductive coupled plasma (ICP) spectrophotometer (Perkin Elmer, Norwalk, CT) for cations/metals.

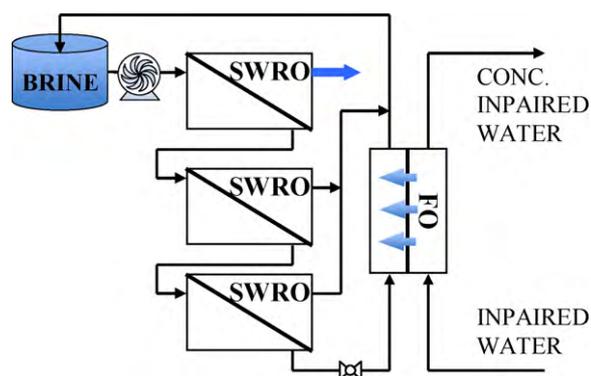


Fig. 3. Flow diagram of the SWRO subsystem utilizing a three-stage membrane array.

Rejection of trace organic chemicals by virgin FO membrane was studied at the bench-scale to determine baseline rejection in the absence of membrane fouling. Deionized feed water was spiked with select organic chemicals, including diclofenac, gemfibrozil, naproxen, and salicylic acid at concentrations of 500 ng/L each. Synthetic seawater (35 g/L sea salt) was used as a draw solution. The system was operated for 48 h to allow sufficient time for accumulation of trace organic chemicals in the draw solution and sufficient volume to produce samples for analysis of the organic compounds using gas chromatography/mass spectroscopy (GC/MS) [31].

2.3. Pilot-scale experiments

2.3.1. Pilot-scale system design and operation

A series of pilot-scale experiments were conducted with continuous streams of impaired water to assess membrane fouling propensity, solute rejection, and any subsequent degradation in performance of the hybrid process.

A pilot-scale system consisting of both FO and SWRO subsystems was constructed and tested at DWRP. The SWRO subsystem was necessary in this investigation because continuous supply of seawater was unavailable at the test site. The SWRO system was used for continuous reconcentration and production of draw solution at constant concentration, and to investigate rejection of nutrient and trace organic chemicals during the operation of the hybrid process. A schematic drawing of the SWRO subsystem is illustrated in Fig. 3. The subsystem was configured in a three-stage array (1-1-1) of 2.5 in. diameter by 40 in. long SWRO membrane elements operated in a semi-closed loop. Diluted draw solution from the FO membrane cell returned to the SWRO feed tank and an RO permeate partial stream (equivalent to the flowrate of water through the FO membrane) was removed from the system (bold single arrow in Fig. 3). A high-pressure, variable speed, positive displacement pump (HydraCell M03, Wanner Engineering Inc., Minneapolis, MN) was used in the RO subsystem.

Sensors were installed in the SWRO subsystem to measure stage 1 permeate and final concentrate (draw solution) conductivities, flow rates of all streams, and operating pressures. Conductivities were measured with two probes; $K = 1 \text{ cm}^{-1}$ for the permeate stream and $K = 10 \text{ cm}^{-1}$ for the concentrate (draw solution) stream (Cole-Parmer, Vernon Hills, IL). Permeate and concentrate flow rates and concentrate pressure were measured with rotameters and diaphragm pressure gauges, respectively. All electronic sensors were connected to the SCADA system.

A large flat sheet FO membrane cell was fabricated with a membrane surface area of 0.266 m². Flat rubber strips were used to create flow channels on the feed solution side of the FO membrane and a turbulence enhancing mesh spacer was installed in the draw solution flow channels. A constant speed rotary vane pump was

used to supply feed at 2.4 L/min to the FO membrane cell. A 50-mesh (279 μm) strainer was installed on the feed line to reduce the loading of suspended solids on the FO membrane.

Synthetic sea salt was dissolved in deionized water and used as feed for the RO system. The sea salt brine was pumped into the SWRO subsystem at a flow rate of 4.4 L/min and concentrated to become draw solution for the FO process with a flow rate of 2.4 L/min. Water flux through the FO membrane was calculated from the rate of accumulation of RO permeate in a separate permeate tank.

Physical cleaning of the FO membrane was performed several times during the course of the pilot testing. The feed flow rate was increased to 4 L/min for approximately three minutes to flush solids that accumulated on the FO membrane out of the membrane cell. Chemical cleaning was performed with a surfactant (Citranox, Alconox Inc., White Plains, NY) diluted to a pH of 3. The cleaning solution was recirculated through the feed channel of the FO membrane cell for approximately 30 min at 2.4 L/min. The pH was then increased to 8 with sodium hydroxide (Fisher Scientific, Waltham, MA), and circulated through the feed solution channel of the FO membrane cell for an additional 30 min, after which the experiments were resumed.

2.3.2. Pilot-scale feed and draw solution

The pilot-scale system was deployed at DWRP for long-term experiments, providing access to both secondary and tertiary treated effluent streams. Secondary treated effluent was drawn from a pipeline that transferred water from the Metropolitan Water Reclamation District's (MWRD, Denver, CO) wastewater treatment plant to DWRP. Tertiary treated water was the DWRP effluent before chlorine disinfection. The average pH was 7.5 for both the secondary and tertiary treated effluents and their average TDS concentration was 400 mg/L. The average TSS concentrations of the secondary and tertiary effluent were 10 and 2 mg/L, respectively.

Pilot-scale experiments were conducted with either secondary or tertiary treated feed water for extended time periods. All experiments with secondary treated effluent were conducted with draw solution concentration of 35 g/L synthetic sea salt. Experiments with tertiary treated effluent were conducted with draw solution concentrations of 20, 35, or 70 g/L sea salt, and were terminated after 200 L of water permeated through the FO membrane. Concentrated seawater (70 g/L) was chosen to represent operation with SWRO brine stream as draw solution. The FO membrane was replaced and the system was rinsed with RO permeate before each experiment.

Analysis of trace organic chemicals required 2 L samples. The impaired water feed and the RO permeate samples were drawn without affecting the system, but when draw solution samples were collected, a large amount of salt was removed from the draw solution loop. To balance the loss of salt, 2 L of stock draw solution were added to the system following sample withdrawal. The loss of other constituents was taken into consideration during calculation of rejection. Samples for analysis of trace organic chemicals were preserved and extracted using solid phase extraction shortly after they were collected [31].

2.4. Preliminary economic model

An economic model was developed using a spreadsheet program (Excel, Microsoft Corp., Redmond, WA). The model assumes that a SWRO facility is treating 757 m³/day (200,000 gallons/day) of seawater at 50% water recovery and that an impaired water stream with similar flowrate is available. The TDS concentrations for the initial seawater and impaired water streams are 35 and 0.5 g/L, respectively.

The model calculates the effect of adding osmotic dilution membrane capacity to osmotically dilute the seawater feed stream with increments of 1% of the impaired water flow rate (i.e., 7.57 m³/day). During each increment the additional membrane area required to achieve an additional 1% recovery of impaired water is calculated based on an empirical model derived from experimental data. The amount of osmotic dilution membrane surface area increases exponentially as the incremental water flux decreases due to loss in driving force. The cost of the osmotic dilution membrane (estimated at US\$12 per m²) is assumed to dominate the capital cost for installing the required osmotic dilution capacity. Membrane costs were uniformly amortized over 5 years with a capital recovery factor assuming 10% interest rate compounded annually.

Conversely, the energy required for the SWRO plant to achieve 50% recovery of the diluted seawater feed is marginally decreased with each increment of dilution. Specific energy demand for the SWRO process as a function of feed water TDS concentration was determined with RO system design software (ROSA version 6.0, Dow Filmtec, Edina, MN). Specific energy demand for desalination of a given feed water concentration was converted to energy cost by multiplying the capacity (assuming 24/7 operation) by the cost of energy (assumed US\$0.10 per kW-h). The benefit-cost ratio (BCR) was calculated by dividing the monetary savings gained from reduced energy consumption by the additional cost incurred from the installation of the osmotic dilution membranes. A BCR value greater than unity indicates a favorable economic return on the investment because each dollar spent on expanding the capacity of osmotic dilution realizes greater than one dollar in energy cost savings from operation of the SWRO system.

3. Results and discussion

3.1. Water permeability

Water permeability experiments were conducted on the FO bench-scale test unit. Results from these experiments are summarized in Table 1 and illustrate that water flux through existing ODMF membranes is relatively low when the draw solution concentration, and thus the osmotic driving force, is low. Operation at higher osmotic pressure produces greater water flux, but water fluxes in FO are still lower than those observed in most RO applications with equivalent hydraulic pressure driving forces. Water flux also deviates from linearity at higher draw solution concentrations. This is due to dilutive internal concentration polarization phenomenon, where the salt in the porous support layer of the membrane is diluted by water diffusing from the feed into the draw solution [20].

The effect of concentration polarization can be seen in the divergence of the specific water flux, as observed from the data summarized in Table 1. Specific flux is the water flux through the membrane divided by the osmotic pressure difference across the membrane ($\Delta\pi$). Application of Eq. (2) to the data presented in

Table 1

Results from bench-scale experiments using deionized water as feed and synthetic seawater draw solution. All bench-scale experiments were conducted with feed and draw solution temperatures and flowrates of 19 ± 0.1 °C and 1.4 L/min, respectively.

| DS conc. (g/L) | Water flux (L/m ² -h) | $\Delta\pi$ (MPa) | Specific water flux (L/m ² -h-MPa) |
|----------------|----------------------------------|-------------------|---|
| 5 | 1.1 | 0.35 | 3.14 |
| 10 | 2.1 | 0.70 | 3.00 |
| 15 | 3.0 | 1.05 | 2.86 |
| 20 | 3.6 | 1.39 | 2.59 |
| 25 | 4.2 | 1.74 | 2.41 |
| 30 | 4.9 | 2.09 | 2.34 |
| 35 | 5.7 | 2.45 | 2.33 |
| 70 | 8.3 | 4.90 | 1.69 |

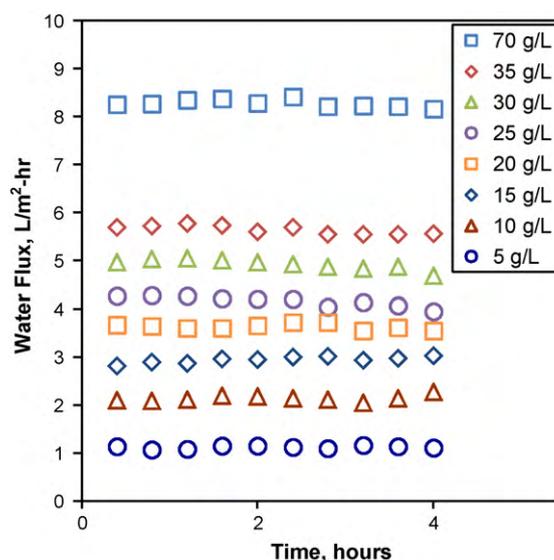


Fig. 4. Water flux as a function of time during bench-scale experiments of different synthetic seawater draw solution concentrations and secondary effluent feed solution.

Table 1, and subsequent linear regression analysis provided a coefficient of determination (R^2) of 0.97, A value of 3.56 L/m² h MPa, and K value of 0.086 m² h/L (3.08×10^5 s/m), which are similar to values published in the literature for the membrane tested [21].

3.2. Short-term membrane fouling tests with impaired water feed

Three sets of bench-scale experiments were conducted with different impaired feed streams and synthetic seawater draw solution at different concentrations. Secondary and tertiary treated wastewater effluents from the DWRP and water from the South Platte River were used as feed solutions in these experiments. During the FO experiments, water flux remained relatively constant—indicating that membrane fouling was minimal. Water flux as a function of time is illustrated in Fig. 4 for experiments conducted with secondary treated effluent feed and synthetic seawater draw solutions at different concentrations. Similar trends were observed during experiments conducted with tertiary treated effluent and South Platte River feed streams. Stable water flux despite operation in an environment with high fouling propensity (i.e., presence of suspended solids, dissolved organic matter, nutrients, and microorganisms) can be attributed to several factors, including low osmotic pressure of the feed solution and limited transport of foulants to the membrane surface due to lower water flux. Yet, it is likely that the combination of a hydrophilic membrane and low operating pressure play a more important role in reducing the rate of membrane fouling during bench-scale experiments [24,28]. It is known that pressure-driven membrane processes (specifically NF and RO) under similar experimental conditions (i.e., water flux, temperature, and feed characteristics) experience more substantial membrane fouling and flux decline [32].

Water flux as a function of draw solution concentration for the different feed solutions is illustrated in Fig. 5. Data presented in Fig. 5 provide a summary of average fluxes obtained during experiments conducted with deionized water (Table 1), secondary treated effluent (Fig. 4), tertiary treated effluent, and river water feeds. The different feed water qualities did not substantially affect the water flux. This is likely due to low TDS concentration (approximately 400 mg/L for the secondary and tertiary treated effluents) of the feed waters compared to the draw solution TDS concentrations. More importantly, unlike pressure-driven membrane processes,

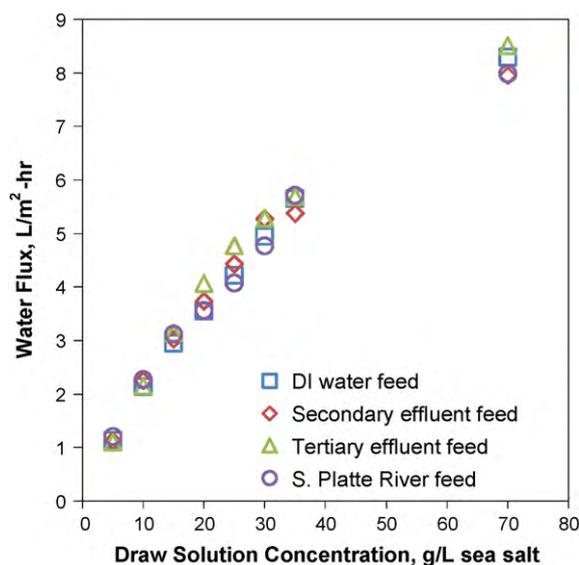


Fig. 5. Water flux as a function of draw solution concentration for short-term bench-scale FO experiments with deionized water, secondary and tertiary effluents, or South Platte River water feed.

short-term, initial flux decline was not observed in these experiments even though the feed streams contained suspended solids, dissolved organic matter, and nutrients [33].

3.3. Pilot-scale experiments

The hybrid FO/SWRO pilot-scale system was deployed at DWRP during the spring and summer of 2008. Initially, the FO membrane cell was installed horizontally with the feed solution flow channel on top and the draw solution flow channel on the bottom; the FO membrane was oriented with the dense active layer facing upward in contact with the feed solution. Initial experiments were conducted with secondary treated effluent feed from MWRD and the draw solution was maintained at a constant seawater salt concentration of 35 g/L. Water flux as a function of time for the first seven-day experiment is illustrated in Fig. 6.

Despite pretreatment with a 50- μm strainer, accumulation of suspended solids was observed in the feed channels of the FO

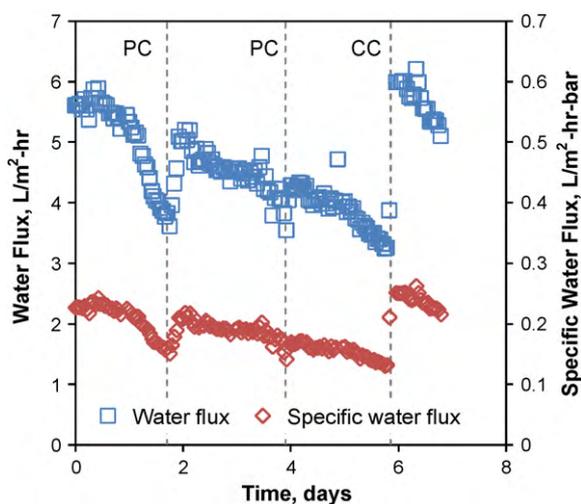


Fig. 6. Water flux and specific water flux as a function of time during a pilot-scale FO experiment with 35 g/L synthetic seawater draw solution and secondary effluent feed stream. PC indicates physical cleaning and CC indicates chemical cleaning. Experiments were conducted with feed and draw solution temperatures of $22 \pm 0.5^\circ\text{C}$ and flow rates of 2.4 L/min. Specific flux data was corrected to 19°C .

membrane cell during the first 3 days of operation. Flux decline was observed in conjunction with the settling of solids on the membrane [33]. Results illustrated in Fig. 6 reveal that the first physical cleaning partially restored water flux that was previously lost due to fouling on the membrane and the second physical cleaning (at the end of the fourth day) was inefficient in restoring the water flux. Prior to chemical cleaning, water flux declined to approximately 50% of its initial level. Results presented in Fig. 6 indicate that chemical cleaning at the end of the sixth day completely restored water flux to its initial value.

Although initial water flux values for the bench-scale and pilot-scale experiments were similar, flux declined more rapidly when the process was tested with a continuous supply of secondary treated effluent that contained and deposited more suspended solids on the FO membrane. Similar to bench-scale results, there was minimal flux decline early in the experiment; yet, flux started to decline at a greater rate at the beginning of the second day. Also, flux declined faster on the second day and after chemical cleaning (seventh day) compared to the other days. In both instances the higher initial flux might have played a role in faster fouling because more foulants are transported to the membrane surface by convection. The accelerated reduction in water flux observed after the period of sustained higher flux may be the result of reverse diffusing of solutes from the draw solution that are becoming immobilized by the fouling layer on the feed side of the membrane. This phenomenon may cause a cake enhanced external concentration polarization whereby reverse diffusing draw solution solutes accumulate on the feed side of the membrane, reducing the effective osmotic pressure difference and by extension the water flux.

Specific flux was calculated and is also presented in Fig. 6. Here specific flux is defined as the water flux normalized to the instantaneous osmotic driving force and corrected for temperature changes due to diurnal fluctuation in feed and draw solution temperatures. These results further confirm that flux decline in the process was due to continuous fouling of the membrane and not due to changes in operating conditions and driving force.

In an attempt to reduce membrane fouling, more experiments were conducted under similar conditions but with the membrane cell oriented with the feed stream flowing below the membrane. Membrane orientation was not changed; the dense active layer was still facing the impaired water feed (now down). It was expected that shear flow and gravity would prevent accumulation of solids on the membrane surface and in the feed channels, reducing flux decline due to membrane fouling. Water flux as a function of time for two experiments is illustrated in Fig. 7. Results revealed that with the new configuration water flux declined minimally compared to flux declined of approximately 10% per day when the feed channel was above the membrane. Solids that did accumulate in the feed channel settled onto the side-plate of the membrane cell and not the membrane itself, resulting in lower membrane fouling. All subsequent experiments were conducted with the new membrane cell orientation.

It is also important to note that membrane fouling was not observed on the SWRO membranes, nor did the RO membranes receive or require cleaning during the three-month deployment and operation of the hybrid pilot system. This observation further emphasizes the unique capacity of the osmotic dilution process to exploit the clean, renewable chemical energy of seawater to achieve high level pretreatment of a highly impaired water stream prior to SWRO.

3.4. Effect of draw solution concentration on process performance

To elucidate the effect of driving force on flux and membrane fouling under the new membrane cell orientation, experiments were conducted with tertiary treated effluent feed and synthetic

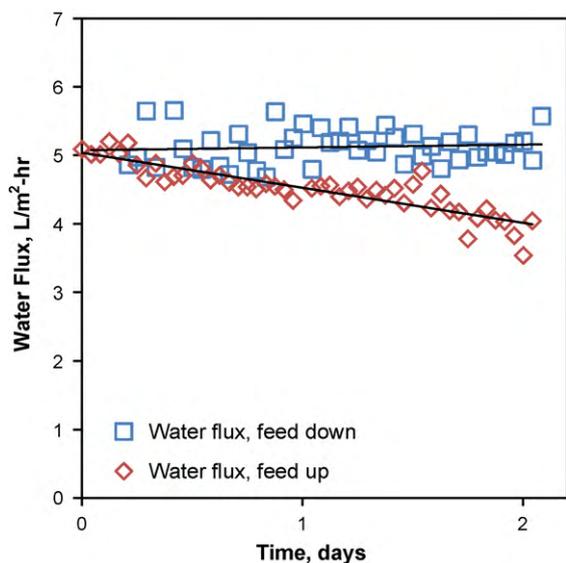


Fig. 7. Water flux as a function of time for pilot-scale FO experiment with the feed channel of the membrane cell facing up and down. Experiment conducted with 35 g/L synthetic seawater draw solution and secondary effluent feed solution.

seawater draw solution at concentrations of 20, 35, and 70 g/L. These draw solution concentrations simulate osmotic dilution of seawater, osmotic dilution of diluted seawater downstream in the process, and osmotic dilution of an RO concentrate in the second stage FO (Fig. 1). Water flux as a function of time is illustrated in Fig. 8 for the three experiments. The experiments were operated continuously for up to 8 days without any membrane cleaning, and with negligible flux decline. All experiments were terminated after approximately 200 L of water were produced.

Water flux during the pilot-scale experiment with draw solution concentration of 35 g/L was approximately 20% higher than flux during bench-scale experiments. Improved mixing in the draw solution channel of the pilot-scale FO membrane cell and operation at higher temperatures have likely reduced external dilutive concentration polarization effects at the draw solution-membrane

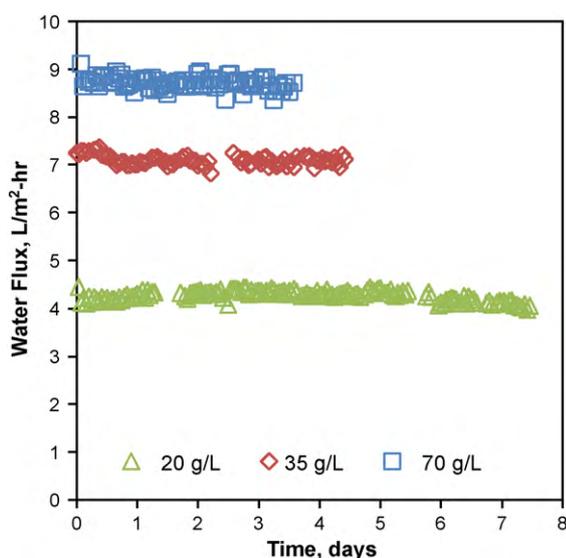


Fig. 8. Water flux as a function of time for pilot-scale FO experiment with 20, 35, or 70 g/L sea salt draw solution and tertiary effluent feed solution. Experiments conducted with the feed channel under the membrane. Experiments were terminated after approximately 200 L of water were processed through the membrane.

interface, promoting higher water flux. The deviation in water flux results between bench- and pilot-scale experiments may also stem from differences in the effective surface area of the membrane and differences in the hydraulic properties of the membrane cells.

3.5. Solute transport during osmotic dilution in the combined FO/RO process

In addition to studying water flux and fouling rates during osmotic dilution treatment of impaired water, solute transport was investigated to determine the capability of the dual barrier membrane system to reject inorganic and organic constituents.

3.5.1. Inorganic solute transport during osmotic dilution

Like most membranes used in separation processes, FO membranes are not perfectly selective barriers. During osmosis, solutes diffuse in both directions across the membrane, from the feed to the draw solution and from the draw solution to the feed [18].

Due to the high concentration of solutes in the draw solution (in this case seawater), transport of inorganic compounds occurs mostly from the draw solution into the impaired feed stream (i.e., reverse solute diffusion). This may contribute to a reduction in the driving force within the final contactor elements of a future osmotic dilution process; yet, the repercussions of reverse diffusion will be substantially reduced because the draw solution will not be operated in a close loop configuration and replenishment of draw solution will not be required.

Specific reverse TDS flux as a function of draw solution concentration is illustrated in Fig. 9 for bench-scale experiments conducted with different feed solutions. Considering the many parameters that can affect reverse solute diffusion during FO and osmotic dilution (e.g., flow velocity on both sides of the membrane, temperatures, pressure difference across the membrane, membrane integrity), results in this study were similar to those observed in previous studies (i.e., 400–500 mg/L) that used NaCl-dominated draw solutions and the same membrane [18,34]. When deionized water was used as feed, reverse TDS diffusion was higher at lower draw solution concentrations, likely due to reduced resistance to solute diffusion created when feed solutes diffuse into the draw solution with the water.

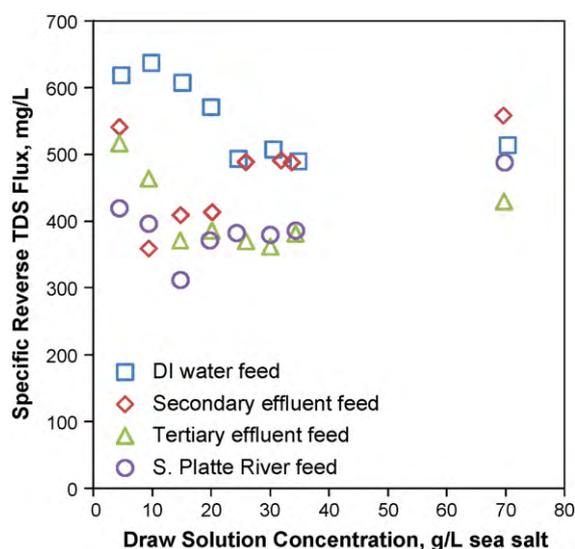


Fig. 9. Specific reverse TDS flux as a function of draw solution concentration for various feed streams during bench-scale FO experiments. Temperatures of the feed and draw solution streams were 19 ± 0.1 °C and feed and draw solution flow rates were 1.4 L/min.

Specific reverse diffusion of major ions was investigated in one bench-scale experiment conducted with 35 g/L synthetic seawater draw solution and deionized water feed. As anticipated, sodium and chloride accounted for the largest fraction of the diffusing TDS (97% of the solute mass flux); this is primarily because they had exhibited the highest concentration gradient. Chloride and sodium were also observed to diffuse at an approximately equal molar-equivalent rate (7.2 meq/L), maintaining electroneutrality of the feed and draw solutions. Specific reverse solute fluxes were also measured for bromide (1.37 mg/L), boric acid (1.24 mg/L), potassium (8.40 mg/L), calcium (1.41 mg/L), and magnesium (2.87 mg/L). Sulfate concentration in the feed was below detection limit at the end of the experiment and its specific reverse solute flux could not be calculated; however, the rate of sulfate diffusion is likely very low based on the physicochemical properties of sulfate ions (i.e., relatively large molecule size, divalent electro-negative charge, low diffusivity) and previous observations [18,35].

3.5.2. Rejection of organic chemicals and nitrogen species

At DWRP, ammonia, nitrate, and total organic carbon (TOC) were present in the secondary treated effluent. Both ammonia and nitrate concentrations were approximately 5 mg/L-N and TOC concentration ranged from 6 to 9 mg/L. Water samples were collected during a 13-day experiment from the feed stream, draw solution, and RO permeate of the pilot system. The concentrations of ammonia and nitrate are summarized in Fig. 10a and b, respectively. Results presented in Fig. 10a suggest that ammonia was present at relatively stable concentrations in the feed stream and over time diffused through the FO membrane into the closed loop draw solution. Ammonia was better rejected by the RO membrane, and consequently accumulated in the closed draw solution loop. Mass transfer calculations based on the concentration of ammonia in the different streams (feed, draw solution, and RO permeate) revealed that ammonia diffused at a constant mass transfer rate through the FO membrane once the concentration of ammonia in the draw solution reached a relatively constant concentration.

After reaching steady state, the concentration of ammonia in the draw solution was found to be slightly higher than in the feed. Because the concentration of ammonia in the feed changes throughout the day and the diffusion of ammonia through the different membranes (FO and RO) is relatively slow, given its low concentration in the feed, the daily samples might not have captured the actual dynamics of mass transport, and the higher concentrations of ammonia in the draw solution might be a result of higher ammonia concentration in the feed (secondary effluent) several hours before sampling. It is important to note that in an open draw solution loop (Fig. 1), ammonia would not accumulate, but instead be diluted and discharged with the concentrated brine after RO. The RO permeate contained very low concentrations of ammonia; thus, illustrating the ability of the hybrid system to consistently produce high quality water.

Nitrate was also found at relatively constant concentrations in the secondary treated effluent feed stream, with the exception of one concentration spike on the third day. Nitrate also accumulated in the draw solution loop; however, unlike ammonia, it reached a similar average concentration in the draw solution as in the feed solution. The concentrations of nitrate and ammonia in the FO and RO permeate were determined by calculating the mass flow rate and dividing by the water flow rate through the FO membrane. Using this information, the rejections of nitrate and ammonia by the FO and RO membranes were calculated and results are summarized in Table 2. These findings indicate that both the FO and RO membranes provided moderate rejection of ammonia and nitrate, but the total system rejection (i.e., combined FO and RO) was high.

UV absorbance at 254 nm was used as a surrogate measurement for TOC, and UV data was used to determine the overall

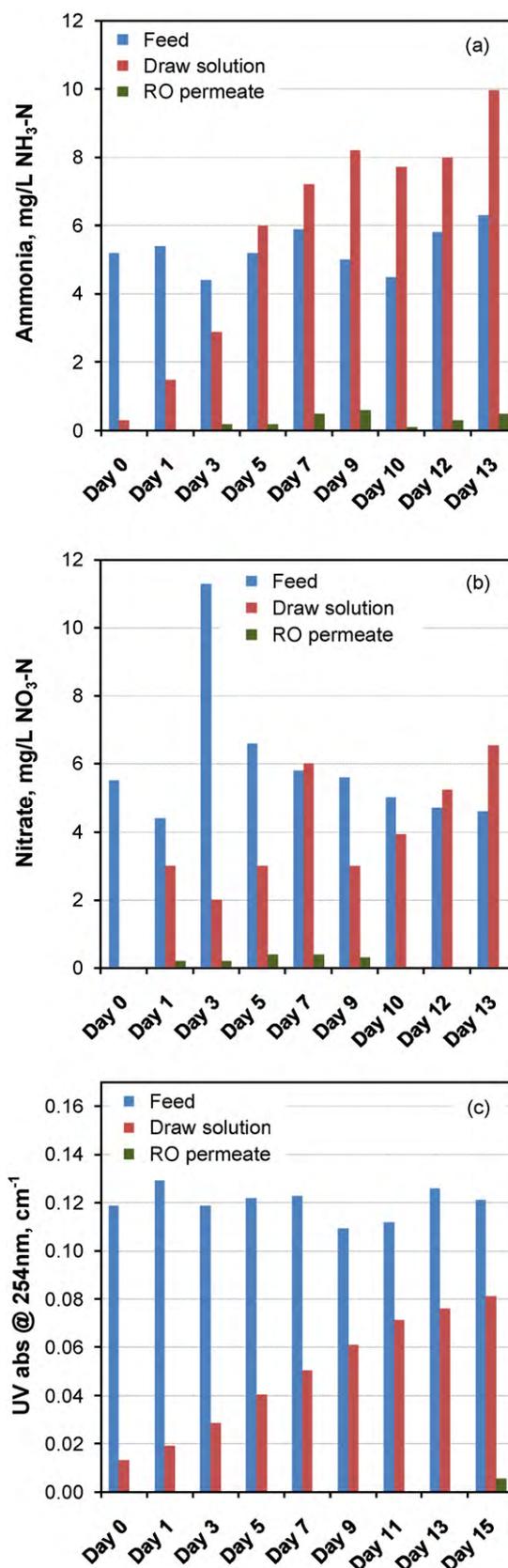


Fig. 10. (a) Ammonia and (b) nitrate concentrations and (c) UV absorbance as a function of time in the feed, draw solution, and RO permeate of the pilot system. Feed was DWRP secondary effluent and the draw solution was synthetic seawater at concentration of 35 g/L TDS.

Table 2

FO, RO, and total ammonia and nitrate rejection for the pilot-scale experiments with 35 g/L sea salt draw solution and secondary effluent feed solution. Values are presented in Fig. 10.

| | FO membrane | RO membrane | Total system |
|---------|-------------|-------------|--------------|
| | % Rejection | | |
| Ammonia | 74 | 77 | 94 |
| Nitrate | 78 | 82 | 97 |
| UV | 85 | 99 | >99.9 |

rejection of organic solutes during the pilot-scale experiments. UV absorbance results as a function of time are illustrated in Fig. 10c for the three different streams in the pilot system. Similar to ammonia and nitrate, UV absorbing compounds were moderately rejected by the FO membrane and further rejected by the RO membrane. UV absorbance was low and close to the detection limit in the permeate stream of the RO system. The UV absorbance of the FO and RO permeates were determined and suggest very high rejection of organic matter containing aromatic moieties through both the FO and RO membranes (Table 2). Phosphate rejection was not investigated in this study, but previous studies have demonstrated that phosphate can be more than 99.6% rejected by FO membranes [24].

3.5.3. Rejection of trace organic chemicals

Concentrations of trace organic chemicals were also measured during select bench- and pilot-scale experiments to determine their rejection by the FO and RO membranes. The compounds that were tested are commonly occurring trace organic chemicals in secondary treated effluent and represent specific groups of emerging pollutants, including pharmaceutical (anti-inflammatory, blood lipid regulators, and metabolites), personal care products, and chlorinated flame-retardants. Bench-scale spiking tests were carried out to determine rejection of the trace organic chemicals by a virgin FO membrane that was not fouled by impaired water. Results indicated that the compounds were well rejected (diclofenac >99%, gemfibrozil >80%, naproxen >90%, and salicylic acid >72%).

To determine the effect of membrane fouling and to evaluate the dual barrier FO–RO approach on rejection of trace organic chemicals, secondary effluent feed and 35 g/L sea salt draw solution were used during a longer-term pilot-scale experiment. The concentrations of six trace organic chemicals in the feed, seawater draw solution, and RO permeate of the pilot system were measured on the eighth day of the experiment and are summarized in Table 3. Feed concentration of individual compounds varied between 100 and 1000 ng/L. Similar to inorganic constituents, all monitored trace organic chemicals slowly accumulated in the draw solution closed loop during the course of the experiment; however, all compounds exhibited concentrations below detection limit in the RO permeate stream.

Table 3

Concentration of trace organic chemicals after 8 days operation of the pilot system with 35-g/L sea salt draw solution and secondary effluent feed.

| | Feed (ng/L) | Draw solution (ng/L) | RO permeate (ng/L) | Rate of transport through the FO membrane (ng/m ² -h) |
|----------------|-------------|----------------------|--------------------|--|
| Diclofenac | 155 | 65 | <1 | 1.3 |
| Gemfibrozil | 960 | 1650 | <2 | 32.3 |
| Ibuprofen | 385 | 255 | <5 | 5.0 |
| Naproxen | 435 | 360 | <1 | 7.0 |
| Salicylic acid | 260 | 85 | <5 | 1.7 |
| TCEP | 800 | 625 | <20 | 12.2 |

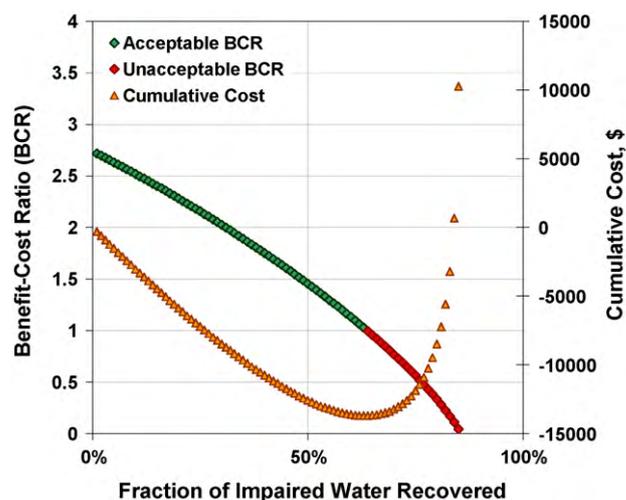


Fig. 11. Results from the economic model. Assumptions: energy cost of 10 ¢/kWh, capital cost of US\$ 12 per m² osmotic dilution membrane, SWRO plant treats 757 m³/day seawater at 50% water recovery. Values for the BCR colored green indicate favorable economic return on the investment, while red symbols indicate that the cost to install additional membrane capacity outweighs the benefits of reduced energy demand in the SWRO process.

3.6. Economic viability assessment

The BCR and cumulative cost of implementing osmotic dilution as a function of the fraction of impaired water recovered by the process is illustrated in Fig. 11 for the scenario of a 757 m³/day seawater (35 g/L TDS concentration) feed flow rate being incrementally diluted by adding an osmotic dilution contactor. As illustrated in Fig. 11 the BCR starts at 2.7 and decreases as the fraction of impaired water recovered increases. The cumulative cost saving of the coupled process decreases initially because the reduction in energy cost for operation of the SWRO subsystem outweighs the cost of installing additional osmotic dilution capacity; however, lower driving force for water flux in later stages of the osmotic dilution process require increasingly more membrane surface area to achieve the desired recovery. Therefore, the cost associated with the amount of membrane area required to achieve higher recoveries of impaired water eventually overcome the energy cost savings realized by the SWRO system. Based on the economic model, the dual barrier process is estimated to achieve favorable economic return (i.e., BCR greater than unity) during operation with up to 63% recovery of the impaired water stream. Beyond 63% recovery the capital cost associated with installing additional osmotic dilution membrane capacity becomes greater than the money saved from reduced energy consumption of the SWRO process. For more concentrated seawaters, the model predicts that under similar conditions, 68 and 71% impaired water recovery will be associated with a favorable BCR for seawater concentrations of 40 and 45 g/L TDS, respectively.

4. Conclusions

During short-term bench-scale experiments, minimal flux decline and low fouling of the FO membrane were observed. Feed water quality was found to have a minimal effect on water flux through the FO membrane; however, fouling was expected to occur when treating impaired water, and thus, short-term, bench-scale investigations might have limited value when evaluating treatability of real impaired water. Results from longer-term pilot-scale experiments demonstrated that under field conditions fouling was occurring and reducing the flux through the FO membrane.

Results from long-term experiments also demonstrated that membrane configuration plays an important role in controlling the rate of membrane fouling during treatment of impaired water. When the FO membrane cell was operated with the feed stream flowing under the membrane, suspended solids did not settle on the membrane and flux decline was negligible. Furthermore, physical and mild chemical cleaning of FO membranes are very effective in reversing flux decline due to membrane fouling in both FO and osmotic dilution processes.

Results from long-term experiments with various draw solution concentrations and tertiary treated wastewater feed qualities revealed that water flux can be sustained and membrane fouling can be minimized even at relatively high water fluxes through FO membranes. Reverse diffusion of TDS through the FO membrane was within the levels reported by other studies. It was demonstrated that rejection of nutrients and organic compounds is greatly improved when treating impaired water with the dual barrier, FO–RO process.

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