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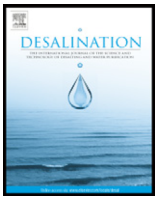

Effects of feedwater pretreatment on the removal of organic microconstituents by a low fouling reverse osmosis membrane

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ABSTRACT

The removal of 16 organic microconstituents by a commercially available low-fouling RO membrane was systematically investigated in this study under different feedwater pretreatment conditions, including magnetic ion exchange (MIEX), alum coagulation and ultrafiltration (UF). It was found that organic microconstituents were consistently removed by RO despite noticeable changes in the composition of feedwater natural organic matter (NOM) as determined by size exclusion chromatography. Sequential treatment of the natural water by MIEX, coagulation, UF, and RO revealed that MIEX removed more than 70% of NOM, but less than 40% of the microconstituents while RO was capable of removing more than 90% of the target organics, regardless of their chemical properties. These findings suggest that RO treatment is effective in removing organic microconstituents in natural water that have molecular weights greater than the molecular weight cutoff of the membrane, while MIEX, coagulation, and UF are not effective means for enhancing organic microconstituent removal by RO. Meanwhile, the fouling of the RO membrane by an ultra-filtered natural surface water was minor and not influenced by the variations in organic composition after different pretreatment schemes, indicating the potential of combining membrane modification and UF pretreatment for effective control of RO membrane fouling.

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1. Introduction

Reverse osmosis (RO) has been increasingly employed in the production of fresh water for drinking water supplies and wastewater reclamation. An important group of substances targeted by RO are trace-level organic microconstituents, including pharmaceuticals and personal care products (PPCPs) and endocrine disrupting compounds (EDCs). These microconstituents have been characterized as emerging contaminants for water treatment due to their potentially adverse health and ecological impacts [10]. A previous study showed that microconstituent removal by conventional water treatment processes are often ineffective [41]. In comparison, several preliminary studies suggested that RO membranes are effective in removing PPCPs/EDCs, as well as other microconstituents [5,9,23,25,24,36].

Similar to microfiltration (MF) and ultrafiltration (UF) membranes employed in fresh water treatment, RO membranes for drinking water augmentation are subject to fouling caused by aquatic substances, especially soil-derived natural organic matter (NOM) [38] and biopolymers produced by aquatic microorganisms [6,13]. RO membrane fouling is also influenced by ionic composition of the source water [15], water temperature [20], and biofilm formation [31]. Organic fouling and biofouling decrease the hydraulic permeability of RO membranes, resulting in loss of productivity and an increase in capital and operational costs for full-scale systems. Therefore, effective fouling control is critically important to the application of RO to freshwater treatment, with most control being achieved primarily through feedwater pretreatment and/or membrane modification [34,35].

Permeation of microconstituents through RO membranes involves adsorption of the organics to membrane surfaces, dissolution of organics into the membrane, and subsequent diffusive transport of dissolved organic molecules through the membrane matrix [37]. This process may be affected by membrane fouling in complex ways due to modification of membrane surface properties by fouling layers and the resulting changes to solute-membrane interactions or solute transport at the membrane-water interfaces. Nghiem and Coleman found that triclosan rejection by a RO membrane and two nanofiltration (NF) membranes increased after the membrane was fouled by bovine serum albumin (BSA), alginate, and humic acids, but was unchanged after the membrane was fouled by hydrophilic silica colloids [30]. Comparatively, Steinle-Darling and co-workers found that N-nitrosoalkylamines with molecular weights smaller than the molecular weight cutoff (MWCO) of a RO membrane were removed to a lesser extent when clean membranes were fouled by alginate while the removal of those

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constituents larger than the membrane MWCO were not affected [37]. Similar results were obtained by Ng for the removal of organics smaller than 100 g/mol by a RO membrane subject to organic fouling [29]. In general, impacts of membrane fouling on microconstituent removal were dependent upon feedwater and membrane properties, as well as the characteristics of organic compounds, and the fouling impacts were often more pronounced for NF membranes than RO membranes due to their differences in the structure of their respective membrane active layers [9,39].

Compared to the reference organic materials used in the aforementioned studies, dissolved organic matter in natural waters are comprised of more complex matrices [28]. Removal of different organic fractions as a result of feedwater pretreatment may change the fouling behaviors of RO membranes [1,7], thereby affecting the rejection of organic microconstituents. Currently, there is little information in the literature regarding the impact of feedwater pretreatment on the rejection of organic microconstituents by RO membranes. As pretreatments are widely integrated in RO treatment of contaminated waters [8], information in this regard is useful for proper selection of pretreatment schemes. A systematic evaluation of the relationships among feedwater pretreatment, membrane fouling, and solute rejection will also provide insights into the complex mechanisms of RO treatment. This served as the major aim of this study. Of particular interest was the application of magnetic ion exchange (MIEX) resin for the pretreatment of feedwater to RO. MIEX resin is an anionic resin designed for the removal of negatively charged NOM species in natural water and consists of magnetic iron oxides particles coated by polymers with positively-charged quaternary amine functional groups. Due to aggregation induced by inter-particle magnetic force, fine MIEX beads can be used to treat source waters in suspended forms and then be rapidly recovered by aggregation and settling of the beads in the absence of hydraulic disturbance. The effectiveness of MIEX in organic removal has been demonstrated in the treatment of natural water and treated wastewater effluent [21,40].

This study had three specific objectives: 1) to evaluate the impacts of MIEX and other pretreatments on the fouling of a low-fouling RO membrane; 2) to investigate the removal of NOM and organic microconstituents by the RO membrane under different fouling conditions; and 3) to assess the efficiency of a sequential pretreatment RO process in removing organic microconstituents.

2. Experiment

2.1. Experimental waters

A natural water sample was collected from the North Bay Regional Water Treatment Plant, California, USA and shipped to the lab overnight. The water sample was stored at 4 °C in the dark. Aliquots of the water sample were analyzed for inorganic and organic constituents using different methods (Table 1). To maintain consistency throughout the experiments, a single large volume sample was used. In order to systematically investigate the effect of NOM on membrane fouling and microconstituent removal, a synthetic model water was prepared based on the inorganic composition of the natural water sample.

2.2. Organic microconstituents

Sixteen PPCPs and EDCs selected in this study were spiked into the synthetic model water or the natural water for RO treatment (Table 2). These compounds were selected based on: 1) capability of established analytical methods, 2) what has historically be detected in the source water and could be expected to be present based on watershed activities, and 3) representing a diverse collection of PPCPs and EDCs having a wide range of properties and applications. In order to eliminate contamination of the natural water by organic solvents, as well as to reduce the cosolvent effect (i.e., increase of water solubility of organic compounds due to the presence of a small concentration of cosolvent), a modified glass-plate method developed in previous studies [23,26] was employed for spiking the microconstituents listed in Table 2, except for caffeine. The final concentration of microconstituents in the spiked water varied between 2 μg/L to approximately 200 μg/L, depending upon the water composition and characteristics of microconstituents. Substantial variations in microconstituent concentrations were also observed in a previous study using similar spiking method [12]. Four of these compounds, atrazine, N,N-Diethyl-meta-toluamide (DEET), lopressor, and propylparaben were only employed in the last run of the RO filtration experiment.

Table 1
Major characteristics of the natural water sample employed in this study.

Parameter	Unit	Value
pH		8.28
Turbidity	NTU	31.1
TOC	mg/L	3.56
Organic colloids	mg C/L	0.07
UV_{254} absorbance	cm^{-1}	0.118
Specific UV absorbance	L/mg·m	3.31
Alkalinity	mg $CaCO_3$/L	85
Conductivity	μS/cm	426
Chloride	mg/L	23.0
Fluoride	mg/L	0.26
Sulfate	mg/L	20.5
Phosphate	mg/L	0.93
Nitrate	mg/L	2.19
Sodium	mg/L	22.4
Magnesium	mg/L	12.5
Potassium	mg/L	6.07
Calcium	mg/L	14.6
Iron	mg/L	0.0026

2.3. Feedwater pretreatment

A concentrated suspension of virgin MIEX resin was obtained from Orica Watercare. The virgin MIEX was applied to the treatment of the natural water for RO experiments at a predetermined optimal dose of 5 mL/L (data not shown). During the pretreatment, MIEX suspension was added to five gallons of natural water and mixed using a laboratory stirrer at 203 rpm for 15 min, and the treated water was allowed to settle for 10 min before collection of the supernatant for subsequent coagulation or UF experiments.

Because a MIEX dose of 5 mL/L is relatively high for full-scale treatment, combination of MIEX and coagulation pretreatment was conducted by lowering the MIEX dose to a practical dose of 2.5 mL/L, followed by coagulation at a predetermined optimal coagulant dose. Reagent-grade aluminum potassium sulfate ($KAl(SO_4)_2 \cdot 12H_2O$) was dissolved in ultrapure water to prepare a stock solution and added to 5 gallons of natural water pretreated by 2.5 mL/L MIEX at a dose of 2.84 mg Al/L. After the addition of coagulant, the water was rapidly mixed at 136 rpm for 2 min and then slowly mixed at 34 rpm for 30 min. The treated water was allowed to settle for 60 min and the supernatant was collected for subsequent UF pretreatment.

The UF membrane employed for pretreatment was a commercially available, hollow fiber membrane fabricated with polyvinylidene fluoride (PVDF) and having a nominal pore size of 20 nm. Natural water with/without other pretreatments was filtered through the UF membrane at a constant flux of 80 L/m^2-h (LMH).

2.4. RO membrane and bench-scale testing system

A Dow Filmtec BW30-400-FR membrane was used throughout this study. This membrane has been employed in full-scale brackish

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