



Influence of naturally occurring dissolved organic matter, colloids, and cations on nanofiltration of pharmaceutically active and endocrine disrupting compounds



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HIGHLIGHTS

- Neutral PhAC/EDC rejections were significantly influenced by water components.
- Ionic compounds exhibited high rejections from all the water matrices examined.
- Presence of naturally occurring DOM caused improved compound rejection.
- Presence of natural colloidal particles had relatively little effect on rejection.
- Colloids provide cation-buffering and minimize the impact of cations on rejection.

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ABSTRACT

This study examined the rejection of selected pharmaceutically active (PhAC) and endocrine disrupting compounds (EDCs) when using nanofiltration as a function of naturally occurring dissolved organic matter (DOM), colloidal particles, cations and their interactions. Lake Ontario water served as a source of natural DOM and colloidal particles. PhAC/EDC rejection experiments were conducted using raw Lake Ontario water and Lake Ontario water that was pre-treated with either ultrafiltration to remove colloidal particles, or fluidized ion exchange resins to remove DOM. Additionally, the concentration of cations (Ca^{2+} , Mg^{2+} , and Na^+) in the raw and pre-treated water matrices was varied. While ionic PhACs and EDCs exhibited high rejections from all the water matrices examined, neutral compounds were most effectively rejected in water containing DOM and no colloids, and least effectively rejected from colloid-containing water with increased cations but no DOM. The presence of DOM significantly improved compound rejection and the increase in cation concentration significantly decreased rejection. The presence of colloids had comparatively little effect except to mitigate the impact of increased cation concentration, apparently providing some cation-buffering capacity. The sequence in which constituents are removed from waters during treatment may significantly impact PhAC and EDC removal, especially of neutral compounds.

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1. Introduction

Nanofiltration (NF) and reverse osmosis (RO) membrane removal of emerging organic micropollutants are influenced by the target compounds' physicochemical properties, membrane surface characteristics and feedwater chemistry (Kimura et al., 2003; Nghiem et al., 2005, 2006; Hu et al., 2007; Yangali-Quintanilla et al., 2009). Some studies (Xu et al., 2006; Nghiem et al., 2008; Chang et al., 2012) have further investigated NF/RO rejection

mechanisms as influenced by compound interactions with membranes pre-fouled by organic and inorganic model foulants. Others (Comerton et al., 2009a; Verliefde et al., 2009) have focused on the impact of solute interactions with source water components including natural organic matter (NOM) and colloidal particles on the rejection of pharmaceutically active compounds (PhACs) and endocrine disrupting compounds (EDCs). Increased rejection of specific compounds has been observed in the presence of NOM in synthetic waters (Hu et al., 2007; Comerton et al., 2009a). Single-sized model colloids have been reported to cause a decrease in rejection of salt and some trace organic contaminants (Hoek and Elimelech, 2003; Ng and Elimelech, 2004; Nghiem et al.,

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2008). The presence of cations may also impact compound rejection when interacting with NOM (Comerton et al., 2009a) or colloidal particles (Lee et al., 2005).

While RO or tight NF membranes (e.g. molecular weight cut-off, MWCO < 200 Da) should provide high removals of target micropollutants, they do so at a cost of high energy requirements to produce and maintain high flux. Looser membranes will require less energy to maintain the same fluxes and may replace RO and tight NF membranes as they can provide efficient removal of PhACs and EDCs. Previous studies (Nghiem et al., 2008; Comerton et al., 2009a; Chang et al. 2012) indicate that looser membranes can remove PhACs and EDCs to varying extents from laboratory-prepared waters that have been separately spiked with model organic matter and colloids. To better understand the production of drinking water from surface water sources, however, studies must now examine removal of PhACs and EDCs using looser membranes when considering representative surface waters, focusing on influences of specific as well as synergistic interactions of typical, indigenous water matrix constituents including dissolved organic matter (DOM), colloidal particles, and cations.

This study investigated the rejection of PhACs and EDCs with low molecular weights ($MW < 300 \text{ g mol}^{-1}$) from raw and pre-treated surface waters using a loose NF membrane (MWCO = 350 Da). The objectives were to confirm the impact of DOM on compound rejection, identify the impact of colloidal and suspended particles on compound rejection, and determine the effect of interactions among DOM, colloidal and suspended particles, and cations on PhAC/EDC rejection.

2. Materials and methods

2.1. Selection and characterization of PhAC/EDCs and NF membrane

Three PhACs (acetaminophen, carbamazepine, sulfamethoxazole) and six EDCs (bisphenol A (BPA), clofibric acid, diethylstilbestrol, estriol, estrone, and 17 β -estradiol), previously detected in US and Canadian surface waters (Chen et al., 2006; Lishman et al., 2006; Servos et al., 2007; Benotti et al., 2009), were examined. These compounds represent a range of physicochemical properties including molecular weight (MW), hydrophobicity (Log D), and dissociation constant (pK_a) (Table 1).

A polyamide NF membrane (NE70, NE 4040-70, Saehan-CSM Membranes, Woongjin Chemical America, CA, USA) was used throughout this study. The membrane's large MWCO (350 Da) (Kim et al., 2009; Lee et al., 2010) relative to the MW of selected PhAC/EDCs ($\leq 288 \text{ g mol}^{-1}$) allowed the investigation of rejection associated with compound interactions and/or association with water matrix constituents, rather than due to reduction of

membrane effective MWCO (Comerton et al., 2009b). NE70 represents a fairly negatively charged and hydrophobic membrane, as indicated by zeta potential (-36.4 mV , measured at pH 8 in 10 mM KCl solution) and contact angle (54.1°), respectively.

2.2. Water matrices examined and characterization of water matrix components

Lake Ontario water, a Great Lakes' source of drinking water for millions of consumers, was selected to serve as a background matrix to examine PhAC/EDC rejection via nanofiltration. To investigate the explicit impact of DOM, Lake Ontario water was pre-treated using an ultrafiltration (UF) membrane (MWCO = 100 kDa, TriSep UE50, TriSep, Goleta, CA, USA) which removed approximately $84 \pm 6\%$ (average \pm standard deviation for 3 measurements of turbidity) of the colloidal and suspended particles while allowing $\geq 84\%$ of the organic matter (expressed as dissolved organic carbon (DOC)) to pass through (Table 2). This resulted in an effectively colloid-free water matrix that retained most of its original DOM. The mean diameter of the particles in the untreated Lake Ontario water, measured using a particle sizing and counting analyzer (Section 2.4), was $6.6 \pm 0.43 \mu\text{m}$ (average \pm standard deviation for 3 measurements). The reduction of turbidity (raw Lake Ontario vs. UF-pretreated Lake Ontario water, Table 2) indicated that very few particles remained in the UF-treated water and the mean diameter measured was $0.66 \mu\text{m}$.

To specifically examine the effect of colloidal particles, Lake Ontario water was pre-treated using a fluidized ion exchange (FIEX) column containing a strong base Type I anion exchange resin (Purolite PPA860S, The Purolite Company, Bala Cynwyd, PA, USA). FIEX pre-treatment resulted in approximately 74% reduction of DOM (as DOC) while allowing over 90% of the colloidal and suspended particles to pass through (Table 2). This water matrix, then, was dominated by naturally occurring colloids and suspended particles, but essentially free of most of DOM.

To investigate the impact of cations on PhAC/EDC rejection, Ca^{2+} , Mg^{2+} , and Na^+ concentrations were approximately doubled when compared to ambient Lake Ontario water. In addition, a laboratory-prepared water (Milli Q, $18.2 \text{ M}\Omega \text{ cm}$ @ 25°C , Millipore (Canada)) was buffered with 1 mM sodium bicarbonate and spiked with the cations at the same concentration as Lake Ontario water to serve as a "control".

In total, seven water matrices were examined including (1) a Milli Q "control", (2) raw Lake Ontario water, (3) raw Lake Ontario water with cation concentration doubled, (4) UF-pretreated (colloid-free) Lake Ontario water, (5) UF-pretreated (colloid-free) water with cations double those of Lake Ontario water, (6) FIEX-pretreated (DOM removed) Lake Ontario water, and (7) FIEX-pretreated (DOM removed) water with cations double those of Lake Ontario water (see Fig. 1).

2.3. PhAC/EDC rejection experimental setup

NF of PhAC/EDCs was conducted using a bench-scale membrane filtration set-up (Fig. 1). Flat-sheet specimens ($19 \text{ cm} \times 14 \text{ cm}$) of the NF membrane with 155 cm^2 effective area were placed into stainless steel cross-flow filtration cells (Sepa CF II, Steriltech, Kent, WA, USA). The spacers in the membrane cells as well as connective piping were made of stainless steel in order to avoid compound losses within the apparatus due to adsorption. Feedwater contained in a storage tank (S) was transferred to a temperature-controlled ($20 \pm 1^\circ\text{C}$) stainless steel 15-L equalization tank (S1) via a stainless steel valveless piston pump (RK-07104-50, Cole-Parmer, Montreal, QC, Canada) with a variable speed drive (RK-07104-31, Cole-Parmer). From the equalization tank, the water was delivered to the membrane cells using a Hydracell

Table 1
Selected PhAC/EDCs and their properties.

Compound	Molecular weight (g mol^{-1})	Log D ^a @ pH 8	pK_a^a	Classification
Acetaminophen	151	0.23	10.2	PhAC
Bisphenol A (BPA)	228	3.86	10.3	EDC
Carbamazepine	236	2.23	14.3	PhAC
Clofibric acid	214	-0.98	4.0	EDC
Diethylstilbestrol	268	4.42	9.7	EDC
Estriol	288	2.45	10.0	EDC
Estrone	270	3.46	10.3	EDC
Sulfamethoxazole	253	-1.43	5.7	PhAC
17 β -estradiol	272	3.35	10.3	EDC

Log D = Octanol–water partition coefficient (ionized and neutral species).

pK_a = Acid dissociation constant.

^a Determined using ACD/I-Labs's physicochemical and ADMET prediction software (Advanced Chemistry Development, Toronto, ON).

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