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Effects of membrane fouling on N-nitrosamine rejection by nanofiltration and reverse osmosis membranes

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ABSTRACT

The impact of fouling on N-nitrosamine rejection by nanofiltration (NF) and reverse osmosis (RO) membranes was investigated in this study. Membrane fouling was simulated using tertiary treated effluent and several model fouling solutions (that contained sodium alginate, bovine serum albumin, humic acid or colloidal silica) to elucidate the changes in rejection behaviour of N-nitrosamines. In general, the rejection of N-nitrosamines increased when the membranes were fouled by tertiary effluent. The rejection of small molecular weight N-nitrosamines was most affected by membrane fouling. In particular, the rejection of N-nitrosodimethylamine (NDMA) by the ESPA2 membrane increased from 34% to 73% after membrane fouling caused by tertiary effluent. The results also indicate that the impact was less apparent for the lowest permeability membrane (i.e., ESPAB), and the rejection of N-nitrosamines by the ESPAB membrane was over 82% regardless of membrane fouling. The effect of membrane fouling caused by model foulants on N-nitrosamine rejection was considerably less than that caused by tertiary effluent. Size exclusion chromatography analyses revealed that the tertiary effluent contains a high fraction of low molecular weight (<500 g/mol) organic substances. It appears that these low molecular weight foulants present in the tertiary effluent can restrict the solute pathway within the active skin layer of membranes, resulting in the observed increase of solute rejection.

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1. Introduction

Augmentation of potable water sources with reclaimed municipal effluent is an important strategy to secure a reliable water supply in regions and countries with severe water scarcity. However, a major concern over this alternative source of water supply is the occurrence of trace organic chemicals which may induce adverse and chronic health effects. Notable amongst these trace organic chemicals is N-nitrosodimethylamine (NDMA) which is an N-nitrosamine that can be formed during the chloramination of the treated effluent [1]. In addition to NDMA, other N-nitrosamines known to occur in treated effluent include N-nitrosomethylethylamine (NMEA), N-nitrosopyrrolidine (NPYR), N-nitrosodiethylamine (NDEA), N-nitrosopiperidine (NPIP), N-nitrosomorpholine (NMOR), N-nitrosodipropylamine (NDPA), N-nitrosodi-n-butylamine (NDBA) [2–4]. Some of these N-nitrosamines have been identified as probable carcinogenic agents and thus their concentrations in drinking water and recycled water intended for potable consumption have been regulated by water authorities around the world [5,6]. The Australian Guidelines for Water Recycling have recommended the maximum value of NDMA, NDEA, and NMOR in recycled water intended for potable supply of 10, 10, and 1 ng/L, respectively [7]. Both reverse osmosis (RO) and nanofiltration (NF) membranes have been frequently used in water reclamation partly to ensure adequate removal of emerging trace chemicals, little is known about their capacity to remove N-nitrosamines in full-scale installations. Reported percentage rejections of NDMA vary greatly in full-scale plants from almost negligible to 86% and the underlying reason for such significant variation in NDMA rejection remains unclear [8–11].

To date, only a few laboratory-scale studies have investigated N-nitrosamine rejection capability of NF/RO membranes using clean matrix solutions [3,12,13]. These studies reported that the rejection of NDMA by RO membranes was in the range from 50% to 70%. The rejection of N-nitrosamines increased in the order of increasing molecular weight and the steric hindrance mechanism

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was identified as a predominant rejection mechanism of N-nitrosamines by NF/RO membranes [3,12,13]. Feed solution characteristics (i.e., pH, ionic strength and temperature of the feed solution) also affected the rejection of NDMA and in some cases other N-nitrosamines [3,13]. In particular, Fujioka et al. [13] reported a significant drop in NDMA rejection (from 49% to 25%) for an increase in feed temperature from 20 to 30 °C. Nevertheless, the variations in these feed solution characteristics explain only some of the variations in NDMA rejections that were reported in the previous full-scale studies.

Municipal wastewater usually contains a large amount of organic and inorganic matter, resulting in the formation of organic and colloidal fouling, bio-fouling and inorganic scales on RO membranes [14,15]. It has been established in the literature that membrane fouling can either increase or decrease the separation efficiency of NF/RO membranes [14,16–18]. However, apart from a laboratory-scale study conducted by Steinle-Darling et al. [3] who investigated the rejection of several N-nitrosamines by an RO membrane (ESPA3) artificially fouled with sodium alginate, to date little attention has been given to the effects of membrane fouling on the rejection of N-nitrosamines. Steinle-Darling et al. [3] reported that membrane fouling by sodium alginate on the ESPA3 membrane caused a reduction in NDMA rejection (from 56% to 37%).

The aim of this work was to provide insights into the effects of membrane fouling on the rejection of N-nitrosamines by NF/RO membranes. The effects of membrane fouling were investigated by comparing the rejections of N-nitrosamines by clean and fouled membranes. Tertiary treated effluent and four different model foulants (namely sodium alginate, bovine serum albumin, humic acid and colloidal silica) were used to induce membrane fouling. The tertiary treated effluent and model foulants were characterised in detail to systematically elucidate the effects of membrane fouling on the rejection of N-nitrosamines by NF/RO membranes.

2. Materials and methods

2.1. NF/RO membranes

Three NF/RO membranes – namely the NF90, ESPA2, and ESPAB – were used in this investigation. These are thin-film composite polyamide membranes with a microporous supporting layer and were supplied as flat sheet samples. Key properties of these membranes are shown in Table 1. The NF90 (Dow Filmtec, Minneapolis, MN, USA) is an NF membrane typically used for softening of brackish water. The ESPA2 (Hydranautics, Oceanside, CA, USA) is a low pressure reverse osmosis membrane that is widely applied for water reclamation applications. The ESPAB (Hydranautics, Oceanside, CA, USA) is also low pressure reverse osmosis but it has been designed to achieve a high boron rejection.

Table 1
Properties of the membranes used in this study.

Membrane	Pure water permeability[a] [L/m^2 h bar]	Conductivity rejection[b] [%]
NF90	11.7 ± 1.1	81.2 ± 2.5
ESPA2	5.5 ± 0.3	98.1 ± 0.3
ESPAB	3.9 ± 0.2	99.3 ± 0.4

[a] Determined with Milli-Q water at 1000 kPa and 20 °C feed temperature. Errors represent the standard deviation of three replicates.

[b] Analysed with feed solution contained 20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$ at permeate flux 20 L/m^{2}h, cross flow velocity 40.2 cm/s, feed pH 8.0 ± 0.1 and feed temperature 20.0 ± 0.1 °C.

2.2. Chemicals

The eight N-nitrosamines used in this study (Fig. 1) were of analytical grade and were purchased from Sigma-Aldrich (St Louis, MO, USA). Their physicochemical properties have been described in detail elsewhere [13]. N-nitrosamine stock solution was prepared in pure methanol with 250 μg/L of each N-nitrosamine. A deuterated surrogate standard was used for each N-nitrosamine under investigation. These surrogate standards include N-nitrosodimethylamine-D6, N-nitrosomethylethylamine-D3, N-nitrosopyrrolidine-D8, N-nitrosodiethylamine-D10, N-nitrosopiperidine-D10, N-nitrosomorpholine-D8, N-nitrosodipropylamine-D14 and N-nitrosodi-n-butylamine-D9, and were purchased from CDN isotopes (Pointe-Claire, Quebec, Canada). A surrogate stock solution containing 100 μg/L of each deuterated N-nitrosamine was prepared in pure methanol. The stock solutions were stored at −18 °C and used within one month of preparation.

Analytical grade NaCl, $CaCl_2$ and $NaHCO_3$ were purchased from Ajax Finechem (Taren Point, NSW, Australia). Sodium alginate (SA), bovine serum albumin (BSA), humic acid (HA) and colloidal silica (Ludox CL, 30% weight suspension in water) were selected as model foulants to simulate polysaccharides, proteins, refractory organic matter and colloidal particles, respectively. These model foulants were purchased from Sigma-Aldrich (St Louis, MO, USA). The Ludox CL is a positively charged silica particle whose surface is coated with a layer of aluminium [19]. The hydrodynamic diameter of the Ludox CL is from approximately 40 nm at below pH 6 to 233 nm at pH 10 due to aggregation effects in different pH solutions [19].

2.3. Tertiary treated effluent

Tertiary treated effluent sample was collected from an advanced water recycling plant in New South Wales, Australia. The treatment train of the plant prior to the sampling point includes screening, bioreactor and sand filtration, and the sample was collected after sand filtration.

2.4. Membrane filtration system

A laboratory-scale cross flow NF/RO filtration system was used in this study (Supplementary material Fig. S1). A detailed description of this system is available elsewhere [13]. The system consisted of a cross flow stainless steel cell with effective membrane area of 4 cm by 10 cm and a channel height of 2 mm. The feed solution was kept in a stainless reservoir and was fed to the membrane cell by a high pressure pump (Hydra-Cell, Wanner Engineering Inc., Minneapolis, MN, USA). The permeate flow and cross flow velocity were regulated by a bypass valve and a back-pressure regulator (Swagelok, Solon, OH, USA). The permeate flow was monitored using a digital flow metre (FlowCal, GJC Instruments Ltd, Cheshire, UK) which was connected to a computer. A stainless steel heat exchanging coil was submerged into the feed reservoir and was connected to a chillier/heater unit (Neslab RTE 7, Thermo Scientific Inc., Waltham, MA, USA) to control the temperature of the feed solution.

2.5. Experimental protocols

Rejection measurement and membrane fouling development were sequentially carried out with four steps: (1) compaction; (2) measuring N-nitrosamine rejection without membrane fouling; (3) fouling development; and (4) remeasuring N-nitrosamine

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