



Rejection of small solutes by reverse osmosis membranes for water reuse applications: A pilot-scale study



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HIGHLIGHTS

- Small solute (N-nitrosamine & boron) rejections were evaluated by a pilot RO plant.
- Results validate findings from lab-scale experiments wrt small solute rejection.
- Permeate flux and temperature could considerably influence small solute rejection.
- Boron & NDMA rejections responded to changes in operating condition in a similar way.
- A strong correlation ($R^2 = 0.86$) between boron and NDMA rejections was obtained.

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ABSTRACT

N-nitrosamines and boron are small solutes of particular concern during water recycling applications. Here, we evaluated the rejection of seven N-nitrosamines and boron under a range of operating conditions and feed solution characteristics. The evaluation was conducted using a pilot-scale reverse osmosis (RO) system to appropriately simulate hydrodynamic conditions of full-scale RO installations. The rejection of seven N-nitrosamines by the pilot RO system varied significantly in the range from 31 to 94%, and rejection increased in the increasing order of their molecular weight. Rejection values obtained from this pilot-scale study were lower than those previously reported in laboratory-scale studies. These discrepancies were attributed to a difference in RO system operating condition (i.e. recovery) between the pilot-scale study (25%) and laboratory-scale study (<0.1%). Nevertheless, rejection data reported here validate the recent findings from laboratory-scale studies with respect to the impact of permeate flux, feed temperature and feed pH on separation efficiencies of N-nitrosamines. Data obtained from this pilot-scale study also validate the strong correlation between boron and NDMA rejection at or below pH 8 regardless of operating conditions and feed solution characteristics. The results suggest that boron rejection can be used as a surrogate for NDMA rejection in full-scale RO installations.

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1. Introduction

Water reuse is an increasingly important strategy in many parts of the world to supplement drinking water resources [1]. Municipal wastewater is abundantly available in the urban environment where water demand is high. In a planned indirect potable water reuse

scheme, secondary or tertiary treated wastewater is reclaimed using advanced treatment technologies to produce high quality water. Reclaimed water can potentially contain pathogenic agents and traces of organic contaminants which may cause adverse impact on human health. Thus, water reuse is subjected to a stringent set of regulations for public health protection. Among a series of water treatment processes in a water reclamation plant, reverse osmosis (RO) filtration serves as an efficient barrier for removing most trace organic chemicals [2]. Nevertheless, a small compound, N-nitrosodimethylamine (NDMA) has been detected in RO permeate at a higher concentration than its regulatory value (i.e. 10 ng/L) [3,4]. Moreover, NDMA rejection evaluated

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in full-scale plants has been reported to be variable and therefore difficult to predict (10–55%) [3–6].

NDMA is frequently detected in secondary treated wastewater [7]. In addition, NDMA concentration can increase during chloramination, which is commonly employed prior to RO filtration. The other N-nitrosamines that can also occur in treated wastewater include N-nitrosomethylethylamine (NMEA), N-nitrosopyrrolidine (NPYR), N-nitrosodiethylamine (NDEA), N-nitrosopiperidine (NPIP), N-nitrosomorpholine (NMOR), N-nitrosodipropylamine (NDPA), N-nitrosodimethylamine (NDBA) [8–10]. NDMA and some other N-nitrosamines have been classified as probable human carcinogens by the US EPA [11]. The permeation of NDMA and other N-nitrosamines through RO membranes has been investigated in many laboratory-scale studies using small membrane coupons [12–16]. These laboratory-scale studies suggest that NDMA rejection by RO membranes is affected by changes in operating conditions (e.g. feed temperature and permeate flux). Nevertheless, findings obtained from these laboratory-scale studies employing flat-sheet membrane specimen cannot be directly used to accurately simulate hydrodynamic conditions of full-scale RO system. Despite the differences between small membrane coupons and spiral wound membrane elements, investigations evaluating the rejection of N-nitrosamines using pilot-scale RO systems remain scarce [5] and no previous pilot-scale RO studies have been conducted to mechanistically elucidate the impact of various operating conditions on N-nitrosamine rejection.

Regularly monitoring N-nitrosamines in aqueous solution at parts per trillion levels is another challenge for most water authorities. Analysis of N-nitrosamine concentrations is generally carried out using an analytical method which comprises solid phase extraction followed by gas chromatography or high pressure liquid chromatography with tandem mass spectrometry detector [17]. The analysis of N-nitrosamines at parts per trillion levels is expensive and not always readily available. Thus, any alternative indicators that simulate the rejection of N-nitrosamines, in particular NDMA, by reverse osmosis could be very useful for monitoring purposes. A recent laboratory-scale study by Tu et al. [18] reported that boron rejections by RO membranes under a range of a specific operating condition (i.e. permeate flux or feed temperature) correlate with NDMA rejection. The similarity in the rejection behaviour between boron and NDMA is very likely due to their similar physicochemical characteristics. In fact, both boron (as a boric acid) and NDMA molecules are small (62 g/mol and 74 g/mol, respectively) and uncharged under operating conditions representative of RO installations (i.e. pH 6–8) [19,20]. It is noteworthy that boron in reclaimed water is also of great concern in places where reclaimed water is used for irrigation. Unlike NDMA, boron is ubiquitously detected in treated wastewater at 0.3–4 mg/L [21] and several commercially available online boron analysers have parts per billion to trillion sensitivity. Thus, boron rejection can potentially be used as a surrogate to estimate NDMA rejection. The use of boron as an NDMA surrogate will not replace compliance monitoring of NDMA in the final product water. However, it will facilitate better system design and reduce the number of analyses, and hence the cost associated with ongoing NDMA monitoring.

To date, the potential use of boron rejection as a surrogate of NDMA rejection has only been demonstrated by laboratory scale experiments using a small membrane sample. The hydraulic and hydrodynamic conditions as well as operating conditions (i.e. water recovery) between a lab-scale RO set-up using small flat sheet membrane coupons and pilot- and full-scale installations using spiral wound membrane elements can differ significantly. Thus, it is warranted to validate whether the observed correlation between boron and NDMA rejections also applies to RO systems employing spiral-wound elements. The aim of this study was to demonstrate the rejection of N-nitrosamines and boron using a pilot-scale RO system simulating hydrodynamic conditions of full-scale RO installations under a range of different operating conditions (i.e. permeate flux, feed pH and feed temperature).

2. Materials and methods

2.1. Chemicals

Analytical grade N-nitrosamines and boric acid (Table 1) were purchased from Sigma-Aldrich (St Louis, MO, USA). These N-nitrosamines include NDMA, NMEA, NPYR, NDEA, NPIP, NMOR and NDPA. A stock solution was prepared in pure methanol at 10 mg/L of each N-nitrosamine. Deuterated N-nitrosamines used for analysis were supplied by CDN isotopes (Pointe-Claire, Quebec, Canada) and these chemicals include N-nitrosodimethylamine-D6, N-nitrosomethylethylamine-D3, N-nitrosopyrrolidine-D8, N-nitrosodiethylamine-D10, N-nitrosopiperidine-D10, N-nitrosomorpholine-D8 and N-nitrosodipropylamine-D14. A surrogate stock solution of 100 µg/L of each deuterated N-nitrosamine was prepared in pure methanol. These stock solutions were kept at –18 °C in the dark and were used within 1 month of preparation. Analytical grade NaCl, CaCl₂ and NaHCO₃ were obtained from Ajax Finechem (Taren Point, NSW, Australia) and used as the background electrolytes during the filtration experiments.

2.2. Pilot-scale filtration system and experimental protocols

The pilot-scale RO filtration system used for this investigation has a permeate design flow of 0.72 m³/h (Fig. 1). The system consists of three 4-inch glass-fibre pressure vessels, a feed reservoir, a centrifugal feed pump (CRN 3-25, Grundfos, Bjerringbro, Denmark), and variable flow meters. Pipe lines in the feed stream were made of stainless steel and those in the permeate stream were made of PVC. Each pressure vessel could hold one RO membrane element (4 in. × 40 in.). Feed solution temperature was controlled in the feed reservoir using stainless steel heat exchanging pipes connected to a chiller unit (Aqua Cooler S360PD-CT, Chester Hill, NSW, Australia). The TFC-HR membrane (TFC 4040HR, Koch Membrane Systems, San Diego, CA, USA) used in the study is a composite polyamide RO membrane designed for brackish water treatment with a membrane area of 7.9 m²/element.

Prior to the pilot-scale filtration experiment, 200 L of RO feed solution was prepared in the feed tank. RO feed solutions used for this investigation were conditioned to obtain 20 mM NaCl, 1 mM CaCl₂ and 1 mM NaHCO₃ in deionised water or actual microfiltration (MF) permeate which was collected from a local full-scale water recycling plant in Australia. The total organic carbon (TOC), conductivity, and pH of the MF permeate were 9.6 mg/L, 0.91 mS/cm, and 6.9, respectively. In addition, N-nitrosamines and boric acid were dosed into the feed solution at approximately 250 ng/L of each N-nitrosamine and 5 mg/L of boron. Subsequently, the pilot RO system was continuously operated under standard conditions (permeate flux 20 L/m²h, feed temperature 20 °C, feed pH 8.0) for 15 h prior to any sampling events. The concentrate and permeate solutions were recirculated into the feed tank. The overall system recovery was maintained at 25% throughout the experiments. For N-nitrosamine analysis, 200 mL of RO feed and permeate samples were collected in amber glass bottles and the N-nitrosamine surrogate stock solution was dosed into each sample to obtain 50 ng/L of each isotope labelled N-nitrosamine. From the same sampling point, 20 mL sample was also collected in plastic bottles for the analysis of boron and sodium ions. Experiments with variable operating conditions started with low permeate flux of 10 L/m²h, which was increased stepwise to 30 L/m²h. When operating conditions were changed, sampling was conducted after at least 1 h of system operation. Experiments with variable feed temperature were first evaluated under low feed temperature (10 °C) and the feed temperature was incrementally increased to 35 °C. Experiments with variable feed pH were conducted by adjusting feed pH (5–9.5) with a dose of either HCl or NaOH.

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