N-nitrosamine rejection by reverse osmosis membranes: a full-scale study

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Keywords
nitrosamine, n, scale, study, full, membranes, osmosis, reverse, rejection

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This study aims to provide longitudinal and spatial insights to the rejection of N-nitrosamines by reverse osmosis (RO) membranes during sampling campaigns at three full-scale water recycling plants. Samples were collected at all individual filtration stages as well as at a cool and a warm weather period to elucidate the impact of recovery and feed temperature on the rejection of N-nitrosamines. N-nitrosodimethlyamine (NDMA) was detected in all RO feed samples varying between 7 and 32 ng/L. Concentrations of most other N-nitrosamines in the feed solutions were determined to be lower than their detection limits (3-5 ng/L) but higher concentrations were detected in the feed after each filtration stage. As a notable exception, in one plant, N-nitrosomorpholine (NMOR) was observed at high concentrations in RO feed (177-475 ng/L) and permeate (34-76 ng/L). Overall rejection of NDMA among the three RO systems varied widely from 4 to 47%. Data presented here suggest that the feed temperature can influence rejection of NDMA. A considerable variation in NDMA rejection across the three RO stages (14-78%) was also observed. Overall NMOR rejections were consistently high ranging from 81 to 84%. On the other hand, overall rejection of N-nitrosodiethylamine (NDEA) varied from negligible to 53%, which was considerably lower than values reported in previous laboratory-scale studies. A comparison between results reported here and the literature indicates that there can be some discrepancy in N-nitrosamine rejection data between laboratory- and full-scale studies probably due to differences in water recoveries and operating conditions (e.g. temperature, membrane fouling, and hydraulic conditions).

Keywords: Water reuse; N-nitrosamines; NDMA; reverse osmosis (RO) membranes; full-scale operation.
1. Introduction

As clean water sources continue to dwindle in many parts of the world, water recycling has been used by water providers to secure safe and reliable supplies of water for municipal, industrial and agricultural uses (Shannon et al., 2008). In many water recycling schemes, reverse osmosis (RO) filtration is deployed as a key process to remove inorganic salts and trace organic chemicals (Bellona et al., 2004; Dolnicar et al., 2010; Shannon et al., 2008; Verliefde et al., 2008). However, among these trace organic chemicals, the rejection of N-nitrosodimethylamine (NDMA) by RO membranes has often been reported to be low and highly variable (Fujioka et al., 2012a). As a result, subsequent treatment by advanced oxidation processes (AOPs), which is commonly based on UV-H$_2$O$_2$, is required where NDMA concentrations in the final product water is stringently regulated (Plumlee et al., 2008; Poussade et al., 2009). In addition to NDMA, several other N-nitrosamines have also attracted scientific and regulatory attention due to their potentially carcinogenic properties (Sedlak and von Gunten, 2011; USEPA, 1993). These N-nitrosamines include N-nitrosomethylamine (NMEA), N-nitrosopyrrolidine (NPYR), N-nitrosodiethylamine (NDEA), N-nitrosopiperidine (NPIP), N-nitrosomorpholine (NMOR), N-nitrosodipropylamine (NDPA), N-nitrosodi-n-butylamine (NDBA) and their occurrence in wastewater and secondary treated effluent has been reported in the literature (Krauss et al., 2010; Reyes-Contreras et al., 2012; Yoon et al., 2012). In particular, NMOR has been reported at up to 12.7 µg/L in secondary treated effluent (Krasner et al., 2009). Potential sources of the high NMOR concentrations in wastewater include toiletries and cosmetics (Spiegelhalder and Preussmann, 1984) and rubber and tire industries (Fajen et al., 1979). For the augmentation of drinking water sources, guideline values of NDMA (10 ng/L), NDEA (10 ng/L) and NMOR (1 ng/L) have been recommended by the Australian Guidelines for Water Recycling (NRMMC et al., 2008). On the other hand, NDMA is the only N-nitrosamine considered in the Australian Drinking Water Guidelines with a recommended guideline value of 100 ng/L (NHMRC and NRMMC, 2011).

The rejection of small neutral solutes such as N-nitrosamines by RO membranes is mainly governed by size exclusion (Fujioka et al., 2012b). In general, the rejection of N-nitrosamines increases in the increasing order of their molecular size (Bellona et al., 2011; Miyashita et al., 2009; Steinle-Darling et al., 2007). Thus, NDMA, which is the smallest molecule among N-
nitrosamines, exhibits the lowest rejection (Fujioka et al., 2012a). Previous laboratory-scale studies (Bellona et al., 2011; Fujioka et al., 2012b; Miyashita et al., 2009) have elucidated the effects of operating conditions and feed water characteristics on the rejection of N-nitrosamines. It is reported that permeate flux can play an important role in the rejection of low molecular weight N-nitrosamines. Fujioka et al. (2012b) also reported that a seven-fold increase in feed ionic strength (from 26 to 182 mM) could lead to some decrease in NDMA rejection (from 51 to 43%) and that pH changes in the feed (from pH 6.5 to 9) could also cause an increased NDMA rejection (from 37 to 51%). In addition to these water quality parameters, a recent laboratory-scale study by Fujioka et al. (2013a) has reported that membrane fouling by tertiary treated wastewater effluent resulted in an increase in the rejection of low molecular weight N-nitrosamines including NDMA.

The rejection of N-nitrosamines by RO membranes has been extensively investigated at the laboratory scale (Bellona et al., 2011; Fujioka et al., 2012b; Miyashita et al., 2009; Steinle-Darling et al., 2007). However, full-scale monitoring data to reaffirm findings from laboratory-scale experiments and to assess the impact of realistic operating conditions on the rejection of N-nitrosamines have rarely been reported in the peer review literature. Plumlee et al. (2008) studied the removal of NDMA by different treatment processes (including RO) at the Interim Water Purification Facility (Orange County, California, USA). NDMA removal by the RO process varied from 24 to 56%. The authors suggested that the variation in NDMA rejection observed in their study might be associated with changing feed conditions and membrane fouling. However, the authors did not monitor the feed and membrane fouling conditions. Farré et al. (2011) reported the fate of NDMA after each treatment process of the Bundamba Water Recycling plant in Queensland, Australia. Because Farré et al. (2011) did not focus on the RO process, only one overall rejection value of NDMA by the RO system can be inferred from their study. Some information about the rejection of NDMA and NMOR by a full-scale RO plant can also be inferred from a study by Krauss et al. (2010), who investigated the fate of N-nitrosamine precursors at the Wulpen/Torreele Water Recycling plant in Belgium. In comparison to NDMA, very little is known about the fate and removal of other N-nitrosamines during RO filtration at full scale. The scarcity of full-scale monitoring and the lack of information regarding operating conditions (e.g. permeate flux and recovery) and feed water characteristics (e.g. temperature, ionic composition) significantly hinder any meaningful data analysis. RO systems for wastewater recycling are typically designed using
three stages to achieve recovery around 85% (Fujioka et al., 2012a). Although the RO feed is further concentrated after each filtration stage, no studies available to date have examined rejection efficiencies for nitrosamines at subsequent stages.

The aim of this study was to assess the removal of eight N-nitrosamines in three full-scale RO plants. N-nitrosamine rejection values obtained at different stages were systematically related to the operating conditions and feed water characteristics. In addition, the difference in N-nitrosamine rejections between a cool and a warm weather period at one plant was also elucidated. Based on the obtained results, implications to water recycling practice were highlighted and discussed.

2. Materials and methods

2.1. RO systems

Samples were collected from three full-scale water recycling plants denoted as A, B and C located in Australia. In these plants, prior to RO filtration, secondary treated effluent is first pretreated by either microfiltration (MF) or ultrafiltration (UF). In all three plants, pre-formed chloramines were added to the process prior to MF or UF filtration to mitigate biofouling on the RO membranes (Figure 1). The RO membranes used in these plants are from three different manufacturers. The membranes used in these three plants are thin film composite with a polyamide skin layer. They were characterized by similar salt (NaCl) rejection and water permeability (Fujioka et al. (2013b). The process flow diagrams of these RO systems are shown in Figure 1. Samples were collected from plant A during cool (A-1) and warm (A-2) weather periods. At plant A, chloramination is normally added downstream of the coagulation process, which was the configuration when sampling campaign A-1 was conducted (Figure 1). During an extended period of warm weather when it is necessary to control algal growth during the coagulation process, chloramination can be added upstream of the coagulation process. Plant A was operated in this configuration when the sampling campaign A-2 took place (Figure 1). Unlike plants B and C, plant A is equipped with a booster pump prior to the third stage to maintain the same average flux at all three stages (Figure 1). All three systems produce reclaimed water for industrial and/or agriculture uses. Plants A and B were designed for a possible indirect potable water recycling application where high quality reclaimed water can be used to replenish an existing reservoir for drinking
water supply. Thus, the UV-H₂O₂ process was also installed after the RO process at these systems for the destruction of residual NDMA in the RO permeate. Similar installation using the UV-H₂O₂ process specifically for the removal of residual NDMA in the RO permeate can also be found elsewhere (Drewes and Khan, 2011; Plumlee et al., 2008).

2.2. Sampling protocol

RO feed and permeate samples were collected from each RO stage (Figure 1). From each sampling point, one sample was collected from plant C in May and December 2012 and duplicate samples were collected in all other sampling events for N-nitrosamine analysis. These samples (500 mL) were stored in amber glass bottles. Deuterated N-nitrosamines corresponding to each target compound were used as isotope labelled surrogates. These deuterated N-nitrosamines were purchased from CDN Isotopes (Pointe-Claire, Quebec, Canada). A surrogate stock solution containing 100 µg/L of each deuterated N-nitrosamines was prepared in pure methanol. Immediately after sample collection, the surrogate stock solution was added to the sampling bottles to obtain 50 ng/L of each isotope labelled N-nitrosamine. Analytical grade sodium thiosulfate (100 mg/L) was also added to the sample as quenching reagent to prevent any further NDMA formation during transportation and sample processing. From each sampling point, 20 mL sample was collected in plastic bottles for the analysis of cations and boron and 100 mL sample was collected in amber glass bottle for the analysis of anions and total organic carbon. Operating conditions and feed temperature of the RO systems on the sampling day are summarised in Table 1. The difference in feed temperature between the entrance and exit of each RO unit was less than 1 °C.

2.3. Analytical technique

An analytical method previously developed for the determination N-nitrosamines in drinking water and treated municipal effluent was employed (McDonald et al., 2012). The method uses solid phase extraction (SPE), gas chromatography and analysis by tandem mass spectrometry with electron impact ionization. Eight N-nitrosamines investigated in this study have molecular weight in the range from 74 to 158 g/mol (Supplementary Material Figure
These eight target N-nitrosamines are hydrophilic in the operating pH range of the RO plants (pH 6-8). Further details of their physicochemical characteristics can be found elsewhere (Fujioka et al., 2012a). The use of direct isotope dilution ensures accurate quantification, accounting for analytical variability that may occur during sample processing, extraction and instrumental analysis. SPE was conducted using Supelclean™ Coconut Charcoal SPE cartridges purchased from Supelco (St Louis, MO, USA). The concentrations of N-nitrosamines were quantified using an Agilent 7890A gas chromatograph coupled with an Agilent 7000B triple quadrupole mass spectrometer. Calibration curves were established for each N-nitrosamine with a range of 0.5-500 ng/L. The NMOR calibration curve was extended to account for the NMOR concentration of over 400 ng/L. The quantitative detection limits of this technique were 3 ng/L for NDMA, NDEA, NPIP, and NMOR, and 5 ng/L for NMEA, NPYR, NDPA, and NDBA.

Total organic carbon (TOC) concentration was determined using a TOC-V CSH analyser (Shimadzu, Japan). Conductivity and pH were measured using an Orion 4-Star Plus pH/conductivity meter (Thermo scientific, USA). Cation and boron concentration were determined using an Inductive Coupled Plasma - Mass Spectrometer (7500CS, Agilent Technologies, Wilmington, DE, USA) following the protocol previously reported by Tu et al. (2011). Anion concentrations were determined using an ion chromatography system (Shimadzu, Tokyo, Japan).

2.4. Calculation

The rejection of N-nitroamines and other solutes in each RO stage and combined RO stages was calculated using the following equations.

\[
R_i [%] = \left(1 - \frac{C_{pi}}{C_{f_i}}\right) \times 100 \quad (1) 
\]

\[
R_T [%] = \left(1 - \frac{C_{pT}}{C_{fT}}\right) \times 100 \quad (2) 
\]
where \( i \) is the number of stage, \( C_{pi} \) is the solute concentration in the RO permeate of the stage \( i \), \( C_{fi} \) is the solute concentration in the RO feed of the stage \( i \), and \( C_{pT} \) is the solute concentration in the combined RO permeate.

### Results and discussion

#### 3.1. Organic and inorganic constituent removal

The feed waters to the three RO systems differed markedly in TOC concentration and salinity (Supplementary Material Table S2). In particular, the feed water to plant A exhibited a relatively high conductivity (salinity) at approximately 2.5 mS/cm. The sewer catchment of plant A is predominantly in a low-lying coastal area and is subjected to seawater intrusion. In fact, due to seawater ingresses, boron concentration in the feed to plant A was also higher compared to plant B and C. Despite the high feed water salinity, the quality of RO permeate at plant A was comparable to that at the other two RO systems. Most common cations and anions in the feed water can be rejected well by the RO membranes. As a result, the permeate at all three RO systems was of high quality with respect to basic water quality parameters. In agreement with the 85% water recovery (Table 1) of these RO systems, TOC and conductivity concentrations in the final concentrates were approximately six times greater than those in the feed waters (Supplementary Material Table S2).

The rejections of TOC, cations (sodium, magnesium, potassium and calcium), anions (chloride, nitrate and sulphate) and boric acid by all three RO systems are summarised in Figure S3 of the Supplementary Material. Divalent ions (i.e. magnesium, calcium and sulphate) were consistently removed over 99%. On the other hand, in agreement with a previous study by Bellona and Drewes (2007), nitrate rejection was slightly lower than that of all other ions. The rejection of boric acid was in the range of 15-30%, which is consistent with the fact that boric acid has a small molecular size and is uncharged at pH below 8 (Tu et al., 2010). The difference between the charged and uncharged solutes observed here can be attributed to the electrostatic interaction and size exclusion rejection mechanisms. In addition to size exclusion, electrostatic repulsion can also play an important role in the rejection of charged solutes by NF/RO membranes (Bellona et al., 2004).
3.2. N-nitrosamine removal

3.2.1. Occurrence of N-nitrosamines in the RO feed water

NDMA was detected in all RO feed water samples (Figure 2). NDMA concentrations (7-16 ng/L) detected in the RO feed solutions were below or only marginally higher than the value (i.e. 10 ng/L) in the final product water stipulated by the Australian Guidelines for Water Recycling, with samples from A-2 being the only exception. In A-2, chloramine was added upstream of the coagulation process and thus resulting in an increase in NDMA formation. Results in Figure 2 are consistent with those obtained from previous studies (Farré et al., 2011; Mitch et al., 2005; Plumlee et al., 2008). For typical water recycling plants where NDMA in raw water can be controlled to similar levels found in this study, reducing NDMA formation in the feed (Mitch et al., 2005) and RO filtration can be implemented to meet the guideline value without relying on an additional subsequent treatment process such as AOP.

In addition to NDMA, several other N-nitrosamines (i.e. NPYR, NDEA, NPIP, NMOR and NDBA) were also detected in some but not all RO feed water samples (Supplementary Material Figure S4). NMEA, which is the second lowest molecular weight compound among the N-nitrosamines investigated here, was not detected during any sampling campaign. Surprisingly, a comparatively high NMOR concentration (177-475 ng/L) was observed in the feed water at plant C. Compared to plant C, NMOR concentrations detected in the RO feed in plants A-2 and B configurations were low. It is noted that NMOR concentrations in A-1, B and C-1 were not reported due to unsatisfactory variation between duplicate samples and poor recovery of the isotopically labelled internal standard. In fact, a sampling program conducted in plant A from 2010 to 2012 revealed low NMOR concentrations (< 21 ng/L) in the RO feed (Supplementary Material Figure S5) which indicates that a very high NMOR concentration like plant C has not been identified in plant A. Likewise, a sampling program conducted in plant B from 2009 to 2011 also showed a relatively low NMOR concentrations in the range from 9 to 57 ng/L in the RO feed (Supplementary Material Figure S5). The results reported here suggest that high NMOR concentrations in RO feed may be site specific and could relate to certain industrial dischargers. The source of NMOR appears to be site specific and further research is necessary to identify sources of NMOR within the catchment of plant C.
After each stage, concentrations of the N-nitrosamines increased to quantifiable levels due to
the concentration effect leading to higher feed concentrations in subsequent stages (Figure 2).
As a result, the highest N-nitrosamine concentration was consistently observed in the final
RO concentrate. For example, NDMA concentrations in the final RO concentrate were two to
six times higher than those in the RO feed. Likewise, NMOR concentrations in the RO
concentrate were approximately six times higher than those in the RO feed.

[Figure 2]

3.2.2. N-nitrosamine concentrations in the RO permeate

NDMA concentrations in the RO permeate were detected above the detection limit (3 ng/L)
at least once in samples from each plant (Figure 3). However, most of these detections did not
exceed the guideline value of 10 ng/L, again with a sample collected from A-2 being the only
exception. Of the seven remaining N-nitrosamines, only NDEA, NMOR and NDBA were
detected in RO permeate samples (Supplementary Material Figure S6). NMOR concentration
in the RO permeate samples of plant C varied between 34 and 76 ng/L, which was
comparatively higher than the other N-nitrosamines. This is because NMOR concentration in
the RO feed of plant C was also higher than all other N-nitrosamines (Figure 3). Once again,
NMOR concentrations in A-1, B and C-1 are noted as not available due to a large variation in
analysed NMOR concentration between duplicate samples and poor recovery of the
isotopically labelled internal standard.

In all cases, the concentration of N-nitrosamines in the RO permeate increased in later stages
due to the increased concentration in the RO feed for each stage (Figure 3). As a result, N-
nitrosamine concentrations in the overall RO permeate (i.e. the combined RO permeate of the
first, second and third stages) were higher than those in the first stage. The results here
indicate that rejection estimates obtained from laboratory-scale systems, which are operated
at low water recovery, may result in an underestimation of N-nitrosamine concentrations in
the RO permeate. Although the permeation of NDMA through RO membranes can be
managed by a subsequent UV-H₂O₂ based AOP, little is known about its removal efficiency
for NMOR and other N-nitrosamines. The results reported here also suggest that, in addition
to NDMA, it is necessary to monitor the concentration of several other N-nitrosamines
particularly NMOR in secondary treated effluent and the corresponding RO permeate.
3.3. Rejections by RO membranes

3.3.1. Overall rejection

Overall NDMA rejections varied significantly with a range of 4-47% among the three RO systems (Figure 4). In plant A, two distinct overall NDMA rejections (36 and 4%) were observed during different sampling occasions. Plant A was operated under the same operating conditions (e.g. recovery and permeate flux) during the two sampling events (i.e. A-1 and A-2), but their feed water temperature differed (19.4 and 28.0 °C) (Table 1). A previous laboratory-scale study revealed that an increase in feed temperature from 20 to 30 °C resulted in a reduction of NDMA rejection from 49 to 24% (Fujioka et al., 2012b).

NDEA rejection at plant A and C varied between zero and 53% (Figure 4). This is considerably lower than the values (86-95%) reported in a recent laboratory-scale study using low pressure RO membranes and synthetic clean water solutions (Fujioka et al., 2012b). Although the mechanism underlining this phenomenon is still unknown, the results reported here indicate that RO filtration in treated wastewater can result in a significant reduction in NDEA rejection. In fact, in a laboratory-scale study Fujioka et al. (2013a) reported a considerable deterioration in NDEA rejection using tertiary effluent as feed water. Overall, NMOR rejection was high and each stage exhibited rejection ranges of 87-91% (Figure 4) which is consistent with previous laboratory-scale studies (Fujioka et al., 2013a; Fujioka et al., 2012b).

3.3.2. Rejection at each stage

In plant B, 16 inch membrane elements were used whereas 8 inch membrane elements were used in plants A and C. Thus, the hydraulic distribution of plant B can differ significantly from that of plant A and C. At plant B, a significant variation in NDMA rejection (14-78%) was observed among the three RO stages (Figure 5). Changes in the permeate flux after each filtration stage may contribute to this variation to some extent (Fujioka et al. 2012b). However, because permeate flux was not monitored in each individual stage at plant B, it was not possible to confirm this hypothesis. Rejection of N-nitrosamines was further investigated...
using the two sampling events at plant A, focusing on the difference in NDMA rejection among the three stages. As RO filtration progressed, feed pH increased slightly and feed conductivity increased significantly for both sampling events (Figure 6). During the first sampling event (A-1) an increase in NDMA rejection from the first stage to third stage was observed. In general, an increase in feed conductivity (or ionic strength) results in a decrease in N-nitrosamine rejection (Fujioka et al., 2012b). However, the current study revealed an opposite trend which indicates that another factor such as membrane fouling may have been developed more extensively in later stages and may have compensated the decreased trend of NDMA rejection. On the other hand, during the second sampling event (A-2), NDMA rejections decreased as RO filtration progressed to later stages (Figure 6). The results reported here indicate that NDMA rejections among three RO stages may vary significantly even when operating conditions (i.e. permeate flux and recovery) were maintained constant.

[Figure 5]

[Figure 6]

4. Conclusions

NDMA was detected in all feed samples at the three full-scale RO trains investigated in this study. Although most other N-nitrosamines were not detected in the RO feed, several N-nitrosamines became detectable as the feed was further concentrated after each filtration stage. N-nitrosamine concentrations in the final RO concentrate were up to six times higher than those in the RO feed. As a notable exception, one of the three plants exhibited high NMOR concentrations (177-475 ng/L) in the feed, resulting in high NMOR concentrations (34-76 ng/L) in the permeate. In most cases, NDMA, NDEA and NDBA were detected below the Australian guideline value in the RO permeate. Overall rejection of NDMA and NDEA among the three RO systems varied significantly with a range of 4-47% and 0-53%, respectively. NDMA rejections among three RO stages also exhibited a significant variation in several cases. These rejection variations may have resulted from the difference in feed temperature and possibly membrane fouling. These findings suggest that N-nitrosamine rejection estimates derived from laboratory-scale flat-sheet membrane studies, which are operated at very low water recovery, may not be representative of full-scale operation. On the other hand, overall NMOR rejections were equally high with a range of 81-84%. The findings of this study provide insights for potential variations in N-nitrosamine rejection among
different RO systems and RO stages. Nevertheless, it is prudent to note the causes of some variations reported here were not sufficiently identified due to changes in multiple parameters during full-scale RO operation. Therefore, additional work is necessary to examine the impact of each cause (e.g. fouling and feed temperature) using a pilot- or full-scale RO systems.

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