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

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Abstract

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-nitrosodimethylamine (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 (3e5 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 (177e475 ng/L) and permeate (34e76 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 (14e78%) 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).

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18   **Abstract**

19   This study aims to provide longitudinal and spatial insights to the rejection of N-nitrosamines
20   by reverse osmosis (RO) membranes during sampling campaigns at three full-scale water
21   recycling plants. Samples were collected at all individual filtration stages as well as at a cool
22   and a warm weather period to elucidate the impact of recovery and feed temperature on the
23   rejection of N-nitrosamines. N-nitrosodimethlyamine (NDMA) was detected in all RO feed
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25   feed solutions were determined to be lower than their detection limits (3-5 ng/L) but higher
26   concentrations were detected in the feed after each filtration stage. As a notable exception, in
27   one plant, N-nitrosomorpholine (NMOR) was observed at high concentrations in RO feed
28   (177-475 ng/L) and permeate (34-76 ng/L). Overall rejection of NDMA among the three RO
29   systems varied widely from 4 to 47%. Data presented here suggest that the feed temperature
30   can influence rejection of NDMA. A considerable variation in NDMA rejection across the
31   three RO stages (14-78%) was also observed. Overall NMOR rejections were consistently
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34   in previous laboratory-scale studies. A comparison between results reported here and the
35   literature indicates that there can be some discrepancy in N-nitrosamine rejection data
36   between laboratory- and full-scale studies probably due to differences in water recoveries and
37   operating conditions (e.g. temperature, membrane fouling, and hydraulic conditions).

38   **Keywords:** Water reuse; N-nitrosamines; NDMA; reverse osmosis (RO) membranes; full-
39   scale operation.

1

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_2O_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-nitrosomethylethylamine (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-

71 nitrosamines, exhibits the lowest rejection (Fujioka et al., 2012a). Previous laboratory-scale

72 studies (Bellona et al., 2011; Fujioka et al., 2012b; Miyashita et al., 2009) have elucidated the

73 effects of operating conditions and feed water characteristics on the rejection of N-

74 nitrosamines. It is reported that permeate flux can play an important role in the rejection of

75 low molecular weight N-nitrosamines. Fujioka et al. (2012b) also reported that a seven-fold

76 increase in feed ionic strength (from 26 to 182 mM) could lead to some decrease in NDMA

77 rejection (from 51 to 43%) and that pH changes in the feed (from pH 6.5 to 9) could also

78 cause an increased NDMA rejection (from 37 to 51%). In addition to these water quality

79 parameters, a recent laboratory-scale study by Fujioka et al. (2013a) has reported that

80 membrane fouling by tertiary treated wastewater effluent resulted in an increase in the

81 rejection of low molecular weight N-nitrosamines including NDMA.

82 The rejection of N-nitrosamines by RO membranes has been extensively investigated at the

83 laboratory scale (Bellona et al., 2011; Fujioka et al., 2012b; Miyashita et al., 2009; Steinle-

84 Darling et al., 2007). However, full-scale monitoring data to reaffirm findings from

85 laboratory-scale experiments and to assess the impact of realistic operating conditions on the

86 rejection of N-nitrosamines have rarely been reported in the peer review literature. Plumlee et

87 al. (2008) studied the removal of NDMA by different treatment processes (including RO) at

88 the Interim Water Purification Facility (Orange County, California, USA). NDMA removal

89 by the RO process varied from 24 to 56%. The authors suggested that the variation in NDMA

90 rejection observed in their study might be associated with changing feed conditions and

91 membrane fouling. However, the authors did not monitor the feed and membrane fouling

92 conditions. Farré et al. (2011) reported the fate of NDMA after each treatment process of the

93 Bundamba Water Recycling plant in Queensland, Australia. Because Farré et al. (2011) did

94 not focus on the RO process, only one overall rejection value of NDMA by the RO system

95 can be inferred from their study. Some information about the rejection of NDMA and NMOR

96 by a full-scale RO plant can also be inferred from a study by Krauss et al. (2010), who

97 investigated the fate of N-nitrosamine precursors at the Wulpen/Torreele Water Recycling

98 plant in Belgium. In comparison to NDMA, very little is known about the fate and removal of

99 other N-nitrosamines during RO filtration at full scale. The scarcity of full-scale monitoring

100 and the lack of information regarding operating conditions (e.g. permeate flux and recovery)

101 and feed water characteristics (e.g. temperature, ionic composition) significantly hinder any

102 meaningful data analysis. RO systems for wastewater recycling are typically designed using

103   three stages to achieve recovery around 85% (Fujioka et al., 2012a). Although the RO feed is

104   further concentrated after each filtration stage, no studies available to date have examined

105   rejection efficiencies for nitrosamines at subsequent stages.

106   The aim of this study was to assess the removal of eight N-nitrosamines in three full-scale

107   RO plants. N-nitrosamine rejection values obtained at different stages were systematically

108   related to the operating conditions and feed water characteristics. In addition, the difference

109   in N-nitrosamine rejections between a cool and a warm weather period at one plant was also

110   elucidated. Based on the obtained results, implications to water recycling practice were

111   highlighted and discussed.

112   2.   Materials and methods

113   *2.1. RO systems*

114   Samples were collected from three full-scale water recycling plants denoted as A, B and C

115   located in Australia. In these plants, prior to RO filtration, secondary treated effluent is first

116   pretreated by either microfiltration (MF) or ultrafiltration (UF). In all three plants, pre-formed

117   chloramines were added to the process prior to MF or UF filtration to mitigate biofouling on

118   the RO membranes (Figure 1). The RO membranes used in these plants are from three

119   different manufacturers. The membranes used in these three plants are thin film composite

120   with a polyamide skin layer. They were characterized by similar salt (NaCl) rejection and

121   water permeability (Fujioka et al. (2013b). The process flow diagrams of these RO systems

122   are shown in Figure 1. Samples were collected from plant A during cool (A-1) and warm (A-

123   2) weather periods. At plant A, chloramination is normally added downstream of the

124   coagulation process, which was the configuration when sampling campaign A-1 was

125   conducted (Figure 1). During an extended period of warm weather when it is necessary to

126   control algal growth during the coagulation process, chloramination can be added upstream

127   of the coagulation process. Plant A was operated in this configuration when the sampling

128   campaign A-2 took place (Figure 1). Unlike plants B and C, plant A is equipped with a

129   booster pump prior to the third stage to maintain the same average flux at all three stages

130   (Figure 1). All three systems produce reclaimed water for industrial and/or agriculture uses.

131   Plants A and B were designed for a possible indirect potable water recycling application

132   where high quality reclaimed water can be used to replenish an existing reservoir for drinking

133 water supply. Thus, the UV-H$_2$O$_2$ process was also installed after the RO process at these

134 systems for the destruction of residual NDMA in the RO permeate. Similar installation using

135 the UV-H$_2$O$_2$ process specifically for the removal of residual NDMA in the RO permeate can

136 also be found elsewhere (Drewes and Khan, 2011; Plumlee et al., 2008).

137 **[Figure 1]**

138 *2.2. Sampling protocol*

139 RO feed and permeate samples were collected from each RO stage (Figure 1). From each

140 sampling point, one sample was collected from plant C in May and December 2012 and

141 duplicate samples were collected in all other sampling events for N-nitrosamine analysis.

142 These samples (500 mL) were stored in amber glass bottles. Deuterated N-nitrosamines

143 corresponding to each target compound were used as isotope labelled surrogates. These

144 deuterated N-nitrosamines were purchased from CDN Isotopes (Pointe-Claire, Quebec,

145 Canada). A surrogate stock solution containing 100 µg/L of each deuterated N-nitrosamines

146 was prepared in pure methanol. Immediately after sample collection, the surrogate stock

147 solution was added to the sampling bottles to obtain 50 ng/L of each isotope labelled N-

148 nitrosamine. Analytical grade sodium thiosulfate (100 mg/L) was also added to the sample as

149 quenching reagent to prevent any further NDMA formation during transportation and sample

150 processing. From each sampling point, 20 mL sample was collected in plastic bottles for the

151 analysis of cations and boron and 100 mL sample was collected in amber glass bottle for the

152 analysis of anions and total organic carbon. Operating conditions and feed temperature of the

153 RO systems on the sampling day are summarised in Table 1. The difference in feed

154 temperature between the entrance and exit of each RO unit was less than 1 °C.

155 **[Table 1]**

156 *2.3. Analytical technique*

157 An analytical method previously developed for the determination N-nitrosamines in drinking

158 water and treated municipal effluent was employed (McDonald et al., 2012). The method

159 uses solid phase extraction (SPE), gas chromatography and analysis by tandem mass

160 spectrometry with electron impact ionization. Eight N-nitrosamines investigated in this study

161 have molecular weight in the range from 74 to 158 g/mol (Supplementary Material Figure

162   S1). These eight target N-nitrosamines are hydrophilic in the operating pH range of the RO

163   plants (pH 6-8). Further details of their physicochemical characteristics can be found

164   elsewhere (Fujioka et al., 2012a). The use of direct isotope dilution ensures accurate

165   quantification, accounting for analytical variability that may occur during sample processing,

166   extraction and instrumental analysis. SPE was conducted using Supelclean^{TM} Coconut

167   Charcoal SPE cartridges purchased from Supelco (St Louis, MO, USA). The concentrations

168   of N-nitrosamines were quantified using an Agilent 7890A gas chromatograph coupled with

169   an Agilent 7000B triple quadrupole mass spectrometer. Calibration curves were established

170   for each N-nitrosamine with a range of 0.5-500 ng/L. The NMOR calibration curve was

171   extended to account for the NMOR concentration of over 400 ng/L. The quantitative

172   detection limits of this technique were 3 ng/L for NDMA, NDEA, NPIP, and NMOR, and 5

173   ng/L for NMEA, NPYR, NDPA, and NDBA.

174   Total organic carbon (TOC) concentration was determined using a TOC-V$_{CSH}$ analyser

175   (Shimadzu, Japan). Conductivity and pH were measured using an Orion 4-Star Plus

176   pH/conductivity meter (Thermo scientific, USA). Cation and boron concentration were

177   determined using an Inductive Coupled Plasma - Mass Spectrometer (7500CS, Agilent

178   Technologies, Wilmington, DE, USA) following the protocol previously reported by Tu et al.

179   (2011). Anion concentrations were determined using an ion chromatography system

180   (Shimadzu, Tokyo, Japan).

181   **[Figure 2]**

182   *2.4. Calculation*

183   The rejection of N-nitrosamines and other solutes in each RO stage and combined RO stages

184   was calculated using the following equations.

185   Each stage rejection R_i [%] $= \left(1 - \dfrac{C_{pi}}{C_{fi}}\right) \times 100$ (1)

186   Overall rejection R_T [%] $= \left(1 - \dfrac{C_{pT}}{C_{f1}}\right) \times 100$ (2)

5

187 where i is the number of stage, C_{pi} is the solute concentration in the RO permeate of the stage

188 i, C_{fi} is the solute concentration in the RO feed of the stage i, and C_{pT} is the solute

189 concentration in the combined RO permeate.

3. Results and discussion

3.1. Organic and inorganic constituent removal

192 The feed waters to the three RO systems differed markedly in TOC concentration and salinity

193 (Supplementary Material Table S2). In particular, the feed water to plant A exhibited a

194 relatively high conductivity (salinity) at approximately 2.5 mS/cm. The sewer catchment of

195 plant A is predominantly in a low-lying coastal area and is subjected to seawater intrusion. In

196 fact, due to seawater ingresses, boron concentration in the feed to plant A was also higher

197 compared to plant B and C. Despite the high feed water salinity, the quality of RO permeate

198 at plant A was comparable to that at the other two RO systems. Most common cations and

199 anions in the feed water can be rejected well by the RO membranes. As a result, the permeate

200 at all three RO systems was of high quality with respect to basic water quality parameters. In

201 agreement with the 85% water recovery (Table 1) of these RO systems, TOC and

202 conductivity concentrations in the final concentrates were approximately six times greater

203 than those in the feed waters (Supplementary Material Table S2).

204 The rejections of TOC, cations (sodium, magnesium, potassium and calcium), anions

205 (chloride, nitrate and sulphate) and boric acid by all three RO systems are summarised in

206 Figure S3 of the Supplementary Material. Divalent ions (i.e. magnesium, calcium and

207 sulphate) were consistently removed over 99%. On the other hand, in agreement with a

208 previous study by Bellona and Drewes (2007), nitrate rejection was slightly lower than that of

209 all other ions. The rejection of boric acid was in the range of 15-30%, which is consistent

210 with the fact that boric acid has a small molecular size and is uncharged at pH below 8 (Tu et

211 al., 2010). The difference between the charged and uncharged solutes observed here can be

212 attributed to the electrostatic interaction and size exclusion rejection mechanisms. In addition

213 to size exclusion, electrostatic repulsion can also play an important role in the rejection of

214 charged solutes by NF/RO membranes (Bellona et al., 2004).

215 *3.2. N-nitrosamine removal*

216 3.2.1. Occurrence of N-nitrosamines in the RO feed water

217 NDMA was detected in all RO feed water samples (Figure 2). NDMA concentrations (7-16

218 ng/L) detected in the RO feed solutions were below or only marginally higher than the value

219 (i.e. 10 ng/L) in the final product water stipulated by the Australian Guidelines for Water

220 Recycling, with samples from A-2 being the only exception. In A-2, chloramine was added

221 upstream of the coagulation process and thus resulting in an increase in NDMA formation.

222 Results in Figure 2 are consistent with those obtained from previous studies (Farré et al.,

223 2011; Mitch et al., 2005; Plumlee et al., 2008). For typical water recycling plants where

224 NDMA in raw water can be controlled to similar levels found in this study, reducing NDMA

225 formation in the feed (Mitch et al., 2005) and RO filtration can be implemented to meet the

226 guideline value without relying on an additional subsequent treatment process such as AOP.

227 In addition to NDMA, several other N-nitrosamines (i.e. NPYR, NDEA, NPIP, NMOR and

228 NDBA) were also detected in some but not all RO feed water samples (Supplementary

229 Material Figure S4). NMEA, which is the second lowest molecular weight compound among

230 the N-nitrosamines investigated here, was not detected during any sampling campaign.

231 Surprisingly, a comparatively high NMOR concentration (177-475 ng/L) was observed in the

232 feed water at plant C. Compared to plant C, NMOR concentrations detected in the RO feed in

233 plants A-2 and B configurations were low. It is noted that NMOR concentrations in A-1, B

234 and C-1 were not reported due to unsatisfactory variation between duplicate samples and

235 poor recovery of the isotopically labelled internal standard. In fact, a sampling program

236 conducted in plant A from 2010 to 2012 revealed low NMOR concentrations (< 21 ng/L) in

237 the RO feed (Supplementary Material Figure S5) which indicates that a very high NMOR

238 concentration like plant C has not been identified in plant A. Likewise, a sampling program

239 conducted in plant B from 2009 to 2011 also showed a relatively low NMOR concentrations

240 in the range from 9 to 57 ng/L in the RO feed (Supplementary Material Figure S5). The

241 results reported here suggest that high NMOR concentrations in RO feed may be site specific

242 and could relate to certain industrial dischargers. The source of NMOR appears to be site

243 specific and further research is necessary to identify sources of NMOR within the catchment

244 of plant C.

245 After each stage, concentrations of the N-nitrosamines increased to quantifiable levels due to
246 the concentration effect leading to higher feed concentrations in subsequent stages (Figure 2).
247 As a result, the highest N-nitrosamine concentration was consistently observed in the final
248 RO concentrate. For example, NDMA concentrations in the final RO concentrate were two to
249 six times higher than those in the RO feed. Likewise, NMOR concentrations in the RO
250 concentrate were approximately six times higher than those in the RO feed.

251 **[Figure 2]**

252 3.2.2. N-nitrosamine concentrations in the RO permeate

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

264 In all cases, the concentration of N-nitrosamines in the RO permeate increased in later stages
265 due to the increased concentration in the RO feed for each stage (Figure 3). As a result, N-
266 nitrosamine concentrations in the overall RO permeate (i.e. the combined RO permeate of the
267 first, second and third stages) were higher than those in the first stage. The results here
268 indicate that rejection estimates obtained from laboratory-scale systems, which are operated
269 at low water recovery, may result in an underestimation of N-nitrosamine concentrations in
270 the RO permeate. Although the permeation of NDMA through RO membranes can be
271 managed by a subsequent UV-H_2O_2 based AOP, little is known about its removal efficiency
272 for NMOR and other N-nitrosamines. The results reported here also suggest that, in addition
273 to NDMA, it is necessary to monitor the concentration of several other N-nitrosamines
274 particularly NMOR in secondary treated effluent and the corresponding RO permeate.

275 **[Figure 3]**

3.3. *Rejections by RO membranes*

277 3.3.1. Overall rejection

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

285 NDEA rejection at plant A and C varied between zero and 53% (Figure 4). This is
286 considerably lower than the values (86-95%) reported in a recent laboratory-scale study using
287 low pressure RO membranes and synthetic clean water solutions (Fujioka et al., 2012b).
288 Although the mechanism underlining this phenomenon is still unknown, the results reported
289 here indicate that RO filtration in treated wastewater can result in a significant reduction in
290 NDEA rejection. In fact, in a laboratory-scale study Fujioka et al. (2013a) reported a
291 considerable deterioration in NDEA rejection using tertiary effluent as feed water. Overall,
292 NMOR rejection was high and each stage exhibited rejection ranges of 87-91% (Figure 4)
293 which is consistent with previous laboratory-scale studies (Fujioka et al., 2013a; Fujioka et al.,
294 2012b).

295 **[Figure 4]**

296 3.3.2. Rejection at each stage

297 In plant B, 16 inch membrane elements were used whereas 8 inch membrane elements were
298 used in plants A and C. Thus, the hydraulic distribution of plant B can differ significantly
299 from that of plant A and C. At plant B, a significant variation in NDMA rejection (14-78%)
300 was observed among the three RO stages (Figure 5). Changes in the permeate flux after each
301 filtration stage may contribute to this variation to some extent (Fujioka et al. 2012b).
302 However, because permeate flux was not monitored in each individual stage at plant B, it was
303 not possible to confirm this hypothesis. Rejection of N-nitrosamines was further investigated

9

304 using the two sampling events at plant A, focusing on the difference in NDMA rejection
305 among the three stages. As RO filtration progressed, feed pH increased slightly and feed
306 conductivity increased significantly for both sampling events (Figure 6). During the first
307 sampling event (A-1) an increase in NDMA rejection from the first stage to third stage was
308 observed. In general, an increase in feed conductivity (or ionic strength) results in a decrease
309 in N-nitrosamine rejection (Fujioka et al., 2012b). However, the current study revealed an
310 opposite trend which indicates that another factor such as membrane fouling may have been
311 developed more extensively in later stages and may have compensated the decreased trend of
312 NDMA rejection. On the other hand, during the second sampling event (A-2), NDMA
313 rejections decreased as RO filtration progressed to later stages (Figure 6). The results
314 reported here indicate that NDMA rejections among three RO stages may vary significantly
315 even when operating conditions (i.e. permeate flux and recovery) were maintained constant.

316   **[Figure 5]**
317   **[Figure 6]**

4. Conclusions

319 NDMA was detected in all feed samples at the three full-scale RO trains investigated in this
320 study. Although most other N-nitrosamines were not detected in the RO feed, several N-
321 nitrosamines became detectable as the feed was further concentrated after each filtration stage.
322 N-nitrosamine concentrations in the final RO concentrate were up to six times higher than
323 those in the RO feed. As a notable exception, one of the three plants exhibited high NMOR
324 concentrations (177-475 ng/L) in the feed, resulting in high NMOR concentrations (34-76
325 ng/L) in the permeate. In most cases, NDMA, NDEA and NDBA were detected below the
326 Australian guideline value in the RO permeate. Overall rejection of NDMA and NDEA
327 among the three RO systems varied significantly with a range of 4-47% and 0-53%,
328 respectively. NDMA rejections among three RO stages also exhibited a significant variation
329 in several cases. These rejection variations may have resulted from the difference in feed
330 temperature and possibly membrane fouling. These findings suggest that N-nitrosamine
331 rejection estimates derived from laboratory-scale flat-sheet membrane studies, which are
332 operated at very low water recovery, may not be representative of full-scale operation. On the
333 other hand, overall NMOR rejections were equally high with a range of 81-84%. The findings
334 of this study provide insights for potential variations in N-nitrosamine rejection among

335 different RO systems and RO stages. Nevertheless, it is prudent to note the causes of some
336 variations reported here were not sufficiently identified due to changes in multiple parameters
337 during full-scale RO operation. Therefore, additional work is necessary to examine the
338 impact of each cause (e.g. fouling and feed temperature) using a pilot- or full-scale RO
339 systems.

5. Acknowledgements

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