



Review

N-nitrosamine removal by reverse osmosis for indirect potable water reuse – A critical review based on observations from laboratory-, pilot- and full-scale studies

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ABSTRACT

N-nitrosodimethylamine (NDMA) and several other N-nitrosamines have been identified as probable human carcinogens. Here, we review key aspects related to the occurrence and removal of N-nitrosamines by reverse osmosis (RO) membranes in the context of indirect potable water reuse. A comprehensive analysis of the existing data reveals significant variations in the rejection of NDMA by RO membranes reported in the literature, ranging from negligible up to 86%. This review article provides some insight into the reasons for such variations by examining the available data on the effects of operating conditions on NDMA rejection. Amongst several operating parameters investigated so far in the literature, feed temperature, membrane permeate flux, feed solution pH and ionic strength were found to have considerable impact on NDMA rejection by RO membranes. In particular, it has been recently shown that seasonal changes in feed temperature (e.g. from 20 to 30 °C) can result in a significant decrease in NDMA rejection (from 49% to 25%). However, the combined effects of all operating parameters identified in the literature to date can only account for some of the variations in NDMA rejection that have been observed in full-scale RO plants. The impacts of membrane fouling and particularly chemical cleaning on the rejection of N-nitrosamines have not been fully investigated. Finally, this review article presents a roadmap for further research required to optimise the rejection of NDMA and other N-nitrosamines by RO membranes.

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Contents

| | |
|--|-----|
| 1. Introduction | 504 |
| 2. Indirect potable water reuse and N-nitrosamines | 504 |
| 2.1. Water reclamation process | 504 |
| 2.2. Occurrence of N-nitrosamines in water recycling schemes | 505 |
| 2.2.1. Presence of N-nitrosamines in wastewater | 505 |
| 2.2.2. NDMA precursors | 505 |
| 2.2.3. NDMA formation | 505 |
| 2.3. Health-based water quality guidelines and standards for N-nitrosamines | 507 |
| 2.4. N-nitrosamine quantification using chemical analysis | 507 |
| 2.5. Removal of N-nitrosamines during water reclamation | 509 |
| 3. N-nitrosamine removal by RO membranes | 509 |
| 3.1. Rejection of N-nitrosamines in laboratory-scale studies | 509 |
| 3.2. Rejection of N-nitrosamines and N-nitrosamine precursors in pilot- and full-scale installations | 509 |
| 3.2.1. Rejection of N-nitrosamines | 509 |
| 3.2.2. Rejection of N-nitrosamine precursors | 510 |
| 3.3. Factors affecting N-nitrosamine rejections | 510 |
| 3.3.1. Feed concentration | 510 |

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| | | |
|--------|--|-----|
| 3.3.2. | Permeate flux | 510 |
| 3.3.3. | Feed pH | 511 |
| 3.3.4. | Total dissolved solids concentration | 511 |
| 3.3.5. | Feed temperature | 511 |
| 3.3.6. | Membrane fouling and membrane ageing | 511 |
| 3.4. | Future research roadmap | 512 |
| 4. | Conclusions | 512 |
| | Acknowledgements | 512 |
| | References | 512 |

1. Introduction

Water reuse has grown significantly in recent years in response to the increasing demand for water brought about by population increase, urbanisation, and diminishing and uncertain availability of freshwater resources. Many water authorities around the world have now recognised the potential value of water reuse after experiencing severe droughts as well as the environmental and economic costs of imported water [1–3]. Since the quality of reclaimed water for potable reuse is stringently regulated, reverse osmosis (RO) treatment has become an increasingly common component of the water reclamation process. RO membranes can successfully remove a wide range of contaminants including inorganic salts and trace organic chemicals [4,5]. However, the rejection of N-nitrosodimethylamine (NDMA) by RO membranes appears to be highly variable [6,7]. N-nitrosamines including NDMA can readily be formed during the disinfection of biologically-treated effluent using chlorine or chloramines [8,9]. Given the probable carcinogenic potency of NDMA and several other N-nitrosamines [10,11], the fate of these compounds in water reclamation applications is of significant interest to both the scientific community and water authority.

For indirect potable water reuse applications involving the use of the RO process, concentration of NDMA in the final product water can be controlled via several strategies. NDMA concentration can be minimised by reducing the formation of NDMA during the chloramination process. This can be achieved by dosing preformed chloramine [12] and reducing the contact time of chloramination [13,14]. However, reducing the NDMA formation may not be sufficient if a higher NDMA concentration than the regulatory level occurs in the inflow of the wastewater treatment plant (WWTP). An alternative approach is to use an additional treatment process for the removal of NDMA. Possible treatment technologies include UV/H₂O₂ treatment process, natural attenuation during aquifer recharge, and RO filtration.

Advanced oxidation using a combination of UV radiation and dosed hydrogen peroxide (H₂O₂) to form hydroxyl radicals has been proven to be effective for the removal of NDMA and has been applied following RO filtration in several water reclamation schemes around the world [6,15]. However, the energy consumption required by UV/H₂O₂ treatment for the control of NDMA is high and can have a negative consequence of increasing the carbon footprint of the water reuse scheme. Moreover, it is still necessary to control the concentration of NDMA by other processes during wastewater reclamation since the removal of NDMA by UV/H₂O₂ treatment is sometimes incomplete [6]. At a water reuse facility in Southern California, there were some periods when reclaimed water after UV/H₂O₂ treatment had to be blended with other non-recycled sources to reduce NDMA concentration in the final product to below the 10 ng/L notification level [7].

Natural attenuation over an extended retention time in an aquifer or surface reservoir has been shown to be effective for the removal of NDMA and other N-nitrosamines [16,17]. Although

natural attenuation is likely to play a significant role as a post RO treatment process for the removal of NDMA and other N-nitrosamines, most water authorities are still reluctant to exclusively rely on this passive treatment technique. A reliable removal efficiency of NDMA and other N-nitrosamines remains a major focus for the control of these contaminants in indirect potable water recycling practices.

RO membranes are widely used for the treatment of reclaimed water for indirect potable reuse and other applications. However, the effectiveness of RO membranes for the rejection of NDMA and other N-nitrosamines is still poorly understood. Broad discrepancy exists in the existing scientific literature regarding the rejection of NDMA by RO membranes. For instance, NDMA rejection by a commonly used RO membrane (TFC-HR, Koch Membranes) was reported to be 50% at the West Basin Municipal Water District water recycling plant in California, USA [6]. At the Scottsdale Water Campus (Arizona, USA), NDMA rejections by the same type of RO membrane (TFC-HR) were reported to be 10% and 70% during two separate sampling events [6]. Compared to NDMA, little is known about the fate of other N-nitrosamines in water reclamation due to the scarcity of sampling data. This paper aims to provide a comprehensive review on the fate of N-nitrosamines and their rejections by RO treatment during water reclamation.

2. Indirect potable water reuse and N-nitrosamines

2.1. Water reclamation process

Indirect potable water reuse is generally performed through a ‘multiple barrier’ approach that incorporates both engineered and natural treatment processes as well as non-treatment measures. These multiple barriers may variably include (1) residential/industrial source control; (2) conventional wastewater treatment; (3) advanced water treatment; (4) environmental buffer and blending; and (5) drinking water treatment [3].

A notable approach for the advanced treatment of reclaimed water is the use of integrated membrane systems (Table 1). Since secondary effluents have high fouling propensity against RO membranes [18], microfiltration (MF) or ultrafiltration (UF) treatment is usually used as a pretreatment step to minimise membrane fouling in the subsequent RO process. The RO process substantially reduces the concentration of dissolved solids including macro-organic molecules and inorganic salts [19]. RO membranes can also achieve an excellent removal of a large range of trace organic chemicals [5,19–21]. Although RO membranes can remove bacteria and viruses [22,23], it is still common to deploy either UV- or chlorine-based disinfection processes as a ‘redundant’ post treatment to inactivate human pathogens (Table 1). Because the rejection of NDMA by RO membranes is highly variable and can be quite low, the advanced oxidation UV/H₂O₂ process may also be used for the destruction of NDMA that can permeate through the RO membrane.

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