



Enhancing organic matter removal in desalination pretreatment systems by application of dissolved air flotation



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HIGHLIGHTS

- Use of DAF for organic matter removal is a good option in desalination systems
- High removal of biopolymers and humic substances were observed using coagulation- DAF
- Optimal coagulant dose was dependent on humic substance concentration

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ABSTRACT

Membrane fouling in reverse osmosis (RO) systems caused by organic matter (OM) remains a significant operational issue during desalination. Dissolved air flotation (DAF) has recently received attention as a pre-treatment option for seawater OM removal; however, only a limited number of studies have been undertaken. This may be because it is difficult to characterise OM in seawater due to the high salt content and low carbon concentration. In this study, DAF pre-treatment experiments were conducted using a model seawater solution, and real seawater and brackish water samples. DAF performance was determined via conventional water quality parameters as well as fluorescence excitation–emission matrix (FEEM) spectroscopy and liquid chromatography with organic carbon detection (LC-OCD). Biopolymers and humic substances were the major organic fractions removed between 38 and 84% and 20–61% depending on the sample, respectively. The optimal normalised coagulant dose (Fe^{3+} to DOC ratio) was observed to be 0.5–4 at pH 5.5 increasing to 4–12 at pH 7.5. At pH 5.5, the optimum coagulant dose increased with increasing humic character of the feed water. Overall, the OM removal efficiency by DAF observed in this study was higher than reported for other membrane-based processes; a combination of DAF and biofiltration is likely to be complementary.

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1. Introduction

Reverse osmosis (RO) is a well-established treatment process for the production of drinking water from brackish water and seawater [1]. Membrane fouling results in a lower production efficiency that can be attributed to a deposition of particles, colloids, inorganic matter (scaling), organic matter and biological products [2]. While scaling can be effectively controlled by pH adjustment [3], problems of organic fouling and bio-fouling persist [2,4,5]. Several pre-treatment options can be implemented to limit bio-fouling in RO, including coagulation–flocculation followed by either dual media filtration (DMF) or, more recently, dissolved air flotation (DAF), in addition to biological filtration (BF) and/or ultra-filtration (UF) [6,3,7]; however, most of

these studies have focussed on comparisons with respect to recovery rather than the removal of specific components [7]. Studies have also investigated hybrid processes, including the incorporation of adsorbents, such as powdered activated carbon (PAC), or coagulation–flocculation with membrane treatment [7]. The pre-treatment process is known to have a significant impact on the organic matter (OM) removal efficiency, not only in terms of relative concentration but also the particular OM fraction removed, for example, coagulation–flocculation-DAF may target larger, charged, hydrophobic natural organic matter, while biofiltration and GAC would be expected to remove smaller, biodegradable fractions [8].

DAF is a well-known treatment process for the removal of OM in fresh water due to its effectiveness at floating the low density OM flocs that are formed; however, its application has been more limited in seawater. It is known that the operating conditions of DAF in saline environments are different to those applied for surface waters due to the high salt concentrations present [9]. For example, a DAF saturator operating pressure of between 4 and 5 bar is generally used for OM

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removal in freshwater [9,10]; however, since, the air solubility and the air transfer efficiency are different in comparison to fresh water, the recycle ratio and pressure should be adjusted to higher values than for fresh water [9]. Furthermore, ferric coagulants are recommended instead of aluminium coagulants which are too soluble in seawater [6]. It is also expected that pre-treatment conditions, such as pH and coagulant dose, will have a significant impact on the DAF performance with respect to OM removal from seawater; however, few studies have investigated this topic [11,12] and hence this remains a knowledge gap.

OM is present in the marine environment at concentrations of approximately 1 to 4 mg/L as carbon (referred to as mgC/L from hereafter) [13–15]. It comprises a range of compounds produced by naturally decaying algae and other marine vegetation as well as terrestrially-derived OM. To date, the relative efficiency of pre-treatment options for RO membranes has been assessed mostly based on bulk organic removal [16–19]. One reason for this is that characterising marine OM is difficult due to the highly diluted mixture of heterogeneous organic compounds in a saline aqueous solution. However, advanced OM characterisation techniques, such as fluorescence excitation–emission matrix (FEEM) spectroscopy [20], as well as size exclusion chromatography with a high sensitivity organic carbon detector [21,22], show promise for the characterisation of low concentrations of OM in saline matrices. For example, FEEM spectroscopy can be used to analyse fluorescing OM fractions in either marine or fresh water at very low OM concentrations. FEEMs can be processed using techniques such as parallel factor analysis (PARAFAC) [23] to assist in identifying components that can be representative of terrestrially-delivered, humic-like, marine/reprocessed-like and protein-like OM based on the excitation/emission spectra [24]. Terrestrial humic-like materials have excitation and emission maxima at longer wavelengths when compared with marine humic-like materials which are less aromatic and presumed to have lower molecular weight [25]. For example, Murphy et al. [20] used PARAFAC of fluorescence spectra and found that coastal and marine dissolved OM fluorescence could be divided into 4–5 humic-like and 3–5 protein-like components. Liquid chromatography with organic carbon detection (LC-OCD) is a size-exclusion chromatographic technique that separates OM fractions according to size, ion interaction and hydrophobic interaction with the former being most dominant mechanism of fractionation [22]. It has an organic carbon detector based on the Grantzel thin-film UV-reactor. It has been suggested that LC-OCD is of sufficient sensitivity to measure water containing low concentrations of OM, for example, RO feed and permeate [22]. For example, it was demonstrated that the seawater OM comprised 50% of humic substances, 10% biopolymers, and 40% building blocks and neutrals [26]. Furthermore, LC-OCD showed that optimised coagulation could remove 95% of hydrophobic compounds from seawater [27]. Therefore, a combination of FEEM spectra and LC-OCD appear to have great potential for assessing the concentration and character of seawater OM.

The aim of this research was to investigate the impact of: (1) DAF operating conditions, including pH and coagulant dose; and (2) influent water quality characteristics in terms of OM character and salinity on DAF performance with respect to OM removal as characterised using FEEM and LC-OCD.

2. Materials and methods

2.1. Feed water

2.1.1. Preparation of model compound seawater solution

A model seawater was prepared for three reasons: 1) for use as a standard solution to ensure a sample of consistent character was always available; 2) to gain insight into the removal of specific fractions; and 3) to ensure that advanced characterisation methods

were able to measure consistently and accurately in the low concentration and high salinity conditions observed for both feed seawater and treated sample. Six model compounds were selected to represent seawater organic matter: sodium alginate (0.11 mgC/L), gum xanthan (0.05 mgC/L), bovine serum albumin (BSA) (0.76 mgC/L), L-tryptophan (7×10^{-5} mgC/L), and oxalic acid (0.06 mgC/L) from Sigma Aldrich (Australia) and Suwannee River humic acid (standard II) (0.48 mgC/L) from the International Humic Substances Society (IHSS) (USA). All chemicals were of analytical grade and were used without any further purification. The model compound solution was prepared to a 1.5 mgC/L concentration in 32 g/L NaCl (Sigma Aldrich, Australia) to provide an organic matter solution of similar concentration to that typically found in seawater (as per [13–15]). The specific concentrations were selected such that distinct signals were obtained when applying advanced organic matter characterisation techniques for analysis after DAF treatment (see SI Figs. S1 and S2).

2.1.2. Real seawater/brackish water samples

Water samples were obtained from various locations to provide a range of feed compositions, particularly with respect to OM concentration and character and salinity, to enable a thorough assessment of the impact of water quality on DAF performance. Specifically, raw seawater samples were obtained from Maroubra Beach and Sydney Harbour and brackish samples from the estuary of Parramatta River in Sydney. These samples were collected at the beach approximately 3–5 m from the shore by the UNSW personnel and were transported to the UNSW laboratory on the same day. Seawater samples from the Gold Coast Desalination Plant (Queensland) and brackish water samples from an estuary at Denham (Western Australia (WA)) and Gascoyne River (WA) were also collected and shipped to the UNSW laboratory overnight. Water was collected in leachate free polypropylene containers to minimise OM leachate from the sample container [28]. Sampling locations were deliberately selected to provide a wide range of water quality characteristics.

2.2. Dissolved air flotation experimental procedure

DAF jar tests were performed using a dissolved air flotation batch jar tester (Model DBT6, EC Engineering, Canada). Ferric chloride (Sigma Aldrich, Australia) was added as coagulant, as recommended by Edzwald and Haarhoff [6], during a 2 min rapid mix (200 rpm) stage after which pH was adjusted to 5.5, 6.5 or 7.5 using 0.1 M HCl or 0.1 M NaOH solution (Chem-Supply, Australia). This was followed by a 10 min flocculation (30 rpm) period and then a 10 min period of flotation using a 14% recycling ratio and saturation pressure of 500 kPa [9]. DAF-treated samples were collected after flotation via sampling ports and filtered through a 0.45 μ m filter (PES, Sartorius Stedim Biotech GmbH, Germany) prior to analysis.

2.3. Analyses

2.3.1. Zeta potential

The charge of the coagulated particles was measured as zeta potential (ZP) using the ZetaSizer Nano (Malvern, Australia). The ZP analysis was performed for all the feed samples and for each dose at different pH values. All samples were measured in triplicate.

2.3.2. Modified fouling index

The fouling potential of feed and DAF-treated samples was evaluated by measuring modified fouling index-UF (MFI-UF) [29]. Membranes used in MFI-UF measurements were first tested using MQ water and then with the feed solution. During the experiment, 1 L of feed water was treated through a 10 kDa polyethersulfone membrane (PES) (Millipore) under 200 kPa of nitrogen gas. The permeate volume was recorded every 10 s.

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