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Formation of disinfection by-products in remineralized desalinated seawater with bacterial materials as precursor

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ABSTRACT

As the essential target of disinfection, bacterial materials should also be considered as one type of disinfection by-products precursors (DBPs). This study investigated the disinfection by-products formation potential (DBPFP) of bacterial materials during chlorination and chloramination. The bacterial materials were harvested from mature biofilm developed in remineralized desalinated seawater with different carbon sources. It was observed that calcium and magnesium which were added to desalinated seawater for remineralization could enhance DBPFP of bacterial materials, with magnesium being more effective. The enhancement effect was related to the ability of calcium and magnesium to shift low molecular weight fractions of bacterial materials to more hydrophobic and larger molecular weight fractions. Bacterial materials developed with humic acid as carbon source contained more hydrophobic and larger molecular weight fractions, hence had higher DBPFP than bacterial materials developed with acetate. DBPs yields of bacterial materials were lower than that of NOMs precursor during chlorination, but during chloramination DBPs yields of bacterial materials were higher. The extracellular polymeric substances (EPS) matrix had higher DBPs yield than biofilm cells. Polysaccharide was more likely to be the main DBPs precursor in the EPS matrix.

1. Introduction

As global fresh water resource is becoming increasingly scarce due to rapid population growth and economic expansion, seawater desalination has become an important option of alternative sources of drinking water in many parts of the world. Over the past few decades, the global production of desalinated seawater has increased dramatically. Seawater desalination had accounted for 94% of water sources in Bahrain and 98% in the United Arab Emirates [1]. In Israel, desalinated seawater contributed to over 20% of the annual fresh water supply in 2009 and is becoming the main water resource in the future [2]. Reverse Osmosis (RO) is a well-developed and commonly practiced process to achieve seawater desalination [3]. However, due to the high rejection rate of salts by RO, the permeate is usually characterized by relatively low pH value, very low buffering capacity and very low hardness [4]. Hence, the raw product of RO is highly aggressive and highly corrosive [5]. This quality of the raw product of RO is not readily for direct usage. Lacking of minimal concentrations of minerals is detrimental to human health [6]. Corrosion problems to traditional metal pipelines can be induced during the transportation of RO permeate in drinking water distribution network [7]. In addition, water quality could also be adversely affected due to the detachment of metal oxide from the inner surface of pipelines into the water [8]. Therefore, post-treatments, such as remineralization, are necessary to stabilize the raw product of RO and improve its quality for the intended purposes [9]. Typical methods include blending with other sources of water and chemical addition of calcium and magnesium compounds [3]. Direct chemical addition of calcium and magnesium compounds could provide greater flexibility and control over the required water quality [9]. The desalinated seawater after remineralization can help to control pipelines corrosion and top up nutrient level.

Although it is known that RO membrane is able to efficiently remove nutrient and microorganism cells, considerable bacterial growth was observed in desalinated seawater in previous studies. Park & Hu [10] had reported significant biofilm and planktonic cells growth in an RO permeate. Biofilm growth was also observed in distribution network transporting desalinated seawater [11]. It was suggested that there might be considerable portion of low molecular weight organic acids that permeated through the RO membrane due to flaws and contaminations in parts of the membrane [12]. Thus, this could provide favorable conditions for microorganism growth. In addition, partial permeation of bacterial cells was suggested, which could act as inoculums for subsequent bacterial growth [10]. Therefore, with sufficient level of nutrient and bacterial inoculums, considerable biofilm in

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desalinated seawater could be observed. Moreover, the process of remineralization to desalinated seawater might further enhance biofilm growth. Blending with other sources of water can introduce organic substances to the low organics containing desalinated seawater, and hence supporting biofilm growth. Addition of calcium and magnesium compounds may also enhance biofilm growth, as calcium and magnesium have long been known to have the potential of influencing biofilm formation [13–15]. The potential effect of remineralization on bacterial activities is a major concern of remineralized desalinated seawater. Therefore, proper measures of biofilm control ought to be adopted to control the biofilm growth in water distribution system and hence ensure safe water quality.

Disinfection is an effective method to control bacterial activities. Free chlorine and chloramines disinfection are commonly applied in desalination plants [16]. However, one big concern of disinfection is the disinfection by-products (DBPs). They had been frequently detected in drinking water distribution network and were raising more and more concerns because they were potentially carcinogenic and associated with other health risks [17]. Two largest groups of DBPs are trihalomethanes (THMs) and haloacetic acids (HAAs) [18]. DBPs are formed though the reaction of disinfectants with DBPs precursors, such as natural organic matters (NOMs), bromide, iodide, etc. [19]. Recently, researchers have also turned their interests to other DBPs precursor, such as biomolecules. Hong et al. [20] reported that chlorinating algal cells could yield THMs and HAAs. It was also reported that soluble microbial products in effluent organic matter from biological wastewater treatment plants could produce THMs and HAAs after chlorination [21]. As the essential purpose of water disinfection is to target the microorganisms in the water, bacterial materials as DBPs precursor should be considered. Wang et al. [22] reported that most common DBPs were detectable during chlorination and chloramination of *Escherichia coli*. A positive correlation between DBPs formation and the log reduction of *E. coli* was observed, confirming that the DBPs were contributed by breaking down of bacterial cells. Meanwhile, it was also observed by Wang et al. [22] that *Pseudomonas aeruginosa*, a typical bacterial species in the pipelines biofilm, could form DBPs.

Therefore, considering the biofilm growth in remineralized desalinated seawater, this study aimed to investigate the formation of DBPs with bacterial materials as precursor. As biofilm cells and extracellular polymeric substances (EPS) matrix are having distinct characteristics and may have different behavior upon remineralization [23,24], the DBPs formation potential (DBPFP) by these two parts were investigated separately. In addition, as it had been reported that carbon source could affect the characteristics of bacterial materials [25], DBPFP of bacterial materials grown with different substrate was compared. Lastly, this study compared the DBPFP of bacterial materials to DBPFP of NOMs.

2. Materials and methods

2.1. Water sampling and preparation

Desalinated seawater used in this study was generated with a lab-scale two phases RO system (Venture Merger, Singapore). Fresh seawater collected from nearby coastal area was firstly passed through a cartridge filter with an absolute pore size of 5 μm before being pumped into the feed tank. Coarse particle were removed to facilitate efficient RO process. Then, the filtered seawater was pumped through the first pass RO unit with a membrane model SWC5-4040 (Hydranautics, USA). A high salt rejection rate of minimum 99.6% was expected at an applied pressure of 5.5 MPa at 25 °C. A permeate recovery rate of 10% was achieved. Subsequently, the permeate from the first pass was pumped through the second pass RO unit with a membrane model ESPAB-4040 (Hydranautics, USA). This membrane achieved a high flux at a low pressure of 1.05 MPa at 25 °C with a salt rejection rate of at least 99.0%. Permeate recovery rate of this membrane was 15%. The permeate from the second pass was collected in a sterilized glassware and stored in dark environment at 4 °C prior to usage. All glassware used in this study were rendered organic-carbon-free by soaking in 0.1 mol/L hydrochloric acid overnight followed by combustion at 550 °C for 4 h. Plastic caps of glassware were cleaned by soaking in 10% (w/v) sodium persulfate at 60 °C for 1 h and rinsed with carbon-free de-ionized (DI) water generated by TKA Smart2Pure Water Purification System (Thermo Fisher, USA). The general water quality parameters of the RO permeate were measured and listed in the Supporting Information (SI) Table S1.

Calcium in the form of calcium chloride and magnesium in the form of magnesium chloride hexahydrate were added to desalinated seawater according to the remineralization guidelines and recommendations [9]. 40 mg/L of calcium ion or 10 mg/L of magnesium ion were added for remineralization.

2.2. Chemicals and solution chemistry

Disinfectants used in this study were free chlorine and monochloramine. Free chlorine was supplied by a stock solution of sodium hypochlorite stored in dark environment at 4 °C. It was freshly diluted to required dosage before being applied. Mono-chloramine was freshly prepared through reaction between free chlorine and ammonium chloride. Sodium hypochlorite solution was slowly poured into ammonium chloride solution with a molar ratio of 1:4. The mixture was left for around half an hour with continuous mixing for complete reaction. Opaque materials were used to avoid photo-degradation of disinfectants. The free chlorine and mono-chloramine disinfectant concentrations and the residue levels were measured with indophenol method, using a DR/890 Portable Colorimeter (Hach, USA) and powder pillows of Freechlor F Reagent and Monochlor F Reagent according to the user manual. The detection range was 0 to 4.5 mg/L as free chlorine. Samples were diluted accordingly before measurement if necessary.

2.3. Bacterial cultivation and extraction

In consideration of the potential release of organic contents from commercial biofilm reactors into desalinated seawater, a new biofilm reactor consisting of only glass material was designed and used in this study. Biofilm was grown in calcium or magnesium remineralized desalinated seawater with 2 mg/L of sodium acetate or humic acid (HA) as a model NOMs under room temperature on coupons in biofilm reactors. After reaching the stationary phase, mature biofilm was harvested. Coupons were removed from the biofilm reactors. DI water was used to wash the coupons gently to remove planktonic cells that might deposit on the surface of biofilm. Then, the coupons were immersed in DI water and biofilm was extracted with a common method of ultrasonication [26]. Five minutes of ultrasonication was sufficient to extract the whole biofilm from the coupons as verified by gram staining on the coupons afterwards. Centrifugation of 10 min at 10000*g* was applied to freshly extracted biofilm in order to separate biofilm cells and EPS matrix.

2.4. Analytical methods

2.4.1. Bacterial materials quantification and characterization

The quantities of biofilm cells and EPS matrix were measured as total organic carbon (TOC) by TOC-L Total Organic Carbon Analyzer (Shimadzu, Japan). It adopted the 680 °C combustion catalytic oxidation method developed by Shimadzu. It had a high level of detection sensitivity with a range of 4 μg/L to 30,000 mg/L. The results were converted to $\mu g/cm^2$ according to the surface area of the coupons. In addition, the composition of EPS matrix was analyzed. Two major groups of components in the EPS matrix, polysaccharide content and protein content were extracted and quantified separately. For polysaccharide, Phenol-Sulfuric Acid Method [27] was used. Two mL of EPS matrix extracted with sodium hydroxide solution of pH 11 was reacted