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# Application of monochloramine for wastewater reuse: Effect on biostability during transport and biofouling in RO membranes

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ABSTRACT

The rising demand for clean and safe water has increased the interest in advanced wastewater treatment and reuse. Reverse osmosis (RO) can provide reliable and high-quality water from treated wastewater. Biofouling inevitably occurs, certainly with wastewater effluents, resulting in RO performance decline and operational problems. Chlorination of feed water has been commonly applied to limit biological growth. However, chlorine use may lead to a loss of membrane integrity of RO systems. In this study the potential of monochloramine as an alternative for chlorine was studied by (i) evaluating the biological stability of a full-scale wastewater membrane bioreactor (MBR) effluent during transport over 13 km to a full-scale RO plant and (ii) assessing the biofouling control potential in membrane fouling simulator (MFS) and pilot-scale RO installation. Microbial water analysis was performed on samples taken at several locations in the full-scale water reuse system (MBR effluent, during transport, and at the RO inlet and outlet) using a suite of tools including heterotrophic plate counts (HPC), adenosine triphosphate (ATP), flow cytometry (FCM), and 16 S rRNA gene pyrosequencing. Growth potential tests were used to evaluate the effect of monochloramine presence and absence on bacterial growth. Results showed limited changes in the microbial water quality in the presence of monochloramine. MFS studies showed that membrane biofouling could be effectively repressed by monochloramine over prolonged time periods. The normalized salt passage in a pilot RO system with monochloramine dosage was constant over a one year period (data of last 130 days presented), demonstrating that no membrane damage occurred. From this study, it can be concluded that monochloramine dosage in wastewater applications is effective in controlling biofouling in RO systems and maintaining a monochloramine residual during water transport provides biologically stable water.

## 1. Introduction

The Environmental Protection Agency 2012 guidelines [1] for water reuse reported that “Treated wastewater is increasingly being seen as a resource rather than simply waste”. Reclaimed water can fulfill most water demands, as long as it is satisfactorily treated to ensure water quality suitable for the intended use [2]. Reverse osmosis (RO) and nanofiltration (NF) membranes produce high-quality water from sources such as brackish or seawater and secondary treated wastewater effluent [3]. The pre-treated feed water or secondary wastewater effluents still contain dissolved organic compounds, microorganisms, and colloidal particles, contributing to membrane fouling [4]. Therefore, membrane fouling is a major constraint for the operation and cost effectiveness of membrane systems [5]. Fouling, severely limits membrane performance, leading to a reduction in permeate quality and quantity, and eventually causing membrane damage. Several types of fouling can occur simultaneously and affect each other [6]. In practice, an extensive pre-treatment can eliminate scaling and particulate fouling but to a lesser extent organic and biological fouling [7,8].

Biofouling or biological fouling is the excessive growth of a biofilm that results in an unacceptable performance decline [7]. In practice, a 10–15% increase in feed channel pressure drop or reduction in permeate flux is considered operationally unacceptable [8,9]. Reducing biological growth and biofouling in transport pipes and membrane

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systems is normally achieved by limiting the essential nutrients for bacterial growth, mainly (but not exclusively) organic carbon [10], and/or dosing disinfectants. Free chlorine is the most commonly used disinfectant to prevent biological growth in water because of its ability to rapidly inactivate most pathogenic microorganisms [2,11]; however residual chlorine has to be removed from the water before entering RO systems with chlorine-intolerant polyamide membranes. Moreover, the reaction of chlorine with organics present in the water results in the formation of halogenated organic by-products such as trihalomethanes (THMs), which are classified as suspect carcinogens for humans [12,13]. Several studies have reported that RO membranes reduce up to 80% of the THM concentration present in the feed water [14,15]; however, using an alternative disinfectant with a lower halogenated organic by-product formation is desirable. Monochloramine dosage was introduced in an attempt to abide by the new THM regulations, due to its weaker tendency to produce halogenated organic reaction products [16]. Monochloramine, the most stable form of chloramine, although a more slowly acting and weaker disinfectant than free chlorine, can be more effective in penetrating and inactivating biofilms [17,18]. In aqueous solution, naturally present ammonia ($NH_3$) or ammonium ions react with chlorine or hypochlorite to form inorganic chloramines [19] through the reaction:

$$HClO + NH_3 \leftrightarrow NH_2Cl \text{ (monochloramine)} + H_2O. \quad (1)$$

The stability and the type of chloramine formed depends on the ratio ammonia/chlorine. Monochloramine formation is a function of the pH and occurs most rapidly at a pH value of approximately 8.3.

For wastewater reuse applications and further treatment of secondary wastewater effluents, preventing bacterial regrowth during transport to tertiary treatment facilities is essential for an optimal RO treatment performance. During transport, several biological processes can occur including biofilm formation on the pipe walls and biofilm detachment [11,20], microbial growth in the bulk water [21], biocorrosion of pipe material [22,23], and proliferation of pathogenic bacteria [24] deteriorating the water quality. Ideally, the goal is to transport biologically stable water where microbial growth is restricted [25]; however, due to the development of more sensitive and accurate microbial analysis techniques changes in microbial presence can be detected without necessarily having a negative impact on the water quality rebuking the earlier definition of biological stability [26,27].

In this study, monochloramine was used to disinfect a membrane bioreactor (MBR) permeate and provide biologically stable effluent water during transport to an RO treatment facility. The treated effluent was transferred by a 13 km long pipe to the RO facility, where monochloramine residual was removed before the water entered the RO membranes. The RO treatment plant suffered from performance decline due to fouling development in the membrane modules. The aim of this study was to investigate the effect of monochloramine dosage on the biological stability of the water during transport. The effectiveness of monochloramine dosage in biofouling control and possible consequences for membrane damage were assessed in lab-scale membrane fouling simulator (MFS) experiments and pilot-scale membrane module experiments. The effect of monochloramine removal and RO filtration on microbial water quality was also examined. A suite of microbial analysis techniques including flow cytometry and pyrosequencing was applied.

## 2. Material and methods

### 2.1. Analysis of water samples from practice

#### 2.1.1. Site description

The RO water treatment facility (DECO) produces demineralized water, cooling tower supply water, and ultrapure water for industrial usage in Terneuzen, the Netherlands (51°20′08″ N, 3°49′40″ E). Since 2010, a membrane bioreactor (MBR) constructed and operated by Evides Industriewater on the site of the municipal wastewater treatment plant (WWTP) (51°17′49″ N, 3°50′14″ E) has been used to produce feed water for the DECO water treatment facility. Table 1 summarizes the average MBR effluent quality. The effluent from the MBR is disinfected using monochloramine and transported over a 13 km long pipe to the DECO facility (residence time of 4 h). Monochloramine was formed by dosing ammonium chloride ($NH_4Cl$ - a 20% solution) and sodium hypochloride (NaClO - a 12.5% solution) according to the following stoichiometric equation (Molar ratio = 1).

**Table 1**
Average MBR effluent quality.

| Parameter | Unit | Value |
|---|---|---|
| Specific conductivity at 25 °C | µS/cm | 1600 ± 450 |
| pH after acid dosage | – | 7.4 ± 0.2 |
| Temperature | °C | 15 ± 4 |
| $O_2$ | mg/L | 9.7 ± 0.2 |
| Total COD | mg/L | 34 ± 8 |
| Total BOD | mg/L | < 3 |
| $Ca^{+2}$ | mg/L | 71 ± 19 |
| $Mg^{+2}$ | mg/L | 22 ± 8 |
| Bicarbonate ($HCO_3^-$) | mg/L | 280 ± 80 |
| TSS | mg/L | 0.3 ± 0.1 |
| TKN | mg/L | 1.9 ± 0.6 |
| $NH_4^+$ | mg/L | 0.9 ± 0.1 |
| Ortho-P | mg/L | 0.8 ± 0.9 |
| $SO_4^{-2}$ | mg/L | 88 ± 22 |

COD: chemical oxygen demand, BOD: biological oxygen demand.

$$NH_4Cl + NaClO \rightarrow NH_2Cl + H_2O + NaCl \quad (2)$$

At the DECO facility, the water is consecutively treated by sulphuric acid dosage ($H_2SO_4$), 50 µm screens, an antiscalant, and sodium bisulfite dosage for monochloramine removal (residual monochloramine ≈ 1 ppm). The water is then fed into the reverse osmosis (RO) system. DOW FILMTEC BW30-400/34i membranes were used. The plant is operated at a minimum capacity of 210 $m^3 h^{-1}$. The recovery of the RO system is 75%. The DECO RO installation has performance decline problems, and membrane cleanings have to be carried out frequently. Cleaning the modules in place (CIP) is done by dosing NaOH up to a pH of 12.

#### 2.1.2. Sampling scheme

The schematic diagram of the treatment train between the MBR and the RO water treatment facility with an overview description of the sampling locations is schematized in Fig. 1. Triplicate samples were taken at each location. Microbial analysis and bacterial community analysis were performed on the samples.

#### 2.1.3. Microbial analysis

*2.1.3.1. Heterotrophic plate counts (HPC) and adenosine triphosphate (ATP) measurements*. For HPC and ATP measurements, water was collected in high-density polyethylene (HDPE) plastic bottles containing 2 mL $L^{-1}$ of a mixed solution of sodium thiosulfate (20 g $L^{-1}$) and nitrilotriacetic acid (25 g $L^{-1}$). HPC was measured by Aqualab Zuid (Werkendam, NL), according to the Dutch standard procedure (NEN-EN-ISO 6222, 1999) [28]. ATP was measured by Het Waterlaboratorium (Haarlem, NL) using a luminometer (Celsis Advance). ATP was first released from suspended bacterial cells with nucleotide-releasing buffer (LuminEX, Celsis) for total ATP measurement, while this step was not performed for assessment of free ATP. Bacterial ATP concentrations were calculated by subtracting free ATP from total ATP concentrations. The detection limit of the method was 1 ng ATP $L^{-1}$.

*2.1.3.2. Flow cytometry*. Measurements of the total and intact bacterial