



Sunlight irradiation triggers changes in the fouling potentials of natural dissolved organic matter

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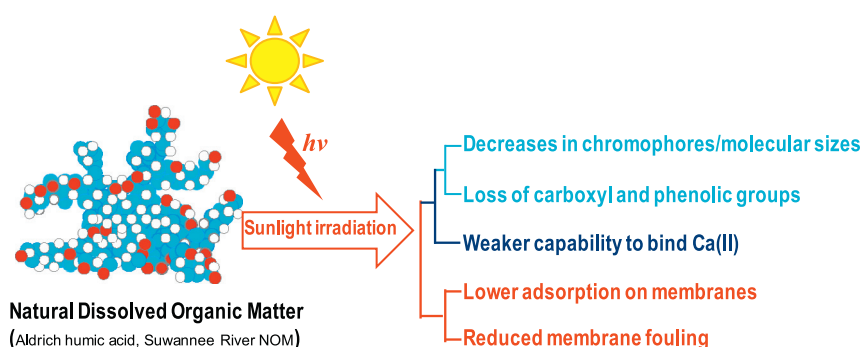
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HIGHLIGHTS

- Three months natural photolysis decreased chromophores and molecular size of DOM.
- Photolysis induced loss of carboxyl and phenolic groups altered Ca-binding of DOM.
- AHA was more susceptible to sunlight irradiation and Ca(II) addition than SRNOM.
- Sunlight exposure reduced membrane fouling potential and rejection degrees of DOM.

GRAPHICAL ABSTRACT



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ABSTRACT

Sunlight-initiated photodegradation has a great impact on the composition and properties of natural dissolved organic matter (DOM) in aquatic environments, which potentially changes the behavior and roles of DOM in water treatment facilities. Here, we explored the effect of sunlight irradiation on membrane fouling behavior of two natural DOM (i.e., Aldrich humic acid (AHA) and Suwannee River DOM (SRNOM)), particularly in the presence of calcium ion (Ca(II)). Results showed that a long-term exposure (3 months) to sunlight during the summer led to decreases in the chromophores and molecular size of both DOM. The characterization by UV–vis spectral parameter $DSlope_{350-400}$ (the slope of the log-transformed absorbance spectra in the range of 350–400 nm) indicated that sunlight-exposed DOM had a weaker Ca(II)-binding ability than unirradiated DOM, which could be attributable to the photochemically induced loss of carboxyl and phenolic groups. Additionally, AHA was found to be more susceptible to sunlight irradiation and Ca(II) addition than SRNOM, likely due to its higher aromaticity. Crucially, dead-end ultrafiltration tests showed that sunlight exposure of both AHA and SRNOM can reduce their fouling potential in the absence of Ca(II) and the presence of low Ca(II) (0.4 mM). In contrast, the addition of higher Ca(II) concentrations (2 and 3.6 mM) led to an increase in their fouling propensities. Overall, sunlight exposure can greatly alter the fouling behavior of natural DOM. This study provides a nexus between the naturally occurring transformation of DOM and its behavior (i.e., membrane fouling) in water treatment facilities.

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1. Introduction

The presence of dissolved organic matter (DOM), which consists of carbon-, nitrogen- and phosphorus-containing functional groups, significantly influences the properties and functions of aquatic systems (Ogawa et al., 2001; Ward et al., 2013). Both terrestrial input and autochthonous production have been recognized as continuous supplies of DOM in natural water (Diem et al., 2013). A large fraction of DOM is composed of the most ubiquitous naturally occurring organic materials, humic substances, which are resistant to biochemical processes but susceptible to photolytic reactions (Ma et al., 2001; Tarr et al., 2001). The chromophores derived from DOM often absorb ultraviolet and visible light, and thus, aquatic biota use them to filter out detrimental sunlight-irradiation, leading to changes in their structures and decreased molecular weights (Rodríguez-Zuniga et al., 2008). For instance, the aromatic compounds (highly unsaturated compounds) in DOM were found to be more readily photodegraded than saturated compounds (Gonsior et al., 2009). Ward and Cory (Ward and Cory, 2016) recently reported that the photochemically induced loss of carboxyl moieties (i.e., photodecarboxylation) accounted for 40–90% of the photomineralization of DOM. Additionally, partial photooxidation incorporates oxygen into some DOM molecules, thus altering the functional and structural groups present within the DOM (Moran and Zepp, 1997; Ward and Cory, 2016). More specifically, moderate amounts of low molecular weight organic acids (acetic, formic, malonic, succinic, oxalic and pyruvic acids) can be formed during sunlight exposure (Brinkmann et al., 2003). As such, sunlight irradiation can result in significant changes to the properties of DOM, even regulating their behavior in engineered water treatment processes. Nevertheless, this issue has not yet been studied sufficiently.

As DOM is responsible for the undesirable color, odor and taste of water and serves as a precursor to harmful disinfection byproducts (DBPs), various DOM removal approaches, such as coagulation/flocculation, advanced oxidation processes (AOPs) and membrane filtration, have been used to treat drinking water (Apell and Boyer, 2010; Sanchez et al., 2014; Lavonen et al., 2015). Membrane filtration treatments, especially low-pressure-driven membrane processes, have been increasingly employed for DOM removal because they produce high quality water and have convenient maintenance requirements (Lee et al., 2004; Zularisam et al., 2006). However, the membrane fouling that is caused by DOM adsorption and pore blockage remains the major obstacle in membrane-based water treatment facilities (Zhang et al., 2003). Operationally, the hydrophobic fractions of DOM accumulate on membranes more readily than the hydrophilic fractions (Ou et al., 2009; Tang et al., 2009; Chon and Cho, 2016). Additionally, there is a strong size-dependence for the fouling behavior of DOM during membrane filtration, e.g., large biopolymers were found to be responsible for membrane fouling during the ultrafiltration of drinking water (Halle et al., 2009). As such, the naturally occurring transformation of DOM, particularly its photochemical degradation, would inevitably alter its properties and fouling potentials during membrane filtration.

In addition to the photodegradation, multivalent metal ions in water, such as calcium and magnesium, play an important role in determining the DOM-DOM and DOM-membrane interactions (Orsetti et al., 2013; Gao et al., 2015; Schartup et al., 2015), and thus membrane performance. Notably, such multivalent cations could induce changes in the charge and configuration of DOM and, hence, the structure and hydraulic resistance of the DOM fouling layer on membranes. Previous studies have shown that calcium ion (Ca(II)) can act as bridges between DOM molecules, resulting in the combination of small molecules to generate large DOM aggregates (Kilduff and Karanfil, 2002; Baalousha et al., 2006). The phenolic and carboxylic groups present in DOM are considered as the major functional groups to which metal ions can bind (Shi et al., 2016). Although other functional groups including quinone, sulfhydryl and amino groups can also form strong complexes with metal ions, the abundance of these groups is too low to contribute to metal

binding (Chen et al., 2013). Overall, the roles and behavior of DOM in engineered systems is substantially determined by the presence of multivalent metal ions in water (Liang et al., 2008). As expected, the photochemically induced changes in DOM, such as the loss of carboxylic groups (Ward and Cory, 2016) and the formation of new molecules (Brinkmann et al., 2003), can change the binding propensity of the DOM molecules with metal ions, which could further complicate the fouling behavior of DOM.

Therefore, the main objective of this study was to reveal the role (s) of sunlight exposure in regulating the fouling behavior of natural DOM. UV-vis spectral parameters and Fourier transform infrared (FTIR) spectroscopy were employed to characterize the changes in the properties of DOM that result from sunlight irradiation. Moreover, the interactions of Ca(II) with the treated DOM and their fouling behavior were explored by the UV-vis spectral parameter and by dead-end filtration tests, respectively. Additionally, membrane adsorption tests were conducted to further investigate the sunlight-induced changes on the fouling propensity of natural DOM. This study is novel in that there are few reports that link the natural transformation of DOM in natural aquatic systems with its behavior in engineered systems.

2. Materials and methods

2.1. Solution preparation and sunlight exposure assay

All chemicals used in the experiments were of analytical grade. Suwannee River DOM (SRNOM, standard number 1R101N) and Aldrich humic acid (AHA) were purchased from International Humic Substance Society (IHSS) and Sigma-Aldrich Corporation, respectively. Stock solutions of these two types of DOM were prepared using DI water and then filtered through 0.45 μm glass fiber membranes to exclude suspended solids. The pH of AHA stock solution was adjusted to 10.0 with a solution of NaOH to improve dissolution and then readjusted to 7.0 before filtration. The AHA and SRNOM solutions were both controlled at 100 mg-C/L (dissolved organic carbon, DOC) and then transferred into 1000-mL acid-washed borosilicate bottles (GL45, Schott Duran, Germany) with a diameter of 101 mm and height of 230 mm for sunlight exposure assay. Moreover, two control bottles of AHA and SRNOM solutions were covered with silver paper to keep them in the dark. Finally, four bottles of the DOM solutions were sealed to avoid evaporation and still placed on the flat roof of our laboratory building to keep them under identical conditions except for sunlight irradiation. We manually shook these bottles every three days and the sunlight exposure test were conducted for three months (June, July and August of 2015) in the summer in Guangzhou, China. Both DOM samples were sterilized by heat treatment at 60 °C for 30 min in a water bath before irradiation, which could have less impact on the DOM properties based on following reasons: i) the humic acid as the main component of AHA and SRNOM exhibited a lower thermal sensitivity (Seredynska-Sobecka et al., 2007), ii) the conformational changes of soil and sediment-derived DOM to the temperature were reversible (Chen et al., 2017). After the exposure, the irradiated and unirradiated DOM samples were collected and stored in the dark at 4 °C until use. The DOC concentrations of these solutions were determined using a TOC analyzer (TOC-L, Shimadzu, Japan). The molecular weight distribution of the treated and untreated DOM was determined using a dead-end filtration system (MSC 300, Mosu Corp., China) with a series of flat-sheet poly(ether sulfone) (PES) membranes (i.e., 100, 50, and 20 kDa) (Microdyn-Nadir Corp., Germany). Meanwhile, a Zetasizer (Nano ZS90, Malvern Corp., UK) was used to monitor changes in the average particle size of the DOM samples after exposed.

2.2. Fourier transform infrared (FTIR) spectroscopy

The unirradiated and irradiated DOM samples were freeze-dried under vacuum at -80 °C for 48 h. Afterwards, the DOM powders were

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