



Solar photolysis of soluble microbial products as precursors of disinfection by-products in surface water

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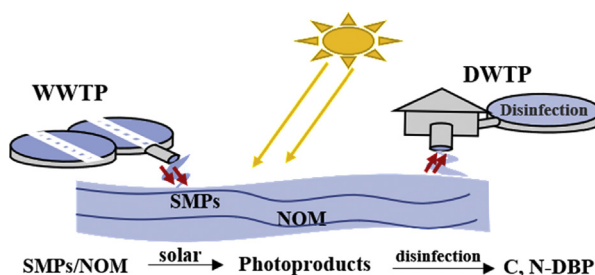
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HIGHLIGHTS

- Solar irradiation induced the increase of disinfection by-products formation.
- Irradiated SMPs were more active than NOM in formation of disinfection by-products.
- Solar irradiation reduced more aromatic functional groups.
- Solar irradiation transformed macromolecular DBP precursors into small MW fraction.

GRAPHICAL ABSTRACT



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ABSTRACT

In the Pearl River Delta area, the upstream municipal wastewater is commonly discharged into rivers which are a pivotal source of downstream drinking water. Solar irradiation transforms some of the dissolved organic matter discharged from the wastewater, also affecting the formation of disinfection by-products in subsequent drinking water treatment plants. The effect of simulated solar radiation on soluble microbial products extracted from activated sludge was documented in laboratory experiments. Irradiation was found to degrade macromolecules in the effluent, yielding smaller, more reactive intermediate species which reacted with chlorine or chloramine to form higher levels of noxious disinfection by-products. The soluble microbial products were found to be more active in formation of disinfection by-products regard than naturally-occurring organic matter. The results show that solar irradiation induced the formation of more trihalomethane (THMs), chloral hydrate (CH) and trichloronitromethane (TCNM), causing greater health risks for downstream drinking water.

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Abbreviations: DOM, Dissolved organic matter; SMPs, Soluble microbial products; UAP, Utilization-associated product; BAP, Biomass-associated product; SRNOM, Suwannee River natural organic matter; DBP, Disinfection by-product; THM, Trihalomethane; CH, Chloralhydrate; HAN, Haloacetonitrile; DCAN, Dichloroacetonitrile; TCNM, Trichloronitromethane; HAcAm, Haloacetamide; DCACAm, Dichloroacetamide; TCACAm, Trichloroacetamide; DOC, Dissolved organic carbon; SUVA₂₅₄, Specific UV absorbance at 254 nm; EEM, Excitation-emission matrix; PARAFAC, Parallel factor; FTIR, Fourier transform infrared spectrometry; SEC, Size exclusion chromatography; MW, .

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1. Introduction

The Pearl River delta (PRD) is a rapidly-developing and densely-populated region in southern China which is running short of drinking water supply. The upstream biologically-treated municipal wastewater is discharged into tributaries of the river that constitute a significant source of downstream drinking water, which is also occurred in worldwide (Mujeriego et al., 2017). Such wastewater usually takes about 2 or 3 days to reach the downstream drinking water treatment plants in PRD areas. Chlorine and chloramine disinfections are then essential for inhibiting pathogenic microorganisms and preventing waterborne diseases before entering drinking water pipe. Abundant organic materials in the wastewater react with chlorine and chloramine to form a variety of toxic and carcinogenic carbonaceous and nitrogenous disinfection by-products (C, N-DBPs), causing the health risks of drinking water use (Chen et al., 2009; Krasner et al., 2009). However, solar irradiation is a major natural process which transforms many micro-pollutants such as fluoroquinolone antibiotics and triclosan, as well as some of the dissolved organic matter (DOM) (Sommaruga, 2001; Wang et al., 2017). Hence, upstream organic precursors of disinfection by-products including trihalomethanes (THMs), haloacetic acids (HAA), chloral hydrate (CH), haloacetonitriles (HANs) and trichloronitromethane (TCNM) can also be influenced by solar irradiation, affecting the downstream DBP formation. This process motivated this study addressing the fate and transport of effluent organic matter in surface water and corresponding alternation of DBP formation.

Previous studies have reported the effect of UV or solar irradiation on natural organic matter containing fluvic-like and humic-like substances (Bazri et al., 2012; Brinkmann et al., 2003a,b; Chow et al., 2008). However, effluent organic matter also contains abundant nitrogen-containing species such as amino acids, which are regarded as the major precursors of more toxic nitrogenous DBPs (N-DBPs). Effluent organic matter typically contains both autochthonous contaminants (e.g. microbial exudates) and allochthonous terrestrial runoff and synthetic organic matter (Jarusutthirak and Amy, 2007; Yu et al., 2015). Soluble microbial products (SMPs) are organic compounds released by the microorganism in biochemical wastewater treatment. They can be classified as utilization-associated products (UAP) associated with substrate uptake and biomass growth and biomass-associated products (BAP) associated with biomass decay (Barker and Stuckey, 1999). In addition, autochthonous SMPs contribute approximately 45% of the organic compounds in effluent organic matter (Yu et al., 2015). It has been suggested that SMPs can increase the DBPs formation in chlorinated water (Liu et al., 2014; Zhang et al., 2015). This research thus considered the effect of solar irradiation on SMPs and NOM, comparing their contributions to subsequent DBP formation for DOM with various origins.

There have been extensive studies investigating the impact of advanced oxidation processes on NOM and the subsequent formation of DBPs (Liu et al., 2012; Meng et al., 2016). DOC and aromatic groups in NOM can be eliminated by solar irradiation, and then THM formation is also depressed during subsequent chlorination (Wang et al., 2015). However, Meng and his colleagues have demonstrated that UV irradiation increases THM formation (Meng et al., 2016). Meanwhile, a few studies have only addressed how the effects of solar irradiation relate to the composition and structure of DOM (Du et al., 2016; Niu et al., 2014; Yang et al., 2014). Du's group has shown that solar irradiation effectively controls subsequent haloacetamide (HACAm) formation. In that work the decreases in DCACAm and TCACAm concentration ranged from 27% to 69% (Du et al., 2017). However, less information is available about effects of sunlight on the reactivity of DOM with respect to DBP formation

during chlorination and chloramination. And no systematic investigation has been conducted to examine the effects of solar photolysis on DBP precursors from various types of sources.

The objectives of this study were (i) to compare the effects of 48 h of solar irradiation on autochthonous SMPs and allochthonous NOM in aqueous solution; (ii) to document any resulting changes in the corresponding by-product formation (especially of THM, CH, DCAN and TCNM) during chlorination and chloramination; (iii) to correlate the properties of the precursors with the DBP formation before and after photodegradation. Such information will help explain the fate of DBP precursors and DBP formation in drinking water treatment plants, both essential for developing techniques to minimize the health risks associated with autochthonous DBPs precursors.

2. Materials and methods

2.1. Samples preparation

A lab-scale sequencing batch reactor contained synthetic wastewater was simulated an activated sludge process to generate SMP samples (Yu et al., 2015). The activated sludge was obtained from the secondary sedimentation tank of the Lijiao wastewater treatment plant in Guangzhou, China. The sludge was cleaned with tap water for 12 h to eliminate residual dissolved organic matter—natural organic matter and synthetic organic compounds. After aerobic incubation for about 130 h at 25 °C, the substrate had been completely consumed to maintain the microorganisms' growth, leaving only SMPs (both utilization-associated and biomass-associated products) as organic matter in the solution (Barker and Stuckey, 1999) (detailed information was shown in Text. S1 in supplementary materials). The solution was then collected and filtered through a 0.22 µm mixed cellulose filter to remove suspended solids and >99.5% of bacteria to obtain SMPs samples (Stedmon et al., 2007; Zhang et al., 2013). The pH of the solution was then 5.6 due to the dissolution of humic and fluvic acids. The pH was adjusted to the neutral range using 0.2 M phosphate buffer and stored in the dark at 4 °C until analyzed. Suwannee River natural organic matter (SRNOM) was obtained from the International Humic Substances Society.

2.2. Simulated solar irradiation

Solar irradiation was performed in the experiments by using a sunlight simulator equipped with an 800 W xenon arc lamp (XPA-7, Xujiang Electromechanical Plant, China) with a wavelength range of 280–800 nm. The total intensity of simulated solar light was 50 ± 3 mW/cm² in the wavelength range 280–800 nm. The irradiation intensities were measured using a CEL-NP2000 spectroradiometer (CEAULIGHT, China). The pH of the samples was maintained at 7.0 ± 0.2 during the irradiation. The samples were irradiated in 50 ml quartz tubes for a total of 48hr, rotating in an out of the irradiation chamber in 8 h intervals. Because pre-experiment of 72 h-irradiation for SMPs was conducted to determine the sunlight exposure time. As shown in Fig. S2, UV absorbance and EEM components of SMP samples decreased notably within 48 h of irradiation, while slight changes were observed within 48–72 h. And the wastewater usually takes about 2 or 3 days to reach the downstream drinking water treatment plants. Therefore, the solar exposure time of DOM was set for 48 h in this study. The dark controls were handled similarly but covered with aluminum foil to eliminate light exposure. All of the glassware and ultra-pure water used were autoclaved for 0.5 h at 121 °C to obtain sterile conditions and to prevent biodegradation. The irradiated samples were then analyzed for DBPFP, DOC content, UV₂₅₄ and UV₂₈₀ absorbance, and

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