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Trihalomethane (THM) formation from synergic disinfection of biologically treated municipal wastewater: Effect of ultraviolet (UV) irradiation and titanium dioxide photocatalysis on dissolve organic matter fractions

Yingjie Meng [a], Yan Wang [a,b,*], Qi Han [c], Nan Xue [a], Yangyang Sun [a], Baoyu Gao [a], Qilin Li [b]

[a] Shandong Key Laboratory of Water Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Shandong University, Jinan 250100, PR China
[b] Department of Civil and Environmental Engineering, Rice University, Houston 77005, USA
[c] Interdisciplinary Graduate School, Singapore Membrane Technology Center, Nanyang Environment and Water Research Institute, Nanyang Technological University, 637141 Singapore, Singapore

HIGHLIGHTS

- THM formation affected by UV/chlorination and UV/TiO_2/chlorination was reported.
- UV irradiation degraded high MW THM precursors into small MW fractions.
- TiO_2 photocatalysis reduced more aromatic functional groups.
- UV/TiO_2 photocatalysis degraded more THM precursors than UV irradiation.

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ABSTRACT

The trihalomethane (THM) formation of two synergic disinfection methods, UV/chlorination and TiO_2 photocatalysis/chlorination, was evaluated to assess the health and ecological risks of biologically treated municipal wastewater. The effect of molecular characteristics of dissolved organic matter (DOM) from membrane bioreactor (MBR) and A^2/O process treated municipal wastewaters was investigated. Results showed that THM formation was elevated in both UV/chlorination and TiO_2/chlorination synergic disinfection processes compared to chlorination alone. The UV/chlorination process formed the most THMs. Both UV irradiation and UV/TiO_2 photocatalysis were found to (i) degrade high molecular weight (MW) THM precursors to small fractions, and (ii) increase the amount of humic acid-like and fulvic acid-like components. Compared with UV irradiation, UV/TiO_2 photocatalysis preferably degraded aromatic protein I, aromatic protein II and soluble microbial byproduct-like. Content of C—O band and C=O band in DOM after UV/TiO_2 photocatalysis was lower than that after UV irradiation. In addition, specific THM formation potential of the precursors in the MBR-treated municipal water was higher than that in the A^2/O-treated municipal water.

1. Introduction

Biologically treated municipal wastewater is considered a reliable and significant source of reclaimed water, and some source waters that are influenced by biologically treated municipal wastewater have become an alternative resource of drinking water due to freshwater scarcity and growing water demand [1]. Generally, biologically treated municipal wastewater must be disinfected by chlorination before being discharged to avoid the potential ecological and health risks. However, this lead to formation of disinfection by-productions (DBPs) because of the high organic precursor concentrations and disinfectant dosage [2]. Volatile DBPs could damage the human health though lung inhalation, and skin absorption. Some DBPs are non-biodegradable and can exist in aquatic environment for a long time [3–5]. Thus, disinfected biologically treated municipal wastewater is considered a potential route of environmental exposure to DBPs [6]. Many researches

* Corresponding author at: Shandong Key Laboratory of Water Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Shandong University, Jinan 250100, PR China.
E-mail address: wangyan_sdjn@aliyun.com (Y. Wang).

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have investigated the properties of DBPs and the relationship with treatment methods of municipal wastewater [7–10].

DBP formation is strongly related to the physicochemical properties of dissolved organic matter (DOM). Li et al. demonstrated that the DBPs formation potential in surface water mainly corresponded to the hydrophobic fraction contained phenolic hydroxyl and conjugated double bonds, but not to the dissolved organic carbon (DOC) value [11]. Farre et al. found that more DBPs were induced from the larger molecular weight fractions (rich humic), and more brominated DBPs were formed by lower molecular weight fractions [12]. However, in many cases, hydrophilic and lower molecular weight fractions also have contributed to DBP formation [13]. In addition, DBP formation is also influenced by disinfection method due to its characteristic reactivity with organic precursors [14,15].

Synergic disinfection, which generally consists of chorine disinfection and other disinfection (e.g. UV irradiation and TiO_2 photocatalysis), are an alternative disinfection methods [16] widely applied in water or wastewater treatment plant to inactivate chorine resistant pathogens [17,18]. UV irradiation used to be used after chlorination. However, because recent studies found that UV following chlorination could form active chlorine species and enhance THMs and HAAs formation [19], UV irradiation before chlorination was investigated recently [20,21]. For example, a wastewater treatment plant with tertiary treatment located in a tourist village on the Mediterranean coast was equipped with UV irradiation and post-chlorination for disinfection [21]. Because disinfection by photocatalysis with supported TiO_2 could significantly improve the efficacy of UV treatment [22]. TiO_2 photocatalysis followed by chlorination was also used to enhance disinfection [23].

However, recent studies also showed that UV irradiation and TiO_2 photocatalysis could change DOM characteristics, including breaking down high molecular weight organic matter and reducing the hydrophobicity fraction [24]. These changes in DOM molecular structure may affect the subsequent DBP formation due to the varying reaction activity of different DOM fractions with chlorine. Chu et al. reported increasing DBP concentration with increasing UV dose in the UV/chlorination process, and ascribed it to the formation of intermediate products [19]. Mayer et al. investigated disinfection byproduct (DBP) formation in TiO_2 photocatalysis with varying influent water quality. They found that photocatalysis exacerbated the production of total trihalomethane (TTHM) and haloacetic acid (HAA) beyond initial levels until economically prohibitive high energy UV lamp was used [25]. On the other hand, Wang et al. found that UV irradiation could eliminate trichloromethane formation but enhanced the formation of dichloroacetic acid [24]. In addition, TiO_2 photocatalysis was found to dramatically decrease TTHM formation potential through the removal of non-biodegradable NOM compounds [26].

Although several researches have investigated the DBP formation in synergic disinfection processes, few have looked at the their effects on DOM properties and the subsequent impact on DBP formation, especially in biologically treated municipal wastewater, which has higher number of DOM compounds and much higher DOM content than surface and drinking waters [27,28] and hence more complex reactions of DBP formation [29]. DBPs in surface water were generally formed by humic acid, fulvic acid and algae derived matters et al. In contrast, DBP precursors in biologically treated municipal wastewater were mainly composed of recalcitrant natural organic matter (NOM), synthetic organic chemicals, soluble microbial products (SMPs), etc. [30].

The research reported here investigated the effect of UV irradiation and TiO_2 photocatalysis on the characteristics of DOM in biologically treated municipal wastewater, including molecular weight, fluorescence and chemical functionality, and the related THMs formation during post chlorination.

2. Materials and methods

2.1. Water samples

The biologically treated municipal wastewater was collected from the secondary effluent of Jinan municipal wastewater treatment plant, which was treated by A^2/O or MBR process, separately. The effluent water samples were filtered through 0.45 μm microfiltration membranes before disinfection experiments. Basic water quality parameters of effluent after 0.45 μm filtration, including pH, UV_{254}, turbidity, DOC and bromide ion concentration, are shown in Table S1.

2.2. Disinfection

The filtered water samples were treated by three disinfection processes: (1) chlorination disinfection, (2) UV followed by chlorination disinfection (UV/chlorination), and (3) UV/TiO_2 photocatalysis followed by chlorination disinfection (TiO_2/chlorination). The schematic diagram of the disinfection experimental process is showed in Fig. S1.

Chlorination disinfection was performed as described in a previous paper [31]. A chlorine dosage of 20 mg/L was selected based on a chlorine demand test, which would provide an adequate free chlorine residual (2.0 mg/L) after 72 h (the APHA standard method [32]).

UV/chlorination was performed by provide UV irradiation before chlorination disinfection. UV irradiation experiment was conducted using standard collimate beam test with a low pressure lamp (253.7 nm, Wuxi Changjiang Medical Drives, China). The average UV dose was approximately 5 kW h m^{-3}. The photoreactor used was described in a previous paper [33]. The UV lamp housed in a quartz tube was placed in the middle of the reactor.

In the TiO_2/chlorination experiment, suspension TiO_2 (3 mg) was added in 300 mL water samples during UV irradiation process according to previous paper [33]. The same UV dosage was used as in the UV/chlorination experiment.

2.3. Analytical measurements

2.3.1. THM precursors

The concentration of DOM, which is thought to be the main THM precursor, was measured by UV_{254}, DOC and specific ultraviolet absorbance (SUVA). UV_{254} and DOC of the DOM samples were analyzed before chlorination using a UV-754 UV/VIS spectrophotometer and a Shimadzu TOC-VCPH analyzer, respectively. SUVA was calculated by normalizing UV_{254} with respect to DOC. Bromide concentration was measured using a Dionex DX-120 ion chromatograph.

The MW distribution, fluorescence and chemical functionality of DOM were also characterized. In order to distinguish THM precursors for three disinfection processes, they were denoted as precursors in raw water, precursors after UV and precursors after UV/TiO_2 according to the disinfection processes. MW distribution of DOM was analyzed with a LC-10ADVP gel filtration chromatograph (GFC) (Shimadzu, Japan) equipped with a TSK G4000PW column (TOSOH Corporation, Japan) and a RID-10A refractive index detector (Shimadzu, Japan). To collect the IR spectra of DOM, the water samples were dried in a vacuum drying oven (FD-1A-50) at −55 °C, and the resulting DOM powder was measured by Fourier Transform Infrared Spectrometry (AVATAR370) [34].

The three-dimensional excitation-emission matrix (3D EEM) spectra measurements were conducted using a luminescence spectrometer (F-4500 FL spectrophotometer, Hitachi, Japan). The EEM spectra were collected with corresponding scanning emission