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# Reductions of dissolved organic matter and disinfection by-product precursors in full-scale wastewater treatment plants in winter



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Shuang Xue<sup>a,\*</sup>, Wujisiguleng Jin<sup>a, b</sup>, Zhaohong Zhang<sup>a</sup>, Hong Liu<sup>a</sup>

<sup>a</sup> School of Environmental Science, Liaoning University, Shenyang 110036, China
<sup>b</sup> The City College of Jilin Jianzhu University, Changchun 130111, China

# HIGHLIGHTS

• Total removal efficiency of DOC, UV-254, THMFP, and HAAFP was higher than 50% in WWTPs.

• Biological treatment was the predominant process responsible for the removal of DOC, THMFP, and HAAFP.

• BAF and SCAS processes achieved better DOM removal because of higher biomass concentration.

• HPO-N and HPI were removed to a higher degree via biological treatment than the other fractions.

• Humic-like fluorescent compounds were not readily eliminated during the biological treatment.

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# ABSTRACT

The reductions of dissolved organic matter (DOM) and disinfection byproduct precursors in four fullscale wastewater treatment plants (WWTPs) (Liaoning Province, China) where different biological treatment processes were employed in winter were investigated. The total removal efficiencies of dissolved organic carbon (DOC), ultraviolet light at 254 nm (UV-254), trihalomethane formation potential (THMFP), and haloacetic acid formation potential (HAAFP) were in the range of 70.3-76.0%, 49.6-57.3%, 54.4-65.0%, and 53.7-63.8% in the four WWTPs, respectively. The biological treatment was the predominant process responsible for the removal of DOC, THMFP, and HAAFP in WWTPs. Differences in the reduction of UV-254 were not significant (p > 0.05) among biochemical reaction pool, secondary sedimentation tank, and disinfection tank. Biological aerated filter and suspended carrier activated sludge processes achieved higher DOM removal than the conventional active sludge and anaerobic-anoxic -oxic processes. Hydrophobic neutral and hydrophilic fraction were removed to a higher degree through biological treatment than the other three DOM fractions. HAAFP removal was more efficient than THMFP reduction during biological treatment. During primary treatment, fluorescent materials in secondary sedimentation tanks were preferentially removed, as compared to the bulk DOM. Humic-like fluorescent compounds were not readily eliminated during biological treatment. The fluorescent materials were more susceptible to chlorine than nonfluorescent compounds.

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## 1. Introduction

Dissolved organic matter (DOM) includes various organic compounds, which consist of aromatic, aliphatic, phenolic, and quinolic functional groups with varying molecular sizes and properties (Chen et al., 2003a, b). DOM plays important roles in the interaction and transport of many toxic organic or inorganic chemicals

\* Corresponding author. *E-mail address:* xueshuang666@sina.com (S. Xue).

http://dx.doi.org/10.1016/j.chemosphere.2017.02.106 0045-6535/© 2017 Published by Elsevier Ltd. throughout the environment (Zhang et al., 2008). DOM also acts as an important precursor of disinfection byproducts (DBPs), such as trihalomethanes (THMs) and haloacetic acids (HAAs), and enables microorganisms to grow in the treatment unit or distribution system (Kim and Yu, 2005).

The widespread use of wastewater treatment plants (WWTPs) has the potential to yield a considerable amount of organic matter such as proteins, polysaccharides, and humic substances, acids, and neutral compounds. Treated wastewater is commonly discharged into rivers, lakes, estuaries, and oceans, and thus, it can be an important source of DOM in the receiving waters (Yang et al., 2014).



Therefore, understanding the fate and removal of DOM in wastewater treatment processes is of great interest to plant design engineers and environmental scientists alike. Some studies investigated the occurrence of DOM and/or DBPs in WWTP effluents. Imai et al. (2002) evaluated the characteristics of DOM in WWTP effluent by comparing DOM fraction distributions, ultraviolet absorption properties, and molecular size distributions. Krasner et al. (2009a) determined the occurrence of DBPs in WWTPs and examined the effect of well-nitrified versus poorly nitrified effluent organic matter (EfOM) on disinfection byproduct (DBP) occurrence. Krasner et al. (2009b) compared different WWTP processes for the control of dissolved organic carbon (DOC), dissolved organic nitrogen (DON), and DBP precursors in EfOM. However, the majority of the research studies were focused on WWTP discharges. Wei et al. (2012) examined the removal and transformation of DOM during a full-scale anoxic/oxic (A/O) WWTP in summer and winter. Removal efficiency of organic micropollutants in WWTPs is often highly dependent on the biological treatment, and it has been indicated that the biological treatment design can affect the overall micropollutant removal (Behera et al., 2011; Luo et al., 2014). However, there remains a lack of sufficient information on the occurrence and fate of DOM and DBP precursors in different treatment units of WWTPs, which use various types of biological treatment processes.

The goal of the present work was to investigate the reductions of DOM and DBP precursors in four full-scale WWTPs in winter. Different biological treatment technologies were used in the four WWTPs: conventional active sludge (CAS) process for WWTP A, biological aerated filter (BAF) for WWTP B, suspended carrier activated sludge (SCAS) process for WWTP C, and anaerobic–anoxic–oxic  $(A^2/O)$  process for WWTP D. The CAS process is one of the most commonly used technologies in municipal and industrial wastewater treatment, because it is cost effective and straightforward (Zhang et al., 2017). The A<sup>2</sup>O process is a sequential process that uses anaerobic, anoxic, and oxic reactors. Currently, the A<sup>2</sup>O process is the most widely used for biological nutrient removal because of its cost effectiveness and high efficiency (Fang et al., 2016). BAF was developed on the basis of biological filters in Europe in the late 1980s and then widely applied worldwide as novel, flexible, and effective bioreactors (Wu et al., 2015). A BAF is a fixed-film biosystem with a small footprint that uses filter media with a high specific surface area and porosity. The filter can promote in-growth biofilms for wastewater treatment (Bao et al., 2016). BAF has the advantages of higher biomass and organic loadings, stronger environmental shock resistance, and less sludge production (Ye and Ni, 2002). The SCAS process is based on the use of plastic carriers with density a little lighter than that of water in which microorganisms form biofilms. Because of air agitation, the carriers are mobilized in suspension of aeration tanks during oxygenation (Wei and Han, 2011). The SCAS process has been successfully used as the favored biological treatment technique during the past two decades, owing to compactness, flexibility, and high-quality effluent production (Wei and Han, 2011). The four types of biological sewage treatment processes are employed on a large scale in China (Wang et al., 2011; Wei and Han, 2011), thereby making them worthy of intensive study. The specific objectives of the present study are as follows: (1) examining the removal of DOM and its fractions isolated by the XAD-8/XAD-4 resin method within different treatment units; (2) investigating the effectiveness of different treatment units for the removal of THM and HAA precursors; and (3) evaluating the impact of different treatment units on the spectroscopic properties of DOM. Such knowledge would be helpful in establishing an optimal treatment strategy for the control of DOM and DBP precursors in WWTPs in cold areas.

#### 2. Materials and methods

#### 2.1. Sample collection

Four municipal WWTPs (WWTP A to WWTP D) in Liaoning Province, China, were chosen for this study. Basic information and process flow charts of the four WWTPs are shown in Table 1 and Fig. 1. Wastewater samples included influents, primary sedimentation tank effluents, biochemical reaction pool (i.e., aeration tank in WWTP A, BAF in WWTP B, aeration tank filled with suspended carrier in WWTP C, and aerobic tank in WWTP D) effluents, secondary sedimentation tank effluents, and disinfection tank effluents. In addition, the V-filter effluents in WWTP D were also sampled. The sampling points are shown in Fig. 1. The general water quality characteristics of influents of the four WWTPs are shown in Table 1. The wastewater samples were collected biweekly in two winters, from December 2013 to February 2014 and from December 2014 to February 2015. Daily-composite samples were obtained by mixing 500 mL sample volumes collected every hour during 24 h. Water samples were collected in acetone-rinsed amber glass bottles with polytetrafluoroethylene (PTFE)-lined caps. To eliminate the variations in wastewater, all the samples were obtained when the plants ran normally. The ambient temperature during the sampling ranged from -24.4-2.1 °C, and there was no snow during the sampling.

# 2.2. DOM fractionation

DOM in wastewater samples was fractionated into five classes: hydrophobic acid (HPO-A), hydrophobic neutral (HPO-N), transphilic acid (TPI-A), transphilic neutral (TPI-N), and hydrophilic fraction (HPI), by using the XAD-8/XAD-4 resin chromatography (Chow et al., 2006). The isolation methods were described in detail by Xue et al. (2009). The recovery rate of the DOC ranged from 93% to 102% in the fraction and isolation procedures in this study.

#### 2.3. Analysis

All wastewater samples were filtered using a 0.45- $\mu$ m cellulose nitrate membrane filter and stored at 4 °C prior to analysis. DOC was analyzed on a Shimadzu TOC-5000 Total Organic Carbon Analyzer (Shimadzu, Kyoto, Japan). Ultraviolet light at 254 nm (UV-254) was measured with a Cary 50 ultraviolet–visible (UV/VIS) spectrophotometer (Varian, Palo Alto, California, USA) at 254 nm using a 1-cm pathlength quartz cell. The instrument was zeroed using Milli-Q water as a blank. Specific ultraviolet light absorbance (SUVA) was calculated as (UV-254/DOC)  $\times$  100.

Trihalomethane formation potential (THMFP) and haloacetic acid formation potential (HAAFP) measurements were performed according to the Standard Method 5710B. The chlorine dosage for each water sample was determined such that a final residual chlorine of  $3-5 \text{ mg L}^{-1}$  remained in the sample after the 7 d of incubation at 25 °C. All samples were adjusted to a pH of 7  $\pm$  0.2 using sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) and sodium hydroxide (NaOH). The neutralized solution was then buffered with a phosphate solution prior to incubation in amber bottles at  $25 \pm 2$  °C for 7 d. At the end of the incubation period, samples were dechlorinated using sodium sulfite (Na<sub>2</sub>SO<sub>3</sub>). THMs were extracted using methyl-tert butyl ether (MTBE) from the chlorinated samples using a modified EPA method 551.1. Five species of HAAs, i.e., monochloroacetic acid (MCAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), monobromoacetic acid (MBAA), and dibromoacetic acid (DBAA), were analyzed in accordance with the EPA method 552.2. MTBE was used as the sole extracting solvent. The concentrations of THMs and HAAs were measured using a gas chromatograph (CP-3800)

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