



# Seasonal evaluation of disinfection by-products throughout two full-scale drinking water treatment plants



Xin Zhong<sup>a</sup>, Chongwei Cui<sup>a,\*</sup>, Shuili Yu<sup>b</sup>

<sup>a</sup> School of Municipal & Environmental Engineering, Harbin Institute of Technology, Harbin, 150090, China

<sup>b</sup> School of Environmental Science and Engineering, Tongji University, Shanghai, 200433, China

## HIGHLIGHTS

- Fate of carbonyl compounds at two full-scale DWTPs.
- Higher carbonyl compounds concentrations were found during warm months.
- For, Gly, M-Gly, Fumar, Benz, protocatechu and 3-OHbenz are the dominant compounds.

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## ABSTRACT

Carbonyl compounds can occur alpha-hydrogens or beta-diketones substitution reactions with disinfectants contributed to halogenated by-products formation. The objective of this research was to study the occurrence and fate of carbonyl compounds as ozonation by-products at two full-scale drinking water treatment plants (DWTPs) using different disinfectants for one year. The quality of the raw water used in both plants was varied according to the season. The higher carbonyl compounds concentrations were found in raw water in spring. Up to 15 (as the sum of both DWTPs) of the 24 carbonyl compounds selected for this work were found after disinfection. The dominant carbonyl compounds were formaldehyde, glyoxal, methyl-glyoxal, fumaric, benzoic, protocatechuic and 3-hydroxybenzoic acid at both DWTPs. In the following steps in each treatment plant, the concentration patterns of these carbonyl compounds differed depending on the type of disinfectant applied. Benzaldehyde was the only aromatic aldehyde detected after oxidation with ozone in spring. As compared with DWTP 1, five new carbonyl compounds were formed (crotonaldehyde, benzaldehyde, formic, oxalic and malonic acid) disinfection by ozone, and the levels of the carbonyl compounds increased. In addition, pre-ozonation (PO) and main ozonation (OZ) increased the levels of carbonyl compounds, however coagulation/flocculation (CF), sand filtration (SF) and granular activated carbon filtration (GAC) decreased the levels of carbonyl compounds.

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## 1. Introduction

Disinfection, the inactivation of viable pathogenic microorganisms, has been used since the early 1900s in drinking water treatment and wastewater treatment. It is used in public systems to protect against waterborne diseases. Many methods of disinfection are currently used and include a variety of chemical oxidants such as ozone, chlorine, chlorine dioxide as well as physical processes

like ultraviolet irradiation. Chlorine, inexpensive and widely available, is the most widely used disinfectant in the world. Ozone is used as a disinfectant as well as an oxidant to control taste and odor problems and can be used to facilitate the removal of iron and manganese (Qi et al., 2013; Rodríguez et al., 2008; Seo et al., 2010; Srinivasan and Sorial, 2011; Von Gunten, 2003; Yuan et al., 2013). Ozone can inactivate most viruses, bacteria, and protozoa better than chlorine. Since ozone decomposes rapidly, it is often followed by a secondary disinfectant such as chlorine to provide a disinfectant residual (Hua and Reckhow, 2013).

Although disinfection is necessary for controlling pathogens and waterborne diseases, it can also produce unwanted by-products. Disinfection by-products (DBPs) form when disinfectants react with natural organic matter (NOM) and other compounds in water.

\* Corresponding author. No.73, Huanghe Road, Nangang District, Harbin City, Heilongjiang Province, China.

E-mail addresses: [zhongxinmagic@163.com](mailto:zhongxinmagic@163.com) (X. Zhong), [18845181188@126.com](mailto:18845181188@126.com) (C. Cui).

NOM is a complex, diverse mixture of organic compounds, with a large portion often composed of humic substances. While hundreds of DBPs have been identified, some major classes include trihalomethanes (THMs), haloacetic acids (HAAs), and aldehydes. Different disinfectants cause different DBPs to form. For example, chlorine produces especially high levels of THMs and HAAs, while ozone produces high concentrations of aldehydes, ketones and carboxylic acids (Huang et al., 2005; Papageorgiou et al., 2014; Jurado-Sánchez et al., 2014).

Richardson (2003) has reported that carbonyl compounds represent about 30% of the ozonation DBPs. Aldehydes may be the source of taste and odor problems in water. Besides, acetaldehyde, formaldehyde, glyoxal and methyl-glyoxal in water have been recognized as possible mutagenic compounds (Can and Gurol, 2003; Richardson et al., 2000). In addition, dicarboxylic acids have been considered potential rodent carcinogens because of potential peroxisome proliferating activity (Moudgal et al., 2000). Meanwhile, as the degradation rates of carboxylic acids are usually lower than their formation rates, they accumulate during disinfection, leading to bacterial re-growth and biofilm formation in distribution systems (Chu and Lu, 2004; Meylan et al., 2007). Carbonyl compounds are not regulated under the EPA, however, some carbonyl compounds are included in the USEPA's Contaminant Candidate List for possible future regulation.

Considering that NOM, as precursor of carbonyl compounds formed during ozonation, is highly variable in terms of concentration and its characteristics even for the same system. It becomes increasingly important to study on the fate of ozonation by-products during water treatment. However, there is limited information regarding the fate of these compounds under the conditions occurred in drinking water treatment plants (DWTPs) (Hammes et al., 2006). Neither survey provides information on the effect of the treatments applied on the formation/removal of carbonyl compounds, nor on the levels found (Richardson et al., 2000; Krasner et al., 2006).

In DWTPs, the occurrence of carboxylic acids varies according to the quality of the water source and the operations carried out. Hence, in a comprehensive study of 6 water treatment facilities using ozone and chlorine dioxide as disinfectants, up to 33 aliphatic and 10 aromatic acids were formed (Richardson et al., 2000). In a similar survey conducted in 12 American full-scale DWTPs, up to 24 aliphatic acids as well as one aromatic acid have been identified as ozone/chlorine and ozone/chloramines DBPs (Krasner et al., 2006). A similar study performed on different steps (raw water, ozonation, sand filtration and finished water) of a DWTP using the ozonation process, which corroborates the findings observed above, since acetic and formic acids are the compounds that experienced a major increase in their concentrations after ozonation along with oxalic and pyruvic acids (up to 48 times). In addition, the acids formed after ozone application can be removed by bio-filtration (Hammes et al., 2006). Vincenti et al. (2010) have compared the occurrence of 6 aliphatic dicarboxylic acids and 9 aromatic (mono- and dihydroxy-) acids in raw and finished water samples, which collected from a DWTP that uses ozone in combination with a chlorinated agent. The results show that 2 new acids (maleic and itaconic) were formed after water treatment, while the levels of some aliphatic acids and all aromatic acids decreased. Jurado-Sánchez et al. (2014) carried on the study of the occurrence of 35 aliphatic and aromatic acids within two full-scale DWTPs, which is the most recently and completely. The results show that 13 carboxylic acids detected in raw water were varied according to the quality of the water source. Besides, the concentration of these compounds differed depending on the type of disinfectant applied in the following treatment steps of each DWTP. When chloramination was used, the levels of the carboxylic acids remained almost

constant and 4 new acids were formed. However, when ozonation/chlorination was used, the total concentration of the carboxylic acids in the raw water sample increased up to 6 times after disinfection and 6 new acids were produced at high levels. Seasonal variations of the carboxylic acids showed in both plants, and maximum levels of all the analytes were reached in the coldest months (autumn and winter), aromatic acids only being found in those seasons. Papageorgiou et al. (2014) studied the presence and fate of carbonyl compounds as ozonation by-products at a DWTP for one year. They detected up to 14 carbonyl compounds at total concentrations of 32.8–110.7 µg/L after ozonation, and found that they were removed by about 64–80% in the treated water. Serrano et al. (2015) studied 11 aldehydes at different points in a DWTP and its distribution network. They found 5 aldehydes in raw water and the concentrations of aldehydes were higher in warmer seasons (spring and summer). Chlorine dioxide caused the formation of 3 new aldehydes. Chloramination raised the levels of 8 aldehydes.

Carbonyl compounds remain a concern due to health hazard, their high biodegradability, and the side-effect that can occur alpha-hydrogens or beta-diketones substitution reactions with disinfectants to form halogenated by-products (Schechter and Singer, 1995). Therefore, the objectives of this study included, namely: (i) the application of accurate and reliable analytical methods for measuring a large number of regulated and non-regulated DBPs including aldehydes and carboxylic acids in two DWTPs treated sequentially with chlorine and ozone-chlorine; (ii) the effect of source water and treatment conditions on their formation and (iii) the effect of seasonal changes on carbonyl compounds concentrations.

## 2. Materials and methods

### 2.1. Materials

The analytical standard containing 15 carbonyl compounds (formaldehyde, acetaldehyde, propanal, butanal, pentanal, hexanal, crotonaldehyde, crotonaldehyde, heptanal, octanal, benzaldehyde, nonanal, decanal, glyoxal and methyl glyoxal) and the derivatization agent O-(2,3,4,5,6-pentafluoro-benzyl)hydroxylamine (PFBOA) were purchased from AccStandard (New Haven, USA). HPLC grade *n*-hexane Anpel (Shanghai, China) was used as a solvent for the liquid-liquid extraction. The PFBOA was prepared daily as an aqueous solution (15 mg/mL) in organic free water. Organic free water was provided by the Synergy UV-Ultrapur Water System (Millipore, Molsheim, France). Standards of the 9 carboxylic acids (>95% purity), formic, acetic, oxalic, malonic, fumaric, protocatechuic, 3-hydroxybenzoic, phthalic and benzoic acid were purchased from J&K (Beijing, China). Stock solutions of the individual acids (10 g/L) were prepared in purified water. All these solutions were stored at 4 °C. Hydrochloric acid, potassium dihydrogenphosphate, sodium hydroxide and orthophosphoric acid were analytical reagent and supplied by Shanghai (Shanghai, China). LiChrolut EN (particle size 40–120 µm) was purchased from Merck (Darmstadt, Germany). Silica-reverse phase sorbent with octadecyl functional groups (Supelclean ENVI-18) was supplied from Supelco (Bellefonte, PA, USA).

### 2.2. Description of DWTPs and sample collection

Two surface DWTPs located in Taihu Lake Region, China. The schematics diagrams of the treatment steps employed in each facility as well as the location of the sampling points are depicted in Fig. 1. DWTP 1 (Fig. 1a) is a conventional water treatment process that involves coagulation/flocculation (CF), sand filtration (SF) and chlorination (CL). DWTP 1 provides 100,000 m<sup>3</sup> of potable water

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