



Characterization of haloacetaldehyde and trihalomethane formation potentials during drinking water treatment



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HIGHLIGHTS

- CH only accounted 25–48% of the total HAs in waters containing 100–150 $\mu\text{g L}^{-1}$ bromide.
- Water treatment with ozone decreased THMFPs, while it significantly enhanced HAFPs.
- Brominated HAFPs after BAC filtration were still higher than those in raw waters.
- Bromine incorporation by THMs and HAs increased with the progress of treatment steps.

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ABSTRACT

Haloacetaldehydes (HAs) are the third prevalent group of disinfection by-products (DBPs) of great health concern. In this study, their formation and speciation during chlorination were investigated for raw and process waters collected at three O_3 -biological activated carbon (BAC) advanced drinking water treatment plants. The results showed that all HA formation potentials (HAFPs) were highly enhanced whenever ozone was applied before or after conventional treatment. Sand filtration and BAC filtration could substantially reduce HAFPs. Trihalomethanes (THMs) were also measured to better understand the role of HAs in DBPs. Very different from HAFPs, THMFPs kept decreasing with the progress of treatment steps, which was mainly attributed to the different precursors for HAs and THMs. Brominated HAs were detected in bromide-containing waters. Chloral hydrate (CH) contributed from 25% to 48% to the total HAs formed in waters containing 100–150 $\mu\text{g L}^{-1}$ bromide, indicating the wide existence of other HAs after chlorination besides CH production. In addition, bromide incorporation factor (BIF) in HAs and THMs increased with the progress of treatment steps and the BIF values of THMs were generally higher than those of HAs. The BAC filtration following ozonation could significantly reduce HA precursors produced from ozonation but without complete removal. The brominated HAFPs in the outflow of BAC were still higher than their levels in the raw water. As a result, O_3 -BAC combined treatment was effective at controlling the total HAs, whereas it should be cautious for waters with high bromide levels.

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1. Introduction

Disinfection has been widely used to kill pathogens in drinking water. However, it is also responsible for the formation of undesired disinfection by-products (DBPs). To date, more than 700 DBPs have been identified in drinking water (Richardson, 2003; Zhao et al.,

2012; Zhai et al., 2014; Yang and Zhang, 2016). Trihalomethanes (THMs) and haloacetic acids (HAAs) are the two most abundant groups of DBPs and have gained significant amount of research efforts. After these, haloacetaldehydes (HAs) are the next prevalent group of DBPs in drinking water (Krasner et al., 2006). Unlike THMs and HAAs, few countries regulated the maximum contaminant level (MCL) for HAs, except 20 $\mu\text{g L}^{-1}$ for chloral hydrate (CH) in Australia and 10 $\mu\text{g L}^{-1}$ for CH in China. Recently, Jeong et al. (2015) reported that HAs had orders of magnitude higher toxicity than the regulated THMs and HAAs. Therefore, the adverse health effects of HAs can be more severe even at lower levels than those of THMs

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and HAAs in finished drinking water. Like HAAs, there are a total of nine species of chlorine and bromine substituted HAs including chloro-(CA), bromo-(BA), dichloro-(DCA), bromochloro-(BCA), dibromo-(DBA), trichloro-(usually exists in its hydrate form as CH), bromodichloro-(BDCA), dibromochloro-(DBCA), and tri-bromoacetaldehyde (TBA). However, little is known on the formation and speciation of these nine HAs in drinking water.

CH is the first identified HAs as DBPs in drinking water and has been determined as one of common monitoring index in treated surface and ground water by the Information Collection Rule since the late 1990s (Blank et al., 2002). Brominated tri-HAs were firstly found during chlorination of fulvic acid solutions containing bromide (Xie and Reckhow, 1996). Later, six HA species, including CA, DCA, BCA, DBA, CH and TBA, were selected as high priority DBPs and monitored at 12 full-scale plants treated waters with high total organic carbon (TOC) and bromide in the 2000–2002 US DBP occurrence study (Krasner et al., 2006). The occurrence of DCA (median = $1 \mu\text{g L}^{-1}$, maximum = $14 \mu\text{g L}^{-1}$) was similar to that of CH (median = $1 \mu\text{g L}^{-1}$, maximum = $16 \mu\text{g L}^{-1}$), whereas the formation of other HAs were low ($<1 \mu\text{g L}^{-1}$). Koudjonou et al. (2008) reported the HA concentrations were in the range of $7.2\text{--}26.8 \mu\text{g L}^{-1}$ at a plant using sequenced $\text{ClO}_2\text{-O}_3\text{-Cl}_2$ disinfection. BDCA and DBCA became the two major species of HAs when the level of bromide was higher than $100 \mu\text{g L}^{-1}$. The brominated HAs were reported to be more toxic than their chlorinated analogues, with TBA being the most cytotoxic and DBA being the most genotoxic (Jeong et al., 2015).

Despite the awareness of the potential health hazards of HAs, previous researches mainly focused on CH. The use of alternative disinfectants such as chloramines, chlorine dioxide or ozone has gained increasing consideration for its effectiveness on minimizing regulated DBP formation (Hua and Reckhow, 2007b; Bougeard et al., 2010; Yang et al., 2012, 2013). However, these strategies may have adverse effect on HA formation and that focusing on CH alone is likely to underestimate the overall formation and speciation of HAs. For example, using ozone as the primary disinfectant suppressed THM and HAA formation successfully, while it would significantly increase CH formation during subsequent chlorination (Yang et al., 2012; Mao et al., 2014). CH formation was negligible by both NH_2Cl and $\text{O}_3\text{-NH}_2\text{Cl}$, while the levels of DCA were higher than that by Cl_2 , and DCA formation was substantially promoted by $\text{O}_3\text{-NH}_2\text{Cl}$ (Krasner et al., 2006). Low molecular weight aldehydes formed during pre-ozonation were considered as the major precursors for CH (McKnight and Reckhow, 1992; Dabrowska and Nawrocki, 2009). Furthermore, for waters containing high bromide levels, pre-ozonation was reported to shift the DBP formation from chlorinated species to higher toxic brominated species (Hua and Reckhow, 2013; Mao et al., 2014). Therefore, understanding the speciation of HAs at water utilities where ozone was applied, especially in the presence of bromide, has important implications.

Ozonation integrated with biological activated carbon ($\text{O}_3\text{-BAC}$) has been increasingly adopted in drinking water treatment plants (DWTPs) to improve the removal of dissolved organic matter (DOM). In China, $\text{O}_3\text{-BAC}$ process had a total drinking water treatment capacity at approximately $20 \text{ million m}^3 \text{ d}^{-1}$ by 2013 (Hong et al., 2013). The $\text{O}_3\text{-BAC}$ process can significantly reduce the precursors of THMs and HAAs (Karnik et al., 2005; Xu et al., 2007; Yan et al., 2010; Chu et al., 2012), and it can also remove the precursors of emerging nitrogenous DBPs such as trichloronitromethane and dichloroacetamide, which can not be removed by conventional treatment (i.e. coagulation-sedimentation-filtration) (Chu et al., 2012). Moreover, BAC filtration can successfully remove ozonation by-products such as aldehydes and carboxylic acids (Krasner et al., 1993; Wert et al., 2007; Mao et al., 2015). However, the effects of $\text{O}_3\text{-BAC}$ process on the formation and speciation of HAs in drinking

water are uncertain.

In this study, formation and speciation of HAs were examined in samples collected from three DWTPs, where all adopted $\text{O}_3\text{-BAC}$ process but contained different levels of bromide. The objective of this study was to investigate the formation and speciation of HAs after different water treatment processes, and to evaluate the effect of $\text{O}_3\text{-BAC}$ process on removal of HA precursors. During the experiments, the formation of regulated THMs was also measured to better assess the roles and contributions of HAs in the total classes of DBPs. THM and HA formation were examined after 24-h chlorination. Seven HA species (i.e. DCA, BCA, DBA, TCA, BDCA, DBCA, and TBA) were analyzed for all the samples.

2. Materials and methods

2.1. Water samples

Water samples were collected from three DWTPs in Jinan (JN), Beijing (BJ) and Jiaxing (JX) in China. Of the three DWTPs, two were located in northern China (i.e. JN and BJ) and the other was located in southern China. As shown in Fig. 1, all adopted $\text{O}_3\text{-BAC}$ process, but there are some differences in the pretreatment and conventional treatment. No pretreatment was used in JN, while pre-ozonation was applied in BJ and submerged aerated filtration (SAF) was used in JX. As for the conventional treatment, high-density sedimentation, high-density sedimentation combined with sand filtration, clarifier combined with sand filtration were respectively used in JN, BJ, and JX. In addition, both JN and BJ used polyaluminum chloride (PACl) as coagulant and polyacrylamide (PAM) as aid, while only PACl was used in JX. The ozone doses applied were in the range of $1\text{--}1.5 \text{ mg L}^{-1}$. The sampling was performed twice between September and November 2015 for all the DWTPs. Samples were collected from the influents of the DWTPs (i.e. raw water (RW)), the effluents of pretreatment (PT), conventional treatment (CT), O_3 , and BAC filtration (Fig. 1). All samples were treated with H_2SO_4 to lower their pH to less than 2.0 for water preservation during transportation to the laboratory at Tsinghua University, and stored in the dark at 4°C after their arrival (MEP, 2009). The water collection from the DWTPs to the laboratory was within 72 h, and the experiments were performed within 24 h when the samples were received at the laboratory.

2.2. Experimental procedure

Samples were readjusted to $\text{pH } 8.0 \pm 0.1$ by the addition of sodium hydrate solution and buffered with 2 mM borate buffer solution ($\text{pH} = 8.0$). Then they were filtered with pre-washed $0.45\text{-}\mu\text{m}$ Durapore membrane filters (Merck, Ireland) prior to the analysis of water quality parameters (i.e. non-purgeable organic carbon (NPOC), UV_{254} , and bromide) and DBP formation potential (DBPFP) tests. The DBPFP tests were designed to determine the extent of HA and THM formation during chlorination. The fresh chlorine stock was prepared by diluting sodium hypochlorite (5–6% available free chlorine) before each experiment. The chlorine doses were with the ratio of chlorine to NPOC of raw water at 3:1 (on a weight basis), which has been commonly used (Bougeard et al., 2010). NPOC was used because the level of inorganic carbon in some samples was too high to enable accurate TOC measurement. Samples were incubated for 24 h at 20°C in the dark. Ascorbic acid at a concentration of 32 mg L^{-1} was used to quench the residual chlorine while not degrading any of the concerned DBPs (Koudjonou and LeBel, 2006; Kristiana et al., 2014). Error bars were calculated from duplicated experiments.

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