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Comparison of the effects of chloramine and chlorine on the aromaticity of dissolved organic matter and yields of disinfection byproducts



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HIGHLIGHTS

- Changes of DOM chromophores caused by chloramine and chlorine are similar.
- Extent of the degradation of DOM chromophores by NH₂Cl is lower than that by Cl₂.
- Cl₂ consumption (ΔCl₂) correlates in both cases with decreases of DOM aromaticity (ΔLnΦ).
- THM, DHAN, DHAA and THAA formed by Cl_2 and NH_2Cl correlate with $\Delta Ln\Phi.$
- \bullet Correlations for ΔCl_2 and DHAA are similar in chloramination and chlorination.

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ABSTRACT

This study compared effects of chlorination and chloramination on the chromophores of dissolved organic matter (DOM) and attendant formation of disinfection by-products (DBPs) in raw and treated surface waters. Comparison of the differential absorbance spectra of chloraminated and chlorinated waters shows that interactions of chloramine with DOM chromophores result in changes that are in many respects similar to those observed for chlorine although the extent of degradation of DOM chromophores and the attendant decrease of DOM aromaticity by chloramine are less pronounced than that caused by free chlorine. The degradation of DOM chromophores caused by the examined disinfectants indicated that in both cases a gradual decrease of DOM took place. Decreases of DOM aromaticity estimated based on the changes of DOM absorbance at 254 or 280 nm were correlated with chlorine consumption in a similar way for both examined disinfectants. Correlations between changes of DOM absorbance and yields of dihaloacetic acids (DHAA) were also similar for chlorination and chloramination. This was interpreted to indicate that the generation of DHAA proceeds via the degradation of the reactive sites associated with DOM chromophores irrespective of whether these sites are engaged by chlorine or chloramine. Correlations between the decrease of DOM aromaticity and formation of other DBP (e.g., trihalomethanes - THM, trihaloacetic acids - THAA and dihaloacetonitriles - DHAN) for

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https://doi.org/10.1016/j.chemosphere.2017.10.063 0045-6535/© 2017 Elsevier Ltd. All rights reserved. chloramine and chlorine were also observed but, as opposed to the observations for DHAA, the correlations between degradation of DOM aromaticity and yields of THM, THAA or DHAN were different for chlorination and chloramination.

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1. Introduction

The occurrence, speciation and mechanisms of generation of disinfection by-products (DBPs) formed in drinking water have been studied since the 1970-ties. Despite the extent of such studies, DBP control remains a major challenge in ensuring the quality of drinking water. As the regulations for trihalomethanes (THMs), haloacetic acids (HAAs) became increasingly tight, many utilities have switched to disinfection with chloramine in whose presence THM and HAA formation is suppressed (Cowman and Singer, 1996; Yang et al., 2008; Kristiana et al., 2009; Lee and Westerhoff, 2009; Krasner et al., 2013). However, formation of dihalogenated HAAs remains significant in chloraminated water (Cowman and Singer, 1996; Duirk and Valentine, 2006; Hua and Reckhow, 2007b; Kristiana et al., 2009). Chloramination also tends to generate considerable levels of nitrogenous DBPs (N-DBPs) (Krasner et al., 2006; Shah and Mitch, 2011; Tian et al., 2013). The fraction of unidentified halogen-containing species contributing to total organic halogen (TOX) is larger in the case of chloramination compared with chlorination (Hua and Reckhow, 2007b; Goslan et al., 2009; Kristiana et al., 2009).

Many studies have compared DBP speciation and yields produced in chlorinated and chloraminated water but the role of the nitrogen-containing groups in dissolved organic matter (DOM) and the nitrogen in chloramine in the generation of N-DBPs (Duirk, 2003; Duirk et al., 2005; Hong et al., 2007) as well as mechanisms of interactions between DOM and chlorine or chloramine are yet to be elucidated in more detail (Yang et al., 2007, 2010; Huang et al., 2012). Difficulties in probing these interactions are associated with the complexity of DOM halogenation reactions (Richardson et al., 2008; Ding and Zhang, 2009; Lu et al., 2009; Tian et al., 2013), DOM polydispersity and its site-specificity (Amy et al., 1987; Nokes et al., 1999; McClellan John et al., 2000; Roccaro et al., 2013, 2014a).

To examine associations between DBP formation and DOM properties, techniques such as DOM fractionation (Hong and Liu, 2007; Hua and Reckhow, 2007a; Korshin et al., 2007a; Lu et al., 2009; Roccaro et al., 2014b), size exclusion chromatography (Zhang and Minear, 2002; Allpike et al., 2005) and in situ approaches such as absorbance and fluorescence spectroscopy have been used (Fabbricino and Korshin, 2004; Boyer and Singer, 2005; Korshin et al., 2007b; Chow et al., 2008; Roccaro et al., 2009; Yang et al., 2008). DOM absorbance and fluorescence are deemed to be primarily associated with the polyhydroxyaromatic (PHA) moiety that is one of the most characteristic features of DOM chemistry. Prior research has demonstrated the existence of strong linear correlations between the aromaticity of DOM (which is the percentage of dissolved organic carbon associated with the PHA moiety) and, on the other hand, specific absorbance of DOM solutions at 254 (SUVA₂₅₄) or 280 nm (ε_{280}) or selected parameters of its fluorescence spectra (Chin et al., 1994; Peuravuori and Pihlaja, 1997; Croué et al., 2000; McKnight et al., 2001; Roccaro et al., 2009; Yang et al., 2008). Recent research has shown that the slopes of logtransformed absorbance spectra of DOM vs. Wavelength are as strongly correlated with DOM aromaticity as it does with the corresponding SUVA values (Roccaro et al., 2015).

Given the association between DOM aromaticity and the generation of DBPs, several surrogate parameters reflecting changes of DOM optical properties and thus its aromaticity (e.g. SUVA₂₅₄, DlnA₃₅₀, ΔA_{272} , ΔA_{280} , $\lambda^{nd}_{0.5}$, $\Delta I_{500/450}$ and integrated fluorescence intensity) have been used to characterize effects of halogenation on DOM chromophores or fluorophores and quantify the formation of individual DBPs, total (absorbable) organic halogen and chlorine consumption (Karanfil et al., 2002; Korshin and Chang, 2008; Roccaro and Vagliasindi, 2010; Roccaro et al., 2011; Yang et al., 2008). Differential absorbance data indicate that the generation of THMs and THAA is closely correlated with the engagement of the slow chromophores and less so with that of the fast DOM chromophores (Korshin et al., 2007b, a).

While effects of chlorine and, in some extent, chloramine on DOM chromophores have been examined in prior research, no attempts have been made to interpret the changes of DOM absorbance and aromaticity induced by its reactions with chloramine in the context of DBP generation.

The goal of this study were: i) to compare effects of chlorine and chloramine on DOM chromophores and its aromaticity; ii) to explore whether correlations between changes in DOM aromaticity and formation of DBP during chloramination exist and iii) to compare such correlations with those observed during chlorination.

2. Materials and methods

2.1. Source waters

Waters used in this study include surface river water from a major source in Campania, Italy (A), a surface water influent (B) and a treated water (C) produced at a drinking water treatment plant located in the Eastern US. The DOC concentration was 3.8 mg/L, 2.0 mg/L and 1.6 mg/L for water A, B and C, respectively. The pH was 7.5 for water A and 7.0 for water B and C. SUVA values were $2.33 \text{ Lm}^{-1} \text{ mg}^{-1}$, 2.46 Lm⁻¹ mg⁻¹ and 1.72 Lm⁻¹ mg⁻¹, for water A, B and C, respectively (Table S1 in Supplementary Data).

Water samples from these sites were shipped in 5-gallon cubitainers to the University of Washington (UW) laboratory. Upon delivery, the samples were passed through 0.45 μ m filters and were stored at 4 °C.

2.2. Experimental and analytical methods

Chlorination and chloramination were carried out in the headspace-free PTFE bags in the dark, at 20 °C, pH 7.0 with 0.03 M phosphate buffer. Chlorine or chloramine doses were 4.2 mg/L, 3.1 mg/L and 2.3 mg/L as Cl₂ for water A, B and C, respectively (Table S1). At desired contact times (from 10 min to 24 h), aliquots were withdrawn from the bags and the residual chlorine was quenched as specified in Standard Method 5710-B. The absorbance of the samples was measured with a PerkinElmer Lamba-18 instrument. Concentrations of THMs, (including CHCl₃, CHCl₂Br, CHClBr₂ and CHBr₃) and haloacetonitriles (HANs, including TCAN, DCAN, BCAN and DBAN) were analyzed using EPA method 551.1. Chlorinated and brominated haloacetic acids HAAs (including

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