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The impact of changes in source water quality on trihalomethane and haloacetonitrile formation in chlorinated drinking water

Chonghua Xue [a], Qi Wang [b,*], Wenhai Chu [a,*], Michael R. Templeton [c]

[a] State Key Laboratory of Pollution Control and Resources Reuse, Ministry of Education Key Laboratory of Yangtze River Water Environment, College of Environmental Science and Engineering, Tongji University, Shanghai 200092, China
[b] College of Life and Environmental Science, Wenzhou University, Wenzhou 325035, China
[c] Department of Civil and Environmental Engineering, Imperial College London, London SW7 2AZ, UK

HIGHLIGHTS

- Reductions in source water DON and DOC led to reduced THM and HAN formation.
- Source water improvements had a greater influence on HANs than THMs.
- Source water bromide reduced but there was increased HAN bromide incorporation.
- DOC may be employed as a surrogate for the impact on THM in source water changes.
- DON is a less reliable surrogate of HAN reduction during source water improvement.

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ABSTRACT

This study examined the formation of disinfection by-products (DBPs), including nitrogenous DBPs, haloacetonitriles (HANs), and carbonaceous DBPs, trihalomethanes (THMs), upon chlorination of water samples collected from a conventional Chinese surface water treatment plant (i.e. applying coagulation, sedimentation, and filtration). Reductions in the average concentrations (and range, shown in brackets) of dissolved organic carbon (DOC) and dissolved organic nitrogen (DON) from 4.8 (3.0–7.3) μg/L and 0.52 (0.20–0.81) μg/L in 2010 to 2.4 (1.4–3.7) μg/L and 0.17 (0.11–0.31) μg/L in 2012, respectively, led to a decrease in HANs and THMs from 5.3 and 28.5 μg/L initially to 0.85 and 8.2 μg/L, as average concentrations, respectively. The bromide concentration in the source water also decreased from 2010 to 2012, but the bromine incorporation factor (BIF) for the THMs did not change significantly; however, for HAN the BIFs increased because the reduction in DON was higher than that of bromide. There was good linear relationship between DOC and THM concentrations, but not between DON and HANs.

1. Introduction

Since their first discovery in the early 1970s, disinfection by-products (DBPs) have been the focus of drinking water regulations, research and water treatment plant (WTP) operations. The number of DBP classes identified in drinking waters has grown from initially the trihalomethanes (THMs) to now a long list of halogenated and non-halogenated organic and inorganic compounds (Bellar et al., 1974; Richardson et al., 2007; Sedlak and Gunten, 2011). Over the last 40 years, many DBP studies have focused on the currently regulated THMs, which commonly occur in water at higher concentrations than most other DBPs. However, in recent years, nitrogen-containing DBPs (N-DBPs) have received increased attention, since some studies suggest that they have higher toxicity than the currently regulated carbonaceous DBPs (C-DBPs) (Plewa et al., 2008).

Haloacetonitriles, a commonly occurring N-DBP class, typically occur at higher concentrations in chlorinated and chloramination drinking waters than many other N-DBPs, e.g., nitrosamines, halonitromethanes,

Abbreviations: BCAN, bromochloroacetonitrile; C-DBPs, carbonaceous disinfection by-products; CF, chloroform; DBAN, dibromoacetonitrile; DBPs, disinfection by-products; DCAN, dichloroacetonitrile; DIN, dissolved inorganic nitrogen; DOC, dissolved organic carbon; DON, dissolved organic nitrogen; HAAs, haloacetic acids; HANs, haloacetonitriles; N-DBPs, nitrogenous disinfection by-products; NOM, natural organic matter; TCAcAm, trichloroacetamide; TCAN, trichloroacetonitrile; THMs, trihalomethanes; TDN, total dissolved nitrogen; UV, ultraviolet; SUVA, specific ultraviolet absorbance; WTP, water treatment plant.

* Corresponding authors. Address: Room 308, Mingjing Building, 1239 Siping Road, Shanghai 200092, China. Tel.: +86 18721871983; fax: +86 21 65986839.
E-mail address: feedwater@126.com (W. Chu).

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and haloacetamides (Krasner et al., 2006; Chu et al., 2009a), and they have significant higher cytotoxicity and genotoxicity than regulated THMs and haloacetic acids (HAAs) (Muellner et al., 2007).

Because of population growth and rising water demand, water utilities are increasingly treating source waters which are more polluted with algae and municipal wastewater. Dissolved organic matter (DOM) in these polluted source waters are a mixture of autochthonous and allochthonous compounds, including humic substances, carboxylic acids, microorganism-associated (e.g. algae, bacteria) products, wastewater effluent organic matters, amino acids and proteinaceous compounds, all of which are potential DBP precursors (Croué et al., 2000; Westerhoff and Mash, 2002; Bond et al., 2011; Gough et al., 2012; Shah and Mitch, 2012; Wang et al., 2013). Also, many of these compounds are characterized by low molecular weight and therefore likely to be poorly removed by conventional WTPs using coagulation–sedimentation–filtration (Chu et al., 2011a; Shah et al., 2012; Gough et al., 2014), and so they may subsequently react with disinfectants to form C-DBPs (esp., THMs) and N-DBPs (esp., HANs).

Generally, DBP control strategies can be divided into three categories: (1) removal of DBP precursors, (2) modification and optimization of treatment and disinfection practices to minimize DBP formation, and (3) removal of DBPs after formation. The addition of pre-treatment to existing WTPs to achieve improved removal of HAN precursors prior to disinfection has been reported (Chu et al., 2011b, 2012; Shah et al., 2012). Efforts have also been made to study HAN formation mechanisms with the purpose of acquiring the knowledge to reduce their formation during chlorination (Bond et al., 2011; Shah and Mitch, 2012). Additionally, the removal of HANs after formation has been investigated (Chu et al., 2009b). However, while these DBP control strategies effectively reduce the formation of some DBPs, they may cause other DBPs to be formed and/or require high operating cost for the water utility. For example, ozone pre-oxidation can be effective for the control of N-nitrosodimethylamine formation during subsequent chloramination (Shah et al., 2012), but promotes the formation of trichloronitromethane and trichloroacetaldehyde (Chu et al., 2012; Shah and Mitch, 2012). An alternative strategy would be protection and improvement of source water quality to reduce subsequent DBP formation in WTP. However, there is relatively little information on the impact of source water quality changes on N-DBP formation.

The aim of this study was to evaluate the impact of source water quality changes on the formation of HANs and THMs in a typical south Chinese WTP which treats water from a surface water reservoir. In 2010, the reservoir was overtaken by blue-green algae, which caused the water to turn a blue color and acquire a foul smell. Because the algae outbreak in the reservoir was attributed to domestic and agricultural wastewaters, the local government took steps to limit the impact of these wastewaters on the reservoir in an attempt to improve the source water quality, specifically by using bioremediation technologies (e.g., stocking silver carp and bighead carp into the reservoir, and limiting the discharge of domestic and agricultural wastewaters) from 2010 to 2012. These measures therefore allowed us to evaluate the effect of source water changes on DBP formation. The DBPs considered in this study included the THMs (chloroform [CF], bromodichloromethane [BDCM], dibromochloromethane [DBCM], bromoform [BF]) and HANs (dichloroacetonitrile [DCAN], trichloroacetonitrile [TCAN], bromochloroacetonitrile [BCAN], dibromoacetonitrile [DBAN]). Changes in dissolved organic carbon (DOC), dissolved organic nitrogen (DON), and bromide in the source water were the main independent variables of interest in this study. Climate change could dramatically alter the quality of source waters (Tang et al., 2013), therefore this study will also provide an indication of the potential scale of impacts of climate change on DBP formation.

2. Materials and methods

2.1. Materials

Standard solutions of the considered DBPs were purchased from Sigma–Aldrich (St Louis, Missouri, USA). Ultrapure water was produced with a Millipore Milli-Q Gradient water purification system (Billerica, MA, USA). All other materials were at least analytical grade and obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China) unless otherwise noted.

2.2. Sample collection

Raw water and filtered water samples were collected from the influent of the mixing tank and the effluent of the clean water tank, respectively, at a surface water treatment plant located in Zhejiang, China every month in 2010 and 2012. The treatment sequence in the WTP was coagulation-sedimentation (15-mg/L polyaluminium chloride, 20-min retention time), sand filtration (8-min retention time) and chlorination (1.5-mg/L chlorine), which is common among Chinese WTPs, as shown in Fig. 1. Raw water and filtered water quality parameters for the WTP in 2010 and 2012 are listed in Table 1. Once collected, disinfectant residuals in the samples were immediately quenched by ascorbic acid at a normality twice the initial normality of the disinfectant added. Glacial acetic acid was used to lower the pH to 4.8–5.5 for the HAN and THM analyses. Subsequently, the samples were transported to the refrigerator (set at 4 °C) in laboratory and analyzed as quickly as possible.

2.3. Sample analyses

Turbidity was measured onsite using a turbidimeter (HACH 2100N, USA). DOC and total dissolved nitrogen (TDN) were measured using a TOC analyzer equipped with a total nitrogen measurement unit (Shimadzu TOC-VCPH, Japan). The DON concentration was the difference in the measured TDN and dissolved inorganic nitrogen (DIN). DIN (ammonia, nitrate, and nitrite), free and total chlorine were measured with a portable spectrophotometer (HACH DR2800, USA). UV_{254} was determined using a UV/Vis double beam spectrophotometer (Unico4802, USA). Bromide concentration was determined by ion chromatography (Dionex 2000, USA)

The concentrations of the THMs (CF, BDCM, DBCM, and BF) and HANs (DCAN, TCAN, BCAN, and DBAN) were quantified using a purge & trap sample concentrator (eclipse4660, OI, USA) and gas chromatography-mass spectrometry (QP2010, Shimadzu, Japan), based on USEPA method 524.2 (USEPA, 1992), as in previous studies (Chu et al., 2011a, 2012) The detection limits for CF, BDCM, DBCM, BF, DCAN, TCAN, BCAN, and DBAN were 0.067, 0.071, 0.087, 0.085, 0.11, 0.091, 0.15, and 0.26 μg/L, respectively.

3. Results and discussion

3.1. DON and DOC

The averages (and ranges) of DOC and DON in raw water decreased from 4.8 (3.0–7.3) μg/L and 0.52 (0.20–0.81) μg/L in 2010 to 2.4 (1.4–3.7) μg/L and 0.17 (0.11–0.31) μg/L in 2012, respectively (Table 1, Fig. 2).

In a survey of US WTPs that were impacted by algae and/or treated wastewater (Mitch et al., 2009), the average DOC/DON ratio (13 mg/mg) of the surveyed waters confirmed that these sources were influenced by elevated levels of DON compared to a previous US study (Lee et al., 2006), where the average DOC/DON ratio in a broader set of surface waters was 18 mg/mg. In the source water in