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Characteristics of molecular weight distribution of dissolved organic matter in bromide-containing water and disinfection by-product formation properties during treatment processes

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ABSTRACT

The characteristics of dissolved organic matter (DOM) and bromide ion concentration have a significant influence on the formation of disinfection by-products (DBPs). In order to identify the main DBP precursors, DOM was divided into five fractions based on molecular weight (MW), trihalomethane formation potential (THMFP) and haloacetic acid formation potential (HAAFP) were determined for fractions, and the change in contents of different fractions and total DBPs during water treatment processes (pre-chlorination, coagulation, sand filtration, disinfection) were studied. Moreover, the relationship between bromide concentration and DBP generation characteristics in processes was also analyzed. The results showed that the main DBP precursors were the fraction with MW < 1 kDa and fraction with MW 3–10 kDa, and the DBP's generation ability of lower molecular weight DOM (<10 kDa) was higher than that of higher molecular weight DOM. During the different processes, pre-chlorination and disinfection had limited effect on removing organics but could alter the MW distribution, and coagulation and filtration could effectively remove organics with higher MW. For DBPs, trihalomethanes (THMs) were mainly generated in pre-chlorination and disinfection, while haloacetic acids (HAAs) were mostly generated during pre-chlorination; coagulation and sand filtration had little effect on THMs but resulted in a slight removal of HAAs. In addition, the results of ANOVA tests suggested that molecular sizes and treatment processes have significant influence on DBP formation. Moreover, with increasing bromide concentration, the brominated DBPs significantly increased, but the bromine incorporation factor in the processes was basically consistent at each concentration.

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Introduction

Disinfection by-products (DBPs) in drinking water formed during chlorination and chloramination processes have drawn worldwide attention. Among the DBPs, trihalomethanes (THMs)

and haloacetic acids (HAAs) are the most representative substances (Aghaba et al., 2016; Ding et al., 2013; Krasner et al., 1989; Richardson and Postigo, 2012; Wang and Zhu, 2010), and many toxicological and biological experiments have indicated that THMs and HAAs could be potentially hazardous to humans

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61 and may lead to carcinogenesis, teratogenesis and mutagenesis
62 (Andrews et al., 2004; Hsu et al., 2001; Mash et al., 2014;
63 Rodriguez et al., 2000). Researchers have proved that dissolved
64 organic matters (DOMs) were the primary precursors of DBPs, as
65 they could easily react with free chlorine to generate THMs and
66 HAAs (Han et al., 2015; Krasner et al., 1989; Yang et al., 2015;
67 Zeng and Arnold, 2014; Zheng et al., 2016), and some studies
68 have also investigated the formation characteristics of different
69 DOM fractions with different molecular size, hydrophilicity and
70 so on (Hua and Reckhow, 2007; Hur et al., 2013; Imai et al., 2003;
71 Lin et al., 2015; Niu et al., 2015b; Panyapinyopol et al., 2005). DOM
72 fractions with different molecular weight (MW) showed mark-
73 edly different physical and chemical properties during water
74 treatment processes, which can make the kinds and amounts of
75 halogenated DBPs remarkably diverse (Collins et al., 1986; Han
76 et al., 2013). Therefore, classifying DOM into different fractions
77 by MW and investigating their variation trends during water
78 treatment processes are important to controlling halogenated
79 DBPs. What's more, brominated DBPs have been of wide
80 concern in recent years, and many studies have been carried
81 out to determine their generation characteristics (Chowdhury
82 et al., 2016; Zha et al., 2014, 2016; H. Zhang et al. 2015).

83 Several studies have reported the effect of the molecular
84 weight of organic matter on DBP formation potential. For
85 example, two source waters from South Carolina and New
86 York were studied comparatively, and the results showed that
87 the production of DBPs increased with the increase of MW in
88 the former water, but the major precursors were fractions
89 with MW < 3 kDa in the latter, indicating that the formation
90 characteristics of DBPs of different MW components differed
91 in different water bodies, and the MW distribution of natural
92 waters had a strong regionalism (Kitis et al., 2002; Lin et al.,
93 2007). Water from the Pearl River was fractioned into different
94 groups based on MW, and it was discovered that the fraction
95 with MW < 0.5 kDa was the main organic component of DOM
96 (at 58%), and that the components with different MW ex-
97 hibited marked differences in reacting with chlorine (Zhao
98 et al., 2006). A study focused on the reaction characteristics
99 of humic acids in disinfection process was conducted, and
100 observed that the main precursors were the smaller molecule
101 matters (Zhang et al., 2005). Detailed analysis of THMs showed
102 that chloroform (TCM) was mainly produced by the fraction
103 with MW of 0.5–10 kDa, while dibromochloromethane (DBCM)
104 and tribromomethane (TBM) were mainly generated from the
105 fraction with MW < 0.5 kDa (Chang et al., 2000).

106 Moreover, in studies focusing on the decrease of DOM it
107 was determined that conventional water treatment technol-
108 ogies, including coagulation processes, filtration processes
109 and disinfection processes, demonstrated clear discrepancies
110 in removal capability for components with different MW
111 (Collins et al., 1986; Han et al., 2013; Wang and Liu, 1999).
112 Experiments showed that the conventional coagulation
113 process mainly removed organic matters whose molecular
114 weights were more than 10 kDa, and pre-oxidation and
115 pre-adsorption processes could effectively improve the removal
116 rate of organics (Han et al., 2013). The slow sand filtration
117 process could remove most DOMs with higher molecular size
118 from river water, and the subsequent photo-Fenton process
119 could strengthen the removal efficiency (Moncayo-Lasso et al.,
120 2008). During disinfection, researchers found that DOM fractions

121 with MW < 2.5 kDa would be decreased more than other frac-
122 tions, and produce higher concentrations of DBPs (Kristiana
123 et al., 2013).

124 The influence of the bromide ion on the speciation and yield
125 of THMs and HAAs in chlorination and other processes was also
126 investigated, and it was found that the brominated DBP (Br-DBP)
127 concentration increased with increasing bromide concentration
128 (Hua et al., 2006; Zha et al., 2014). Experiments showed that with
129 increasing bromide concentration, the total yields of THMs and
130 HAAs increased (Bond et al., 2014); six bromide concentrations
131 ranging from 0.05 to 4.0 mg/L were studied during chlorination,
132 and the results showed that the speciation of THMs and HAAs
133 gradually shifted from chlorinated species to mixed bromochloro
134 species during chlorination with the increase of bromide con-
135 centration (Uyak and Toroz, 2007).

136 To date, the primary precursors of DBPs have been deter-
137 mined to be different in different water bodies (Chang et al.,
138 2000, 2001; Zhang and Minear, 2002), and the ability of organic
139 fractions to generate DBPs is also different (Zhao et al., 2013).
140 Meanwhile, few studies simultaneously considered the rela-
141 tionship between fractions with different MW and the contents
142 of DBPs in water treatment processes, and fewer integrated the
143 effects of bromide ion.

144 The purpose of this study is to analyze the organic fractions
145 with different MW in Yuqiao Reservoir, study the disinfection
146 by-product formation potential (DBFP) (including trihalometh-
147 ane formation potential (THMFP) and haloacetic acid formation
148 potential (HAAFP)) of the source water, and also analyze the
149 change characteristics of organic fractions and contents of DBPs
150 during sequential water treatment processes. In consideration
151 of the bromide ion pollution in the source water, we enhanced
152 the bromide concentration and set several levels to study the
153 relationship between bromide ion concentration and DBP
154 generation characteristics in treatment processes. Through
155 this work, we hope to identify the primary precursors of THMs
156 and HAAs of Yuqiao Reservoir, to provide information on the
157 change characteristics of organic fractions with different
158 molecular size, the relationship between bromide and DBP
159 generation characteristics in treatment processes, and finally,
160 to provide suggestions for the control of DBPs in water
161 treatment processes.

1. Materials and methods 163

1.1. Sample collection 164

165 All samples were collected from Yuqiao Reservoir located in
166 Tianjin, North China. Yuqiao Reservoir is the most important
167 water resource for Tianjin in North China, with a capacity of
168 1.559 billion m³, supplying drinking water for 15.17 million
169 people in Tianjin (H. Zhang et al., 2015; Y. Zhang et al., 2015).
170 The water quality of Yuqiao Reservoir has a strong impact on
171 the drinking water quality of Tianjin. In recent years, 171
172 researchers have conducted various studies on the water
173 quality of Yuqiao Reservoir. For example, the dynamic change
174 of chlorophyll (Liu et al., 2014), correlation analysis of the
175 richness of phytoplankton and the main water factors (Zhang
176 et al., 2013), the heavy metals in the surface sediment (Wu
177 et al., 2011), and eutrophication forecasting and management

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