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# Comparison of chlorination and chloramination in carbonaceous and nitrogenous disinfection byproduct formation potentials with prolonged contact time



Hiroshi Sakai <sup>a, \*</sup>, Shunsuke Tokuhara <sup>b</sup>, Michio Murakami <sup>c, e</sup>, Koji Kosaka <sup>d</sup>, Kumiko Oguma <sup>a</sup>, Satoshi Takizawa <sup>a</sup>

<sup>a</sup> Department of Urban Engineering, School of Engineering, The University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo, 113-8656, Japan

<sup>b</sup> Hitachi, Ltd., 6-6, Marunouchi 1, Chiyoda-ku, Tokyo, 100-8280, Japan

<sup>c</sup> Fukushima Medical University, 1, Hikariga-oka, Fukushima City, 960-1295, Japan

<sup>d</sup> National Institute of Public Health, 2-3-6 Minami, Wako-shi, Saitama 351-0197, Japan

<sup>e</sup> Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo, 153-8505, Japan

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## ABSTRACT

Due to decreasing water demands in Japan, hydraulic retention times of water in piped supply systems has been extended, resulting in a longer contact time with disinfectants. However, the effects of extended contact time on the formation of various disinfection byproducts (DBPs), including carbonaceous DBPs such as trihalomethane (THM) and haloacetic acid (HAA), and nitrogenous DBPs such as nitrosodimethylamine (NDMA) and nitrosomorpholine (NMor), have not yet been investigated in detail. Herein, we compared the formation of these DBPs by chlorination and chloramination for five water samples collected from rivers and a dam in Japan, all of which represent municipal water supply sources. Water samples were treated by either filtration or a combination of coagulation and filtration. Treated samples were subjected to a DBP formation potential test by either chlorine or chloramine for contact times of 1 day or 4 days. Four THM species, nine HAA species, NDMA, and NMor were measured by GC-ECD or UPLC-MS/MS. Lifetime cancer risk was calculated based on the Integrated Risk Information System unit risk information. The experiment and analysis focused on (i) prolonged contact time from 1 day to 4 days, (ii) reduction efficiency by conventional treatment, (iii) correlations between DBP formation potentials and water quality parameters, and (iv) the contribution of each species to total risk. With an increased contact time from 1 day to 4 days, THM formation increased to 420% by chloramination. Coagulation-filtration treatment showed that brominated species in THMs are less likely to be reduced. With the highest unit risk among THM species, dibromochloromethane (DBCM) showed a high correlation with bromine, but not with organic matter parameters. NDMA contributed to lifetime cancer risk. The THM formation pathway should be revisited in terms of chloramination and bromine incorporation. It is also recommended to investigate nitrosamine formation potential by chloramination.

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### 1. Introduction

Disinfection byproducts (DBPs) that result following drinking water treatment are serious concerns. Their presence in drinking water was first reported in 1974 (Rook, 1974), and numerous subsequent studies on DBPs have been performed. In 1977, fulvic acid was found to be a major precursor of trihalomethanes (THMs)

\* Corresponding author. E-mail address: h\_sakai@env.t.u-tokyo.ac.jp (H. Sakai).

http://dx.doi.org/10.1016/j.watres.2015.11.002 0043-1354/© 2015 Elsevier Ltd. All rights reserved. (Rook, 1977). THMs and haloacetic acids (HAAs) are two major classes of carbonaceous DBPs (C-DBPs), and are known as possible human carcinogens (Nieuwenhuijsen et al., 2000; Muellner et al., 2007). Nitrosamines (NAs) are also considered important and emerging nitrogenous DBPs (N-DBPs) in water (Mitch et al., 2003). Nitrosodimethylamine (NDMA) and nitrosomorpholine (NMor) are members of this extremely potent class of carcinogens, which impose  $10^{-6}$  cancer risk even at ng/L levels (US EPA, 2015).

Natural organic matter (NOM) is considered a possible DBP precursor, both for carbonaceous and nitrogenous DBPs (Goslan et al., 2009). NOM is a complex of various organic materials, the

exact chemical identities of which are unknown (Bond et al., 2009). Thus, various analyses have been performed to determine its characteristics in DBP production. Use of Ultraviolet (UV) absorbance (Korshin et al., 1997) has been used to characterize organic matter. Specific UV absorbance (SUVA) was proposed (Weishaar et al., 2003) as an indicator of reactivity in DBP formation. Fractionation and molecular size distribution of target organic matter were also analyzed to determine the relationship with DBP formation potential (Imai et al., 2003).

Climate change is expected to impact source water quality, especially with regard to organic matter and DBP formation potential (Delpa et al., 2009). As the Japanese islands range from cooltemperate, temperate, to sub-tropical regions, it is expected that source waters will contain a variety of organic matter. Therefore, we examined samples from major cities in a temperate region and a remote island in a sub-tropical region to elucidate the differences. Social factors were also taken into consideration. Recently, Japanese water utilities have encountered decreased water demand due to decreasing populations in water supply areas (Musiake and Koike, 2009; Koike and Oki, 2009). Decreased water demand is expected to result in prolonged retention times in the water supply system.

Compared with chlorine, chloramine is a more stable agent for continuous disinfection, even after a prolonged retention time in the water supply system (Vikesland et al., 2001). Although a great deal of information is available on DBP production by chlorination, there is little information regarding the production of DBPs by chloramination, especially C-DBPs. Although few studies have compared chlorine and chloramine in terms of DBP formation potential (Bougeard et al., 2010; Goslan et al., 2009), they did not investigate the differences in behavior of DBP species or the effects of prolonged contact time. Therefore, in the study described herein, we compared chlorination and chloramination with a short contact time (1 day) to a longer contact time (4 days).

Both carbonaceous and nitrogenous DBPs were investigated. The C-DBPs examined were four-THMs and nine-HAAs, while the N-DBPs examined were NDMA and NMor. Coagulation-filtration treatment or filtration treatment was also applied to source water to determine the potential for reduction by conventional water treatment processes. Conventional water treatment systems remove organic matter and reduce DBP formation potential (Black et al., 1996; Sohn et al., 2006). This manuscript further discusses differences in reduction ratios between THM and HAA species, with a focus on bromine by either chlorination or chloramination.

Fluorescence excitation-emission matrix (EEM) analysis is a simple method to determine the presence of certain types of organic matter. One way to quantify the results by dividing the matrix into five regions has been proposed (Chen et al., 2003): (i) tyrosine-like, (ii) tryptophan-like, (iii) fulvic-like, (iv) soluble microbial products, and (v) humic-like. However, only a limited number of studies have examined the relationship between each EEM region and DBP formation potential (Hua et al., 2007, 2010). While lake water was analyzed in previous studies, river water as a municipal water supply source has not been investigated in detail.

In the present study, DBP formation potential and its relationship to organic matter components were investigated. Lifetime cancer risk was calculated based on the Integrated Risk Information System unit risk information (USEPA, 2015) for six measured DBPs to compare the performance of chlorination and chloramination with prolonged contact times.

Two disinfectants, chlorine and chloramine, were used to evaluate the risk by DBPs. Three types of DBPs, THM, HAA and NA, were measured as C- and N-DBPs. The experiment and analysis focused on (i) prolonged contact time from 1 day to 4 days, (ii) reduction efficiency by conventional treatment, (iii) correlations between DBP formation potential and water quality parameters, and (iv) the contribution of each species to total risk.

#### 2. Materials and methods

#### 2.1. Water samples

Water samples were collected from rivers or reservoirs used as municipal water supply sources. Of the five water samples used in this study, four were collected from two major cities in Japan located in a temperate region. Three temperate samples were from one city (Tem11, 12 and 13), and the other temperate sample was from another city (Tem21). The other sample (Tro11) was collected on a remote island in a sub-tropical region. Samples were treated either by filtration through glass fiber filters (GF/C) with a pore size of 1.2  $\mu$ m (Whatman), or by coagulation and GF/C filtration. At each treatment step, basic water quality parameters as well as fluorescence EEM were measured. Treated and untreated samples were subjected to a DBP formation potential test with either chlorine or chloramine for a contact time of 1 day or 4 days.

#### 2.2. Coagulation and filtration

Water samples were treated by either filtration or a combination of coagulation and filtration. To minimize the impact of contaminating organic matter, all filters were baked for 2 h at 400 °C prior to filtration. Polyaluminum chloride (PAC) was used as a coagulant. PACI dose was determined by a jar test for each water sample. The coagulation process began with rapid mixing at 120 rpm for 3 min, followed by slow mixing at 30 rpm for 15 min, and ended with a settlement process for 20 min. After the jar test, the turbidity of the supernatant was measured to determine the optimum PAC dose.

#### 2.3. Organic matter analysis

Organic matter was characterized by dissolved organic carbon content (DOC), UV absorbance at 254 nm (UV<sub>254</sub>), specific UV absorbance (SUVA), and fluorescence EEM. DOC was measured with a TOC analyzer (TOC-V; Shimadzu), UV<sub>254</sub> was measured with a spectrophotometer (UH-5300; Hitachi), and EEM was analyzed with a fluorescence spectrophotometer (F-4500; Hitachi).

#### 2.4. DBP formation potential test

#### 2.4.1. Chlorination or chloramination

Samples were treated with either chlorine or chloramine and subjected to a DBP formation potential test under uniform formation condition. Sodium hypochlorite solution (Kishida Kagaku) was used for chlorination. Chloramine solution was prepared by reaction of ammonium chloride with sodium hypochlorite solution. The concentration of monochloramine was confirmed using a colorimeter (DR890; HACH). The chlorination/chloramination reaction was initiated by the addition of chlorine or chloramine to 800 mL of water sample. The pH of the solution was adjusted to  $7.0 \pm 0.2$ . After the addition of disinfection agent, samples were transferred into 500-mL amber glass bottles to fill the bottle without a headspace. Chlorinated samples were placed in an incubator for 1 day or 4 days at a constant temperature of 20 °C. The concentration of chlorinating agent was adjusted so that it remained within a certain concentration range after a reaction time of 1 day or 4 days, taking the concentrations of organic matter and ammonium nitrogen into consideration; free chlorine concentration ranged from 1.0 to 2.0 mg/L and combined chlorine concentration ranged from 2.5 to 3.5 mg/L, with reference to our previous report (Kosaka et al., 2009).

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