



A comparison of iodinated trihalomethane formation from iodide and iopamidol in the presence of organic precursors during monochloramination



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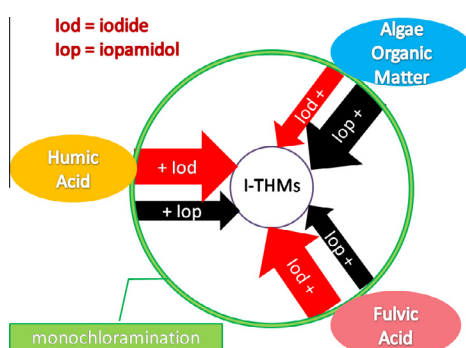
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HIGHLIGHTS

- I-THMs formation from iopamidol favored AOM rather than HA/FA during monochloramination.
- Br⁻ exhibited higher activity to form I-THMs from I⁻ pathway than iopamidol pathway.
- I-THMs formed rapidly from iodide but relatively slow from iopamidol.
- I-THMs yields positive correlated with SUVA in I⁻ pathway while iopamidol negative.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 29 May 2014

Received in revised form 15 July 2014

Accepted 16 July 2014

Available online 27 July 2014

Keywords:

Algae organic matter

Humic acid

Iodinated contrast media

Bromide

Disinfection by-products

ABSTRACT

The formation of iodinated trihalomethanes (I-THMs) from iodide and iopamidol, two major iodine sources to form iodinated disinfection by-products (I-DBPs) during monochloramination, was compared in the presence of different organic precursors including algae organic matter (AOM), Suwannee River humic acid (HA), fulvic acid (FA) and 2 raw waters. Effect of bromide, reaction time and pH on I-THM formation was also studied. The results showed that in the presence of HA and FA, monochloramination of iodide exhibited much higher I-THM formation than that of iopamidol, while in the presence of AOM, monochloramination of iopamidol formed more I-THMs than that of iodide. The presence of bromide increased I-THM formation from iodide with iodoform as the dominant species, while bromine-containing I-THM species increased significantly from iopamidol with bromochloroiodomethane as the predominant species. The reaction between monochloramine and iopamidol is relatively slow and long-lasting compared to that between monochloramine and iodide. I-THM formation from iodide in the presence of HA was maximum at pH 7 but significantly inhibited at pH higher than 7.5 from iopamidol in the presence of AOM. The correlation between the ratio of I-THMs to dissolved organic carbon concentration and the specific ultraviolet absorbance value of raw waters was positive for iodide but negative for iopamidol during monochloramination.

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1. Introduction

Disinfection has been applied in drinking water treatment process to kill waterborne pathogens and safeguard human health starting from the utilization of chlorine in the early 1900s [1]. About 70 years later, the identification of chloroform and other trihalomethanes (THMs) raised concerns of risk caused by these disinfection by-products (DBPs) to human health [2,3]. Although many researchers have studied DBP formation mechanisms and the US Environmental Protection Agency issued a regulation to control 11 DBPs, including 4 THMs, 5 haloacetic acids, bromate and chlorite [4], there are still lots of emerging DBPs not been investigated thoroughly [5]. As a group of them, iodinated DBPs (I-DBPs) have been detected in developed countries [6] and shown to be more cytotoxic and/or genotoxic than their chlorinated and brominated analogs [7,8].

Natural iodide (I^-) was found in raw waters at average concentration of 20 $\mu\text{g/L}$ [9], and the concentration as high as 104.2 $\mu\text{g/L}$ was found in a drinking water treatment plant (DWTP) [8]. It is believed to be the primary iodine source for the formation of I-DBPs. However, Duirk et al. [10] found that iodinated X-ray contrast media (ICM) were another iodine source. ICM are widely used pharmaceuticals to image soft tissues such as organs and blood vessels during diagnostic tests, which are metabolically stable in the body and can be rapidly discarded through urine and feces [11]. However, municipal waste water treatment plants can not remove ICM effectively [12] and leave them to be an important iodine source in the aquatic environment. In an occurrence study of source waters from 10 cities in the US, 4 kinds of ICM were recognized and iopamidol was the most frequently detected one with concentrations up to 2700 ng/L [10]. Furthermore, their study indicated that iopamidol can form the most appreciable level of I-DBPs compared to iohexol and iopromide, another two species of ICM [10].

Some researchers have studied the formation of I-DBPs using different disinfectants including chlorine, chloramine, ozone, potassium permanganate, chlorine dioxide and manganese dioxide [13] and [14]. Bichsel and von Gunten [13] proved that chloramine can form significant amount of I-DBPs because it cannot oxidize the reactive hypiodous acid (HOI) to the stable iodate so that the reaction between HOI and natural organic matter (NOM) leads to the formation of I-DBPs. Hua and Reckhow [15] found hydrophilic and low molecular weight (MW) organic precursors produced more iodinated trihalomethanes (I-THMs) than corresponding hydrophobic and high MW precursors during chloramination. Bromide (Br^-) concentration also plays an important role in I-THM formation. Jones et al. [16] revealed that Br^-/I^- ratio had an important effect on the speciation of I-THM. These researches were aimed at the formation of I-THMs from iodide; however, investigations on I-THM formation from ICM (especially iopamidol) are limited. Furthermore, the effect of different kinds of organic precursors on I-THM formation has not been studied yet.

The objectives of this study were (1) to investigate and compare the formation of 6 I-THMs from iodide and iopamidol, respectively, during monochloramination in the presence of different organic precursors, including algae organic matter (AOM), Suwannee River humic acid (HA), fulvic acid (FA) and 2 raw waters and (2) to study the effect of bromide, reaction time and solution pH on the formation of I-THMs from iodide + HA and iopamidol + AOM, respectively, during monochloramination.

2. Materials and methods

2.1. Chemicals and reagents

Iopamidol was purchased from US Pharmacopeial Convention (Rockville, MD) at purity of 99.6%. Five I-THM standards, including

chlorodiiodomethane (CHCl_2 , 90–95%), dichloriodomethane ($\text{CHCl}_2\text{I} \geq 95\%$), bromochloriodomethane ($\text{CHBrClI} \geq 95\%$), dibromiodomethane (CHBr_2I , 90–95%) and bromodiiodomethane (CHBrI_2 , 90–95%), were obtained from CanSyn Chem. Corp. (Toronto, ON, Canada). Methyl *tert*-butyl ether (MtBE) and methanol were purchased from J.T. Baker (Phillipsburg, NJ). Potassium iodide (KI), potassium bromide (KBr), potassium dihydrogen phosphate (KH_2PO_4), sodium hydroxide (NaOH), sodium hypochlorite (NaClO available chlorine 4.00–4.99%), ammonium chloride (NH_4Cl), sodium sulfite (Na_2SO_3) and iodoform (CHI_3) standards were purchased from Sigma–Aldrich (USA). All of the solutions were prepared with ultra-pure water produced using a Milli-Q water purification system (Millipore, USA).

2.2. Water sources

Three kinds of artificially synthesized waters and 2 raw waters from DWTPs were used in this study. Suwannee River HA and FA were purchased from the International Humic Substances Society (IHSS) to act as NOM precursors. The stock solutions of HA and FA were prepared by dissolving 50 mg of HA or FA in 1 L purified water at pH 7.5, which were adjusted using 1 N H_2SO_4 or NaOH, mixed for 4 h, filtered through 0.45 μm cellulose acetate membranes (Millipore Corp., Billerica, MA) to remove suspended solids and then stored at 4 $^\circ\text{C}$ in darkness until use. AOM is different from NOM with the principal components of protein-like substances instead of humic-like matters [17,18]. *Microcystis aeruginosa*, one of the most common blue-green species found in fresh water bloom [19], was selected as the representative AOM and used as the third organic precursor to make the synthesized water. *M. aeruginosa* was purchased from the Institute of Hydrobiology, Chinese Academy of Sciences and cultured in BG11 media in an incubator at 25 $^\circ\text{C}$ [18]. The algae solution was harvested during its exponential period and filtered through 0.45 μm cellulose acetate membranes prior to usage.

Raw waters of Yangtze River and Huangpu River were collected from Yangshupu DWTP (YDWTP, Shanghai, China) and Minhang No. 2 DWTP (MDWTP, Shanghai, China), respectively, in the autumn of 2013 and filtered through 0.45 μm membranes prior to usage. The characteristics of these 5 water sources are summarized in Table 1.

2.3. Experimental procedures

The monochloramination experiments were conducted in duplicate under headspace-free conditions in 40-mL amber glass screw-cap vials with PTFE-lined septa at room temperature (25 ± 1 $^\circ\text{C}$). The effect of 4 factors on I-THM formation from iodide and iopamidol were evaluated, including different types of organic precursors, bromide ion, reacting time and solution pH. The stock solution of monochloramine (NH_2Cl) was prepared freshly by mixing sodium hypochlorite with ammonium chloride at N/Cl molar ratio of 1.2. We utilized the preformed monochloramine rather than prechlorination followed by ammonia addition because I-THM formation was generally lower for the latter process due to

Table 1
Characteristics of five water sources.

Source	HA	FA	AOM	YDWTP	MDWTP
DOC (mg C/L)	5.00	5.00	5.00	1.80	4.26
UV ₂₅₄ (cm^{-1})	0.315	0.244	0.010	0.032	0.107
SUVA ^a (L/mg m)	6.29	4.88	0.2	1.77	2.51
Br^- ($\mu\text{g/L}$)	ND ^b	ND	ND	16	95

^a SUVA: the UV₂₅₄ absorbance divided by the DOC concentration.

^b ND: not detected.

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