



Formation of iodo-trihalomethanes, iodo-acetic acids, and iodo-acetamides during chloramination of iodide-containing waters: Factors influencing formation and reaction pathways



Shaogang Liu^{a,b}, Zhenlin Li^b, Huiyu Dong^a, Bernard A. Goodman^c, Zhimin Qiang^{a,*}

^a Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, University of Chinese Academy of Sciences, Chinese Academy of Sciences, 18 Shuang-qing Road, Beijing, 100085, China

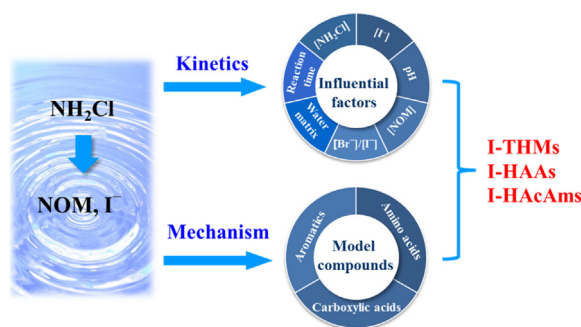
^b Guangxi Colleges and Universities Key Laboratory of Food Safety and Pharmaceutical Analytical Chemistry, Guangxi Key Laboratory of Chemistry and Engineering of Forest Products, School of Chemistry and Chemical Engineering, Guangxi University for Nationalities, Nanning, 530006, Guangxi, China

^c College of Physical Science and Engineering, State Key Laboratory for Conservation and Utilization of Subtropical Agro-Bioresources, Guangxi University, Nanning, 520004, Guangxi, China

HIGHLIGHTS

- Kinetic model for HOI/I₂ evolution during chloramination was developed.
- Maximum I-DBPs formation and HOI/I₂ consumption occurred at pH 8.0.
- I-HAcAms, once formed, could be further hydrolyzed to corresponding I-HAAs.
- Low-SUVA₂₅₄ NOM favored I-DBPs formation compared to high-SUVA₂₅₄ NOM.
- Reaction pathways for I-DBPs formation during chloramination were proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigated systematically the factors influencing the formation of iodinated disinfection by-products (I-DBPs) during chloramination of I⁻-containing waters, including reaction time, NH₂Cl dose, I⁻ concentration, pH, natural organic matter (NOM) concentration, Br⁻/I⁻ molar ratio, and water matrix. Among the I-DBPs detected, iodoform (CHI₃), iodoacetic acid (IAA), diiodoacetic acid (DIAA), triiodoacetic acid (TIAA), and diiodoacetamide (DIACAm) were the major species produced from reactions between reactive iodine species (HOI/I₂) and NOM. A kinetic model involving the reactions of NH₂Cl auto-decomposition, iodine species transformation and NOM consumption was developed, which could well describe NH₂Cl decay and HOI/I₂ evolution. Higher concentrations of CHI₃, IAA, DIAA, TIAA, and DIACAm were observed in chloramination than in chlorination, whereas IO₃⁻ was only formed significantly in chlorination. Maximum formation of I-DBPs occurred at pH 8.0, but acidic conditions favored the formation of iodinated haloacetic acids and DIACAm. Increasing Br⁻/I⁻ molar ratio from 1 to 10 did not increase the total amount of I-DBPs, but produced more bromine-substituting species. In addition, chloramination of 18 model compounds indicated that

* Corresponding author.

E-mail address: qiangz@rcees.ac.cn (Z. Qiang).

low-SUVA₂₅₄ (specific ultraviolet absorbance at 254 nm) NOM generally favored the formation of I-DBPs compared to high-SUVA₂₅₄ NOM. Finally, potential pathways for I-DBPs formation from chloramination of NOM were proposed.

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1. Introduction

The formation of disinfection by-products (DBPs) during water treatment is of great concern because of their potential adverse health effects. To comply with the increasingly stringent regulatory limits for trihalomethanes (THMs) and haloacetic acids (HAAs) [1], many water utilities have switched from free chlorine to monochloramine (NH₂Cl) as a secondary disinfectant, because the latter leads to lower concentrations of regulated THMs and HAAs. However, the use of NH₂Cl can promote the formation of a large number of other potentially harmful DBPs, such as haloacetoneitriles (HANs), *N*-nitrosodimethylamine, and iodinated disinfection by-products (I-DBPs) [2,3]. Therefore, DBPs formation can still be problematic during chloramination of drinking water.

During the disinfection processes, iodide (I⁻) in source waters is rapidly oxidized to hypiodous acid (HOI), which can either react with natural organic matter (NOM) to form various I-DBPs, or be further oxidized to iodate (IO₃⁻) [4–6]. Apart from iodinated trihalomethanes (I-THMs) [7], many other I-DBPs have also been found in disinfected drinking water in recent years, including iodinated haloacetic acids (I-HAAs) [2], polar I-DBPs [8], and iodinated haloacetamides (I-HAcAms) [9]. The I-DBPs are generally more toxic than their chlorinated and brominated analogues [10], and recent mammalian cell toxicity results have shown that iodoform (CHI₃) is 60 and 146 times more cytotoxic than bromoform (CHBr₃) and chloroform (CHCl₃), respectively [11,12]. I-HAAs are frequently found in chloraminated water, which represent a new class of highly toxic DBPs in drinking water [2,13]. In addition, new nitrogenous I-DBPs, such as I-HAcAms [e.g., chloroiodoacetamide (ClIAcAm), diiodoacetamide (DIAcAm), and iodoacetamide (IAcAm)], are believed to be more toxic than their carbonaceous analogues [14]. By comparing the I-DBPs formed in oxidative treatments of I⁻-containing waters using chlorine [8,15–17], NH₂Cl [3,7], chlorine dioxide [18,19], ozone [20], and potassium permanganate [21], it turns out that NH₂Cl favors the formation of I-DBPs [4,22,23]. However, most previous studies on I-DBPs in drinking water have only focused on I-THMs, while little is known about the formation and speciation of I-HAAs and I-HAcAms during chloramination.

In this study, we systematically investigated the factors influencing the formation of three typical classes of I-DBPs (i.e., I-THMs, I-HAAs, and I-HAcAms) during chloramination of I⁻-containing waters, including reaction time, NH₂Cl dose, I⁻ concentration, pH, NOM concentration, Br⁻/I⁻ molar ratio, and water matrix. A kinetic model, which involved the reactions of NH₂Cl auto-decomposition, iodine species transformation and NOM consumption, was developed to describe NH₂Cl decay and HOI/I₂ evolution. Moreover, 18 model compounds (i.e., amino acids, carboxylic acids, and aromatic compounds) were chloraminated to help clarify the reaction mechanisms for I-DBPs formation.

2. Materials and methods

2.1. Chemicals

All chemicals were of the highest purity available. Individual I-THMs including bromodiiodomethane (CHBrI₂), bro-

mochloriodomethane (CHBrClI), chlorodiiodomethane (CHClI₂), dibromiodomethane (CHBr₂I) and dichloriodomethane (CHCl₂I), I-HAAs including bromoiodoacetic acid (BrIAA), chloroiodoacetic acid (ClIAA), diiodoacetic acid (DIAA) and iodoacetic acid (IAA), and I-HAcAms including bromoiodoacetamide (BrIAcAm), ClIAcAm, DIAcAm and IAcAm were all purchased from CanSyn Chem. Co. (New Westminster, BC, Canada). A standard mixture of four regulated THMs [THM4, including bromodichloromethane (CHCl₂Br), CHBr₃, CHCl₃ and dibromochloromethane (CHClBr₂)] and an internal standard of 1,2-dibromopropane were purchased from J&K Chemical Co. (Beijing, China). CHI₃ (99%), dichloroacetamide (DClAcAm, 99%), dichloroacetoneitrile (DClAN, 99%), iodoacetoneitrile (IAN, 99%), *o*-iodophenol (99.8%), *p*-iodophenol (99.8%) and triiodoacetic acid (TIAA, 90%) were obtained from Sigma Aldrich (St. Louis, MO, USA). Suwannee River NOM isolate (2R101N) was obtained from the International Humic Substances Society. Ammonium sulfate (99%), potassium bromide (99%), potassium iodide (99%) and NaOCl (5% of active chlorine) were obtained from Sinopharm Chemical Reagent Co. (Shanghai, China). In addition, 18 model compounds (Sinopharm Chemical Reagent Co.) were used to mimic the reactivity of a variety of NOM moieties during disinfection, whose chemical structures and molecular formulae are listed in Table S1. All reaction solutions were prepared with ultrapure water produced by a Milli-Q system (Advantage A10, Millipore, Billerica, MA, USA).

2.2. Experimental procedures

The formation of I-DBPs during chloramination of I⁻-containing waters was studied as a function of reaction time, NH₂Cl dose, I⁻ concentration, pH, NOM concentration, Br⁻/I⁻ molar ratio, and water matrix. All experiments were performed in triplicate using 40-mL amber glass bottles with polytetrafluoroethylene-lined septum screw-caps under headspace-free conditions. The pH of all reaction solutions was controlled at 7.0 ± 0.1 with 5 mM phosphate buffer except when the effect of pH was examined. Prior to each experiment, preformed NH₂Cl solution was freshly prepared by adding a desired amount of NaOCl stock solution to an (NH₄)₂SO₄ solution (10 mM bicarbonate buffer, pH 8.5) at a Cl/N molar ratio of 0.7:1 according to previously published procedures [24]. For comparison, a set of experiments using free chlorine was also conducted under the same conditions. Similar experimental procedures were adopted when testing the formation of I-DBPs during chloramination of the model compounds. After certain reaction times, water samples were withdrawn and added to an acidified sulfite solution ([HSO₃⁻]:[NH₂Cl] molar ratio = 1.5) to quench the residual oxidant. To prevent potential hydrolytic loss of byproducts during storage, water samples were extracted immediately after quenching.

2.3. Analytical methods

Reactive iodine species (i.e., HOI/I₂) in aqueous solutions were quenched with an excess of phenol and then analyzed as iodophenols by high-performance liquid chromatography (HPLC) as previously described [5]. Dissolved organic carbon (DOC) concentration was measured with a TOC-VCPN analyzer (Shimadzu, Japan). Br⁻, I⁻, and IO₃⁻ concentrations were analyzed by an

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