



# Identification, toxicity and control of iodinated disinfection byproducts in cooking with simulated chlor(am)inated tap water and iodized table salt



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## ABSTRACT

Chlorine/chloramine residuals are maintained in drinking water distribution systems to prevent microbial contamination and microorganism regrowth. During household cooking processes (e.g., soup making), the residual chlorine/chloramines in tap water may react with the iodide in iodized table salt to form hypoiodous acid, which could react with remaining natural organic matter in tap water and organic matter in food to generate iodinated disinfection byproducts (I-DBPs). However, I-DBPs formed during cooking with chloraminated/chlorinated tap water are almost completely new to researchers. In this work, by adopting precursor ion scan of  $m/z$  127 using ultra performance liquid chromatography/electrospray ionization-triple quadrupole mass spectrometry, many new polar I-DBPs formed during cooking with chloraminated/chlorinated tap water were detected and proposed with structures, of which 3-iodo-4-hydroxybenzaldehyde, 3-iodo-4-hydroxybenzoic acid, 3-iodo-4-hydroxy-5-methylbenzoic acid, diiodoacetic acid, 3,5-diiodo-4-hydroxybenzaldehyde, 3,5-diiodo-4-hydroxybenzoic acid, 2,6-diiodo-4-nitrophenol, 2,4-diiodo-6-nitrophenol, and 2,4,6-triiodophenol were confirmed with standard compounds. With the aid of ultra fast liquid chromatography/ion trap-time of flight-mass spectrometry, molecular formula identification of five new I-DBPs ( $C_8H_5O_4I$ ,  $C_7H_4NO_4I$ ,  $C_8H_5O_5I$ ,  $C_7H_4NO_5I$ , and  $C_8H_6O_3I_2$ ) was achieved. A developmental toxicity with a recently developed sensitive bioassay was conducted for the newly identified I-DBPs, suggesting that phenolic I-DBPs (except for iodinated carboxyphenols) were about 50–200 times more developmentally toxic than aliphatic I-DBPs. The major I-DBPs in a baseline simulated cooking water sample were determined to be from 0.72 to 7.63  $\mu\text{g/L}$ . Polar I-DBPs formed under various disinfection and cooking conditions were compared, and suggestions for controlling their formation were provided.

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## 1. Introduction

In drinking water distribution systems, chlorine residual is maintained to provide a primary barrier against microbial contamination from leaks/cross-connections/breakdowns and against microorganism regrowth/biofilm growth (White, 1999; Mays, 2000). Chlorine residual exists in two forms: free chlorine residual (i.e.,  $\text{HOCl}/\text{OCl}^-$ ) and combined chlorine residual (i.e., chloramines with  $\text{NH}_2\text{Cl}$  as the dominant species). Although free chlorine is more effective for disinfection than chloramines, chloramines are less reactive towards natural organic matter and form

less regulated disinfection byproducts (DBPs), and also they are more stable and persist longer in distribution systems. As a result, utilities have been increasingly switching to chloramines as a more reliable secondary disinfectant (Singer, 1994; Zhang et al., 2000; Krasner et al., 2006; Xie, 2004; Seidel et al., 2005; Hua and Reckhow, 2007; Bond et al., 2011; Tian et al., 2013; Chuang et al., 2013). The U.S. EPA has established guidance that disinfectant residual must be maintained in a public water distribution system and the running annual average of the measured free chlorine or chloramine residual in the distribution system must not exceed 4.0 mg/L as  $\text{Cl}_2$  (U.S. EPA, 2007). A higher chlorine residual in tap water may cause chlorinous taste and odor. In Europe, the chlorine residual in tap water can be much lower than 4.0 mg/L as  $\text{Cl}_2$ .

For human beings, iodine is essentially required by the thyroid

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gland to generate indispensable hormones. A low-iodine diet is the main cause of iodine deficiency, which will induce iodine deficiency disorders, such as stillbirth, cretinism, miscarriage as well as goiter (an abnormal thyroid gland enlargement) (Bleichrodt et al., 1980; Hetzel, 1983). To avoid health problems caused by iodine deficiency, programs on universal salt iodization are implemented in most countries with iodine deficiency (WHO, 2007; UNICEF, 2008). Table S1 in Supporting Information (SI) shows an incomplete list of countries (including 28 countries from Asia, Europe, North America, and Oceania) using iodized table salt (Mannar and Dunn, 1995; WHO, 2000; Delange et al., 2002; De Jong, 2004; ICCIDD, 2004; Dasgupta et al., 2008). Most countries used potassium iodide (KI) and potassium iodate ( $\text{KIO}_3$ ) as iodine fortifiers in table salt, and the iodization levels were at 5–77 mg I/kg salt, depending on the iodine contents in local water and food supplies in a region/country.

A cooking process (e.g., soup making) generally means that tap water, iodized table salt, flour/starch/meat, and seasonings/sauces/oil are mixed together in a cooking device, and then heated to boiling and kept boiling gently for a few minutes to a couple of hours. The iodized table salt in Canada might contain up to 77 mg KI as I per kg salt. If 1 L of tap water was dosed with 2 g of such salt, it would produce a water solution with an iodide level of 154  $\mu\text{g/L}$ , which is substantially higher than the iodide levels in raw water (usually below 10  $\mu\text{g/L}$ ). During cooking, the residual chlorine/chloramines in tap water may react with the iodide in iodized table salt to generate hypiodous acid (HOI), which could react with the remaining/decomposed natural organic matter in tap water and the organic matter in flour/starch/meat to form iodine-containing organic compounds. Since the iodine-containing organic compounds formed during cooking are assumed to result from tap water residual disinfectants, they are referred to as cooking water iodinated DBPs (I-DBPs).

Drinking water I-DBPs have drawing increasing concern in the past decade because they are generally of much higher cytotoxicity, genotoxicity, developmental toxicity, and growth inhibition than their bromo- and chloro-analogues (Plewa et al., 2004; Cemeli et al., 2006; Richardson et al., 2008; Yang and Zhang, 2013; Liu and Zhang, 2014). Four groups of I-DBPs have been previously reported to exist in drinking water worldwide, including iodinated trihalomethanes, aliphatic acids, haloacetamides, and haloacetoneitriles (Plewa et al., 2004; Krasner et al., 2006; Richardson et al., 2008; Chu et al., 2012; Luo et al., 2013; Wei et al., 2013). However, relative to drinking water I-DBPs, cooking water I-DBPs are almost completely new to researchers. Becalski et al. (2006) reported the formation of chloroiodoacetic acid and iodoacetic acid in simulated cooking water. This is the only report on cooking water I-DBPs that we are aware of in the literature, and the only two cooking water I-DBPs in the report were identified with gas chromatography/mass spectrometry (GC/MS). Since a majority of cooking water I-DBPs are suspected to be polar or highly polar that are hard to be detected by GC/MS, a reasonable choice for their detection should be liquid chromatography/MS.

Recently, drinking water polar I-DBPs have been fast selectively detected through a newly developed precursor ion scan (PIS) method (PIS 127) using electrospray ionization-triple quadrupole mass spectrometry (ESI-tqMS) (Ding and Zhang, 2009). Further through ultra performance liquid chromatography (UPLC)/ESI-tqMS analysis, structural information of new polar I-DBPs can be obtained. For some polar I-DBPs whose molecular formulae have different combinations, a high resolution mass spectrometry analysis using ultra fast liquid chromatography/ion trap-time of flight-mass spectrometry (UFLC/IT-TOF-MS) was applied to identify their molecular formulae through accurate  $m/z$  value determination.

This study was designed to reveal whole spectra of polar I-DBPs formed during cooking with chloraminated/chlorinated tap water and iodized table salt, and identify their structures/formulae using UPLC/ESI-tqMS and UFLC/IT-TOF-MS. Comparative developmental toxicity of newly identified I-DBPs to the embryos of *Platynereis dumerilii* was evaluated. This bioassay is a sensitive metric and has been successfully applied in determining the comparative developmental toxicity of dozens of individual DBPs in wastewater, chlorinated drinking water, and tap water (Yang and Zhang, 2013, 2014; Liu et al., 2015). Moreover, formation of polar I-DBPs under various disinfection and cooking conditions was examined.

## 2. Materials and methods

### 2.1. Materials

A NaOCl stock solution was diluted from a reagent grade NaOCl solution and titrated according to a standard procedure (APHA et al., 1995). Suwannee River natural organic matter (SRNOM, 1R101N) was purchased from the International Humic Substances Society. Wheat flour was purchased from a common local supermarket (Parkshop) in Hong Kong. Iodoacetic acid, 2,4,6-triiodophenol, and 3-methyl-4-hydroxybenzoic acid were purchased from Sigma–Aldrich. 3-Iodo-4-hydroxybenzaldehyde was purchased from Meryer Chemical Technology Corporation Limited. 3-Iodo-4-hydroxybenzoic acid was purchased from Apollo. Chloroiodoacetic acid and diiodoacetic acid were obtained from Orchid Cellmark. 2,6-Diiodo-4-nitrophenol, 3,5-diiodo-4-hydroxybenzoic acid, and 3,5-diiodo-4-hydroxybenzaldehyde were obtained from Alfa Aesar. Synthesis, isolation, and quantification of 3-iodo-4-hydroxy-5-methylbenzoic acid and 2,4-diiodo-6-nitrophenol were performed in the laboratory (SI and Figs. S1 and S2) since they did not have commercially available standard compounds. All other materials were purchased at reagent grade or higher from Merck or Sigma–Aldrich.

### 2.2. Simulated cooking water sample preparation

Simulated cooking water was prepared so that the whole experiment can be repeated by other researchers, which allowed for comparisons of DBPs formation between different labs, systems, and treatment processes. Simulated raw water was prepared by dosing 3 mg/L SRNOM as C and 90 mg/L  $\text{NaHCO}_3$  as  $\text{CaCO}_3$  to ultrapure water. To prepare simulated tap water with different types and levels of disinfectant residual, the simulated raw water was chlorinated or chloraminated (by dosing NaOCl or  $\text{NH}_2\text{Cl}$ ) in darkness with aimed disinfectant residual levels of 0.5, 1.0, 2.0, 3.0, and 4.0 mg/L as  $\text{Cl}_2$  after a contact time of 12 h.  $\text{NH}_2\text{Cl}$  was generated via a reaction of NaOCl and  $\text{NH}_4\text{Cl}$  at ammonia-to-chlorine molar ratio of 1.0:0.8 (Zhai et al., 2014) just before use. The relatively high disinfectant residual levels were applied for better detection of I-DBPs formed during cooking. To prepare a simulated cooking water sample, 1 L of the simulated tap water was transferred to a glass beaker and added with different amounts (2, 4, and 8 g) of iodized table salt (which was prepared by following the same dry method as that for the production of real iodized table salt, i.e., fully mixing 40 mg as I of powder KI or  $\text{KIO}_3$  and 0.50 kg of powder high-purity sodium chloride to avoid other impurities in table salt) and different amounts (0.002, 0.02, 0.05, 0.1, 0.2, 0.4, 0.5, and 0.8 g) of wheat flour. The relatively high amount of iodized table salt was dosed to amplify I-DBP formation during cooking. The mixture in the beaker was heated on a stirring hot plate with an aluminum foil cover to different temperatures (20, 50, 80, and 100 °C), and kept cooking at the specific temperature for different time lengths (10, 20, 30, 40, and 60 min). Different sequences (sequential addition of

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