



Spatial variations in the occurrence of potentially genotoxic disinfection by-products in drinking water distribution systems in China[☆]



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ABSTRACT

We investigated the occurrence of disinfection by-products (DBPs) with genotoxic potential in plant effluent and distribution water samples from four drinking water treatment plants in two Chinese cities using comprehensive two-dimensional gas chromatography–quadrupole mass spectrometry. We tested the samples for 37 DBPs with genotoxic potential, which we had previously identified and prioritized in water under controlled laboratory conditions. Thirty of these DBPs were found in the water samples at detection frequencies of between 10% and 100%, and at concentrations between 3.90 and 1.77×10^3 ng/L. Of the DBPs detected, the concentrations of 1,1,1-trichloropropan-2-one were highest, and ranged from 299 to 1.77×10^3 ng/L with an average of 796 ng/L. The concentrations of 6-chloro-2-N-propan-2-yl-1,3,5-triazine-2,4-diamine and 2,6-ditert-butylcyclohexa-2,5-diene-1,4-dione were also much higher, and ranged from 107 to 721 ng/L, and from 152 to 504 ng/L, respectively. Concentrations of 1,1,1-trichloropropan-2-one, 2-chloro-1-phenylethanone, 2,2-dichloro-1-phenylethanone and 6-chloro-2-N-propan-2-yl-1,3,5-triazine-2,4-diamine were highest at or near the treatment plants and decreased with increasing distance from the plants. Patterns in the concentrations of benzaldehyde, 2-phenylpropan-2-ol, and 1-methylnaphthalene differed between plants. The levels of DBPs such as 4-ethylbenzaldehyde, (E)-non-2-enal, and 1-phenylethanone were relatively constant within the distribution systems, even at the furthest sampling points ($20 \text{ km} < d < 30 \text{ km}$). A risk assessment showed that there was no risk to human health. It is, however, important to note that, because of limited availability of toxicity data, only five DBPs were evaluated in this study. The risks to health associated with exposure to the target potentially genotoxic DBPs should not be ignored because of their prolonged existence in drinking water.

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1. Introduction

There is increasing concern about the generation of disinfection by-products (DBPs) during water processing procedures because of their potential to cause adverse health effects (Nieuwenhuijsen et al., 2000; Richardson et al., 2007; Villanueva et al., 2004; Waller et al., 1998). Two classes of DBPs, trihalomethanes (regulated limit of 80 or 100 µg/L) and haloacetic acids (regulated limit of 60 µg/L), are currently regulated in many countries because of the potential health risks (EC, 1998; Richardson et al., 2007; USEPA,

2006). However, epidemiological studies of drinking water consumption have shown that the regulated DBPs cannot explain the observed conditions, such as bladder and colon cancer (Bull, 2006; Bull et al., 2001).

With the goal of identifying new DBPs that might be toxicologically relevant to the relationship between exposure to drinking water and elevated cancer risk, we previously developed a non-targeted screening method using comprehensive two-dimensional gas chromatography–quadrupole mass spectrometry (GC × GC-QMS) combined with OECD QSAR Toolbox Ver. 3.2 to allow unknown volatile and semi-volatile DBPs in drinking water to be identified and prioritized (Li et al., 2016). We found, from our earlier study, that 58 out of the 170 newly-identified DBPs were actual or potential genotoxicants. Some, including 1,1,1-trichloropropan-2-

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one, chloromethylbenzene, 1-bromopropan-2-one, benzaldehyde, 4-ethylbenzaldehyde, 1-phenylethanone, 2,6-ditert-butylcyclohexa-2,5-diene-1,4-dione, and 1-methylnaphthalene, were on the list compiled by Richardson (2011). Of these DBPs, 1,1,1-trichloropropan-2-one is the one most frequently detected in drinking water, and the concentration is usually at a low $\mu\text{g/L}$ level (Golfopoulos and Nikolaou, 2001; 2005; Golfopoulos et al., 2003; Serrano et al., 2014, 2015b). 1,1,1-Trichloropropan-2-one was included in the Information Collection Rule of the United States Environmental Protection Agency (USEPA, 1996) because of potential risks to human health (Jeong et al., 2012; Le Curieux et al., 1994). Chloromethylbenzene, that received the highest ranking for potential carcinogenicity (Woo et al., 2002) and was not already included in the Information Collection Rule, was selected for the US Nationwide DBP Occurrence study (Weinberg et al., 2002). However, there have been very few studies of the occurrence of these previously-reported DBPs in drinking water, let alone the newly-detected DBPs with genotoxic potential.

The main aim of this study, therefore, was to find out if the DBPs with genotoxic potential that we identified in our previous study were present in actual drinking water samples. We tested plant effluent and distribution water samples from four drinking water treatment plants (DWTPs) in two Chinese cities. We examined how the concentrations of the detected DBPs varied within distribution systems, and we also estimated the risks to human health.

2. Materials and methods

2.1. Materials

Thirty-seven commercially available DBP standards were listed in the Supporting Information (SI) Table S1, and were purchased from AccuStandard (New Haven, CT, USA), Alfa Aesar (Ward Hill, MA, USA), TCI (Tokyo, Japan), J&K Scientific (Beijing, China) and Acros (Belgium). High-performance liquid chromatography-grade methyl *tert*-butyl ether (MTBE) was purchased from Thermo Fisher Scientific (Waltham, MA, USA). Ultrapure water (18.2 M Ω cm) was prepared by a Milli-Q purification system (Millipore, Billerica, MA, USA). Anhydrous sodium sulfate (99.5%, guaranteed reagent, Sinopharm Chemical Reagent, Beijing, China) was heated at 450 °C for at least 4 h, allowed to cool naturally, and then stored in desiccators.

2.2. Sampling

Plant effluent and distribution water samples were collected from four DWTPs in May 2016. Three DWTPs (A, B, and C) were investigated in City L, and one (D) in City S (B and C represent two treatment processes using the same source water). A total of 40 distribution water samples (20 samples in each city) were collected from all the treatment plants. Six distribution water samples were collected from Plant A (about 1–3 km from the treatment plant). Eight distribution water samples were collected from Plant B (about 1–30 km from the treatment plant). Six distribution water samples were collected from Plant C (about 3–20 km from the treatment plant). Twenty distribution water samples were collected from Plant D (about 1–10 km from the treatment plant). The sampling locations are shown in SI Figs. S1–S2, and water treatment scheme of each plant is described in SI Table S2. One water sample was collected in each sampling point. For samples from the distribution systems, taps were run for 5 min to eliminate stagnant water from pipes and ensure that distribution system water was being sampled. All water samples were filtered through a 0.45- μm glass fiber filter (Millipore, Billerica, MA, USA) before analysis. The basic water quality parameters of water samples are summarized in SI

Table S3.

2.3. Extraction procedure

Procedures for water sample pretreatment were carried out according to our previous study (Li et al., 2016). Briefly, a 2 L of each water sample was concentrated by passing it through a 100 mg Supelclean LC-C18 solid phase extraction cartridge (Supelco, Bellefonte, PA, USA) coupled to a 500 mg Oasis HLB cartridge (Waters, Milford, MA, USA). The cartridges were conditioned before use with 10 mL each of MTBE, methanol and water. The sample was passed through the tandem cartridges at a flow rate of 3–5 mL/min. The analytes absorbed onto the solid phase extraction cartridges were eluted using 10 mL of MTBE, and then the eluents were dried over using anhydrous sodium sulfate. Finally, the organic eluents were collected and concentrated to ca. 100 μL under high-purity nitrogen, and reconstituted with *n*-hexane to 500 μL for GC \times GC-qMS analysis.

2.4. Analytical methods

Dissolved organic carbon (DOC) analyses were carried out using a Teledyne Tekmar TOC Fusion analyzer (Mason, Ohio, USA) according to Standard Method 5310 B (APHA, 1998). The ultraviolet absorbance at 254 nm (UV_{254}) was measured by a HACH DR5000 UV spectrophotometer (HACH, Loveland, CO, USA). The inorganic anions (Cl^- , Br^- , NO_2^- , NO_3^- , and SO_4^{2-}) were determined using a Dionex ICS-1500 ion chromatography system (Sunnyvale, CA, USA) following the method described in Standard Method 4110B (APHA, 1998). The chlorine and chloramine residual were measured using a HACH Pocket Colorimeter™ II (HACH, Loveland, CO, USA).

The method used to analyze the DBPs in the drinking water samples were based on the method we developed in a previous study (Li et al., 2016). Briefly, DBP analyses were carried out using an Agilent 7890A gas chromatograph instrument (Agilent Technologies, Palo Alto, CA, USA) equipped with an Agilent 5975C electron ionization quadrupole mass spectrometer instrument (Agilent Technologies, Palo Alto, CA, USA) and a ZX-2 LN2 cooled-loop thermal modulator (Zoex, Lincoln, NE, USA). Modulation was carried out every 8 s and the duration of the hot pulse was 400 ms. The first-dimension column was a DB5-MS (30 m \times 0.25 mm \times 0.25 μm ; Agilent Technologies, Palo Alto, CA, USA), and the second-dimension column was a BPX 50 (2 m \times 0.1 mm \times 0.1 μm ; SGE, Melbourne, Australia). High-purity helium (99.999%) was used as the carrier gas in constant flow mode at 1 mL/min. Sample injection of 1 μL was made in splitless mode, and the injector temperature was set at 280 °C. The temperature program began at 40 °C with a hold time of 3 min, and then increased at 2.5 °C/min to 260 °C with a hold time at 260 °C for 5 min. The electron impact ion source temperature was set to 200 °C. Mass spectra were collected from m/z 45–350 at 20 spectra/s after a solvent delay of 8 min. Data processing was conducted using the GC Image 2.1 software (Zoex, Lincoln, NE, USA). Identification of compounds was confirmed using the authentic standards (SI Table S1).

2.5. Human health risk assessment

Human health risk assessment is based on the assumption that humans may be exposed to organic pollutants in water mainly through ingestion and dermal absorption (USEPA, 1989, 1997). In this study, non-cancer hazard quotients (HQs) and lifetime cancer risk (LCR) were assessed by calculating the exposure dose (D) of selected DBPs through the ingestion pathway (USEPA, 1989, 2008). The following equations were used for estimating the D, LCR and

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