



Spatial and temporal evaluations of disinfection by-products in drinking water distribution systems in Beijing, China

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ARTICLE INFO

Article history:

Received 26 February 2010

Received in revised form 17 June 2010

Accepted 19 June 2010

Keywords:

Drinking water

Disinfection by-products

Spatial and seasonal variations

ABSTRACT

Disinfection by-products were determined in 15 water treatment plants in Beijing City. The effects of different water sources (surface water source, mixture water source and ground water source), seasonal variation and spatial variation were examined. Trihalomethanes and haloacetic acids were the major disinfection by-products found in all treated water samples, which accounted for 42.6% and 38.1% of all disinfection by-products respectively. Other disinfection by-products including haloacetonitriles, chloral hydrate, halo ketones and chloropicrin were usually detected in treated water samples but at lower concentrations. The levels of disinfection by-products in drinking water varied with different water sources and followed the order: surface water source > mixture water source > ground water source. High spatial and seasonal variation of disinfection by-products in the drinking water of Beijing was shown as a result.

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1. Introduction

Chlorination has been the most common disinfectant process for domestic drinking water for many years in Beijing. Chlorination could dramatically reduce the incidence of waterborne diseases such as typhoid, cholera, and hepatitis, as well as gastrointestinal illness (Morris and Levine, 1995). However, chlorine can also react with natural materials in the raw water to form disinfection by-products (DBPs) that are hazardous to health (White, 1992; WHO, 2004; Uyak et al., 2008). Some epidemiologic studies (IARC, 1991; WHO, 1996; Singer, 1999; Magnus et al., 1999; Nieuwenhuijsen et al., 2008) have shown an association between long-term exposure to disinfection by-products and increased risk of cancer and potential adverse reproductive effect. Trihalomethanes (THMs) and haloacetic acids (HAAs) are the most important groups of DBPs in chlorinated finished water. Others are haloacetonitriles (HANs), chloral hydrate (CH), halo ketones (HKs) and chloropicrin (CP). THMs include chloroform, bromodichloromethane (BDCM), dibromochloromethane (DBCM) and bromoform. Total THMs (TTHM) refer to the sum of these four substances above. HAAs include nine substances, dichloroacetic and trichloroacetic acids were the most common ones among them, while the other compounds were found generally at lower levels. In the US, the regulated haloacetic acids, known as HAA5, are: monochloroacetic acid (MCAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), monobromoacetic acid (MBAA), and dibromoacetic acid (DBAA). HANs include dichloroacetonitrile (DCAN), bromochloroacetonitrile (BCAN), dibromoacetonitrile (DBAN) and trichloroacetonitrile (TCAN). The sum of these HANs was defined as

total HANs (THAN). HKs include 1,1-dichloropropane (1,1-DCP) and trichloropropane (1,1,1-TCP) and the total HKs (THK) were regarded as the sum of the above two halo ketones.

Beijing, the capital of China, is located in the Haihe river basin. It is of the semi-arid and semi-humid continental monsoon climate, which inherits its less precipitation. Its average precipitation is only 585 mm (Pan, 2006). The water from Miyun and Guanting Reservoirs, two main surface water sources for the municipality, provides two-thirds of the city's surface water supply, with half of which coming from Miyun Reservoir. The groundwater is also used as a raw water supply for Beijing City. There are many water treatment plants in Beijing City. These plants use surface water, ground water and mixture water (surface water mixed with ground water) as their water sources.

In Beijing, the main focus of the disinfection by-products is trihalomethanes (THMs). Only limited researches are conducted on other DBPs such as haloacetic acids, haloacetonitriles, halo ketones, chloropicrin and chloral hydrate. In order to understand the consequences of the future application of DBPs and to monitor and control the DBPs of drinking water in Beijing, a survey of disinfection by-product occurrence in Beijing city was conducted at 15 drinking water treatment plants for the first time. Particular attention was focused on the distribution of the levels of DBPs with an emphasis on their seasonal and spatial evolutions in a water distribution system.

2. Material and methods

2.1. Sample collection

Water samples used in this study were collected from the treatment plants in Beijing cities. Sampling sites were shown in Fig. 1. A total of 15

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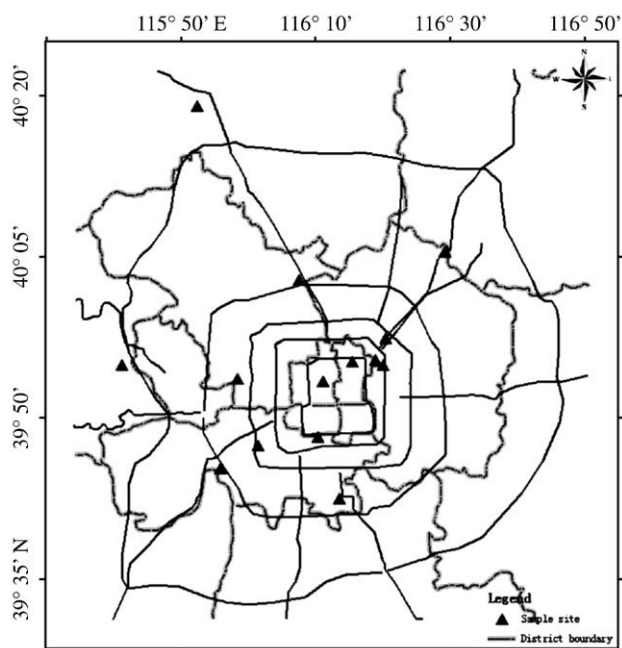


Fig. 1. Selected sampling sites in Beijing City.

treatment plants were sampled in spring, summer, autumn and winter. For all water treatment plants, four samples (raw water, finish water, water between the treatment plant and the system extremities and water at the extremity point of the distribution system) were selected for the purposes of water quality parameters and DBP measurements. Because most of tap waters come from the water supply network in Beijing, a group of the latter two samples could be shared by two or more water treatment plants. Only 7 groups (56 samples) of these two samples were selected. And a total of 176 samples were collected for all the treatment plants. The selection of sampling points was selected from the monitor points of water quality provided by the Beijing Centers for Diseases Control and Prevention. The four THM species and the five regulated HAAs were analyzed. Other DBPs including haloacetonitriles (HANs), chloral hydrate (CH), halo ketones (HKs) and chloropicrin (CP) were also determined.

2.2. Analytical method

Measurements of free chlorine were conducted using the DPD titrimetric method (Standard method 4500-Cl-F) with a DR-700 colorimeter from Hach. Total organic carbon (TOC) was analyzed using a Shimadzu TOC analyzer (model 5000). Water pH and temperature were measured on site using a solid selective electrode (electrolytic gel). The USEPA Method 551.1 (USEPA, 1998) was used to determine the THMs, HANs, HKs, CH and CP. After the liquid–liquid extraction, analyses of these DBPs were performed by gas chromatography using a Varian Vista 6000 GC equipped with an electron capture detector, an on-column injector and a J&W DB-5 capillary column. The HAAs were analyzed following derivatization with acidic methanol using a micro liquid–liquid extraction gas chromatographic method based on EPA Method 552.2 (USEPA, 1995).

2.3. Quality assurance and quality control

In order to monitor the precision and reliability of analytical results, no less than 50% replicate samples were examined in DBP analysis. Field blanks, which are accompanied with samples to the sampling sites were used to determine any background contamination.

Method blanks and spiked blanks (standards spiked into solvent) were analyzed and were subtracted from the analytical results to remove the contribution of contamination in laboratory. The result of recovery was shown in Table 1.

3. Results and discussion

3.1. Descriptive statistics

Table 2 listed the TOC, residual chlorine, water temperature and pH in raw water and treated water from water treatment plants in Beijing City. The TOC level as a surrogate of a DBP precursor was not so high, ranging from 0.55 to 3.47 mg/L. The variation of the mean value of TOC level was winter > autumn > spring > summer in raw water. It could be explained by the flush period of Beijing. Most of the rainfall concentrates in summer, which occupies 85% of the annual rainfall (Pan, 2006). However, it seldom rains in the winter in Beijing. The TOC concentrations decreased with an increase of rainfall due to dilution by rainwater. The water temperature exhibited a strong seasonal variation as: summer > autumn > spring > winter. The concentration of residual chlorine ranged from 0.05 to 0.80 mg/L in the treated water. The pH value of water ranged from 6.82 to 8.55, and was higher in autumn than in other seasons.

The concentrations of DBPs in water samples were summarized in Table 3. The means and medians were calculated in order to illustrate what the central tendency for each compound concentration could be. Trihalomethanes (THMs) and haloacetic acids (HAAs) were the major disinfection by-products found in all treated water samples, which accounted for 42.6% and 38.1% of all DBPs respectively. TCM was the most abundant THMs with a concentration range from not detected to 29.41 µg/L. DCAA and TCAA were the main components of HAAs. The highest DCAA and TCAA levels were 13.02 µg/L and 20.10 µg/L, respectively. MCAA and MBAA were not detected in any samples. Other DBPs including halogenated acetonitriles (HANs), halogenated ketones (HKs), chloral hydrate (CH) and chloropicrin (CP) were usually detected in treated water samples but at lower concentrations. Concentrations of HANs were found to be much lower than THMs and HAAs. The mean THAN concentration of 2.69 µg/L was similar to the concentrations measured in Melbourne, Australia (2–7 µg/L, Simpson and Hayes, 1998), but was lower than IZMIR, Turkey (0.25–88.4 µg/L, Baytak et al., 2008). The mean concentrations of 1,1-DCP and 1,1,1-TCP were 1.27 µg/L and 0.62 µg/L. The CH and CP levels were very low, and the mean concentrations of them were 0.93 µg/L and 0.31 µg/L, respectively.

Table 1
Descriptive statistics of the result of quality control.

| Group | Compounds | Recovery (n = 6) | | |
|-------|-----------|------------------|---------|----------------|
| | | Spike (µg/L) | RSD (%) | Recoveries (%) |
| THMs | TCM | 20 | 4.9 | 75.1–115 |
| | BDCM | 20 | 3.6 | 87.2–115 |
| | DBCM | 20 | 4.7 | 81.2–117 |
| | TBM | 20 | 3.1 | 85.7–116 |
| HAAs | MCAA | 50 | 7.2 | 74.4–93.3 |
| | MBAA | 30 | 9.6 | 88.7–103 |
| | DCAA | 30 | 6.0 | 96.9–111 |
| | TCAA | 30 | 3.5 | 95.7–114 |
| | DBAA | 30 | 4.1 | 99.8–112 |
| HANs | TCAN | 20 | 5.6 | 71.0–108 |
| | DCAN | 20 | 5.7 | 79.2–122 |
| | BCAN | 20 | 4.8 | 75.1–115 |
| | DBAN | 20 | 3.4 | 80.1–123 |
| HKs | DCP | 20 | 6.4 | 72.8–116 |
| | TCP | 20 | 5.4 | 88.2–121 |
| CH | CH | 20 | 5.7 | 73.1–105 |
| CP | CP | 20 | 4.5 | 76.1–96.1 |

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