Formation of disinfection byproducts in typical Chinese drinking water

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Abstract

Eight typical drinking water supplies in China were selected in this study. Both source and tap water were used to investigate the occurrence of chlorinated disinfection byproducts (DBPs), and seasonal variation in the concentrations of trihalomethanes (THMs) of seven water sources was compared. The results showed that the pollution level for source water in China, as shown by DBP formation potential, was low. The most encountered DBPs were chloroform, dichloroacetic acid, trichloroacetic acid, and chlorodibromoacetic acid. The concentration of every THMs and haloacetic acid (HAA) compound was under the limit of standards for drinking water quality. The highest total THMs concentrations were detected in spring.

Key words: disinfection byproducts (DBPs); haloacetic acids; trihalomethanes

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Introduction

Drinking water safety has been a key focus of modern society. Disinfection of drinking water was introduced to ensure that our water supply was microbiologically safe through the removal of pathogens to prevent waterborne disease transmission. The use of chlorination was first introduced more than 100 years ago and is still the most widely used disinfectant due to its low cost, easy acquisition and persistent effect (Golfinopoulos and Nikolaou, 2005). Increasing research on disinfection byproducts (DBPs) has, however, raised significant health concerns. Trihalomethanes (THMs) were firstly identified in 1974 (Rook, 1974). Additionally, in 1976 the National Cancer Institute (USA) discovered that chloroform is carcinogenic to animals (National Cancer Institute, 1976). In 1980, haloacetic acids (HAAs) were identified and classified as another important group of chlorinated DBPs (Quimby et al., 1980). Approximately 600–700 DBPs have been reported (Krasner et al., 1989), with THMs and HAAs accounting for more than 80% of the total amount.

To ensure drinking water safety, many countries developed guidelines or standards to regulate and limit the concentration of DBPs in drinking water. China implemented the new “Standards for Drinking Water Quality” in 2006, which increased the number of DBP related items from one to thirteen compared (GB5749-2006) to

the 1985 edition. Despite this, limited DBP research has been carried out in China on either the source or tap water, especially in relation to the impact of rapid economic development and environmental pollution. Our research selected eight typical source water supplies from different locations in China to investigate the occurrence of chlorinated disinfection byproducts (CDBPs) in both source water and corresponding drinking water (after treatment).

This study is important in terms of standardizing sampling protocols and chlorination conditions, and to establish a link between water qualities parameters and DBP formation. The information obtained will enhance the future development of raw water source selection and classification of sources with different water qualities and treatments required for the goal of controlling CDBPs in drinking water. While similar surveys from other countries have been reported in the literature, this is the first study of its kind on Chinese water sources. This study also compared water quality in China with developed countries.

1 Materials and methods

1.1 Water systems and sample collection

Eight water sources, BJ, XHD, XS, NT, NSBD, DY, SC and MPS (Fig. 1), from five major river systems in China, Pearl River, Qiantang River, Yangtze River, Yellow River, and Songhua River, were selected for this survey. Eight source water samples were collected in January 2009. The

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Fig. 1 Maps of sampling locations.

BJ and XHD samples were collected from Guangdong Province in south China, XS sample was collected from the Qiantang River, the NT sample was collected from Jiangsu Province, the NSBD sample was collected in Shijiazhuang from the South-to-North Water Diversion Project, the DY sample was collected in the estuary of the Yellow River, and the SC and MPS samples were collected from Harbin City in northeast China.

Treated water sampling of the corresponding treatment plant was performed in autumn (November in 2008), winter (January in 2009), and spring (April in 2009). All seven plants used conventional treatment processes with chlorine as the disinfectant. Plant chlorine doses were determined based on network chlorine residual performance. Samples were collected at the product storage tank (24 hr retention time). As the typical detention time for Chinese distribution systems is approximately 36 hr, samples collected after a 24 hr detention time was considered a good representation of the sample at the customer taps in the distribution system. The sampling bottles were pre-dosed with sodium hyposulfite to quench residual chlorine.

The collected samples were delivered to the laboratory immediately. All samples were stored in amber bottles at 4°C to minimize constitution changes before analysis.

1.2 Materials

Four species of THMs (chloroform (CF), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform (BF)), were purchased from AccuStandard (USA) with purity above 98%. Nine species of HAAs, (monochloroacetic acid (MCAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), monobromoacetic acid (MBAA), dibromoacetic acid (DBAA), tribromoacetic acid (TBAA), chlorodibromoacetic acid (CDBAA), bromochloroacetic acid (BCAA), and bromodichloroacetic acid (BDCAA)) and internal standard 1,2-dibromopropane were also purchased from AccuStandard with purity above 96%. Methanol, methyl-tert-butyl ether (MTBE), and hexane were of HPLC grade. Guaranteed reagent grade sodium sulfite anhydrous was heated at 450°C for three hours; acidic methanol was freshly prepared. Stock solutions of chlorine (3000 mg/L) were prepared from 5% sodium hypochlorous (NaClO) and periodically measured with DPD (N,N-diethyl-*p*-phenyldiamine) spectrophotometry (State Environmental Protection, 2002). Phosphate buffer

solution (50 mmol/L KH_2PO_4 -NaOH, pH 7.0) was prepared using MilliQ water (18.2 Ω/cm).

All glass vials were washed with detergent, rinsed with tap water, immersed in 15% chromic acid lotion for 24 hr, and then rinsed with ultra pure chemical free MilliQ water three times. Finally, these glass vials were heated in an oven at 500°C for 4 hr. Glass apparatus with scales were washed with detergent and chromic acid lotion then rinsed with deionized water and dried at room temperature.

1.3 Chlorine disinfection

Chlorination experiments were carried out in glass vials with polypropylene screw caps and Teflon-lined septa. Source water samples were filtrated with 0.45 μm cellulose acetate membranes to eliminate suspended solids before chlorination. The pH of all samples was kept at 7.0 ± 0.2 with phosphate buffer solution. The stock chlorine solution was then added to the samples at differing doses (mg/L) BJ 7.50, XHD 33.75, XS 8.13, NT 12.50, NSBD 8.13, DY 6.88, SC 15.75, and MPS 11.23). The selected chlorine dose, determined by Standard Methods (APHA, 1998), ensured the presence of a measurable residual in all samples after the entire chlorination period. Finally, the samples were kept at 25°C in the dark. After a contact time of 24 hr, all samples were dechlorinated with sodium sulfate and the concentrations of THMs and HAAs were measured immediately.

1.4 Analytical methods

Water quality parameters including pH, TOC, and UV_{254} were analyzed in accordance with Standard Methods (APHA, 1998). Dissolved organic carbon (DOC) was detected with a TOC analyzer (TOC-V_{CPH}, Shimadzu, Japan); pH was measured with a pH meter (MP220, Mettler Toledo, Switzerland); and UV absorbance at 254 nm (UV_{254}) was determined with a spectrophotometer (UV-Vis8500, Tianmei, China).

The THMs and HAAs were analyzed by gas chromatography (Agilent 6890N, USA) with the electron capture detector (ECD) according to US EPA methods 551.1 and method 552.3 (US EPA 1990, 2003). The temperature program was (1) THMs: detector, 250°C; injector, 200°C; initial column temperature 35°C for 4 min, then increased to 260°C by 20°C/min, and hold 4 min; (2) HAAs: detector, 290°C; injector, 250°C; initial column temperature 35°C for 4 min, then increased to 70°C by 2°C/min. The

injection volume was 1 μL .

2 Results and discussion

2.1 Water quality parameters

Samples were analyzed for water quality parameters, including the concentrations of DOC, UV_{254} , specific UV absorbance (SUVA), pH, and disinfection byproducts. The results are shown in Table 1.

Dissolved organic carbon (DOC) concentration can be used to identify the level of pollution and the concentration of organic carbon in water, and is also a key factor for forecasting disinfection byproducts formation. Table 1 shows that the concentration of DOC in raw water varied from 1.3 to 4.9 mg/L (BJ was the lowest and XHD was the highest), which were generally considered as low, while DOC in drinking waters ranged from 1.2 to 4.4 mg/L (BJ was the lowest and XHD was the highest). It is worth noting that the MPS samples showed the largest difference of 46% after treatment and the DOC concentrations in the raw water samples collected in the south were lower than those in the north. Measuring UV_{254} shows the amount of aromatic chemicals or conjugated chemicals in water and is very important in forecasting levels of natural organic materials, such as humic acid (Bolto et al., 1999). Our data show that UV_{254} in the eight raw water samples ranged from 0.028 to 0.116 cm^{-1} (NSBD was the lowest and MPS was the highest), while drinking water ranged from 0.016 to 0.076 cm^{-1} . Specific UV Absorbance ($100 \times \text{UV}_{254}/\text{DOC}$) is defined as the proportion of aromatic organic carbon or conjugated chemicals to total organic chemicals in water, therefore it is a common indication parameter of the aromatic contents of water samples and is often represented as the precursors of DBPs in water (Ates et al., 2007). The SUVA from the eight raw water samples ranged from 1.0 to 2.9 L/(cm·mg), with NSBD the lowest and NT the highest. The pH of the raw water samples

varied from 7.3 to 8.2 and from 7.0 to 7.8 in drinking water. There was no significant difference in the pH among these samples.

2.2 Concentrations of DBPs

The total concentrations of THMs (TTHMs) were measured for seven of the eight drinking waters (sample for NSBD was not analyzed because this sample was not collected) and ranged from 3.2 to 42.4 $\mu\text{g/L}$, with XHD the lowest and DY the highest (Table 2). The concentrations of the THMs for all drinking water samples were below the guideline values from the “Standards for Drinking Water Quality” (GB5749-2006), namely the concentration of all four THMs divided by their regulated values were less than 1 as following equation.

$$\text{GB} = \frac{C_{\text{CF}}}{60} + \frac{C_{\text{DCBM}}}{60} + \frac{C_{\text{CDBM}}}{100} + \frac{C_{\text{BF}}}{100}$$

The concentrations were also below the 80 $\mu\text{g/L}$ safe level set by the US EPA Disinfectant/Disinfection Byproduct Rule (I) (US EPA, 1998). The concentrations of THMs in raw water samples varied from 68.3 to 295.3 $\mu\text{g/L}$, with BJ the lowest, MPS the highest, and CF the dominant compound, accounting for up to 81% of total THMs. The distribution of THMs species was $\text{CF} > \text{DCBM} > \text{DBCM} > \text{BF}$, which was consistent with previous work (Golfonopoulos and Nikolaou, 2005; Deng et al., 2008).

According to Table 3, the total concentrations of five HAAs (THAAS: MCAA, DCAA, TCAA, MBAA, and DBAA) in raw water ranged from 88.7 to 654.0 $\mu\text{g/L}$ (XS was the lowest and MPS was the highest), while the total concentrations of nine HAAs (THAA9) ranged from 282.8 to 1154.4 $\mu\text{g/L}$ (XS was the lowest and XHD was the highest). The sum of CDBAA, DCAA, and TCAA amounted for more than 79% for all nine HAAs. The total concentrations of the five HAAs in drinking water ranged from 29.6 to 86.8 $\mu\text{g/L}$ (BJ was the lowest and MPS was the highest), while the total concentration of nine HAAs

Table 1 Water quality parameters of raw waters and corresponding drinking waters

Sampling points	pH	UV_{254} (cm^{-1})	DOC (mg/L)	SUVA (L/(cm·mg))	Br^- ($\mu\text{g/L}$)	TTHM ($\mu\text{g/L}$)	THAA5 ($\mu\text{g/L}$)	THAA9 ($\mu\text{g/L}$)	THM/DOC ($\mu\text{g/mg}$)	THAA5/DOC ($\mu\text{g/mg}$)	THAA9/DOC ($\mu\text{g/mg}$)
Raw water											
BJ	7.8	0.031	1.3	2.4	20.3	68	97	409	52	74	310
XHD	7.3	0.094	4.9	1.9	88.0	252	467	1154	52	96	236
XS	7.3	0.037	1.6	2.4	59.6	82	89	283	53	57	183
NT	7.9	0.051	1.8	2.9	32.5	130	182	313	72	101	174
NSBD	8.0	0.028	2.7	1.0	86.6	109	89	372	41	33	139
DY	8.2	0.080	4.7	1.7	50.0	165	162	848	35	34	180
SC	7.8	0.082	4.5	1.8	6.6	229	489	558	51	109	124
MPS	7.4	0.116	4.4	2.6	5.6	295	654	673	67	148	152
Drinking water											
BJ	7.3	0.017	1.2	1.4	4.9	16	30	112	13	25	93
XHD	7.5	0.076	4.4	1.7	45.3	3	59	129	1	13	29
XS	7.2	0.016	1.5	1.0	36.8	15	50	76	10	33	50
NT	7.5	0.040	1.7	2.3	26.1	4	48	85	2	28	50
NSBD	—	—	—	—	—	—	—	—	—	—	—
DY	7.8	0.040	3.3	1.2	59.1	42	30	106	13	9	32
SC	7.0	0.047	3.3	1.4	0	13	72	99	4	22	30
MPS	7.6	0.027	2.4	1.1	0	23	87	110	10	37	46

— No sample.

Table 2 Concentration of THMs in raw and drinking water samples (unit: $\mu\text{g/L}$)

Sampling points	Raw water					Drinking water					
	CF	BDCM	DBCM	BF	TTHMFP	CF	BDCM	DBCM	BF	TTHM	GB
BJ	60.1	9.9	1.8	ND	68.3	12.2	3.6	2.2	0.4	16.3	0.288
XHD	207.5	51.5	11.7	0.8	252.0	1.9	1.1	0.8	ND	3.2	0.058
XS	56.1	26.9	9.3	1.0	81.5	6.5	7.2	5.1	1.3	15.4	0.293
NT	111.0	21.9	4.1	0.3	129.6	2.6	1.2	0.9	ND	4.0	0.072
NSBD	68.9	40.0	17.9	1.9	109.3	—	—	—	—	—	—
DY	94.1	43.8	24.6	3.1	165.6	19.5	18.6	14.0	2.7	42.4	0.802
SC	218.0	14.4	1.6	ND	229.5	12.6	1.1	ND	ND	13.4	0.228
MPS	290.9	5.4	0.8	ND	295.3	20.5	3.4	0.4	ND	23.2	0.402

ND: not detected; —: no sample.

Table 3 Concentrations of HAAs in raw and drinking water samples (unit: $\mu\text{g/L}$)

Sampling points	Raw water										
	MCAA	CDBAA	MBAA	DCAA	TBAA	TCAA	CBAA	DBAA	BDCAA	THAA5	THAA9
BJ	6.2	268.5	4.7	33.0	14.9	46.1	17.0	6.9	11.7	96.9	409.0
XHD	57.2	490.7	14.6	155.7	7.9	215.1	141.4	24.7	47.1	467.3	1154.4
XS	8.9	122.5	1.6	29.6	4.7	44.3	37.4	3.9	29.5	88.7	282.8
NT	6.1	64.9	1.1	73.2	1.8	100.3	43.5	1.6	20.6	182.3	313.1
NSBD	10.7	212.9	2.1	37.2	8.1	33.6	43.3	5.2	18.7	88.8	371.8
DY	15.9	566.6	6.5	57.3	46.6	69.1	73.2	13.0	31.4	162.0	848.2
SC	17.5	18.8	0.7	119.8	0.8	350.4	33.1	1.2	15.9	489.5	558.0
MPS	20.5	4.6	0.4	144.1	ND*	488.5	ND	0.6	14.3	654.0	672.8

Sampling points	Drinking water										
	MCAA	CDBAA	MBAA	DCAA	TBAA	TCAA	CBAA	DBAA	BDCAA	THAA5	THAA9
BJ	ND	36.5	0.5	3.2	5.2	21.4	19.3	4.5	21.3	29.6	111.8
XHD	ND	7.0	1.6	5.6	6.8	30.9	39.5	20.9	17.1	59.0	129.3
XS	3.0	13.1	35.5	0.3	3.3	10.6	ND	0.9	9.6	50.3	76.3
NT	ND	5.8	1.7	3.2	6.0	31.2	10.4	11.6	15.7	47.6	85.4
NSBD	—	—	—	—	—	—	—	—	—	—	—
DY	4.6	26.0	0.2	1.6	4.9	19.2	28.2	4.3	16.9	29.9	105.8
SC	55.3	3.6	1.1	5.7	5.9	8.4	6.7	1.5	11.4	71.8	99.4
MPS	50.8	3.4	1.1	10.9	1.2	23.2	5.7	0.8	12.7	86.8	109.8

ND: not detected; —: no sample.

varied from 76.3 to 129.3 $\mu\text{g/L}$ (XS was the lowest and XHD was the highest). HAA concentration decreased after water plant treatment. According to the “Standards for Drinking Water Quality” (GB5749-2006), the guideline value for DCAA and TCAA is 50 $\mu\text{g/L}$ and 100 $\mu\text{g/L}$, respectively. While all analyzed drinking water samples were below these limits, total concentration of the five HAAs in the drinking water of SC and MPS exceeded the maximum contaminant level of 60 $\mu\text{g/L}$ proposed by the Disinfectant/Disinfection Byproduct Rule (I) of US EPA (1998).

Numerous studies have investigated the DBPs levels all over the world. An analysis of THMs and HAA5 in 31 drinking water samples in Finland showed a large fluctuation, ranging from 6.2 to 265.5 $\mu\text{g/L}$ and from 1.15 to 102.8 $\mu\text{g/L}$, respectively (Nissinen et al., 2002). Research on drinking water has shown that the concentration of THMs and THAA5 in Greek drinking water were below 50 and 70 $\mu\text{g/L}$, respectively (Golfonopoulos and Nikolaou, 2005), the mean concentration of THMs in Canadian drinking water was 13.3 $\mu\text{g/L}$ (Koudjonou et al., 2008), and the mean concentration of THMs in Australian drinking water was below 200 $\mu\text{g/L}$, relatively high as THM concentrations prior to treatment were from 400 to 600 $\mu\text{g/L}$ (Drikas et al., 2008). In our research, the THMs and THAA5 in drinking waters were from 3.2

to 42.4 $\mu\text{g/L}$ (Table 2) and 29.9 to 86.8 $\mu\text{g/L}$ (Table 3), respectively, while the THM formation potential in source waters ranged from 68 to 295 $\mu\text{g/L}$ (Table 1). Compared to the countries listed above, chlorinated DBPs pollution in China appears to be in relatively good condition.

According to the analysis above, DBPs levels in Chinese drinking waters are within regulated values. The results also demonstrate that China's current water plants control DBPs by removing DBP precursors to ensure drinking water safety.

2.3 Relationship between UV_{254} and THMs, HAAs

In Fig. 2, the correlation between UV_{254} and DBPs in raw waters was high ($R^2 > 0.8$), while the correlation between DOC, SUVA, and DBPs was very low (correlation efficient less than 0.1). It would appear, therefore, UV_{254} is more representative of the amount of disinfection by-product precursor in raw water.

As the Disinfectant/Disinfection Byproduct Rule (I) of US EPA (1998) stipulates maximum contaminant level (MCL) of five HAAs, we summed the five HAAs to represent the level of HAAs in water. Figure 3 shows the high correlation between THMs and the five HAAs in raw water. In the eight raw water samples, the formation potential of THMs paralleled that of the five HAAs. That is to say, if the THM formation potential of source water

Fig. 2 Relationship between UV_{254} and THM, HAA in raw water.

Fig. 3 Relationship between THMs and five HAAs in raw waters.

was high, the formation potential of the five HAAs was high accordingly.

All seven drinking water plants in this study used conventional water treatment, but the dose of chloride added to the drinking water differed. We analyzed the relationship between chloride dose and the formation of DBPs in the seven drinking waters. From Fig. 4 it can be seen that the correlation between chloride dose and THM was much higher than that between chloride dose and THAA5. The formation of DBPs is affected by many factors, such as water quality and water treatment (Ates et al., 2007). In this study, a higher dose of chloride resulted in more THM. For THAA5, however, the correlation was small. This highlights that the effect of chloride dose is complicated.

2.4 Specific-DBP in raw water

Specific-DBP was defined as the ability of DBPs formation per DOC in water. In raw waters, the lowest specific-THM, specific-THAA5 and specific-THAA9 were all found in NSBD (Fig. 5). Results showed that NT, MPS, and BJ had relatively high specific-THM, specific-THAA5, specific-THAA9 and SUVA.

According to Table 1, we can group the eight raw water samples into polluted and un-polluted source water: the SC, XHD and MPS samples had high DOC concentration,

Fig. 4 Relationship between dose of chloride and the formation of THM, HAA in drinking water.

Fig. 5 Specific total DBP formation potential (DBPFP/DOC) in raw waters.

UV_{254} absorbance, and DBP formation potential (DBPFP), while these parameters for BJ and XS were low. From another perspective, NT, MPS, and BJ had high concentrations of SUVA and specific-DBP, while these parameters were low for NSBD and DY.

The MPS sample was collected in a reservoir surrounded by flourishing forest, a likely source of humic acid which it is primarily derived from plant remains; SC was sampled in Songhua River, which was badly polluted by a leak of benzene compounds in 2005 and continued pollution from chemical and petroleum plants along the river. This has influenced that high amount of aromatics in this water, particularly high DBPFP. For the XHD sample, large quantities of untreated domestic sewage flood into the body water and total coliform exceeds the regulated limit for drinking water standards. Taking these factors into consideration, we determined SC, MPS and XHD to be polluted sources.

MPS, SC and XHD were sampled from polluted water sources. These results accord well with our study as the specific-DBPs of these three source water samples were higher than those samples collected in the unpolluted area. Total THM in this study was lower than those in Australian raw water (Drikas et al., 2008), but specific-DBPs were all much higher, although below 40 $\mu\text{g/L}$. It is considered likely that the conditions in Australia involving

Fig. 6 Concentration of specific-TTHMs in raw (a) and drinking water (b) in autumn, winter, and spring.

the presence of high DOC influenced these differences.

2.5 Seasonal variation in concentrations of specific-TTHMs in drinking water

The concentrations of specific total THMs in the seven drinking water samples collected in autumn, winter and spring are shown in Fig. 6b. The specific-TTHMs concentration was highest in spring, and almost similar in autumn and winter. The concentrations of specific-TTHMs in the north were generally higher than in the south. This was thought to be due to the relatively high water temperature in spring, which can accelerate the formation of DBPs, and the higher dose of chlorine added to the water in spring than in winter (Baytak et al., 2008). In Fig. 6a, the highest total THMs was also in spring, while total THMs in autumn was much lower than in winter as the chloride dose in autumn is lower by 20%–50%.

3 Conclusions

The amounts of TTHM, DCAA, and TCAA in the seven drinking water samples were all below the “Standards for Drinking Water Quality” (GB5749-2006). The proportion of CF to four THMs accounted for up to 81%, and the four THMs species were in the order of CF > DCBM > DBCM > BF. Total concentrations of five HAAs in SC and MPS drinking water exceeded the maximum contaminant level of 60 µg/L proposed by the Disinfectant/Disinfection by-product Rule (I) of US EPA (1998).

The correlation between UV₂₅₄ and DBPs in raw waters was high and the formation potential of THMs paralleled that of the five HAAs.

The eight raw water samples were grouped into polluted and un-polluted source water, with SC, MPS, and XHD considered as the polluted sources.

The highest concentration of THMs in drinking water and source water were both detected in spring and the DOC concentration in south was lower than that in north.

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