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

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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, haloketones 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.

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), haloketones (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 haloketones.

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, haloketones, 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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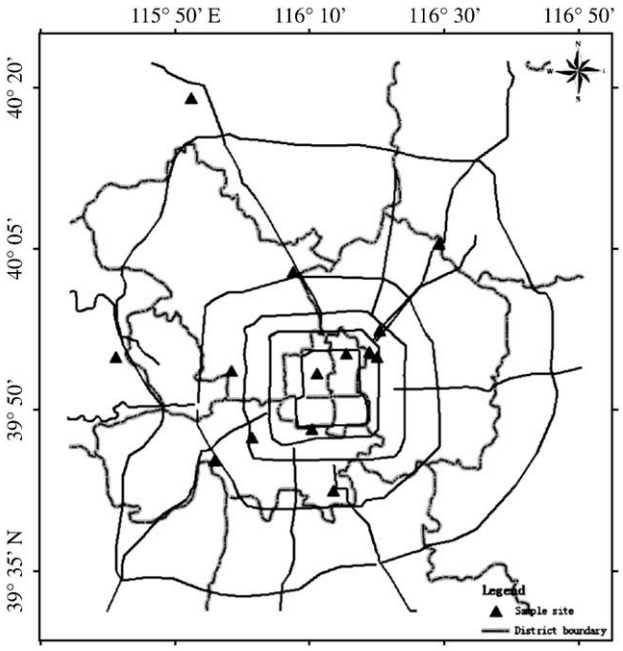

Fig. 1. Selected sampling sites in Beijing City.

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), haloketones (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

Table 2
Characteristics of water quality and operational parameters of Beijing City.

		TOC				Water temperature				Residual chlorine				pH			
		Aut	Win	Spr	Sum	Aut	Win	Spr	Sum	Aut	Win	Spr	Sum	Aut	Win	Spr	Sum
Raw	Min	0.74	0.27	0.55	0.68	15.00	5.00	10.00	15.00	–	–	–	–	7.41	7.13	7.01	6.97
	Max	7.75	3.61	3.11	3.36	24.50	16.5	17.50	28.00	–	–	–	–	8.60	7.96	8.35	8.55
water	Mean	1.94	2.13	1.80	1.66	17.90	14.2	15.30	18.80	–	–	–	–	8.00	7.42	7.44	7.53
	Std	2.29	1.18	0.91	0.97	2.77	3.76	2.24	4.15	–	–	–	–	0.34	0.29	0.49	0.48
Treated	Min	0.63	0.71	0.60	0.64	17.00	2.00	10.00	17.00	0.10	0.05	0.20	0.05	7.02	7.19	6.98	6.82
	Max	3.47	3.23	3.12	2.84	24.00	15.00	17.50	27.00	0.80	1.10	1.50	2.00	8.33	7.90	7.66	7.72
water	Mean	1.76	2.03	1.74	1.54	20.50	8.75	15.30	22.10	0.40	0.49	0.70	0.64	7.90	7.32	7.41	7.46
	Std	0.89	0.88	0.87	0.74	2.53	4.89	1.71	3.78	0.21	0.26	0.41	0.46	0.32	0.20	0.18	0.26

Aut: autumn; Win: winter; Spr: spring; and Sum: summer.

As shown in Table 4, the correlation was observed between each group of DBPs. There was a strong correlation between each group of DBPs. Some correlation coefficients of them were higher than 0.6. The correlation was also observed between DBPs and water qualities. A low but definite with a small relationship ($r=0.287$) was obtained between HAA formation and TOC, and a moderate correlation with a substantial relationship ($r=0.415$) was obtained between HAN formation and TOC. However, there was no apparent correlation between TOC and THMs. In general, greater DBP levels are formed in waters with higher concentrations of TOC (USEPA, 2006). TOC removal may be a possible reason that is used to explain the lack of an apparent correlation between TOC and DBPs. There were likely a lot of variations in the TOC removal between the various plants of Beijing City, which obviously impacted by-product formation. Another reason was that TOC is an indicator of mass organic substance and does not differentiate between the various chemical compounds that make up the precursor compounds (USEPA, 2001). Water temperature was strongly correlated with CH, CP and THMs. But there was no apparent correlation between water temperature and other DBPs. The rates of formation of most DBPs increase with increasing temperature. However, high temperature conditions in the distribution system promote the accelerated depletion of residual chlorine, which can mitigate DBP formation and promote biodegradation of DBPs (especially HAAs) unless chlorine dosages are increased to maintain high residuals (Singer and Reckhow, 1999). All of these reasons affect the relationship between water temperature and DBPs. A strong positive correlation existed between applied chlorine dose and most groups of DBPs except CH. As the concentration of chlorine or chloramines increases, the production of DBPs increases. Formation reactions continue as long as precursors and disinfectant are present (Krasner, 1999). Chlorine dosage which had no apparent correlation with CH may be may be due to the quality of water source, water temperature and also the material properties of the water pipes. No significant correlations were observed between DBP levels and pH. In general, THM formation increases with increasing pH, whereas the formation of HAAs and other DBPs increases with decreasing pH (USEPA, 2006). However, in Beijing City, the pH of water sample ranged from 6.82 to 8.55. And the pH only changed slightly. The pH value could hardly affect the formation of DBPs.

3.2. Distribution of DBP concentration in different water sources

As shown in Fig. 2, variation of all DBPs concentration in different water sources was surface water > mixture water > ground water. The drinking water of Beijing comes from three main sources: surface water, ground water and mixture water (surface water mixed with ground water). For the two most important disinfection by-products, the range of concentration for THMs was 9.83–69.98 μg/L in surface water, 5.15–28.76 μg/L in mixture water and 1.72–28.64 μg/L in ground water and for HAAs in surface water, mixture water and ground water were 3.87–30.36 μg/L, 0.21–30.36 μg/L and 0.00–18.68 μg/L, respectively. The reason why the DBP level was lowest in ground water was that the drinking water from ground water sources often comes from individual wells. The water from these wells is usually not treated. The applied chlorine dose was in an order of

Table 3
The concentrations of DBPs for the treated waters of Beijing City (μg/L).

		N	Mean	Median	Minimum	Maximum	Std
THMs	TCM	104	6.51	4.75	ND	29.41	6.51
	BDCM	104	3.70	4.32	ND	9.90	2.80
	DBCM	105	2.35	2.25	ND	6.36	1.40
	TBM	105	1.93	1.5	ND	9.82	1.69
	TTHM	104	14.49	14.12	ND	44.28	9.73
HAAs	DCAA	98	2.69	1.57	ND	13.02	3.28
	TCAA	82	3.38	1.61	ND	20.10	4.12
	DBAA	99	0.59	0.49	ND	2.86	0.56
	HAA5	82	6.50	4.67	ND	30.36	6.88
HANs	TCAN	104	0.72	0.11	ND	5.14	1.07
	DCAN	70	0.80	0.59	ND	3.43	0.85
	BCAN	105	1.03	0.88	ND	3.80	0.85
	DBAN	101	0.97	0.86	ND	4.70	0.73
	THAN	66	2.69	2.44	ND	7.80	1.65
HKs	1,1-DCP	98	1.27	0.72	ND	10.03	1.73
	1,1,1-TCP	105	0.62	0.45	ND	2.48	0.54
	THK	98	1.84	1.41	ND	12.16	2.02
CH	CH	104	0.93	0.50	ND	10.44	1.37
CP	CP	95	0.31	0.21	ND	2.08	0.35

Total number of sample = 116.
N: sample size, ND: not detected, and Std: standard deviation.

Table 4
The correlation matrices for water quality parameters and DBPs in water samples.

		CH	CP	THMs	HAAs	HANs	HKs
CH	*r*	1.000					
	p						
CP	*r*	0.325**					
	p	0.000					
THMs	*r*	0.498(**)	0.334(**)				
	p	0.000	0.000				
HAAs	*r*	0.711**	0.215*	0.693(**)			
	p	0.000	0.011	0.000			
HANs	*r*	0.419**	0.404**	0.866(**)	0.680**		
	p	0.000	0.000	0.000	0.000		
HKs	*r*	0.223**	0.564**	0.359(**)	0.470**	0.388**	
	p	0.004	0.000	0.000	0.000	0.000	
TOC	*r*	0.127	−0.157	0.176	0.287*	0.415**	0.111
	p	0.211	0.138	0.082	0.011	0.001	0.292
Water temperature	*r*	0.205*	0.443**	0.253(*)	−0.053	−0.073	−0.163
	p	0.042	0.000	0.011	0.642	0.569	0.118
Chlorine dosage	*r*	0.163	.265(*)	.329(**)	.233(*)	.293(*)	.309(**)
	p	0.106	0.012	0.001	0.040	0.020	0.003
pH	*r*	0.167	−0.045	−0.195	0.143	0.008	0.132
	p	0.112	0.688	0.063	0.226	0.952	0.225

r: Pearson correlation; *p*: sig. (2-tailed).
** Significant at the 0.01 level (2-tailed).
* Significant at the 0.05 level (2-tailed).

surface water > mixture water > ground water. Moreover, the mean value of TOC level for three water sources followed the sequence: surface water (2.28 mg/L) > mixture water (1.50 mg/L) > ground water (1.23 mg/L). In general, formation reactions of DBPs continue as long as precursors and disinfectant are present. Surface water sources such as reservoirs and streams were more likely to have higher disinfection by-products levels than ground water sources. And the disinfection by-product level of mixture water was higher than the ground water but lower than the surface water.

3.3. Spatial variations of DBPs

The spatial variations of DBPs were shown in Fig. 3. The total DBP levels were higher in the west than in the east of Beijing. A possible reason came from the different water source. The water treatment plants that used ground water as their water source are almost in the west of Beijing and the ground water source produced lower DBPs than mixture water and surface water. Another reason was caused by the rainfall. The general trend of rainfall increases from west to east and from north to south in Beijing (Pan, 2006). The DBP precursor decreased with an increase of rainfall due to dilution by rainwater. In the spatial variation of THMs, the variation of major components TCM and DCBM was similar to TTHM. But the concentration of DBCM and TBM did not exhibit a clear spatial trend (Fig. 3b). This could be explained by the bromide level of water sample. In most water samples of Beijing, bromide ion was determined (Ye, 2007). The bromide ion level ranged from 0.019–0.135 mg/L for ground water and 0.016–0.121 mg/L for surface water. The mean value of bromide ion concentration in ground water was higher than in surface water. The patterns of the four THM species with increased Br^- concentrations are as follows: TCM and DCBM decrease continuously; DBCM increases initially and then decreases, TBM increases continuously (Chang et al., 2001). In variation of HAAs, the variation trend of TCM and DCBM was similar to the variation of HAA5. DBAA level also did not exhibit a clear spatial variation. That could also be explained by the bromide level of water sample. The DBAA level increased with increased Br^- concentrations (Nikolaou et al., 1999; Chang et al., 2001). In the variation of HANs, the variation trend of DCAN was similar to the variation of total HANs. TCAN BCAN and DBAN did not exhibit a clear spatial variation. The main reason also was caused by bromide concentration. With increased Br^- concentrations, BCAN and DBAN increased and TCAN and DCAN decreased instead. The spatial variation of 1,1-DCP, 1,1,1-TCP, CH and CP was similar to the spatial variation of TTHM, HAA5 and THAN.

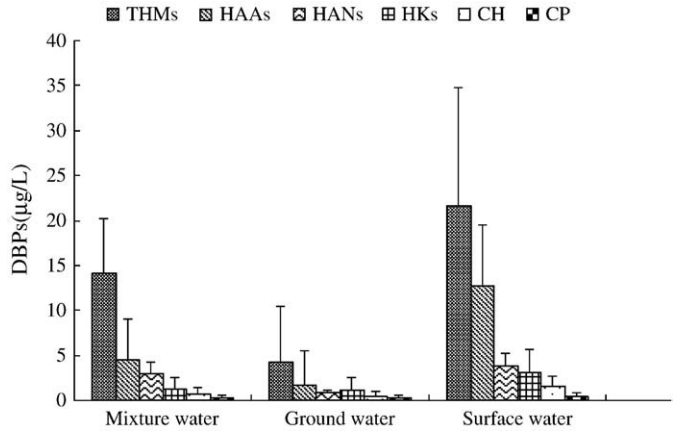

Fig. 2. Species distribution of the DBPs of different water sources.

3.4. Seasonal variations of DBPs

There are significant variations in the seasonal data of DBPs, and the distribution of DBP concentration in each season is summarized in Fig. 4. THM levels were high in summer and decreased in spring, autumn and winter. The main reasons could be explained by water temperature and chlorine residue. THM levels generally increase with the temperature of water. Moreover, higher level of chlorine residual may also favour the formation of THMs (Nikolaou et al., 1999). The variation of THMs was consistent with many studies that reported maximum THM formation in summer (Rodriguez et al., 2004; Toroz and Uyak, 2005). HAAs exhibited a different seasonal variation than THMs. The variation of HAA level was in an order of winter > spring > autumn > summer. This could be explained by the precursor concentrations (TOC levels) of the different seasons in Beijing water (Table 2). The HAA formation is known to increase with increasing

precursor concentrations. The TOC level of water sample in autumn was a little higher than in spring. However, HAAs are known to biodegrade over time when the disinfectant residual is low. The residual chlorine level in spring was higher than that in autumn. And the HAA biodegradation in spring was lower than that in autumn. For these reasons, the HAA level in spring was higher than that in autumn. The trend for seasonal HAA variability was similar to some studies (Rodriguez et al., 2004; Rodriguez et al., 2007). The variation of HANs was consistent with HAAs. In the variation of other classes of DBPs, the highest HK levels were observed in winter, CH is observed in autumn, and CP is highest in summer. These classes of DBPs exhibited different seasonal variations as compared to THMs and HAAs. Haloacetonitriles, haloketones, chloropicrin and chloral hydrate presented at lower concentrations in water samples. There were limited researches on these classes of DBPs.

4. Conclusion

A survey of disinfection by-products occurrence in Beijing City was conducted at 15 drinking water treatment plants for the first time in Beijing. THMs and HAAs were the major DBPs found in all water treatment plants. The variation of all DBPs concentration in different water sources was as follow: surface water > mixture water > ground water. In spatial variation, the total DBPs levels were higher in the west than in the east of Beijing. The spatial variation of TCM, DCBM, DCAA, TCAA, DCAN, 1,1-DCP, 1,1,1-TCP, CH and CP was very similar to

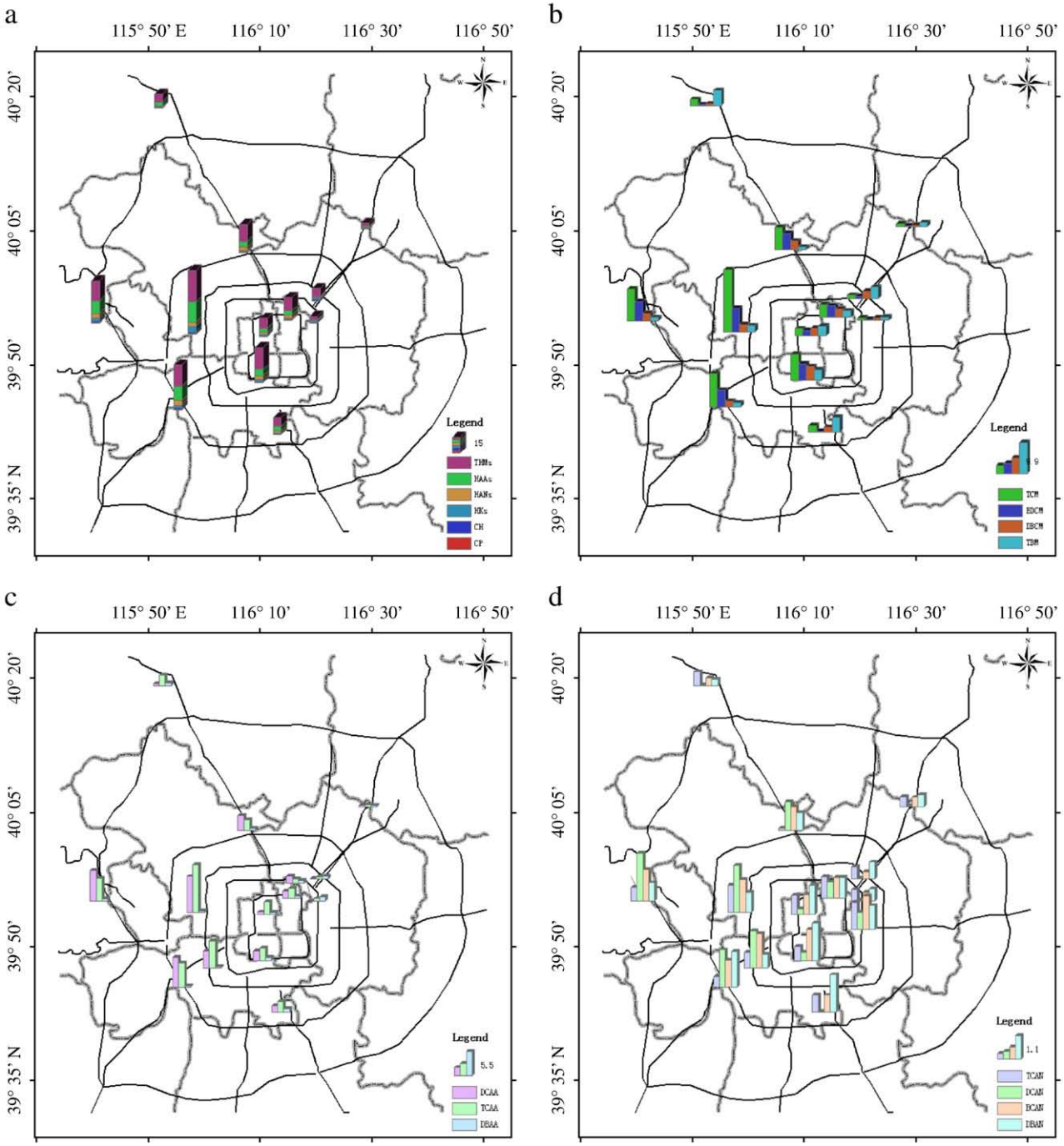

Fig. 3. Spatial variations of DBPs in water samples (a. DBPs; b. THMs; c. HAAs; d. HANs; e. HKs; f. CH and CP).

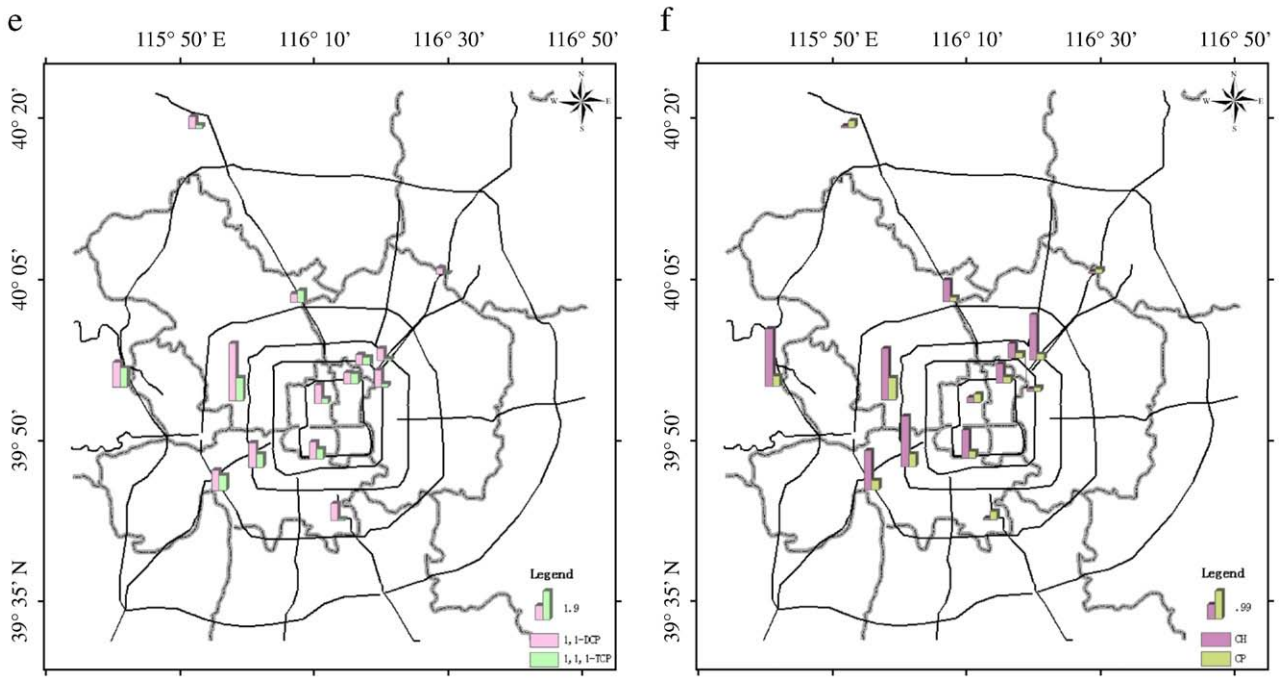

Fig. 3 (*continued*).

that of total DBPs. But, DBCM, TBM, DBAA, TCAN, BCAN and DBAN did not show significant spatial variation. In the seasonal variation, THM levels were high in summer and decreased in spring, autumn and winter. The variation of HAA levels is in the order of winter > spring > autumn > summer. The variation of HANs was consistent with HAAs. In the variation of other classes of DBPs, the highest HK levels were observed in winter, CH is observed in autumn, and CP is highest in summer.

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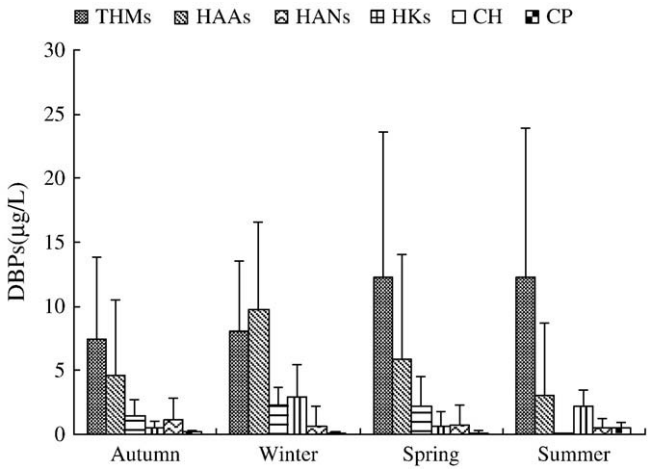

Fig. 4. Seasonal variation of DBPs in water samples.

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