

Characteristics of Disinfection By-Products Formation in Chlorination of Principal Raw Waters for Drinking Water of Jeju Island, Korea

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Abstract

This study was carried out to investigate the characteristics of disinfection by-products (DBPs-trihalomethanes (THMs), haloacetic acids (HAAs) and haloacetonitriles (HANs) formation in chlorination of principal raw waters used for drinking water on Jeju Island, Korea. The domestic water supply of other area and humic acid solution (HA) were used as a reference point. The effects of chlorine contact time, solution temperature and pH on DBPs formation potential (DBPFP) were investigated for raw waters. In addition, the effect of Br⁻ was studied for HA. The DBPFP (THMFP, HAAFP and HANFP) were increased with increasing chlorine contact time. Comparing the individual DBPFPs for raw waters, they decreased in the order of HAAFP > THMFP ≥ HANFP. As the solution temperature was increased, the THMFP, HAAFP and HANFP increased. With increasing the solution pH, the THMFP was increased, but HAAFP and HANFP were decreased. With the addition of 0.3 mg/L Br⁻ for HA, the DBPFP was increased and the major chemical species changed: from trichloromethane to dibromochloromethane and tribromomethane for THMs; from dichloroacetic acid and trichloroacetic acid to tribromoacetic acid for HAAs; and from dichloroacetonitrile to dibromoacetonitrile for HANs.

Key Words : Disinfection by-products, Trihalomethanes, Haloacetic acids, Haloacetonitriles, Chlorination of raw waters, Jeju Island

1. Introduction

Chlorine is the most common disinfectant used in water treatment to inactivate pathogenic microorganisms, improve coagulation and filtration efficiency, prevent algal growth in sedimentation basins and filters, prevent biological regrowth in the water distribution systems and oxidize reduced inorganic solutes, such as sulfide and ferrous ions (Agus et al., 2009; Golfinoopoulos and Nikolaou, 2005). However, water chlorination can produce disinfection by-products

(DBPs) that pose threats human health or aquatic ecosystems. The most common DBPs are trihalomethanes (THMs), haloacetic acids (HAAs) and haloacetonitriles (HANs). The THMs formed during chlorination, representing between 5% and 20% of the DBPs, are trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM) and tribromomethane (TBM). Total trihalomethanes (TTHMs) refer to the sum of these four chemical substances. HAAs include the following nine compounds: monochloroacetic acid (MCAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), monobromoacetic acid (MBAA), dibromoacetic acid (DBAA), tribromoacetic acid (TBAA), bromochloroacetic acid (BCAA), dibromochloroacetic acid (DBCAA), bromodi-

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chloroacetic acid (BDCAA), the first five compounds are regulated by the current Disinfection/Disinfectant By-Products (D/DBP) Rule due to limited formation and occurrence data for some of the species (USEPA, 1998). The sum of above nine compounds is referred as total haloacetic acids (THAAs). HANs, which are small, nitrogenous haloorganics, include monochloroacetonitrile (MCAN), dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), monobromoacetonitrile (MBAN), dibromoacetonitrile (DBAN), tribromoacetonitrile (TBAN) and bromochloroacetonitrile (BCAN). The sum of above seven compounds is referred as total haloacetonitriles (THANs).

It was reported that DBPs in drinking water are affected by the organic precursors, disinfectant dosage, water quality parameters (pH, solution temperature, concentration of $\text{NH}_3\text{-N}$ and Br^- , etc) and the reaction time between the organic precursors and disinfectant (Chen and Weisel, 1998; Uyak and Toroz, 2007). The organic precursors include natural organic matters (NOM), and are generally characterized by total organic carbon (TOC), dissolved organic carbon (DOC), ultraviolet absorbance at 254 nm (UV_{254}) or specific ultraviolet absorbance (SUVA, defined as the UV absorbance expressed as per meter of absorbance per unit concentration of DOC, in mg/L). The content of humic substances in surface water has been found to be a major factor affecting the formation of organic by-products during drinking water disinfection. Humic substances include humic and fulvic acids, the concentrations of which vary depending on the vegetation near the water source, the concentration of algae in water and the time of the year. A number of studies have been focused on the reaction of humic and fulvic acids with chlorine and the subsequent DBPs yield.

The studies on DBPs in Jeju Island of Korea were only few, and included those of THMs (Song, 1997) and TOXs (Yoon, 2001). The purpose of this study was to investigate the characteristics of DBPs (THMs,

HAAs and HANs) formation in chlorination of principal raw waters for drinking water of Jeju Island, Korea. As a comparison, raw water of other domestic area and humic acid solution (known as a strong hydrophobic acid) were used. The effects of chlorine contact time, solution temperature and pH on the formation potential of DBPs were investigated for these raw waters. In addition, the effect of Br^- was studied for HA.

2. Materials and methods

The raw water samples of Jeju Island used in this study were as follows: Iho springwater (I-SW), Oedo springwater (O-SW) and Wolsan groundwater (W-GW), supplying Wolsan Water Treatment Plant (WTP), Jeju City; Samyang 3 springwater (S-SW), supplying Doryeon WTP, Jeju City and used after being mixed with other raw water; Eoseungsaeng springwater (E-SW), supplying Eoseungsaeng WTP, Jeju City; Gangjeong springwater supplying Gangjeong WTP, Seogwipo City; Topyeong groundwater (T-GW) supplying Topyeong WTP, Seogwipo City. As the reference water samples, Nakdong river water (N-RW) and humic acid solution (HA) were used. N-RW was sampled at the reservoir for Chilseo WTP, Gyeongsangnamdo. HA (Aldrich Chem. Co., USA) was prepared as 0.3 mg/L solution, by dissolving it in 500 mL of 0.1 N NaOH, filtering through a qualitative filter paper and a prewashed 0.45 μm cellulose nitrate filter paper, and diluting with water. It was adjusted with to pH 6.5-8.5 with 0.1 N HNO_3/NaOH . Samples were filtered through a prewashed 0.45 μm cellulose nitrate filter paper and were used for the measurements of dissolved organic carbon (DOC) and absorbance at 254 nm (UV_{254}).

The trihalomethanes formation potential (THMFP), haloacetic acids formation potential (HAAFP) and haloacetonitriles formation potential (HANFP) through chlorination of various raw water samples, were analyzed separately. The halogenated organics formation

range of 0.008 ~ 2.182 mg/L, except for HA. NH₃-N which was related to chlorine consumption, was not detected for all the water samples.

3.2. Characteristics of DBPs formation

3.2.1. Effect of chlorine contact time

It is found that the DBPs formation potential (DBPFP) upon the reaction of raw water and chlorine depends on the organic matter characteristics and chlorine contact time (Huang and Yeh, 1997). In order to investigate the DBPFP (THMFP, HAAFP and HANFP) with chlorine contact time, a chlorine concentration of about ten times the DOC content of raw waters was added to water samples used in this study and they were incubated at 20°C for 0.5, 1, 3, 6, 18, 24, 48, 72, 96, 120, 144, 168, 192 h, the results of which are shown in Fig. 1.

As shown in Fig. 1, the THMFP, HAAFP and HANFP were increased with increasing chlorine contact time, although they differed according to the characteristics of each raw water. Comparing DBPFPs at 72 h and 168 h with that at 24 h chlorine contact time, it was 1.1 ~ 2.7 times higher in the case of the former and 1.6 ~ 7.0 times higher in the case of the latter for THMFP, 1.2 ~ 1.4 times higher in the case of the former and 1.4 ~ 3.3 times higher in the case of the latter for HAAFP, and 1.4 ~ 1.7 times higher in the case of the former and 1.6 ~ 2.7 times higher in the case of the latter for HANFP. The maximum THMFP for the raw waters of Jeju Island, N-RW and HA were in the range of 2.2 ~ 8.4 µg/L, 39.8 µg/L and 14.5 µg/L, respectively, indicating that that for N-RW was 4.7 ~ 18.1 times higher than those for the raw waters of Jeju Island, and 2.7 times higher than that for HA. However, the maximum THMFP per unit of DOC was 13 ~ 50 µg/mgC for the raw waters of Jeju Island, 52 µg/mgC for N-RW and 73 µg/mgC for HA, indicating that that for N-RW was only 1.0 ~ 4.0 times higher than those for the raw waters of Jeju Island, and lower than that for HA. The

maximum HAAFP for the raw waters of Jeju Island, N-RW and HA were in the range of 9.1 ~ 29.3 µg/L, 51.0 µg/L and 43.5 µg/L, respectively, indicating that that for N-RW was 1.7 ~ 5.6 times higher than those for the raw waters of Jeju Island, and 1.2 times higher than that for HA. However, the maximum HAAFP per unit of DOC was 66 ~ 140 µg/mgC for the raw waters of Jeju Island, 67 µg/mgC for N-RW and 219 µg/mgC for HA, indicating that that for HA was the highest, followed by those for the raw waters of Jeju Island, then that for N-RW, differently from THMFP/DOC. The maximum HANFP for the raw waters of Jeju Island, N-RW and HA were in the range of 3.5 ~ 7.0 µg/L, 45.7 µg/L and 5.1 µg/L, respectively, indicating that that for N-RW was 6.5 ~ 13.1 times higher than those for the raw waters of Jeju Island, and 13.1 times higher than that for HA. The maximum HANFP per unit of DOC was 7.8 ~ 64 µg/mgC for the raw waters of Jeju Island, 60 µg/mgC for N-RW and 26 µg/mgC for HA, indicating that that for N-RW was 0.9 ~ 7.7 times higher than those for the raw waters of Jeju Island and 2.3 times higher than that for HA, and that for HA was the lowest, compared with THMFP/DOC and HAAFP/DOC. Comparing the DBPFP for raw waters, it decreased in the order of HAAFP > THMFP ≥ HANFP (data was not shown). The difference in DBPFP or DBPFP/DOC for the raw waters is likely due to complex factors involving several water quality parameters, such as the concentration and characteristics of DOC, pH, Br⁻ concentration, etc.

The major chemical species of DBPs formed under the same conditions as above, were examined. The results for the ratio of each DBPFP to total DBPFP at the condition of chlorine contact time for 48 h are shown in Table 2 ~ Table 4, although they are a little different with chlorine contact time. As shown in Table 2, the major chemical species for THMs, were TCM for HA and N-RW, TBM for S-SW, DBCM and TBM for I-SW, DBCM for W-GW,

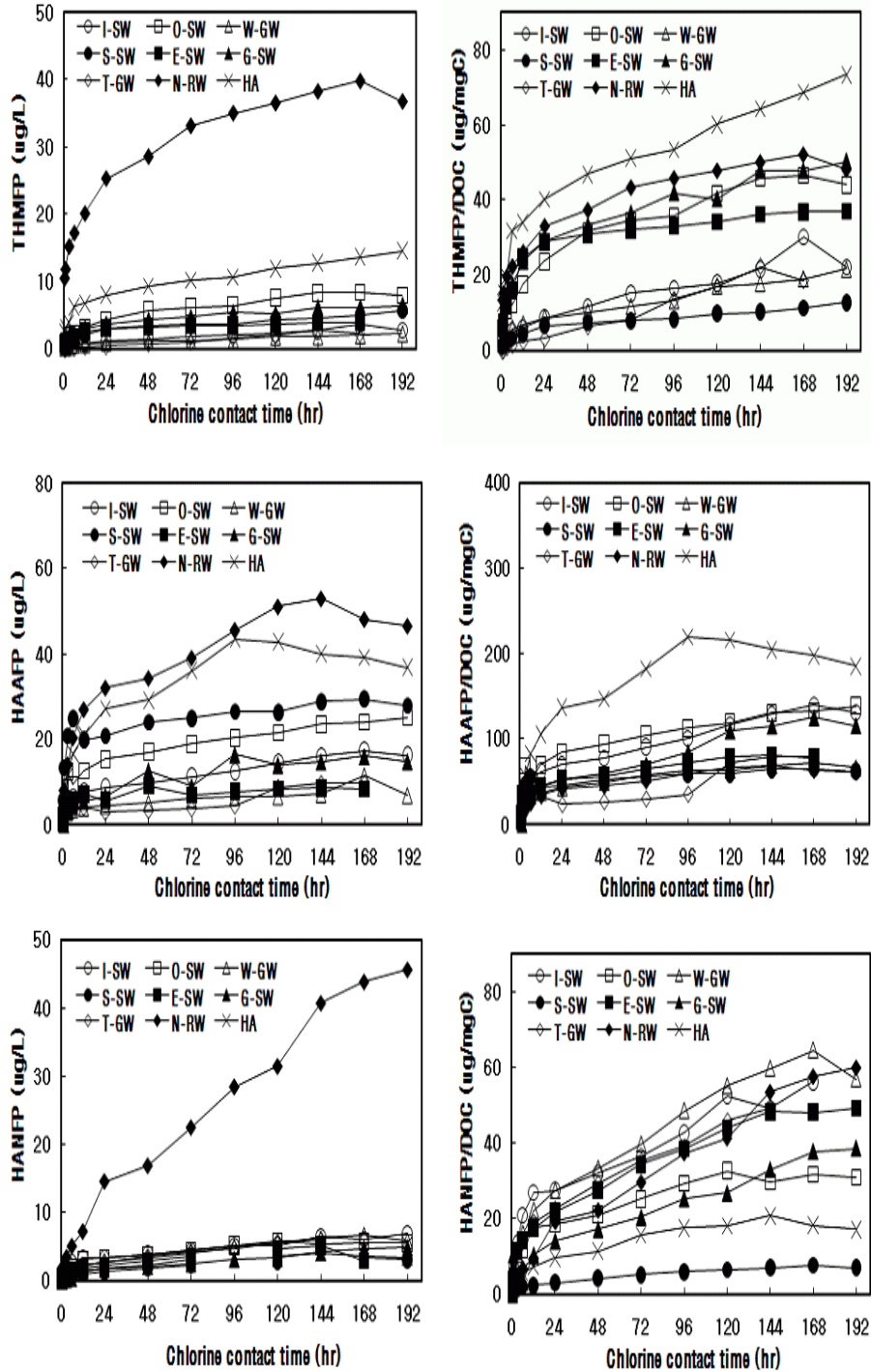


Fig. 1. DBPPF and DBPPF/DOC with chlorine contact time for the raw waters used in this study.

Table 2. The ratio of each THMFP to total THMFP for the raw waters used in this study at the condition of chlorine contact time for 48 h

Raw water source	TCMFP/THMFP(%)	Brominated THMFP/THMFP(%)		
		BDCMFP/THMFP(%)	DBCMPF/THMFP(%)	TBMFP/THMFP(%)
I-SW	0	9	49	42
O-SW	17	34	41	8
W-GW	5	22	58	15
S-SW	0	0	6	94
E-SW	37	38	25	0
G-SW	25	39	36	0
T-GW	23	36	39	2
N-RW	51	27	19	3
HA	100	0	0	0

DBCM and BDCM for O-SW, G-SW and T-GW, TCM and BDCM for E-SW, which may be mainly due to the the concentration ratio of Br⁻/DOC. With its higher ratio (eg., I-SW, W-GW, S-SW), much more DBCM and/or TBM were formed. Similar results are shown for HAAs and HANs (Table 3 and Table 4). For HAAs, they were DCAA and TCAA for HA, E-SW, G-SW and T-GW, TBAA for S-SW, I-SW and W-GW, DCAA, BCAA and CDBAA for O-SW. For N-RW, they were TBAA and TCAA, differently from the pattern of those for THMs, which

may be due to their formation mechanism and the concentration ratio of Br⁻/DOC, and etc. For HANs, they were DCAN for HA, DBAN for S-SW, I-SW, W-GW and T-GW, DBAN and BCAN for O-SW, DCAN and DBAN for E-SW and G-SW, MCAN, DCAN and DBAN for N-RW. DBAN was formed for all the raw waters, and MCAN was only detected for N-RW. In addition, TCAN, MBAN and TBAN examined in this study were not detected for all the raw waters, which may be due to their instability, although their formation mechanism was not clear.

Table 3. The ratio of each HAAFP to total HAAFP for the raw waters used in this study at the condition of chlorine contact time for 48 h

Raw water source	DCAAFP/HAAFP (%)	TCAAFP/HAAFP (%)	BCAAFP/HAAFP (%)	BDCAAFP/HAAFP (%)	DBAAFP/HAAFP (%)	CDBAAFP/HAAFP (%)	TBAAFP/HAAFP (%)
I-SW	21	0	7	12	10	0	50
O-SW	18	8	22	9	14	20	9
W-GW	17	10	5	21	8	0	39
S-SW	0	0	1	3	8	0	88
E-SW	32	30	17	14	7	0	0
G-SW	35	31	15	10	9	0	0
T-GW	27	39	11	13	10	0	0
N-RW	8	19	14	8	13	12	26
HA	64	36	0	0	0	0	0

Table 4. The ratio of each HANFP to total HANFP used in this study at the condition of chlorine contact time for 48 h

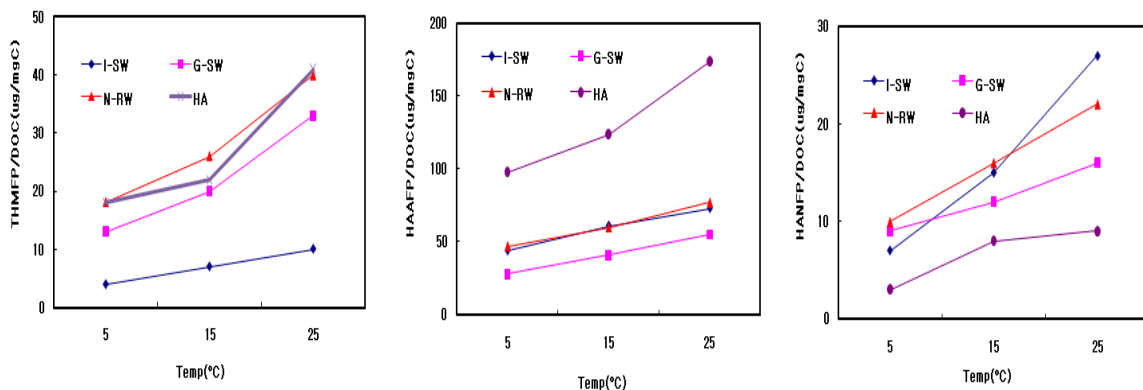
Raw water source	MCANFP/HANFP(%)	DCANFP/HANFP(%)	BCANFP/HANFP(%)	DBANFP/HANFP(%)
I-SW	0	5	12	83
O-SW	0	0	39	61
W-GW	0	0	18	82
S-SW	0	0	0	100
E-SW	0	47	12	41
G-SW	0	32	23	45
T-GW	0	13	15	72
N-RW	37	22	19	22
HA	0	100	0	0

3.2.2. Effect of solution temperature and pH

It is known that the DBPFP increases with increasing the solution temperature, but differed according to the solution pH (Peters et al., 1980). In order to investigate the effect of solution temperature on the DBPFP, a chlorine concentration of about ten times the DOC content of raw water, was added to the samples (I-SW, G-SW, N-RW and HA), the waters were incubated at 5 °C, 15 °C and 25 °C for 24 h, and the results are shown in Fig. 2. The THMFPP, HAAFP and HANFP at 25 °C, as compared with those at 5 °C were 2.5 times, 1.7 times and 3.9 times higher for I-SW, 2.5 times, 2.0 times and 1.8 times higher for G-SW, 2.2 times, 1.6 times and 2.2 times

higher for N-RW, and 2.2 times, 1.8 times and 3.0 times higher for HA, showing similarity to those reported by Peters et al. (1980), although their increasing rate differed a little according to the characteristics of raw waters.

In order to examine the effect of solution pH on the DBPFP, samples were treated as above, except that they were changed to pH 6.5, 7.5 and 8.5, and were incubated at a uniform temperature of 20 °C. With increasing the solution pH, the THMFPP was increased, the opposite was observed for HAAFP and HANFP. It was demonstrated that the THMFPP was increased with increasing pH as the THMs were formed by basic catalytic process, but HAAs were


Fig. 2. Effect of solution temperature on THMFPP/DOC, HAAFP/DOC and HANFP/DOC for the raw waters (I-SW, G-SW, N-RW and HA).

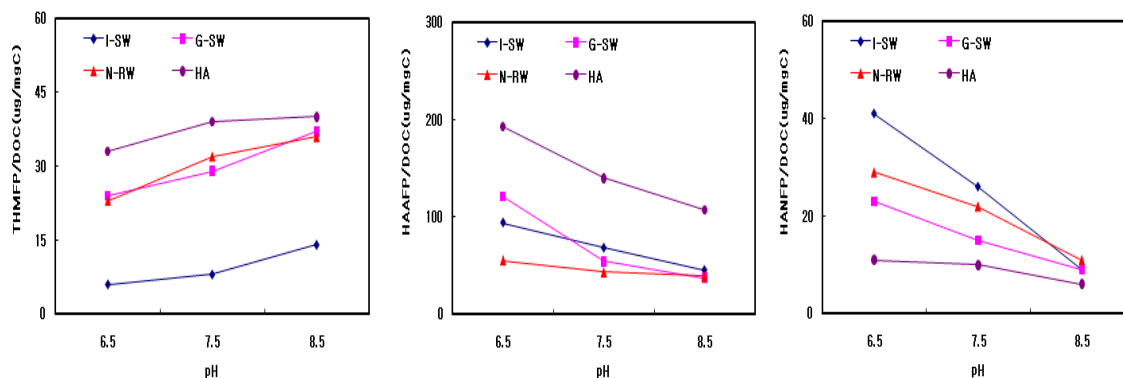


Fig. 3. Effect of solution pH on THMFP/DOC, HAAFP/DOC and HANFP/DOC for the raw waters (I-SW, G-SW, N-RW and HA).

formed in acidic conditions more readily (Miller and Uden, 1983; Peters et al., 1980). It is considered that HANs are formed in acidic conditions in the same way as HAAs, although the formation mechanism was not evident from this work.

3.2.3. Effect of bromide ion

It was reported that when raw waters containing Br^- were chlorinated, brominated DBPs were formed and their formation potential (DBPFP) were different, depending on the Br^- concentration and the characteristics of organic matter within a given sample (Huang and Yeh, 1997). In order to examine the effect of Br^- concentration on the DBPFP, 0.3 mg/L Br^- was added to HA (0.3 mg/L), and the solution was chlorinated

with a chlorine concentration of about ten times the DOC content in the solution for 24~192 h. The results are shown in Table 5~Table 8. As shown in Table 5, each of THMFP/DOC, HAAFP/DOC and HANFP/DOC in the presence of 0.3 mg/L Br^- addition, was 2.6~3.9 times, 1.4~1.8 times and 2.5~4.1 times higher than in the absence of Br^- addition, respectively, indicating that DBPFP increased with increasing concentrations of Br^- . This result was similar to that of Song's (1997), where 0.1~3.2 mg/L Br^- were added to the chlorinated HA for 24 h, THMFP in the presence of Br^- additions was 1.4~6.9 times higher than in its absence.

Table 6~Table 8 show the effect of Br^- (0.3 mg/L)

Table 5. Effect of bromide addition (0.3 mg/L) on THMFP/DOC, HAAFP/DOC and HANFP/DOC at different chlorine contact time for HA (0.3 mg/L)

Chlorine contact time(hr)	THMFP/DOC ($\mu\text{g}/\text{mgC}$)		HAAFP/DOC ($\mu\text{g}/\text{mgC}$)		HANFP/DOC ($\mu\text{g}/\text{mgC}$)	
	in the absence of Br^- addition	in the presence of Br^- addition	in the absence of Br^- addition	in the presence of Br^- addition	in the absence of Br^- addition	in the presence of Br^- addition
24	40	154	137	237	10	25
48	47	157	147	267	11	34
72	51	165	182	278	16	40
96	53	170	220	298	18	43
120	60	181	216	305	18	54
144	64	198	205	331	21	58
168	69	205	198	340	18	65
192	73	190	186	317	17	70

Table 6. Effect of bromide addition (0.3 mg/L) on the ratio of each THMFP to total THMFP at different chlorine contact time for HA (0.3 mg/L)

Chlorine contact time(hr)	in the absence of Br ⁻ addition		in the presence of Br ⁻ addition		
	TCMFP/THMFP(%)	TCMFP/THMFP(%)	BDCMFP/THMFP(%)	DBCMFP/THMFP(%)	TBMFP/THMFP(%)
24	100	4	13	40	43
48	100	4	13	40	43
72	100	4	14	40	42
96	100	4	14	39	43
120	100	4	13	39	44
144	100	4	12	38	46
168	100	4	13	39	44
192	100	4	13	39	44

on the ratio of each DBPFP to total DBPFP at different chlorine contact time for HA (0.3 mg/L). It was observed that the chemical species in the presence of 0.3 mg/L Br⁻ addition changed greatly, compared with those in the absence of Br⁻ addition. For THMs, 100% of TCM in its absence changed to 4% of TCM, 12~14% BDCM, 38~40% of DBCM and 42~46% of TBM in the presence of Br⁻ additions, indicating that the major chemical species in its presence changed from TCM to DBCM and TBM. For HAAs, 56~64% of DCAA and 36~44%

of TCAA in its absence changed to 10~4% of DCAA, 3~1% of TCAA, 3~6% BCAA, 13~10% BDCAA, 10~12% CDBAA and 54~62% of TBAA in its presence, indicating that the major chemical species in its presence changed from DCAA and TCAA to TBAA. For HANs, 100% of DCAN in its absence changed to 17~6% of DCAN, 15~10% of BCAN and 68~84% of DBAN in its presence, indicating that the major chemical species changed from DCAN to DBAN.

Table 7. Effect of bromide addition (0.3 mg/L) on the ratio of each HAAFP to total HAAFP at different chlorine contact time for HA (0.3 mg/L)

Chlorine contact time(hr)	in the absence of Br ⁻ addition				in the presence of Br ⁻ addition				
	DCAAFP/HAAFP (%)	TCAAFP/HAAFP (%)	DCAAFP/HAAFP (%)	TCAAFP/HAAFP (%)	BCAAFP/HAAFP (%)	BDCAAFP/HAAFP (%)	DBAAFP/HAAFP (%)	CDBAAFP/HAAFP (%)	TBAAFP/HAAFP (%)
24	64	36	10	3	3	5	13	11	55
48	64	36	11	3	4	4	13	11	54
72	59	41	10	2	4	4	13	11	55
96	63	37	8	1	4	5	13	12	57
120	58	42	5	1	5	6	13	12	58
144	58	42	5	1	5	6	11	10	62
168	59	41	4	1	6	6	11	11	61
192	56	44	4	1	6	7	10	11	61

Table 8. Effect of bromide addition (0.3 mg/L) on the ratio of each HANFP to total HANFP at different chlorine contact time for HA (0.3 mg/L)

Chlorine contact time(hr)	in the absence of Br ⁻ addition		in the presence of Br ⁻ addition	
	DCANFP/HANFP(%)	DCANFP/HANFP(%)	BCANFP/HANFP(%)	DBANFP/HANFP(%)
24	100	17	15	68
48	100	13	13	74
72	100	12	12	76
96	100	11	13	76
120	100	8	14	78
144	100	8	12	80
168	100	7	11	82
192	100	6	10	84

4. Conclusions

The values of specific UV absorbance (SUVA) for the raw waters used in this study, were in the range of 1.14~9.39. It was considered that the DOCs of O-SW, I-SW, N-RW and G-SW were composed of largely aquatic humics, and that they were relatively hydrophobic, aromatic and of high molecular weight, whereas those of S-SW and T-GW were composed largely of non-humic materials which were relatively hydrophilic, less aromatic and of lower molecular weight, and those of W-GW and E-SW were composed of weak hydrophobic acids or hydrophilic neutrals.

The THMFP, HAAFP and HANFP were increased with increasing chlorine contact time, although they differed depending on the characteristics of raw water. Comparing the maximum DBPFP and DBPFP/DOC among the raw waters, the maximum THMFP, HAAFP and HANFP were obtained from N-RW, but the maximum THMFP/DOC, HAAFP/DOC and HANFP/DOC were obtained for HA, HA and N-RW, indicating that the highest DBPFP was formed for N-RW with the highest DOC content, but the highest THMFP/DOC and HAAFP/DOC, lower HANFP and HANFP/DOC were formed for HA. Comparing the DBPFP for raw waters, it decreased in the order of

HAAFP > THMFP ≥ HANFP, which is thought to be due to complex factors involving several water quality parameters, such as the concentration and characteristics of DOC, pH, Br⁻ concentration, etc.

The major chemical species for THMs after the chlorine contact time for 48 h, were TCM for HA and N-RW, TBM for S-SW, DBCM and TBM for I-SW, DBCM for W-GW, DBCM and BDCM for O-SW, G-SW and T-GW, and TCM and BDCM for E-SW. For HAAs, they were DCAA and TCAA for HA, E-SW, G-SW and T-GW, TBAA and TCAA for N-RW, TBAA for S-SW, I-SW and W-GW, and DCAA, BCAA and DBCAA for O-SW. For HANs, they were DCAN for HA, DBAN for I-SW, S-SW, W-GW and T-GW, DCAN and DBAN for E-SW and G-SW, DBAN and BCAN for O-SW, MCAN, DCAN and DBAN for N-RW.

As the solution temperature for I-SW, G-SW, N-RW and HA was increased from 5°C to 25°C after 24 h chlorine contact time, each of THMFP, HAAFP and HANFP at 25°C, as compared with those at 5°C, were 2.2~2.5 times, 1.6~2.0 times and 1.8~3.9 times higher, respectively. With increasing the solution pH, the THMFP was increased, HAAFP and HANFP were decreased.

Each of THMFP/DOC, HAAFP/DOC and HANFP/DOC in the presence of 0.3 mg/L Br⁻ addition for HA, was 2.6~3.9 times, 1.4~1.8 times and 2.5~4.1 times higher than each of those in the absence of Br⁻ addition, respectively (indicating that DBPFP increased in the presence of Br⁻), and the major chemical species changed from TCM to DBCM and TBM for THMs, from DCAA and TCAA to TBAA for HAAs, and from DCAN to DBAN for HANs.

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