

## Variations of Disinfection By-products in a Chlorinated Drinking Water Distribution System

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The chlorination of municipal drinking water supplies leads to the formation of so-called disinfection by-products (DBPs), many of which have been reported to cause harmful health effects based on animal studies. This study was conducted: 1) to observe seasonal changes in the major DBPs at four sampling sites on a drinking water distribution system located in Chunchon, Kangwon Do; and 2) to examine the effects of major water quality parameters on the formation of DBPs. During the field sampling, the water temperature, pH, and total and free chlorine residuals were all measured. The water samples were then analyzed for total organic carbon (TOC) and eight disinfection by-products in the laboratory. Chloroform, dichloroacetic acid, and trichloroacetic acid were the major constituents of the measured DBPs. The concentrations of the total DBPs were highest in fall, particularly in October, and lowest in summer. The concentrations of the total DBPs increased with increasing TOC concentrations. Multiple regression analyses showed that the concentrations of chloroform, bromodichloromethane, and chloral hydrate were linearly correlated with the pH. Other water parameters were not included in the regression equations. Accordingly, these results suggest that TOC and pH are both important factors in the formation of DBPs.

Key words : drinking water, disinfection by-products, water quality parameters, total organic carbon, pH, formation

### 1. Introduction

Municipal water supplies are disinfected with chlorine ( $\text{Cl}_2$ ) or chlorine-containing chemicals to suppress the growth of microbial pathogens. However, such a treatment process has been reported to cause the formation of a wide variety of so-called disinfection by-products (DBPs), from the reaction of residual chlorine (mostly HOCl and OCl<sup>-</sup>) with organic materials such as humic substances<sup>1,2</sup>. Some studies have suggested that people who have regularly ingested chlorinated tap water develop a higher risk of bladder and rectal cancers than others<sup>3,4</sup>. Therefore, many countries currently regulate the levels of DBPs in drinking waters and the Korean Ministry of Environment have also limited the sum of the concentrations of four trihalomethanes (THMs) in drinking water to below 100  $\mu\text{g/l}$  since 1991.

Many studies have been conducted to elucidate

the variation in DBPs at different locations along a distribution system. However, complete kinetic models for the prediction of DBP concentrations have not yet been established, probably due to the complexity of the reactions between the organic matter and the chlorine residual. Laboratory and field studies have suggested that the formation of DBPs during the chlorination of drinking water depends on several factors, including reaction time, pH, water temperature, chlorine residual, bromide ion concentration, and organic matter content. Chen and Weisel<sup>5</sup> showed that THMs increased with an increasing contact time between chlorine residual and organic matter, yet that the concentrations of non-THMs, such as haloacetonitriles (HANs), halo ketones (HKs), chloropicrin, and haloacetic acids (HAAs), decreased with an increasing residence time. Stevens *et al.*<sup>6</sup> found in a laboratory experiment that 60% of DBPs are formed within a 4-hour reaction period and that

non-THMs decrease with an increasing pH. In addition, it was suggested that any increase in the chlorine residual or total organic carbon(TOC) in water also increases the production of DBPs. In summer, the concentrations of THMs, HANs, and HKs are higher than in other seasons, thereby suggesting that a higher water temperature favors the formation of DBPs.<sup>5,7)</sup> However, many studies have reported results that are inconsistent with one another, probably because of the use of dechlorinating reagents, such as  $\text{Na}_2\text{S}_2\text{O}_3$  or  $\text{NH}_4\text{Cl}$ , and variations in the water sampling time. For instance, laboratory studies<sup>8,9)</sup> have shown an increasing formation of THMs relative to the residence time, while Otson *et al.*<sup>10)</sup> reported the opposite in a field study.

This study was conducted over 10 months: 1) to determine the seasonal and spatial variations of major DBPs along a municipal drinking water distribution system; and 2) to examine the effects of major water quality parameters on the formation of DBPs.

## 2. Materials and Methods

### 2.1. Chemicals

The bromodichloromethane(BDCM), ( $\pm$ )-2-bromopropionic acid(BPA), *t*-butyl methyl ether (MTBE), chloral hydrate(CH), dichloroacetic acid (DCAA), methanol(MeOH), sodium sulfate ( $\text{Na}_2\text{SO}_4$ ), sodium thiosulfate( $\text{Na}_2\text{S}_2\text{O}_3$ ), sulfuric acid( $\text{H}_2\text{SO}_4$ ), and trichloroacetic acid(TCAA) were all purchased from Fluka; the chloroform(CF) from Junsei; the 1-bromo-3-chloropropane(BCP), dichloroacetonitrile(DCAN), and 1,1-dichloro-2-propanone(DCP) from Aldrich; and the 1,1,1-trichloropropanone(TCP) from Supelco. All these chemicals were used without further purification.

### 2.2. Field Measurements and Sampling

Four sampling sites(Fig. 1) along a municipal water distribution system were visited once a month from August 1998 to May 1999. The distribution system originates from the Yongsan water treatment plant, which was established in 1980 and supplies approximately 25,000 tons of drinking water per day to about 50,000 citizens in Chunchon, Kangwon Do. The treatment plant draws surface

water from the effluent of the Chunchon Dam and employs chlorine( $\text{Cl}_2$ ) for disinfection. The average flow rate of the water within the system was calculated to be 0.70 m/s and the total length from sampling site S-1 to S-4 was approximately 6.5 km. There was no storage tank between S-1 to S-4. Based on this data, the total residence time of the water in the distribution line was estimated to be 2.6 hours.

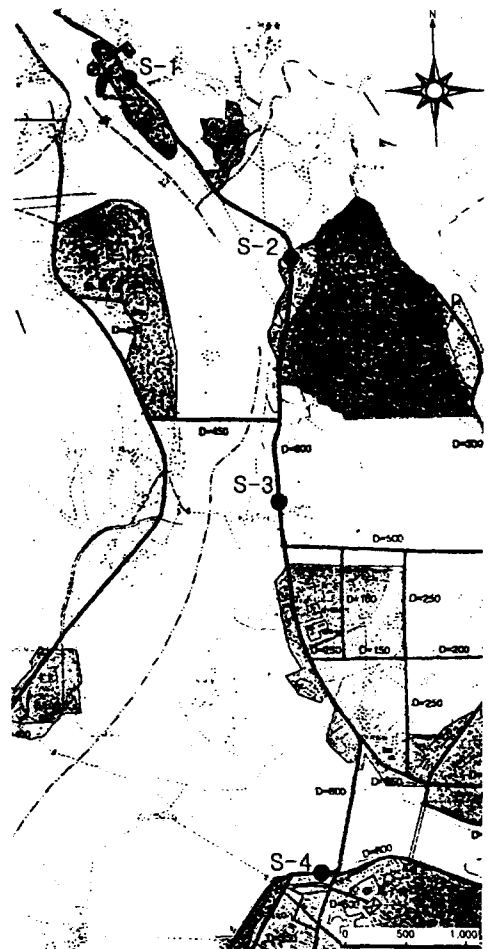


Fig. 1. Location of sampling sites(S-1 to S-4). Each sampling site was located near the main drinking water distribution system.

Some water quality measurements were made during the field trip. The pH and water temperature were measured using a portable pH meter(Model 71P, Isteck), and the total(T-Cl) and free chlorine

residuals(F-Cl) were measured using a CP-15 Chlorine Photometer(HF Scientific, Inc). The water samples were collected in 40 ml EPA glass vials containing 4 mg of sodium thiosulfate( $\text{Na}_2\text{S}_2\text{O}_3$ ) with zero head space, following at least a five-minute flushing. Care was taken not to form any bubbles in the vials. The vials were then placed in an ice box containing blue ice packs, carried to the laboratory, and stored at 4°C until extracted within 2 days.

### 2.3. Analysis of Water Samples for TOC and DBPs

The TOC in the collected water samples was measured using a TOC analyzer(5000A, Shimadzu), 24 hours after maintaining 10 ml of each water sample with 0.1 ml of 2 N HCl at 4°C.

The CF, BDCM, CH, DCAN, DCP, and TCP were all determined as follows. Twenty milliliters of water were transferred into a 40 ml glass vial and 2 ml of MTBE were added. BCP was spiked into the water as the internal standard and the mixture was vigorously shaken by hand for approximately 2 minutes following the addition of 4 g of anhydrous sodium sulfate. After the two layers were clearly separated, 1  $\mu\text{l}$  of the upper MTBE solution was injected into a gas chromatograph(GC) equipped with an electron capture detector(ECD). The analysis of the water samples for DCAA and TCAA was conducted following the procedure as described by Kim *et al.*<sup>11)</sup> except for the use of hands for shaking. ( $\pm$ )-2-bromopropionic acid(BPA) was used as the internal standard for the analysis.

The GC employed was Varian 3400CX and the ECD was equipped with  $^{63}\text{Ni}$ . The injection port was maintained at 200°C and the ECD temperature was 280°C. The Rtx-5(Restek) capillary column(30 m  $\times$  0.32mm  $\times$  0.50  $\mu\text{m}$ ) was used. Nitrogen gas (UHP grade) was used as both the column carrier(1.0 ml/min) and the ECD makeup gas with a total flow rate of 30 ml/min. The GC oven temperature programming was as follows: an initial temperature of 40°C for 8 minutes; a ramp of 6°C/min up to 150°C that was held for 4 minutes; and a second ramp of 15°C/min up to 250°C that was held for 12 minutes. The total run time was 49 minutes.

Duplicate field blank samples containing distilled and deionized water were also analyzed along with the field samples to check for any background contamination during the field trip and storage. To adjust for the extraction efficiencies of volatile DBPs and haloacetic acids, BCP or BPA was spiked into each extraction vial as the internal standard. A five-point calibration curve for each compound was prepared using the GC response(area) of the analyte divided by that of the internal standard in the chromatogram. The estimated limits of the quantification for CF, BDCM, CH, DCP, TCP, DCAN, DCAA, and TCAA were 0.61, 0.20, 0.81, 0.32, 0.17, 0.070, 1.1, and 0.16  $\mu\text{g/l}$ , respectively, based on a signal-to-noise ratio of 3.

### 2.4. Statistical Analysis

SPSS 7.5 for Windows(Korean version) was used for the statistical analysis of the data. The significance of each test was determined at a confidence level of 95%.

## 3. Results and Discussion

### 3.1. Descriptive Statistics on Water Quality Parameters and DBPs

For the eight DBPs measured, the means, medians, and standard deviations are shown in Table 1. CF, DCAA, and TCAA were identified as the major components of the measured DBPs during the study period, whereas BDCM, CH, DCAN, DCP, and TCP were only minor constituents. These results are consistent with previous reports published by Shin *et al.*<sup>12)</sup> who measured the concentrations of major DBPs at 15 water treatment plants in Korea, and Nieminski *et al.*<sup>13)</sup> who analyzed the drinking water from 35 treatment plants in Utah of the USA for DBPs. All the DBPs measured were widely distributed in their seasonal variations. The entire data set of the concentration composition of the eight DBPs is graphically shown in Fig. 2. CF was the primary constituent(31%), followed by DCAA(26%), TCAA(21%), CH(6.3%), and BDCM(5.8%).

### 3.2. Monthly Variations of DBPs

The monthly variations in the eight DBPs at each sampling site are shown in Fig. 3. Overall,

Table 1. Concentrations( $\mu\text{g/l}$ ) of disinfection by-products for entire water samples

	CF	BDCM	CH	DCAN	DCP	TCP	DCAA	TCAA
Mean	11	2.1	2.3	1.2	1.1	1.2	9.2	7.6
Median	9.5	1.9	1.6	0.89	0.32	0.90	7.9	6.5
Standard Deviation	7.7	1.1	1.7	0.89	1.7	0.98	7.3	5.7
Minimum	0.79	0.31	0.80	0.12	0.32	0.29	1.1	0.91
Maximum	42	4.9	6.4	4.8	8.7	4.8	35	30

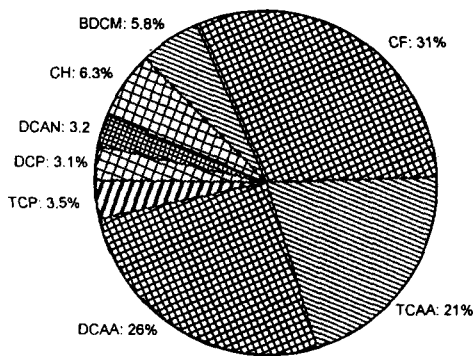


Fig. 2. Composition of eight disinfection by-products for entire data set. Chloroform, DCAA, and TCAA were identified as the major DBP constituents.

the concentrations of CF, DCAA, and TCAA were higher than the other DBPs throughout the sampling period. Interestingly, the October data showed a remarkable rise in DBPs, especially CF, DCAA and TCAA, at S-1, S-2, and S-3. However, at S-4, only CF showed a sharp elevation in October, as the DCAA and TCAA levels remained as low as the other DBPs. Also at site S-4, there were no substantial variations in the levels of CF, DCAA, and TCAA from January to May and the levels stayed relatively high. At sites S-1 and S-2 the CF, DCAA and TCAA levels were also elevated in March. Moreover, at S-3 the DCAA and CF levels were slightly higher than the other DBPs yet the TCAA level was low. All the other DBPs stayed at low levels.

### 3.3. Seasonal and Spatial Variations

The data were grouped into four seasons based on the water temperature: summer(Aug & Sept), fall(Oct & Nov), winter(Dec, Jan, & Feb),

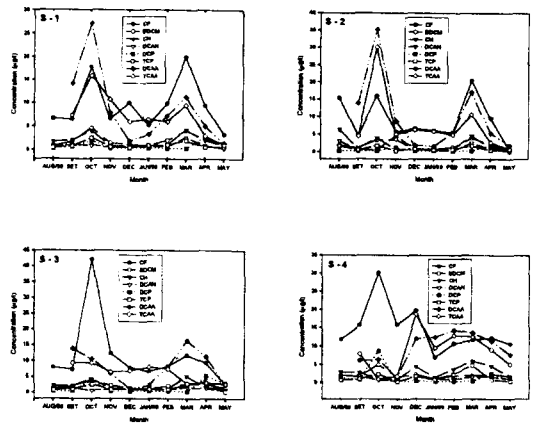


Fig. 3. Monthly variations of eight disinfection by-products at four sampling sites.

and spring(Mar, Apr, & May). The seasonal mean values, median values, and standard deviations of the measured water quality parameters along with the DBP concentrations are listed in Table 2. The mean water temperatures in the summer, fall, winter, and spring were 26, 14, 6.7, and 14°C, respectively. The pH values stayed at neutral levels (~7) in all seasons. The average TOC value was highest in winter, however, the median TOC value was highest in fall. The average and median values of the CF, BDCM, and TCAA concentrations were highest in fall. The average DCAA concentration was highest in fall, however, its median value was highest in summer. The highest CH level was recorded in winter. The average and median values of the total DBPs measured were highest in fall.

To identify any statistical differences in the TOC and DBPs among the four seasons and among the four sites, a two-way analysis of a variance (ANOVA) test was performed at a level of significance ( $\alpha$ ) of 0.05(SPSS for Windows 7.5).

The TOC, total DBPs, CF, BDCM, CH, DCAN, DCP, TCP, DCAA, and TCAA concentrations were included as dependent variables, whereas the sites and seasons were included as factors, with assumptions of normal distributions. The test results are shown in Table 3. There was no statistical difference in the mean values of any of the dependent variables at any of the four sampling sites ( $p > 0.05$ ). However, not all the mean values of the variables CF, BDCM, and the total DBPs were equal among the four seasons, thereby indicating that CF, BDCM, and the total DBPs were statistically highest in fall.

The seasonal variations shown in this study were not consistent with earlier studies,<sup>5,13</sup> where the highest DBP concentrations were recorded in the summer season. This implies that water temperature is not the only factor affecting the formation of DBPs and that other parameters such as the TOC and T-Cl may also be important in the process. The reason why there was no statistical difference

among the DBP levels at the four sampling sites is probably because the residence time (~2.6 hours) in the distribution system was too short to produce any statistical differences in the concentrations. In the study conducted by Chen and Weisel<sup>5</sup> where the total residence time was 3 days, increases were found in the THM concentrations and decreases in the non-THM concentrations with an increasing residence time. It has also been previously established that in a controlled experiment, when aquatic humic materials are chlorinated at pH 7 and 20°C, the concentrations of THMs, DCAA, and TCAA increase relative to the reaction time, whereas the DCAN and TCP concentrations decrease<sup>14,15</sup>.

#### 3.4. Relationship Between Total DBPs and Water Quality Parameters

The correlation between the TOC and total DBPs was examined. Since no statistical differences were observed among the four sampling sites, all the data were included in the analysis except for the

Table 2. Quarterly mean (median) values and standard deviations of water quality parameters and disinfection by-products ( $\mu\text{g/l}$ ) in chlorinated drinking water

Parameters or DBPs	Summer	Fall	Winter	Spring
Water temperature (°C)	26 (26) ± 1	14 (14) ± 4	6.7 (7.1) ± 1.8	14 (16) ± 3
pH	6.9 (7.0) ± 0.3	7.0 (6.9) ± 0.4	6.7 (6.5) ± 0.7	7.2 (7.1) ± 0.1
T-Cl (mg/l)	0.95 (0.94) ± 0.43	0.64 (0.73) ± 0.27	0.82 (0.87) ± 0.30	0.84 (0.88) ± 0.29
F-Cl (mg/l)	—*	—*	0.54 (0.59) ± 0.24	0.55 (0.56) ± 0.26
TOC (mg/l)	0.93 (0.92) ± 0.16	1.2 (1.2) ± 0.19	1.4 (1.1) ± 1.0	0.76 (0.76) ± 0.24
THMs				
Chloroform	9.5 (7.7) ± 4.3	18 (16) ± 12	8.7 (6.8) ± 4.7	13 (12) ± 5
Bromodichloromethane	1.6 (1.8) ± 0.5	3.2 (3.5) ± 1.3	1.2 (1.1) ± 0.5	2.8 (2.4) ± 0.9
Chloral hydrate	2.7 (2.1) ± 1.6	2.1 (1.0) ± 1.7	1.4 (0.90) ± 1.3	3.7 (4.5) ± 1.8
Dichloroacetonitrile	1.5 (1.3) ± 0.8	1.2 (0.67) ± 1.5	0.84 (0.81) ± 0.37	1.7 (1.6) ± 0.7
HKs				
1,1-Dichloropropanone	0.58 (0.36) ± 0.43	2.1 (0.80) ± 2.9	0.34 (0.32) ± 0.05	1.7 (1.2) ± 1.8
1,1,1-Trichloropropanone	0.86 (0.72) ± 0.42	1.4 (1.4) ± 1.0	0.95 (0.92) ± 0.38	2.1 (1.9) ± 1.6
Haloacetic acids				
Dichloroacetic acid	12 (14) ± 4	13 (8.3) ± 12	6.0 (4.4) ± 4.8	8.8 (9.6) ± 5.5
Trichloroacetic acid	7.3 (7.6) ± 2.0	10 (8.0) ± 9	8.5 (6.6) ± 4.3	6.7 (6.1) ± 4.2
Total DBPs	26 (28) ± 14	51 (43) ± 27	28 (21) ± 14	34 (34) ± 18

\* Not measured.

two extreme data points. Although a normality test employing the Shapiro-Wilk test showed that the TOC values were normally distributed ( $p=0.496$ ) whereas the DBPs were not ( $p=0.031$ ), it was still assumed that both variables were normally distributed. The test results showed that the TOC values and total DBPs were statistically correlated to each other with a Pearson correlation coefficient ( $r$ ) of 0.468 and  $p$ -value of 0.00527 (Fig. 4). This shows that the organic compounds in the source water are important precursors of DBPs. Likewise Krasner *et al.*<sup>16)</sup> reported a positive relationship between the concentrations of total THMs and the TOC concentration.

In addition, the correlations between each DBP were examined. Because none of the DBPs measured were normally distributed according to the Shapiro-Wilk test ( $p \leq 0.05$ ), the nonparametric correlations were examined and the Spearman correlation coefficients ( $r_s$ ) calculated (Table 4). BDCM was significantly correlated with most other DBPs, except for DCAA, and with a negative correlation with TCAA. CF was correlated with BDCM ( $r_s=0.728$ ), CH ( $r_s=0.654$ ), DCAN ( $r_s=0.441$ ), and TCP ( $r_s=0.560$ ). DCAA and TCAA were correlated with each other ( $r_s=0.590$ ). The finding that the concentrations of BDCM and CF were correlated with DCAN is consistent with the report by Krasner *et al.*<sup>16)</sup> who observed that the total THM concentration was positively correlated with the total HAN concentration. Accordingly, based on the above results it appears that the formation of these two types of DBP is affected by similar water quality parameters.

Table 3. Two-way analysis of variance (ANOVA) table for determining differences among sites and among seasons

Variable	$p$ -value		Variable	$p$ -value	
	Site	Season		Site	Season
TOC	0.442	0.056	DCAN	0.486	0.391
Total DBPs	0.644	0.038*	DCP	0.385	0.060
CF	0.287	0.022*	TCP	0.430	0.389
BDCM	0.976	0.001*	DCAA	0.938	0.203
CH	0.467	0.240	TCAA	0.824	0.331

\* Statistically significant at a confidence level of 95%.

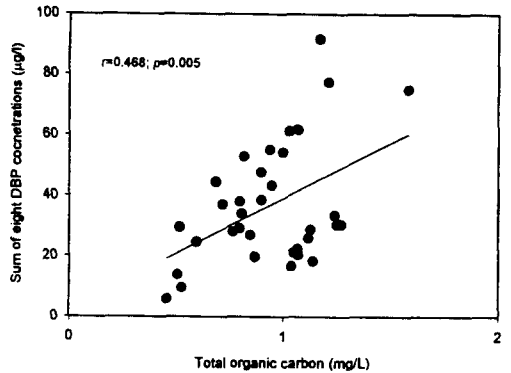


Fig. 4. Correlation between TOC and sum of eight disinfection by-products. Two variables were significantly correlated to each other with a Pearson correlation coefficient ( $r$ ) of 0.468 ( $p < 0.05$ ).

### 3.5. Factors Affecting DBP Concentrations

Based on the assumption that all the data were normally distributed, stepwise multiple regression analyses were performed to examine the effect of water quality parameters on the level of each DBP. The water temperature, TOC, T-Cl, and pH were all included as independent variables and each DBP was a dependent variable. Only pH was included in the regression models for CF (Fig. 5), BDCM, and CH (Fig. 6) at a significance level of 0.05. The Pearson correlation coefficients ( $r$ ) and  $p$ -values for CF, BDCM, and CH were 0.328 ( $p=0.039$ ), 0.366 ( $p=0.0201$ ), and 0.394 ( $p=0.012$ ), respectively. The other DBPs were not affected by any of the water quality parameters investigated in this study.

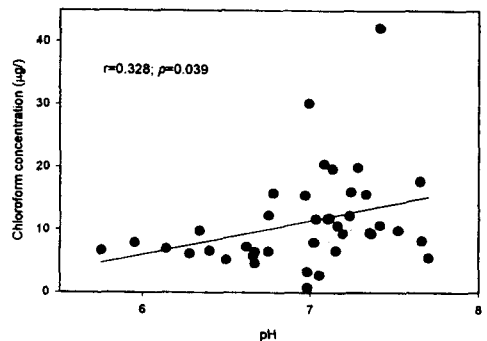


Fig. 5. Correlation between pH and chloroform concentration. Two variables were weakly yet significantly correlated each other with a  $r$  value of 0.328.

Table 4. Nonparametric correlations among concentrations of disinfection by-products

	BDCM	CF	CH	DCAA	DCAN	DCP	TCAA	TCP
BDCM	1.000							
CF	.728* (.000)	1.000						
CH	.511* (.001)	.654* (.000)	1.000					
DCAA	-.312 (.064)	-.203 (.234)	.265 (.118)	1.000				
DCAN	.497* (.001)	.441* (.004)	.665* (.000)	-.059 (.730)	1.000			
DCP	.513* (.001)	.212 (.189)	.149 (.360)	-.297 (.078)	.227 (.158)	1.000		
TCAA	-.365* (.028)	-.264 (.120)	-.082 (.635)	.590* (.000)	-.250 (.127)	-.222 (.193)	1.000	
TCP	.506* (.001)	.560* (.000)	.589* (.000)	-.005 (.976)	.358* (.024)	.060 (.712)	-.147 (.392)	1.000

\* Significant at  $\alpha=0.05$ .

\*\* Numbers in each cell indicate Spearman correlation coefficients ( $r_s$ ) and  $p$ -values (in parenthesis).

These results are consistent with the study by Reckhow *et al.*,<sup>14</sup> who observed that CF formation is facilitated by a high pH, whereas the formation of TCAA and TCP is hindered by a high pH. They proposed that CF and TCAA are the major species at basic and neutral pHs, respectively. They also showed that TCP and CH are the precursors of CF and TCAA, respectively. Therefore, it would appear that pH is an important water quality parameter in controlling the composition of DBPs in chlorinated water.

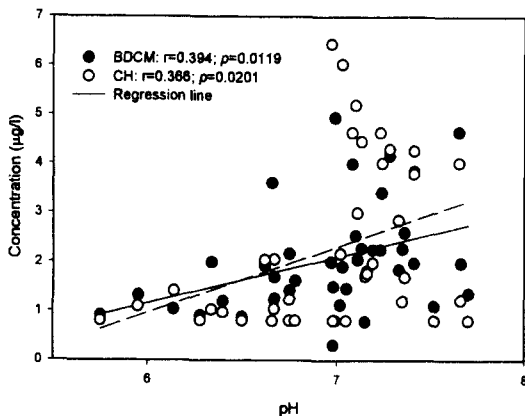


Fig. 6. Correlation between pH and BDCM/CH. The two DBPs were weakly yet significantly correlated to pH with  $p$ -values of less than 0.05.

#### 4. Conclusions

The following conclusions were drawn from this study:

- 1) Chloroform, dichloroacetic acid, and trichloroacetic acid are the major components of disinfection by-products (DBPs) in chlorinated drinking water.
- 2) This study revealed that the levels of total DBPs were highest in fall rather than in summer, probably due to a high organic carbon content, thereby indicating that water temperature is not the sole critical factor in the formation of DBPs.
- 3) The formation of DBPs is facilitated by high levels of organic precursors.
- 4) pH is an important determinant of the composition of DBPs. A high pH favors the formation of THMs and CH.

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