



Characteristics of DBPs reduction of AOM by dissolved air flotation

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ABSTRACT

Algae increase the concentration of dissolved organic carbon (DOC), which causes unpleasant tastes and odors in water. This DOC is known as algal organic matter, which comprises extracellular organic matter and intracellular organic matter. Carbonaceous disinfection by-products (C-DBPs) and nitrogenous DBP (N-DBPs) are produced from DOC by disinfection processes, and N-DBPs are about 140 times more toxic than C-DBPs. We compared the levels of N-DBPs when dissolved air flotation (DAF) or conventional gravity sedimentation was used to remove algae before/after chlorination. The degradation of algal cells by chlorine treatment increased the DOC level, which was attributable to DBPs/DBP formation precursors (DBPFPs). The levels of N-DBPs such as dechloroacetonitrile DCAN and C-DBPs such as trihalomethanes increased with the amount of chlorine applied. Haloacetonitriles increased continuously after treatment because the residual chlorine reacted with dissolved organic nitrogen. C-DBPs and N-DBPs were reduced by inter-chlorination rather than pre-chlorination. Extracting algae from source waters is effective for reducing the chlorine dose and the levels of DBPFPs in drinking water treatment processes. The application of DAF prior to inter-chlorination greatly reduced the N-DBP levels in source water containing algae.

Keywords: Algal organic matter; Carbonaceous disinfection by-product; Disinfection by-product formation precursor; Dissolved air flotation; Nitrogenous disinfection by-product

1. Introduction

The excessive growth of algae in intake reservoirs and rivers is associated with eutrophication, and it is an increasing problem that affects drinking water

treatment processes. Algae increase the concentration of dissolved organic matter, which has negative effects on coagulation processes and it produces algal toxins [1–3], as well as unpleasant tastes and odors [4–6]. The algal metabolites also increase the requirement for coagulants, reduce the length of filter runs because of

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clogging, and increase the need to manage disinfection by-products with variable chlorine demands [7–9].

Conventional treatment processes cannot remove algae and algal by-products completely, so the presence of algae needs to be minimized [10]. In particular, trihalomethanes (THMs) that originate from soluble or biodegradable organic matter may be attributable to algae. Various methods have been investigated for removing algae, such as dissolved air flotation (DAF), pre-ozonation coupled with flotation/filtration, and pre-oxidation. However, the by-products produced during the pre-oxidation of algal organic matter (AOM) are harmful to human health [11].

AOM comprises extracellular organic matter (EOM) and intracellular organic matter (IOM), which is present at high concentrations in some water sources. EOM comprises the metabolites excreted by algal cells into the surrounding environment [12]. IOM may be released after algal cells die and subsequently lyse. The disinfection of algae-laden water by ozonation and chlorination produces many disinfection by-product DBPs, including carbonaceous DBPs (C-DBPs) and nitrogenous DBPs (N-DBPs) [13–15]. The dissolved organic nitrogen produced by the breakdown of algal cells via oxidation can increase the production of N-DBPs [16]. It has also been reported that N-DBPs are about 140 times more toxic than C-DBPs [17].

Various techniques have been used for algae removal to minimize the levels of DBPs produced by drinking water treatment processes [18]. DAF may be a solution for collecting algae without breaking the algal cells [19]. The short process time of DAF treatment is effective for removing dissolved organic matter, which is followed by rapid filtration [20].

In the present study, we compared the production of N-DBPs when DAF or conventional gravity sedimentation (CGS) was used to remove algae before/after chlorination. *Microcystis* sp. and mixtures of algae were selected and cultured for 30 d in the laboratory. Chlorine was then injected into the two algal solutions to monitor DBP production before/after DAF and CGS treatments.

2. Material and methods

2.1. Algae culture

A mixture of algae was cultivated in Jaworski's medium. In natural systems, *Oscillatoria* spp. and *Microcystis* spp. are the dominant species. Thus, these were mixed with *Pediastrum* spp. and *Scenedesmus* spp. in the algae mixture solution. The solution was cultivated for 30 d at a temperature of $27 \pm 2^\circ\text{C}$ with light,

i.e. 1,300–1,500 lux, for 14 h and darkness for 10 h each day. The initial concentration of chlorophyll-a (Chl-a), i.e. 102 $\mu\text{g/L}$, increased to 700 $\mu\text{g/L}$ after subculture (1:4) for 20 d.

After sieving the mixed algae solution through a 75 μm filter, the filtered algae were rinsed with distilled water five times. The residue was mixed with 300 mL distilled water and placed in a darkened area of a refrigerator.

Microcystis spp. was cultured from 112.3 $\mu\text{g/L}$ Chl-a to 400 $\mu\text{g/L}$ for 30 d. The culture was centrifuged and concentrated to obtain a high concentration stock. The algae mixture and *Microcystis* spp. were discarded when the dissolved organic carbon (DOC) concentration began to degrade after 36 d.

2.2. Carbon and nitrogen composition of EOM/IOM in algae

The stock was distributed in seven 1 L volumetric flasks with doses ranging from 1 to 7 mL, with 1 mL increments. The flasks were filled with distilled water and mixed in a shaking incubator for 4 h at 25°C and 150 rpm, and these solutions are referred to as AOM solutions. Next, 200 mL of the supernatant water were filtered using a 0.45 μm syringe filter and the filtrates are referred to EOM solutions (Non-filter solution are referred IOM).

The AOM and EOM solutions were used to analyze the total organic carbon (TOC; TOC-VcpH SSM-5000A; Shimadzu Co., Japan), UV absorbance at 254 nm (UV spectrophotometer, DR5000TM, Hach Co., USA), and Chl-a was analyzed according to standard methods [21].

2.3. Analysis of DBP formation precursors produced from algae during chlorination

The concentrations of C-DBPs and N-DBPs were monitored while chlorine was added to the EOM and IOM solutions. Chl-a is used as index to measure identically both *Microcystis* spp. and algae mixtures. The Chl-a concentrations of the *Microcystis* spp. and algae mixtures were determined after diluting to 150 $\mu\text{g/L}$, respectively. A NaOCl 1,000 mg/L stock solution was used to chlorinate both solutions with doses of 1, 3, 5, 7, and 10 mg Cl_2/L . The levels of DOC, dissolved total nitrogen (DTN), THMs, and haloacetonitriles (HANs) were analyzed after shaking for 4 h and filtering through a 0.45 μm membrane filter. The UV₂₅₄, total nitrogen (TN), and DTN were measured with a UV spectrophotometer (DR5000TM, Hach Co. USA). Chl-a was analyzed according to

standard methods. The turbidity was measured with a turbidimeter (2100N, Hach Co., USA). THMs and HANs were detected by GC-MS (Purge and Trap, OI Analytical, Shimadzu Co., Japan) and GC-ECD (GC-ECD, Shimadzu Co., Japan), respectively.

2.4. Comparison of DAF and CGS

The efficiencies of the reduction of DOC, DTN, THMs, and HANs by DAF and CGS were evaluated at different sampling times after chlorination using the IOM and EOM solutions. Alum ($\text{Al}_2(\text{SO}_4)_3 \cdot 16\text{--}18\text{H}_2\text{O}$) was used as a coagulant with a 0.1 M stock solution. Previous studies suggest that 2 mg Al/L is the optimum dose for coagulation [22,23]. In the present study, the optimum dose of alum was determined by a jar test where the solution had a Chl-a concentration of $150 \pm 5 \mu\text{g/L}$. NaOCl (i.e. 0, 5, and 10 mg Cl_2/L) was used to dose the solution with rapid mixing for 10 min at 150 rpm and slow mixing for 15 min at 40 rpm. The coagulant was injected before rapid mixing, where the pH was maintained at 7.0 ± 0.5 with 0.1 N NaOH or 0.1 N HCl. For DAF, the recycling rate of the reactor and the pressure in the saturator were 10% and 5 atm, respectively. Samples were collected at 0.5, 1, 3, 5, 10, 30, and 60 min with DAF and CGS. The TOC, TN, turbidity, and Chl-a results were compared at different sampling times for the two processes.

2.5. DBP reduction at different chlorine injection points

Pre-chlorination and inter-chlorination are used to oxidize metal ions and dissolved organics, and to enhance coagulation during water treatment processes. If mixed algae are present in the source water, it is necessary to evaluate the levels of DBPs and DBP formation precursors (DBPFPs) produced at different chlorine injection points with the same $C \times T$ value. In the present study, C-DBPs, N-DBPs, and DBPFPs were monitored based on the levels of TOC, TN, THMs, and HANs. Table 1 shows the experimental conditions at different chlorine injection points.

The chlorination stages and the four sampling points are shown in Fig. 1 [24]. The experiment was

performed in the laboratory at 25°C. DAF or CGS was applied before or after chlorine injection at different points using solutions with Chl-a contents of 150 $\mu\text{g/L}$. The conditions used for DAF and CGS were the same as those employed in the experiments described above. Rapid filtration was performed, where the sand filtration depth was 50 cm and its surface loading rate was 122 $\text{m}^3/\text{m}^2/\text{day}$. Samples were taken and analyzed to determine the levels of DOC, DTN, THMs, and HANs at points 1–4.

3. Results and discussion

3.1. C and N compounds in EOM/IOM from algae

Fig. 2 shows the concentrations of EOM and IOM, which comprised C and N compounds from cells, with increasing levels of Chl-a. The C:N ratio was 1:4 for IOM and 1:1 for EOM. The concentration of EOM was lower than that of IOM because the mixed algae solution was diluted with distilled water. The EOM:IOM carbon ratio was 1:6 and that of nitrogen was 1:2.5. The C and N compounds in IOM and EOM were attributable to DBP and DBPFPs [12].

3.2. C and N DBPs and DBPFPs produced by chlorination

The concentrations of DOC and DTN were changed by chlorination in the mixed algae and *Microcystis* spp solutions. Table 2 shows the Chl-a, DOC, and DTN levels with different chlorination dosages.

The initial DOC concentrations in the mixed algae and *Microcystis* spp. solutions were 2.21 and 1.86 mg/L, respectively. However, the concentrations increased to 2.55 and 2.21 mg/L, respectively, after dosing with 1 mg/L chlorine. The increase was greater during the initial stage than the final stage in both solutions. The application of a chlorine dosage of 10 mg/L to the mixed algae solution increased the DOC level by 1.3 times and the DTN level by 2.3 times. Chl-a was oxidized by chlorine and the levels of DOC and DTN increased. The degradation of algal cell after the injection of 1 mg/L chlorine increased the DOC concentration, which was attributable to DBPFPs. Table 3 shows that the chlorophyll was discolored and the cells were

Table 1
Experimental condition depending on different point of chlorine injection

Point no.	Process	Mode 1	Mode 2	Mode 3	Mode 4
Point 1	Pre-chlorination	10 mg/L Cl	10 mg/L Cl	0 mg/L Cl	0 mg/L Cl
Point 2	Flotation/sedimentation	Flotation	Sedimentation	Flotation	Sedimentation
Point 3	Inter-chlorination	0 mg/L Cl	0 mg/L Cl	10 mg/L Cl	10 mg/L Cl
Point 4	Rapid filtration	Surface loading rate 122 $\text{m}^3/\text{m}^2/\text{day}$			

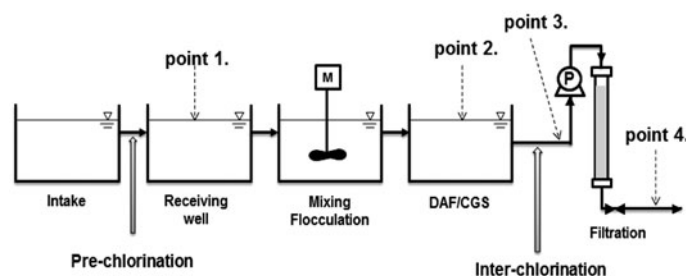


Fig. 1. Sampling point of treatment process on chlorine injection.

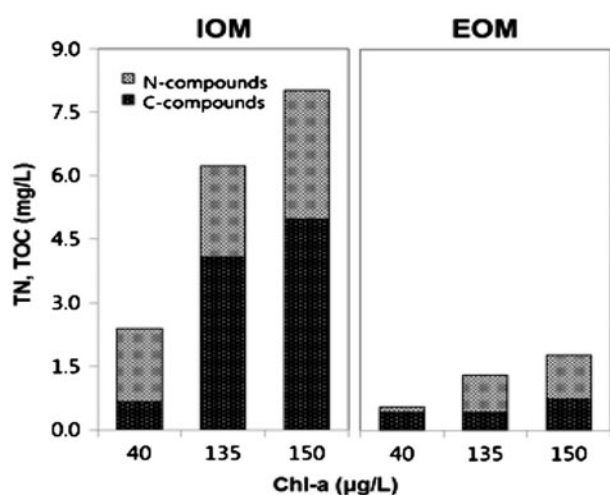


Fig. 2. C and N compounds depending on chlorophyll-a concentration.

broken as the chlorine concentration increased. The levels of dichloroacetonitrile (DCAN), THMs increased with chlorine treatment in a linear manner, as shown in Fig. 3. The ratio of C-DBPs relative to N-DBPs was 100:1.

The levels of THMs and HANs increased to 240 and 2.4 µg/L, respectively. Thus, C-DBPs were produced more readily compared with N-DBPs initially. However, the final N-DBP levels were 1.7 times greater than those of C-DBPs. The DOC:DTN ratio remained the same as the level of chlorine increased.

3.3. Comparison of the effects of DAF and CGS at different sampling times


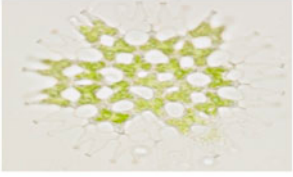

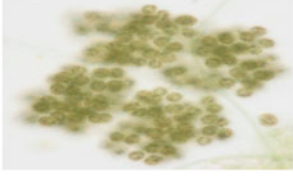
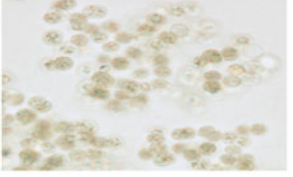
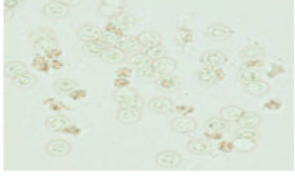



The effects of DAF and CGS without chlorination at different sampling times are shown in Fig. 4. The levels of Chl-a, TOC, and TN were analyzed with an influent Chl-a concentration of 150 ± 5 µg/L. We found that 83% of the Chl-a was removed within 1 min when DAF was applied to the sample whereas a reduction of 81% was achieved within 5 min with CGS. DAF showed 80% removal rate within 2.5 min whereas CGS showed 30% removal rate. TN was also shown similar result with TOC. It means that TN and TOC could be seemed to be same as result by DAF and CGS. Algae comprising characteristics of various EOM and IOM fractions shows that TOC and TN can be happened to DBP formation [24]. Recent studies [25] have reported that 98% of blue-green algae were removed by DAF but only 70–94% were removed by CGS. Edzwald [19] also reported that DAF removed 90–99% of various types of algal cells whereas CGS had a removal rate of 60–90%.

The coagulation chemistry is the most important operational control variable that affects the flotation performance. Without coagulation, the algae possess a negative charge and they are often hydrophilic, which means that bubble attachment is poor. Good coagulation chemistry depends on the pH and the coagulant dose. The optimum coagulation conditions with an appropriate pH and coagulant dose produce flocs with neutral charge and high hydrophobicity. A high

Table 2
Contents of algae mixture and *Microcystis* sp.

Solutions	Algae mixture			<i>Microcystis</i> sp.		
	0 mg/L	1 mg/L	10 mg/L	0 mg/L	1 mg/L	10 mg/L
Chlorophyll <i>a</i> (µg/L)	147	79	8.7	150.1	44	7.9
DOC (mg/L)	2.21	2.55	2.98	1.86	2.21	2.62
DTN (mg/L)	0.44	0.86	1.00	N.D	0.35	0.80

Table 3
Damage and discoloration of algae cell by chlorine dosage

	Chlorine 0 mg/L	Chlorine 5 mg/L	Chlorine 10 mg/L
<i>Pediastrum</i> sp.			
<i>Microcystis</i>			
<i>Scenedesmus</i> sp.			

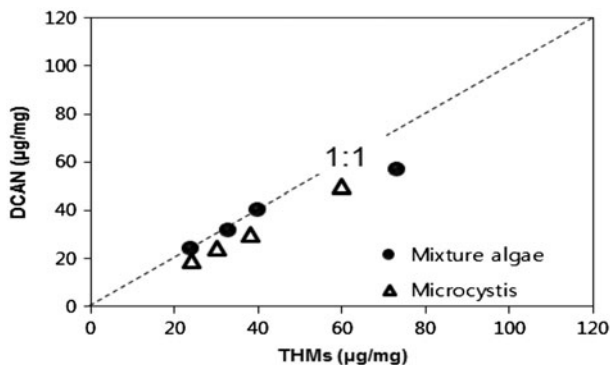


Fig. 3. THM/CF (C-DBP) vs. DCAN (N-DBP) production by chlorination.

SUVA (Specific UV Absorbance) value indicates the hydrophobicity of a source water after chlorine dosing. It also can evaluate whether TOC and DBP precursor concentrations could be correlated with UV_{254} . Fig. 5 shows that the SUVA decreased from 4.48 to 3.99 L/mg/m, which indicated that a high coagulant dosage was required. UV_{254} has found increasing acceptance and is now widely used for predictor of TOC and THM precursors. It was also used for setting coagulant dosages for water supply in which TOC control coagulation [26].

Dosages of 0, 5, and 10 mg Cl_2 /L were injected into the mixed algae solution and coagulated in a jar

test. Without chlorination, the algae were flocculated with an Al dose of 0.5 mg, as shown in Fig. 5. With chlorine levels of 5 and 10 mg/L, flocs were formed with Al doses of 1.25 and 1.8 mg, respectively. Thus, the requirement for Al was increased with high concentrations of chlorine because the TOC increased, as shown in Fig. 6. A previous study showed that 90% of the TOC produced by *Chlorella zofingiensis* was removed using DAF by controlling coagulation using alum [27].

The decomposition of algae by chlorination increases the DOC level and produces DBPFPs. Thus, extracting algae from source waters is an effective approach that reduces the chlorine dosage required and the levels of DBPFPs in drinking water treatment processes.

3.4. DBP production by pre-chlorination or inter-chlorination in water treatment processes

The chlorine injection stage affects DBP production if algae are present in a water source. Fig. 7 shows the DTN concentrations at different sampling points. The DTN concentration was lower with inter-chlorination compared with pre-chlorination. This is because DAF and CGS have important roles in removing algae as nitrogen sources before degrading the cells by chlorination. Pre-chlorination of the algal cells increased the levels of DOC and DTN. The reduction in the DTN

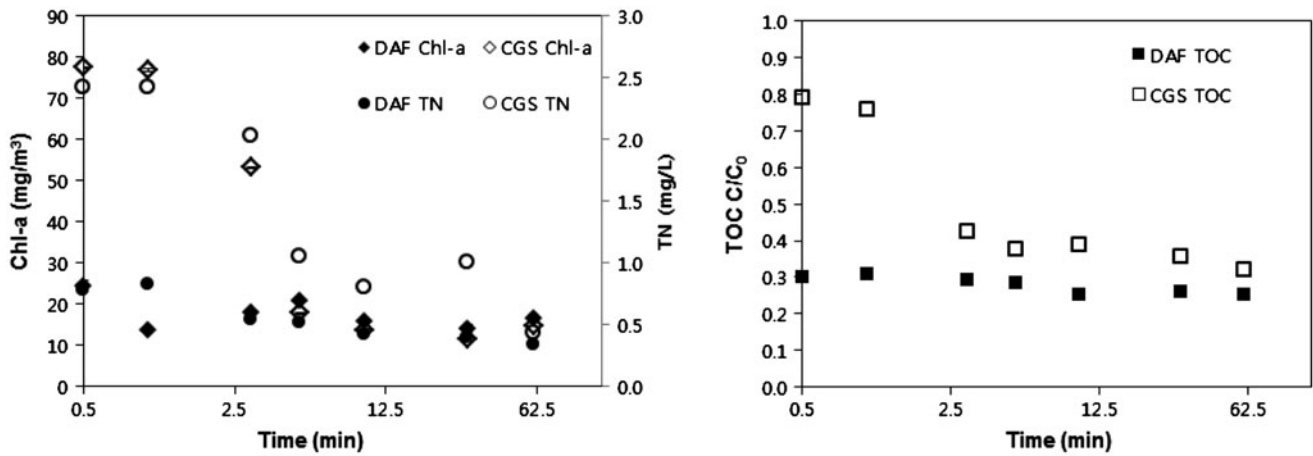


Fig. 4. Chl-a, TN, and TOC removal on retention time by DAF/CGS.

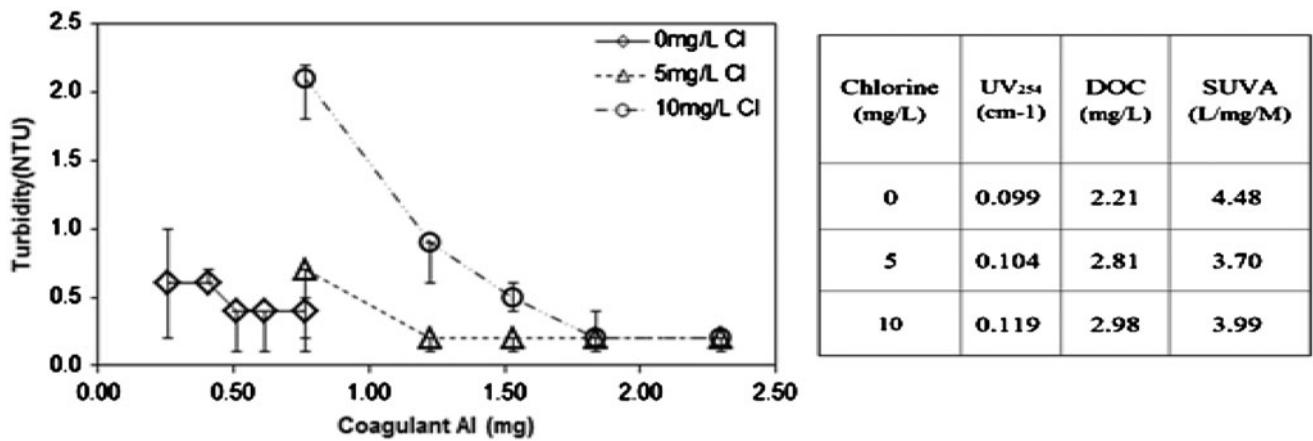


Fig. 5. Al, UV₂₅₄, DOC, SUVA, and turbidity on chlorine dose.

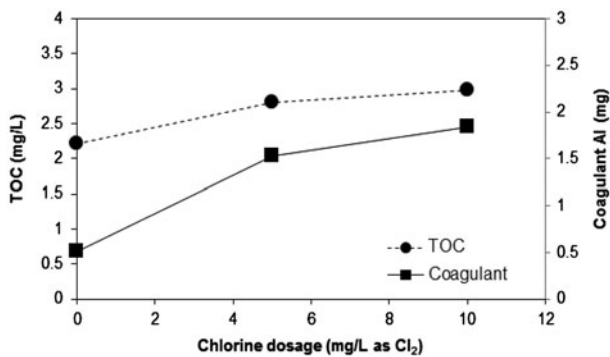


Fig. 6. Optimum coagulant dosage and TOC changes in chlorine concentration.

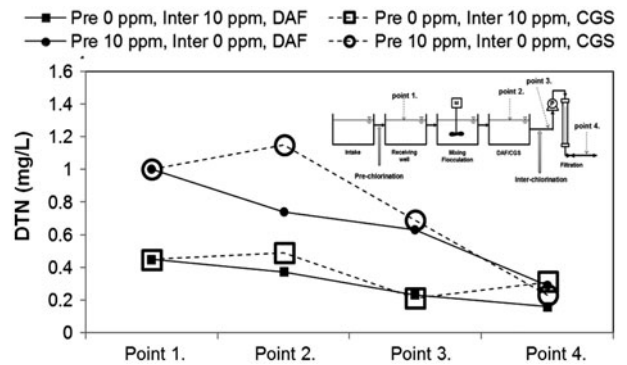


Fig. 7. DTN changes on chlorine injection.

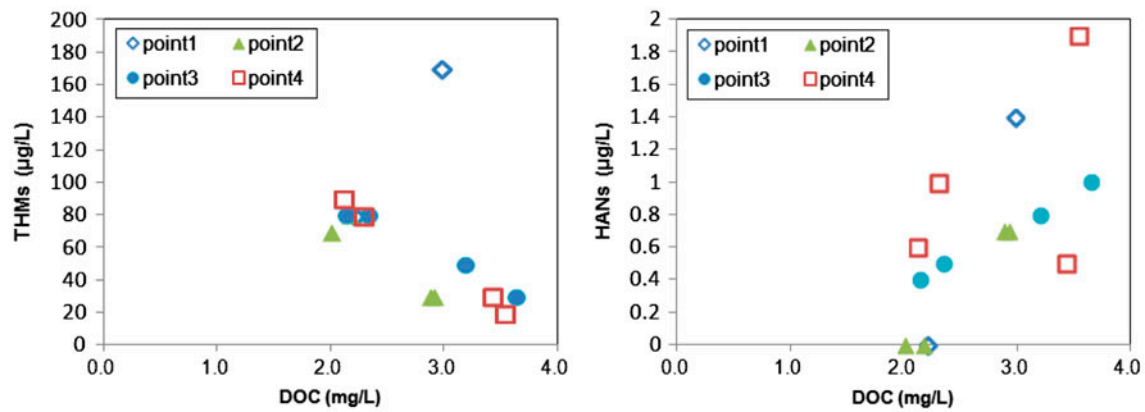


Fig. 8. THMs/HANs vs. DOC.

level was higher with DAF than that with CGS when using pre-chlorination. However, there was little difference between the two processes when using inter-chlorination. The DTN level decreased continuously with time because of the oxidation of residual chlorine, even after the removal treatment was complete.

The production of C-DBPs and N-DBPs varied among sampling points, as shown in Fig. 8. Sampling points 3 and 4 had lower concentrations of C-DBP but higher concentrations of N-DBP. This is because algae

oxidized from chlorine were decomposed to dissolved organic matter. DAF can remove algae contained by N-DBP effectively rather than CGS in the post process. The characteristics of DCAN formation were consistent with our expectations based on a presumed proteinaceous precursor. The by-products formed slowly over a period that ranged from hours to days and their formation was highly dependent on the chlorine concentration. The THMs increase at high pH because many hydrolysis reactions actually lead to THM

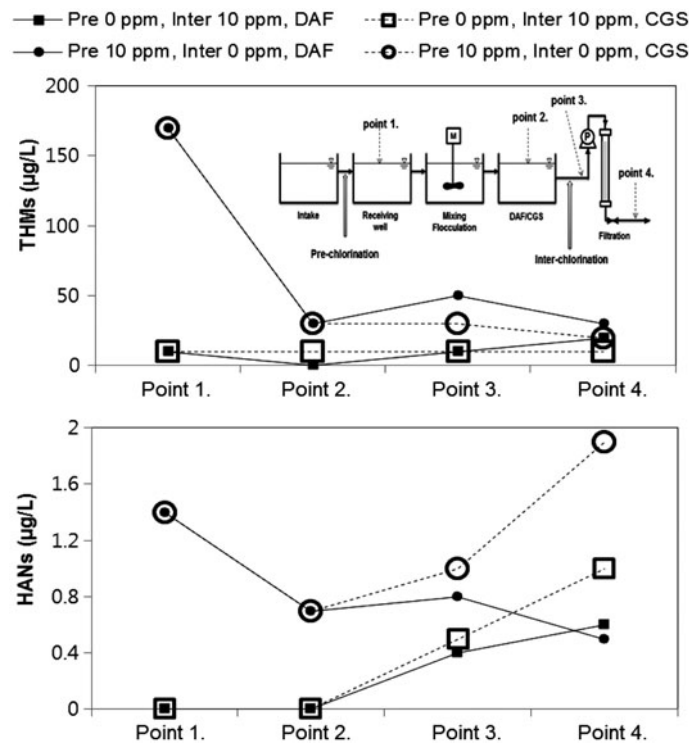


Fig. 9. C and N-DBPs on chlorine injection.

formation. The reactions for producing C-DBPs proceeded at a faster rate with a higher pH [28], however, pH should be kept at the initial condition (7.0 ± 0.5). It means that there were no specific changes during experiment. Fig. 7 shows that the DTN concentration decreased continuously because of these reactions. However, the algae were removed by DAF or CGS before inter-chlorination, which explains why the initial concentration of DTN was low. The DTN level decreased continuously as N-DBPs were produced after treatment with DAF or CGS.

A Chl-a concentration of $145 \mu\text{g/L}$ reacted with 10 mg/L chlorine to yield $170 \mu\text{g/L}$ THMs. The THMs produced comprised 97–98% chloroform and 2–3% bromodichloromethane. In addition, 90% of the Chl-a was removed by chlorination and the levels of the DBPs increased 17 times. Chlorination produced $1.4 \mu\text{g/L}$ DCAN, which comprised 99% of the HANs. As a general way, disinfection dose operates a greater role in DBP formation during primary disinfection than during secondary disinfection. When chlorine is added from 3 to 5 mg/L , THMs can be produced in direct proportion to that dose, however, when chlorine excesses over 5 mg/L , the extent of THM formation with increasing dose levels off. In a previous study, the level of DCAN production was also higher than that of trichloroacetonitrile among the HANs produced by AOM oxidation of *Microcystis aeruginosa* using chlorine [29].

Amino acids and fatty acids can react with dissolved organic nitrogen to produce monochloramine (RNHCl) and dichloramine (RNCl_2). Haloaldehydes and DCAN may be produced as by-products of the chloramine production process [30,31]. Fig. 9 shows that the concentration of HANs was decreased after the DAF process, whereas it increased after the CGS process. This was because more DOC and DTN were removed by DAF compared with CGS. The inter-chlorination experiment shows that the levels of HANs still increased after CGS.

4. Conclusion

For IOM, the ratio of C:N was 1:4 and that of EOM was 1:1. The EOM:IOM carbon ratio was 1:6 and that of nitrogen was 1:2.5. The algal cells were degraded by chlorine injection and the increase in the DOC was attributable to DBPs and DBPFPs. The levels of N-DBPs such as DCAN and C-DBPs such as THM (e.g. chloroform) increased constantly with the chlorine concentration. The SUVA value indicates the hydrophobicity of a source water after chlorine dosing. In this study, an increase in the amount of

chlorine reduced the SUVA value and decreased the dosage of coagulant required. The levels of HANs increased continuously after treatment because residual chlorine reacted with dissolved organic nitrogen. The DTN concentration was reduced more by DAF than CGS. The levels of C-DBPs and N-DBPs were decreased more by inter-chlorination than pre-chlorination. Extracting algae from source waters is effective for reducing the chlorine dosage required and the DBPFP levels during drinking water treatment processes. The application of DAF prior to inter-chlorination reduced the levels of N-DBPs greatly in treated water.

Acknowledgments

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