Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2013.826327

52 (2014) 990–998
January

Taylor & Francis
Taylor & Francis Group

Pilot study on the removal of TOC, THMs, and HAAs in drinking water using ozone/UV–BAC

Vu Nha Trang, Nguyen Phuoc Dan*, Lai Duy Phuong, Bui Xuan Thanh

Faculty of Environment, Ho Chi Minh City University of Technology (HCMUT), Building B9,
268 Ly Thuong Kiet st., District 10, Ho Chi Minh City, Vietnam
Email: npdan@hcmut.edu.vn

Received 9 March 2013; Accepted 13 April 2013

ABSTRACT

The contaminated water is a main source of many waterborne diseases such as cholera, typhoid, etc. causing many serious public health crises, especially in developing countries with the low hygiene condition. In Vietnam, previous studies have reported that industrial and domestic waste cause adverted pollution on Saigon River that supplies water for 30% of 10 million residents in Ho Chi Minh City. Annual monitoring water quality data during 2006–2011 showed that concentration of ammonia and organic matter has been significantly increased and over the allowable limit (COD > 10 mg/L). In order to reduce these pollutants in the finished water and for disinfection, a large dose of chlorine (about 3–6 mg/L) needs adding in the treatment process at Tan Hiep water treatment plant. A negative effect of using too much chlorine is the formation of disinfection by-products (DBPs) such as trihalomethanes (THMs) and haloacetic acids (HAAs). The concentration of THMs in finished water of Tan Hiep plant ranged from 50 to 200 μg/L. In this study, a pilot-scale plant using Ozone/UV process combined with biological activated carbon filter (BAC) was studied on eliminating the organic matters and the DBPs formation in drinking water. With the feed water taken after sedimentation, the combination of Ozone/UV–BAC proved to be very effective in TOC, DOC, UV_{254}, and SUVA removals with percentage reduction of 19.1, 17.6, 30.7, and 16.4%, respectively. The removal efficiency of the combined process was always higher than the sum of treatment efficiency of each process (Ozone/UV or BAC) alone. It was confirmed that the pretreatment of Ozone/UV could help to increase the organic removal of BAC filter. According to the organic reduction, the concentration of DBPs in water has decreased considerably with 70.6% (THMs) and 67.7% (HAAs) removal. DBPs formation potential decreased 39.3% (THMFP) and 46.1% (HAAFP).

Keywords: Ozone/UV; Biological activated carbon; THM; HAAs; Natural organic matter

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*Corresponding author.

Presented at the Fifth Annual International Conference on "Challenges in Environmental Science & Engineering—CESE 2012" Melbourne, Australia, 9–13 September 2012

1. Introduction

Disinfection is a key process to produce safe drinking water without waterborne diseases for consumers. This stage is very necessary to reduce natural organic matters (NOM) presented in raw water and prevent the regrowth of bacteria in the distribution system [1]. However, the common use of chlorine as a disinfectant could lead to accelerate the formation of disinfection by-products (DBPs), when chlorine species (OCl^-/HOCl) react with the dominant fraction of aquatic NOM comprised of humic and fulvic substances [2,3]. The significant increase in NOM in raw water will descend the efficiency of water treatment processes, increase the demand of coagulant or disinfectant and enhance the formation of problematic biofilms in bacteria reproduction. On the other hand, it is well known that most of NOM is not easily removed by the conventional treatment process of coagulation–sedimentation followed by rapid sand filter due to its various composition, size, polarity, and reactivity.

The presence of DBPs in drinking water is of great concern because of their potentially carcinogenic, and adverse effects on human health. Major halogenated DBPs in water commonly identified are trihalo-methanes (THMs), haloacetic acids (HAAs), haloacetonitriles (HANs), cyanogen halides, etc. Among them, species of THMs which are comprised of chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$), dibromochloromethane (CHBrCl2), and bromoform (CHBr3) are considered the most conventional issue of researches. Because of the serious health risk which is represented by THMs, regulatory action has been taken to control the levels of them in treated drinking water. The US Environmental Protection Agency (USEPA) has set a maximum contaminant level of 80 μg/L for total THM (TTHM) in the stage 1 of disinfection process [2,4]. The Commission of European Communities has also proposed a Council Directive with parametric values of 40 μg/L $CHCl_3$ and 15 μg/L $CHBrCl_2$, while other countries may have different limits [5]. In Vietnam, the standard level of TTHM in drinking water is 460 μg/L ($CHCl_3$ 200 μg/L). Previous studies have shown the importance of several parameters for the formation of THMs such as chlorine, contact time, residual chlorine, bromide, ammonia, pH, temperature, content, and type of NOM. It means that levels of THMs were largest at the extremity of the distribution system corresponding to the longest retention time. Besides, THMs once formed are very difficult to eliminate. Thus, it may be compulsory for any effective treatment strategy removing THMs level in drinking water to focus on hindering their formation by mitigating or degrading the precursors.

Advanced oxidation processes (AOPs) have been employed in drinking water treatment to eliminate the organic pollutants as well as remove DBP precursors for decades [6,7]. Various combinations of oxidants, radiation, and catalyst has been studied and developed for NOM removal. These processes involve the generation of highly reactive powerful oxidizing species, especially the hydroxyl radicals (OH) which are very nonselective oxidants. Generally, these oxidation processes are not used for the total oxidation of organic matters into carbon dioxides. However, they have changed the characteristic of NOM and impact their reactivity with chlorine [7]. The large molecular weight constituents of NOM were broken into smaller and more biodegradable compounds such as aldehydes or carboxylic acids during partial oxidations. Among the various AOPs, it has been well established that ozonation in water would result in considerable organic matter reduction and biodegradability improvement. In particularly, the photo-catalytic oxidation is considered effective in degrading refractory organic substances, and photochemical treatment has been applied to increase the biodegradability of various recalcitrant pollutants [8]. Compared with other AOPs, the processes based on the combination of ozone (O_3), hydrogen peroxide, ultraviolet light (UV), Fenton's reagent, etc. were proven more effective in removing organic pollutants than treatment of each oxidant alone [7]. The combined O_3/UV has been observed to result in a significant mineralization of DOC and reduction of formation potential of THM and HAA [9]. It has also been reported that O_3/UV also known as photolytic ozonation offered a relatively higher in biodegradability improvement than others. The O_3/UV combination effectively destroys organic contaminants largely because of the very high oxidation potential of the hydroxyl radical formed, when ozone decomposed in water [10,11].

Furthermore, AOPs may find better application in combination with other treatments to enhance their efficiency for NOM removal. It is well known that AOPs have significant potential in improving the biodegradability of NOM. Therefore, AOPs can be used as a pretreatment to reduce NOM, when the resulting intermediates are easily degraded in a further biological treatment. It been suggested that AOP pretreatment followed by biodegradation minimizes chlorine dosage and diminishes bacteria regrowth in the distribution system. The more biodegradable compounds are readily utilized as major food sources by micro-organisms in the biological activated carbon (BAC) filtration [6]. Besides BAC treatment has been used widely for various processes to purify drinking water, because it brings lower cost and higher performance than normal activated carbon adsorption treatment. Recent researches have shown that BAC process reduces effectively biodegradable organic matters, and DBP precursors mostly by bacteria metabolism beside adsorption treatment by activated carbon. It is considered that BAC treatment combined with AOP is especially effective, and this process has been applied at several municipal water treatment plants to mitigate dosage of chlorine for disinfection, to manage taste and odorous substances and to avoid the formation of DBPs. The combination of O_3/UV based on AOP and BAC treatment shows significant reduction of TOC, DOC, UV_{254}, and DBPs compared with the O_3/UV or BAC treatment alone. Hence O_3/UV-BAC process has been suggested a promising treatment for the reduction of DOC, harmful DBPs as well as their precursors.

However, few studies have been reported on applying these advanced processes in drinking water treatment in Vietnam. Previous researches shown that the water quality of Saigon River has been degraded with high content of NOM, and high demand of chlorine used; thus, the increase in DBP formation regard to long retention time of chlorinated water in distribution system could harm the public health [12]. This study aimed to evaluate NOM and DBPs, DBP formation potential (DBP-FP) removal of a pilot-scale plant using O_3/UV coupled with BAC with capacity ranging from 6 to 10 m^3/d. The feed water was taken from the effluent after sedimentation tank at Tan Hiep water treatment plant, Ho Chi Minh City. The effectiveness of each unit operation (O_3/UV or BAC) was examined in terms of TOC, DOC, and UV_{254}, and DBPs (THM, THM formation potential [THMFP], HAA, and HAA formation potential [HAAFP]).

2. Materials and method

2.1. Pilot-scale experiment

A pilot-scale experiment with capacity ranging from 6 to 10 m^3/d was set up at Tan Hiep Water Treatment Plant which takes raw water from Saigon River and providing daily 300,000 m^3 of clean water for Ho Chi Minh City use. The experiment included two separate lines: (1) Line I: an ozone/UV reactor followed by a BAC filter, and (2) Line II: only a BAC column, as a comparative Lines (Fig. 1).

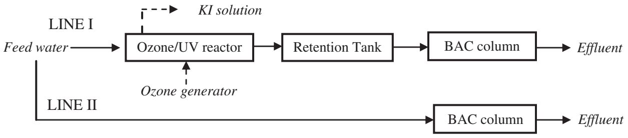

Fig. 1. Schematic diagram of the pilot-scale plant.

The ozone/UV reactor has a working volume of 127 L (diameter × height = 450 mm × 800 mm). The feed ozone dosage used was 2.7–3.0 mg/L using an ozone generator with capacity of 50 W, equivalent to the ozone production of 1.0 g/h. Concentration of UV irradiance was 240 mW s/cm^2 by three UVC lamps (Philip-Holland) with capacity of 15 W per 0.6 m lamp. The ozone gas was produced from pure oxygen by the ozone generator (Lino, Vietnam), and was continuously bubbled into water through a porous air diffuser. The ozone flow rate was adjusted by flow meter.

The BAC column of Line I has bed volume of 106 L of GAC (diameter × total height = 300 mm × 2,200 mm and that of GAC bed = 1,500 mm), whereas the BAC column of Line II has bed volume of 26.5 L of GAC (diameter × total height = 150 mm × 2,200 mm and that of GAC bed = 1,500 mm). Granular activated carbon using in this study was manufactured in Vietnam from coconut shells which has the average diameter of 1–2 mm, bulk density of 530 kg/m^3, and specific density of 1.2 kg/L.

A retention tank with volume of 300 L, corresponding to hydraulic retention time of 20–30 min, was followed by ozone/UV reactor in order to remove the excess ozone to minimize bad effects on bacteria growth in the BAC column. A 5% KI solution tank was used for absorbing ozone released from the surface.

2.2. Feed water

The processes used in the Tan Hiep WTP include prechlorination, coagulation, sedimentation, rapid sand filtration, and disinfection. Large amount of chlorine has been used for prechlorination at the raw water intake point and at the receiving box at Tan Hiep WTP are about 1–2 and 2–4 g/m^3, respectively. Feed water was effluent from a fluidized sludge bed tank, prior to the rapid sand filter of Tan Hiep WTP. The characteristics of feed water are shown in Table 1.

Table 1 shows that ammonia nitrogen concentration was high. As DOC of 2.61 ± 0.15 mg/L, THMFP was 356 ± 23 μg/L, which is higher than the allowable limit value of THMs of USEPA drinking water standards (80 μg/L).

Table 1
Characteristics of feed water

Parameters	Unit	Range	Mean value ($n = 12$)
pH	–	6.7–7.1	7.0 ± 0.2
Turbidity	FAU	0.0–3.0	2.5 ± 0.9
Color	Pt-Co	13.0–41.0	24.3 ± 9.4
Alkalinity	mg $CaCO_3$/L	23.2–35.6	25.7 ± 2.5
Ammonia nitrogen	mgN/L	0.12–0.80	0.34 ± 0.12
Nitrate	mgN/L	0.217–0.573	0.33 ± 0.18
TN	mgN/L	0.763–1.692	1.06 ± 0.249
COD_{Mn}	mg/L	2.81–4.56	3.43 ± 0.78
UV_{254}	cm^{-1}	0.068–0.139	0.093 ± 0.01
TOC	mg/L	2.270–3.554	2.75 ± 0.22
DOC	mg/L	2.137–3.394	2.61 ± 0.15
BDOC	mg/L	0.221–0.395	0.31 ± 0.04
THMFP	µg/L	292–411	356 ± 23

2.3. Operating conditions

The pilot-scale experiment was run continuously during 6 months (from October 2010 to April 2011) at Tan Hiep WTP. The pilot experiment of Line I (O_3/UV-BAC) was conducted at 4 runs via combinations between contact time of O_3/UV reactor and EBCT of BAC column (Table 2). Each run was continuously operated at least 30 days (Table 3).

To promote the attached growth biomass on GAC of the pilot-scale experiment, two BAC columns were run for 90 days at the laboratory of Ho Chi Minh University of Technology using the tap water mixed with the effluent from the domestic wastewater treatment plant to obtain the feed water with TOC of 10–15 mg/L. These BAC columns were backwashed using the tap water at the flow rate of 8–12 L/s m^2 to obtain bed expansion of 25–30%.

2.4. Analytical methods

All the analytical parameters were measured according to standard methods [13]. Chemical oxygen demand test is conducted by procedure of TCVN 4565–88 with kali permanganate as a strong oxidant under acidity condition at boiling temperature. The excess $KMnO_4$ is then titrated with oxalic acid 0.1 N. Ultraviolet absorbance was measured at 254 nm (UV_{254}) using a Varian Cary 50 UV/Visible spectrophotometer in a 1 cm quartz cuvette, in accordance with Method 5910. It was used as a surrogate parameter to monitor the changes in chemical structure of NOMs during the water treatment processes. While SUVA which is the absorbance at 254 nm wavelength divided by DOC ($SUVA = UVA_{254} \times 100/DOC$, in

Table 3
Operating conditions of runs of Line I (O_3/UV–BAC)

Run	Contact time of O_3/UV, min	EBCT of BAC, min
P1	15	25
P2	20	20
P3	25	20
P4	20	15

Table 2
Operating conditions of Line I and Line II experiments

	Line I			Line II
	Ozone/UV reactor	BAC L1		BAC L2
Reaction time, min	25; 20; 15	EBCT, min	25; 20; 15	25; 20; 15
Upflow velocity, m/h	1.6; 2.0; 2.7	Hydraulic loading rate, m^3/m^2/h	3.6; 4.5; 6.0	3.5; 4.5; 5.9
Flow rate, m^3/d	6.1; 7.6; 10.2	Flow rate, m^3/d	6.1; 7.6; 10.2	1.5; 1.9; 2.5
UV dose, mW s/cm^2	300; 240; 180			
Applied ozone rate, mg/L of reactor	3.63; 2.90; 2.18			
Ozone/TOC, gO_3/gTOC	0.42; 0.62; 0.91			

L/mg m). TOC was measured according to Method 5310C, using Shimadzu Total Organic Carbon Analyzer TOC-V CPH equipped with auto sampler ASI-V. Potassium hydrogen phthalate standard solutions of 0.5, 1, 2, 5, 10 mg/L were used to calibrate the instrument. The samples were filtered through a 0.45-μm membrane filter and poured into 25 mL TOC vials to determine DOC value with the same TOC analyzer above. DOC is classified into BDOC and NBDOC based on biodegradability, BDOC is defined as the organic carbon which is biodegradable regardless to its absorbability, while NBDOC is the organic carbon which is not biodegradable. BDOC measurement was similar to method of Servais et al. [14]. 75 g saturated activated carbon (GAC) was placed in 225 mL of sample. The GAC used was taken from the GAC filter which was operated for more than one year. The mixture was incubated in darkness in 7–10 days at 30˚C. The removed DOC was defined as biodegradable DOC. Then, NBDOC was determined by difference of (DOC–BDOC).

To prevent THM formation after sampling, water samples were de-chlorinated on-site with Na_2SO_3 0.01 N solution before using liquid–liquid extraction Method 6232 by pentane (C_5H_{12}) [13]. Analyses of THMs and HAAs were performed using a Shimadzu QP2010-Plus Gas Chromatographer—Mass Spectroscopy equipped with a Ni^{63} electron capture detector and a Shimadzu auto injector AOC-20i, following a modified version of EPA Method 502.2 [15] and EPA Method 552.2 [16], respectively. Helium gas carried the volatilized sample through a 30 m (length) × 0.25 mm (ID) × 0.25 μm capillary column housed within the GC oven which is subjected to the following temperature program: start temperature at 45˚C hold for 7 min, end temperature at 200˚C for 3 min, and gradient 15˚C/min.

THMFP and HAAFP analyses were conducted in accordance with Method 5710B [13]. The samples were chlorinated and incubated in dark at a neutral pH at 25 ± 2˚C for 7 days. Chlorine, in the form of sodium hypochlorite 5% solution, was added to each sample with appropriate amounts. A free chlorine concentration at least 3 mg/L and not more than 5 mg/L at the end of a 7 day reaction period was maintained to ensure that chlorine was not the rate limiting factor. After the incubation period, the chlorinated samples were de-chlorinated by Na_2SO_3 0.01 N solution and analyzed for THMs or HAAs with the above procedure. THMFP is difference between THMs concentration of the sample incubated with chlorine for 7 days (THM_7) and that of the initial sample (THM_0). Determination of HAAFP is similar to that of THMFP.

3. Results and discussion

3.1. Effect of operational parameters on organic matter reduction

3.1.1. O_3/UV process

The results show that the DOC removal in O_3/UV process has increased with the raise of contact/reaction time because of correlative increases in ozone and UV irradiance dose. TOC or DOC removal was not high, whereas UV_{254} or SUVA value of the effluent has reduced significantly by O_3/UV treatment. It confirmed that the aromatic structure in NOMs have been broken down during the advanced oxidation process [7]. TOC and DOC removal efficiencies at contact times (CT) of 20 and 25 min were not different. Therefore, CT of 20 min is considered as the suitable contact time due to low-energy consumption and low investment cost. Fig. 2 describes effects of the contact time on changes of

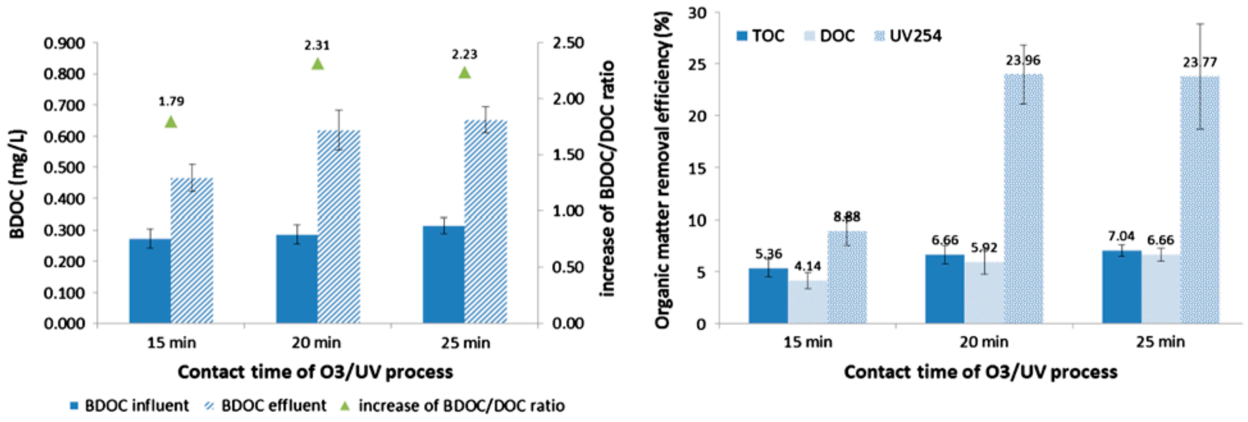

Fig. 2. Effects of contact time on changes of BDOC/DOC ratio (left) and organic removal efficiencies (right).

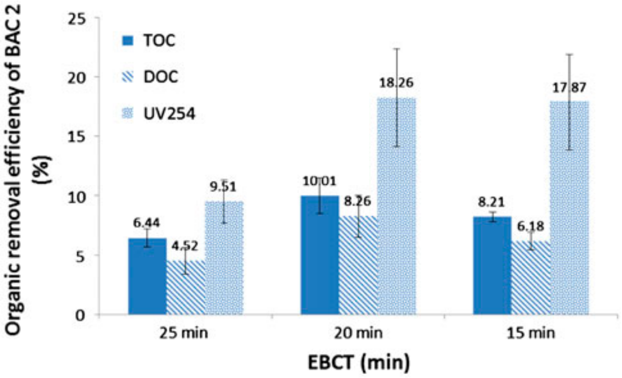

Fig. 3. Effects of EBCTs on organic removal efficiency in BAC alone (Line II).

BDOC/DOC ratio, and Fig. 3 shows the organic removal efficiencies at various CT.

At contact time of 20 min, removal efficiencies on TOC, DOC, UV_{254}, and SUVA of O_3/UV process were 7.04, 6.66, 23.77, and 17.91%, respectively. The biodegradability of effluent was increased via increase in the ratio BDOC/DOC, which was 2.31 times in comparison with the one in feed water.

3.1.2. BAC alone

The BAC column (Line II), without any pretreatment, was examined at EBCT of 25, 20, and 15 min, corresponding to the filtration velocity of 3.5, 4.5, and 5.9 m/h, respectively. Fig. 3 describes the organic removal efficiencies at different EBCTs of BAC column. When EBCT reduced from 25 to 20 min, the organic removal efficiency increased. This could be explained that higher flow velocities may enhance turbulence that helps good contact of contaminants on the surface of BAC grains. Organic matters could be transported into the spores of the carbon grains, and the growth of attached microorganism may increase. However, when EBCT reached 15 min, the removal efficiency has dropped because the attached biofilm was lost through strong turbulence at high velocity (5.9 m/h). At EBCT of 20 min, the removal efficiencies on TOC, DOC, and UV_{254} of the BAC column were 10, 8.3, and 18%, respectively.

3.1.3. O_3/UV-BAC combination

Different runs of contact time of O_3/UV reactor and EBCT of BAC Line I (with the pretreatment using advanced oxidation process) were tested to determine the suitable operational parameters for the pilot plant including O_3/UV process followed by BAC. The experimental results present that the highest organic removal was obtained at the contact time of 20 min and EBCT of 20 min. Table 4 presents effects of various CT and EBCTs on organic removal efficiency.

3.1.4. Organic removal

Table 5 shows that the combination of O_3/UV-BAC obtained higher organic removal efficiencies than those of each process in terms of TOC or DOC removal. It confirmed that O_3/UV is suitable to use as a pretreatment prior to BAC column to eliminate organic matters.

Table 4
Effects of various CT and EBCTs on organic removal efficiency ($n = 10$)

Run	TOC removal (%)	DOC removal (%)	UV_{254} removal (%)
P1	11.63 ± 0.93	9.95 ± 1.57	18.42 ± 3.35
P2	19.13 ± 1.80	17.80 ± 2.73	40.06 ± 2.14
P3	18.54 ± 0.98	16.83 ± 0.78	34.46 ± 5.17
P4	14.86 ± 1.05	12.88 ± 1.47	31.73 ± 5.79

Table 5
Organic removal efficiencies of O_3/UV, BAC alone, and O_3/UV-BAC process

Parameter	O_3/UV	O_3/UV-BAC	BAC alone	Δ%
TOC	7.05 ± 1.64	19.10 ± 2.29	9.85 ± 1.75	2.20
DOC	6.52 ± 1.71	17.63 ± 3.18	7.96 ± 2.03	3.15
UVA_{254}	20.92 ± 6.67	30.66 ± 5.57	17.43 ± 4.15	−7.69
SUVA	15.87 ± 2.34	16.42 ± 2.75	10.45 ± 1.98	−9.90

Note: Δ% = O_3/UV-BAC – O_3/UV-BAC alone.

Table 6
Average capacity of BAC column on DOC removal

Type	$\Delta DOC_{removed}$, mg/L	V_{carbon}, L	$V_{treated\ water}$, m^3/day	DOC removal rate, g $DOC_{removed}/m^3$ of activated carbon/day
O_3/UV-BAC (Line I)	0.438 ± 0.079	91.89	6.62	31.55 ± 5.69
BAC alone (Line II)	0.200 ± 0.054	22.97	1.65	14.36 ± 3.88

3.2. *Effect of O_3/UV pretreatment on BAC process*

Table 6 shows DOC removal rate of BAC column with and without pretreatment using O_3/UV process. BAC with pretreatment using O_3/UV obtained higher organic removal rate. The O_3/UV oxidation process improves biodegradable fraction in influent organics by breaking down the complex organics into the simple ones. Besides the biomass attached on GAC would also increase with time. The latter contributed to the increase in the DOC removal per gram of GAC.

Fig. 4 presents the changes of DOC fraction during O_3/UV, BAC alone, and combination of O_3/UV-BAC.

3.3. *DBP removal*

Fig. 5 shows that DBPs such as total THMs (THM_0) has been eliminated significantly by treatment processes used in the pilot study. THM removal of O_3/UV, BAC alone, and combination of O_3/UV-BAC were 33, 31, and 71%, respectively. THM compounds which were already formed in prechlorination may be reduced by evaporation, microorganism activities of the attached biofilm in the activated carbon beds. In fact, previous researches also claimed that chloroform, the main THM compound in drinking water, is easily evaporative and biodegradable [1]. Besides, the THM-precursors which mainly are NOMs and chlorine concentration may be removed significantly by these processes.

THMFP in the finished water was mitigated significantly. The average ratio of THMFP/DOC has been decreased from 136.4 to 92.2 μg THM/mg DOC. THMFP removals of O_3/UV, BAC alone, and O_3/UV-BAC process were 16, 25, and 40%, respectively (Fig. 5 (b)). The O_3/UV process may not oxidize completely organics, however, this process breaks down or changes the structure of certain NOMs so that they

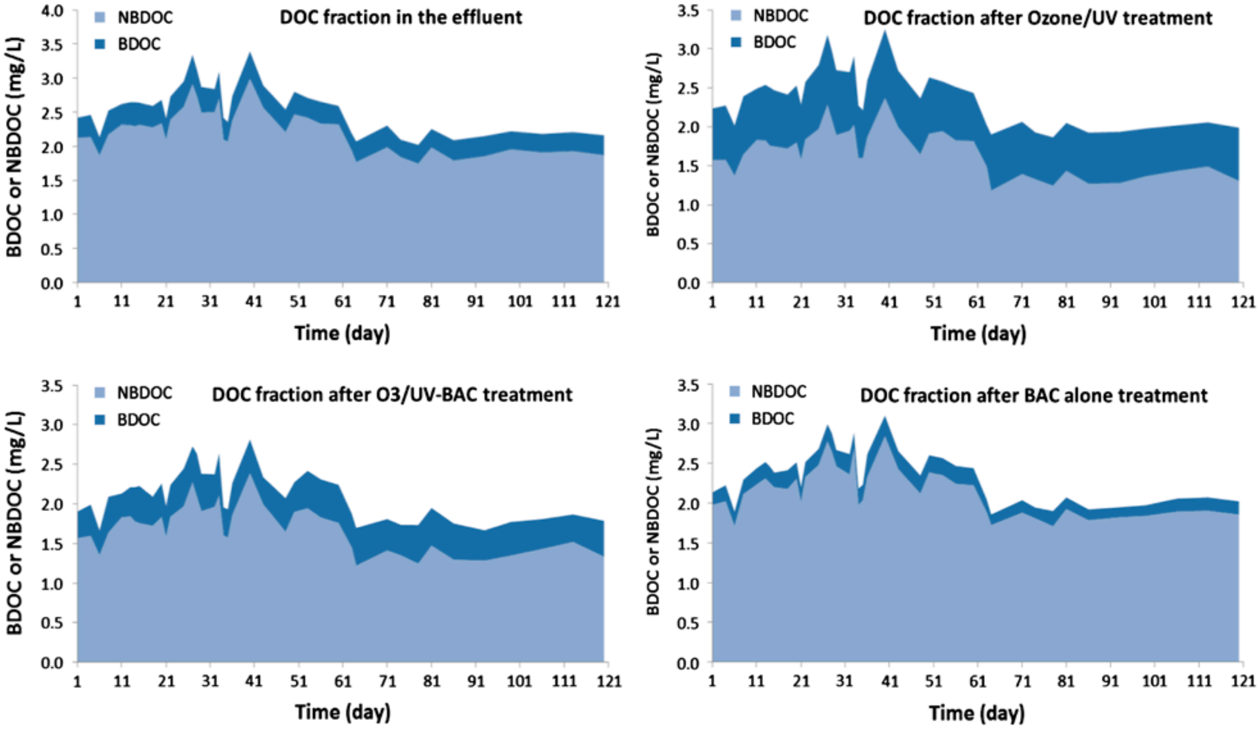

Fig. 4. Changes of DOC fraction during O_3/UV, BAC alone, and combined O_3/UV-BAC treatment.

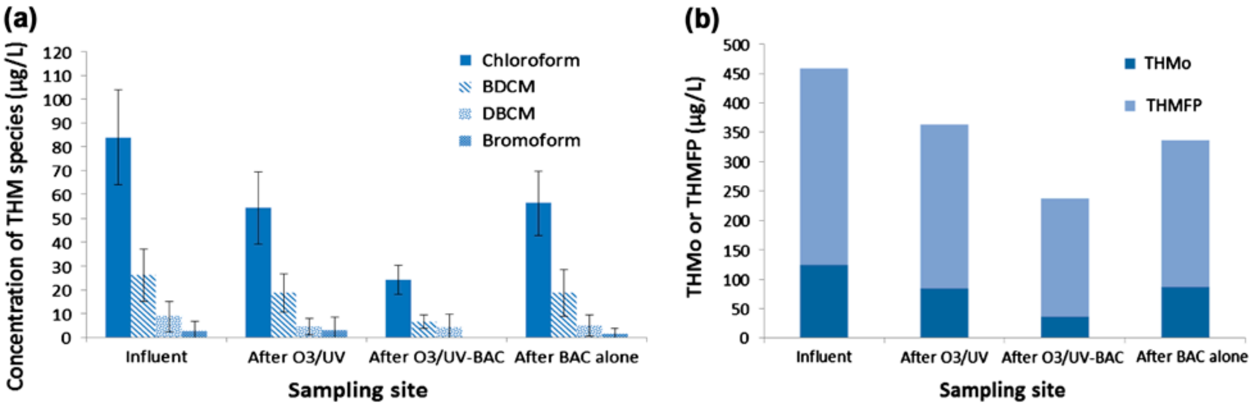

Fig. 5. (a) Changes in concentration of THM species (left), (b) THM_o and THMFP at each treatment process.

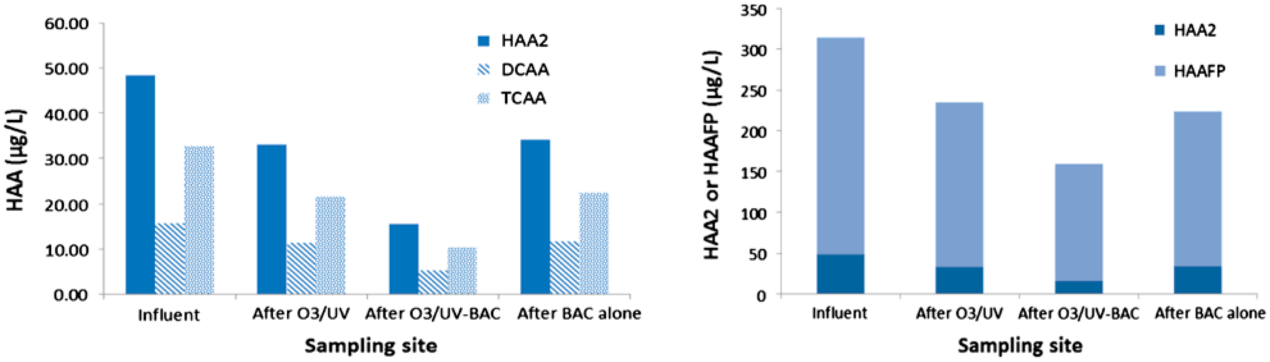

Fig. 6. Changes in concentration of each HAA species (left) and HAA2 and HAAFP (right) at each treatment process.

could not react with chlorine to form chloroform and other THM species [9,17].

Similarly, another DBP group, HAAs also was reduced in these treatment processes. There are some species of HAA group existed in drinking water as one of the main DBPs. However, total concentration of two HAA species including DCAA and TCAA (HAA2) was selected to examine, because they are regulated in Vietnamese standard for drinking water quality. HAA removals of O_3/UV, BAC alone, and O_3/UV-BAC combination process were 32, 29, and 68%, respectively. Moreover, HAAFP was also decreased because of the drop of NOMs and chlorine concentration in water. HAAFP removals of O_3/UV, BAC alone, and O_3/UV-BAC process were 24, 29, and 46%, respectively (Fig. 6). Although the amount of NOMs did not changed much in O_3/UV process, strong oxidant agents formed in this process has changed the structure of these NOMs and make them not easily react with chlorine to form HAAs [9,17].

4. Conclusion

The contact time of 20 min of O_3/UV process was appropriate for breaking down the complex organic compounds in terms of high treatment efficiency and low-energy consumption. Besides, EBCT of 20 min of BAC achieved high organic removal efficiency. The combination of O_3/UV-BAC has proved that pretreatment using O_3/UV is absolutely necessary to accelerate the organic removal rate as well as DBPs removal of BAC process in terms of TOC, THMs, HAAs, etc. O_3/UV followed by BAC obtained the better treatment efficiency than each process alone. Removal efficiencies of TOC, DOC, UV_{254}, and SUVA through pilot study of O_3/UV-BAC were 19, 18, 31, and 16%, respectively. The concentration of DBPs has decreased considerably. THMs removal of 71% and HAAs removal of 68% were achieved, whereas decrease in DBP formation potential were 39.3% for THMFP and 46.1% for HAAFP.

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