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Characteristics of disinfection by-products reduction in the processes of drinking water treatment system using Nakdong river water

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ABSTRACT

In this study, batch and continuous experiments were conducted to investigate the influential factors on the distribution and formation of aldehydes, bromate and bromide in Nakdong river water and to investigate the behaviour of disinfection by-products (DBPs) in the processes of drinking water treatment system. The mean influent concentrations were 3.1 mg/L (2.4–3.9 mg/L) for dissolved organic carbon (DOC), 57.6 ug/L (42.0–85.7 ug/L) for total aldehydes (TA) and 55.7 ug/L (17.4–89.3 ug/L) for bromide. As a result, TA and bromide levels in the Nakdong river water increased with the decreasing water flow rate and decreasing water temperature. In pre- and post-ozonation, the DBPs concentration increased with increasing bromide level, ozone dosage and pH value. In particular, the concentration of DBPs formation in both pre-ozonation and winter was relatively higher than that in both post-ozonation and summer. In coagulation and sedimentation, polyaluminium sulphate organic magnesium coagulation resulted in slightly less DBPs removal than polyaluminium silicate chloride coagulation. TA was significantly reduced at the top of sand and biological activated carbon (BAC) filters, whereas bromate was effectively reduced in a three-month-old BAC filter.

Keywords: Disinfection by-products; Aldehydes; Bromate; Ozonation; Drinking water treatment processes

1. Introduction

Most drinking water treatment (DWT) plants taking raw water from surface water in Korea have adopted the typical conventional water treatment processes consisting of pre-chlorination, coagulation–sedimentation, rapid sand filtration and disinfection, which are designed for particle separation such as turbidity removal [1]. In recent years, micropollutant loads as well as conventional pollutant from the effluents of wastewater treatment plant and farming land due to industrialization, urbanization and annul farming in the basin of Nakdong river have been gradually increased. The composition of contaminants has been shifting from inorganic to organic materials such as total organic carbon (TOC). As a means of complying with current and anticipated water quality regulations as well as the control of increasing micropollutants in

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the river water, advanced water treatment system has added ozonation and granular activated carbon (GAC) processes to conventional water treatment system have been widely adopted. Based on the previous studies for advanced water treatment processes, pre-ozonation was needed to enhance the removal of TOC and the reduction of total trihalomethane formation potential when it was used in combination with coagulants such as aluminium sulphate (alum) and polyaluminium chloride (PACl) [2]. In coagulation, the improvement of coagulation efficiency has become the main target to decrease the use of a final chlorine dose and the formation of disinfection by-products (DBPs) [3]. In filtration, it was reported that DBPs formation potentials reduction and degradation were effective due to biological activity in the sand filter (SF) with a relatively long hydraulic retention time (HRT) [4]. The combination of ozonation and biological activated carbon (BAC) is one of the most promising processes among advanced DWT processes. The main advantage of BAC filtration is that it removes biodegradable compounds comprising the most undesirable fraction of organic matter in water [5]. Ozonation and GAC processes have been demonstrated to be efficient in the oxidation and adsorption of pharmaceuticals as well as other micropollutants in drinking water [6–8]. Ozonation in drinking water results in the formation of both organic by-products (assimilable organic carbon, aldehydes, carboxylic acids and ketones) and inorganic by-products (bromate) [9,10]. von Gunten [11] reported that bromate formation can become a serious problem for bromide levels above 100 ug/L. In this study, we focused on the investigation of the behaviour of aldehydes and bromate as the DBPs in raw water and the treatment processes. Therefore, the objectives of this study are to investigate the influential factors on the formation of the DBPs in the Nakdong river water and to investigate the DBPs behaviour in the processes of the DWT system using the Nakdong river water.

2. Materials and methods

2.1. Experimental apparatus and materials

The pilot-scale plant (100 m^3/d) used for efficient DWT consisted of pre-ozonation contactor, coagulator/flocculator/cross-flow settler, SF, post-ozonation contactor and GAC filters (Fig. 1). The pre-ozonation contactor followed by rapid mixing basin was 0.15 m in width (*W*), 0.65 m in length (*L*) and 6.5 m in height (*H*) and the post-ozonation contactor followed by GAC filters was *W* 0.24 m, *L* 0.96 m and *H* 6.50 m, which was fabricated using a transparent acrylic plate. The rapid mixing basin (*W* 0.35 m, *L* 0.35 m, *H* 0.90 m) was followed by the flocculation basin (*W* 1.20 m, *L* 2.10 m, *H* 1.20 m) and parallel sedimentation basins. The sedimentation basin (*W* 1.20 m, *L* 5.00 m, *H* 2.40 m) was equipped with sampling ports at an interval of 1 m and with solids removal devices that transfer settled residuals to appropriate processing facilities. All basins except the ozone contactor were made of stainless steel. The design of sand and GAC filters allowed for water sampling over the filter bed height, as well as influent and effluent water samples. The SF (*W* 0.60 m, *L* 0.60 m, *H* 3.90 m) was packed to 0.9 m with sand media with an effective size of 1.0 mm. GAC filters (*W* 0.30 m, *L* 0.30 m, *H* 6.00 m) were packed to 2.5 m with two different carbons with an effective size of 0.9 mm. The two carbons were coal-based carbon (Coal-C) (Calgon F-400, USA) and coconut-based carbon

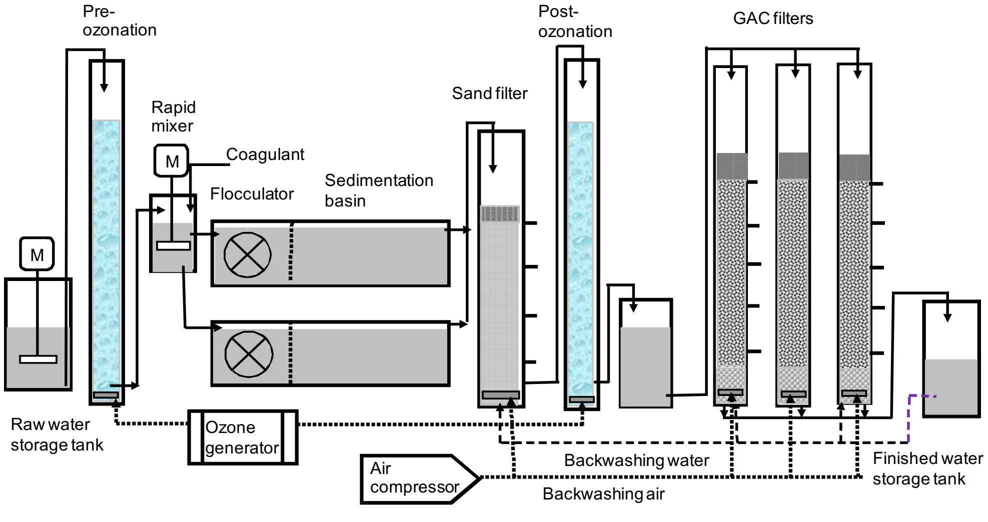

Fig. 1. Schematic of pilot-scale drinking water treatment plant.

Table 1
Experimental conditions of pilot-scale drinking water treatment plant

Unit process	Range	Unit process	Range
Ozonation	Pre-/post-O_3	Sand filtration	
Ozone dosage (mg/L)	0.5–5.0/0.5–5.0	Media depth/size (mm)	900/1.0
Contact time (min)	4–6/7–10	Filtration velocity (m/d)	130 ± 3
Coagulation/sedimentation		GAC filtration	
Rapid mixing/flocculation/sedimentation time (min)	1.3/33.8/194.3	Media depth/size (mm)	2,500/0.9
Coagulant	PACl/PASCl/PASOM	EBCT (min)	15 ± 0.5
Coagulant dosage (mg/L)	1.0–5.0 as Al_2O_3		

(Coconut-C) (Samchully, Korea). Pore volume and specific surface area of the carbons were 0.548 cc/g and 1,100 m^2/g for the Coal-C and 0.495 cc/g and 1,230 m^2/g for the Coconut-C, which were measured by the Autosorb-1 MP (Quantachrome, USA). The sand and GAC filters were equipped with backwashing devices and sampling ports at intervals of 15 and 50 cm.

2.2. Operational conditions

The operating conditions of DWT system are summarized in Table 1. The influent was fed by downflow into the top of the pre- and post-ozone contactors, the SF and the GAC filters. PACl (Al_2O_3 11.0%), polyaluminium sulphate organic magnesium (PASOM; Al_2O_3 7.6%) and polyaluminium silicate chloride (PASCl; Al_2O_3 8.4%) as a coagulant were added in the rapid mixing basin. Ozone was produced from air by ozone generator (OZAT CFS-1A, Switzerland) and the concentration of ozone dosage was controlled and monitored by an ozone control system (PCI, USA). Batch ozonation experiments were conducted to evaluate the effect of Br^- concentration, pH and ozone dosage on the DBPs formation. The batch ozonation experiments were conducted in a bubble column (inner diameter: 10 cm, height: 250 cm) built from an acrylic cylinder adapted with a sample port at the bottom of the column. The pre- and post-ozonation contactors were used for downstream treatment and received the raw water and the water treated by coagulation/sedimentation/sand filtration process, respectively. Both pre- and post-ozone dosage ranged from 0.5 to 5.0 mg/L. In batch coagulation experiment, the effectiveness and dosage of coagulants were optimized for minimum settled water turbidity and pH using conventional jar testing procedures. The jar teat was carried out in a jar test apparatus (Phipps & Bird PB-700). The contents of the jars were rapidly mixed at a paddle speed of 100 r/min for 30 s, followed by slow mixing at 40 r/min for 10 min and settling for 20 min.

The raw water used for this study was the river water collected from lower region of the Nakdong river. The ranges of raw water quality parameters with mean values in parentheses were: water temperature 4.0–27.8°C (16.3°C); pH 7.4–9.5 (8.5); biochemical oxygen demand (BOD) 1.4–4.9 mg/L (2.6 mg/L); dissolved organic carbon (DOC) 2.4–3.9 mg/L (3.1 mg/L); alkalinity 35.5–60.9 mg/L as $CaCO_3$ (51.5 mg/L as $CaCO_3$); total aldehydes (TA) 42.0–85.7 ug/L (57.6 ug/L) and bromide (Br^-) 17.4–89.3 ug/L (55.7 ug/L). Samples of the influent and the effluents from each process were collected, preserved and refrigerated at 4°C until analysed.

2.3. Analytical methods

DOC was measured by the combustion-infrared method using total organic carbon analyser (TOC-5000, Shimadzu, Japan). Seven aldehydes (formaldehyde, acetaldehyde, propanal, butanal, pentanal, glyoxal and methylglyoxal) were analysed by gas chromatography (HP 5890 Series II Plus, USA) with electron capture detection (GC/ECD) according to USEPA Method 556 [12]. The concentrations of bromide and bromate were determined with the ion chromatography and an UV detector by the post-column derivatization method using o-dianisidine [13].

3. Results and discussion

3.1. Characteristics of Nakdong river water

Fig. 2 shows the seasonal variation of TA and bromide concentrations in the Nakdong river water. The average concentrations of TA and Br^- in raw water were 55.8 ug/L ranging from 42.0 to 85.7 ug/L and 55.7 ug/L ranging from 17.4 to 89.3 ug/L, respectively. Water temperature fluctuated from 4.0 to 27.8°C during the monitoring period. When the water temperature dropped to less than 10°C in winter, the TA and Br^- concentrations were frequently above 71.2 and 74.1 ug/L, respectively. The lowest water flow rate

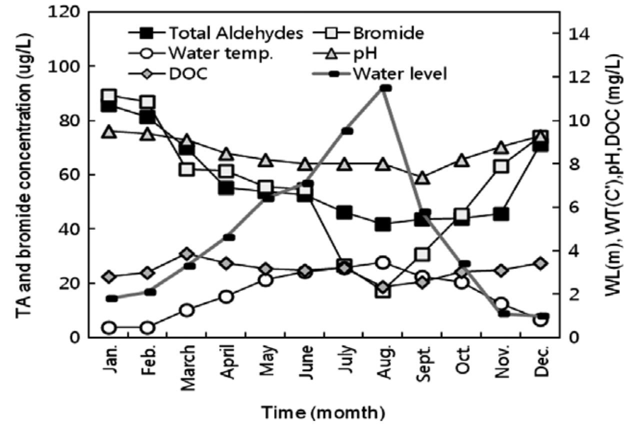

Fig. 2. Seasonal variation of aldehydes and bromide concentration in Nakdong river water.

and highest TA and Br$^-$ concentration in the Nakdong river water during the winter season (November–February) were attributed to the large water volume transferred from the river to cover the water demand as well as the lowest rainfall. The data showed that TA and bromide levels increased with the decrease in both water flow rate and water temperature. High Br$^-$ level in the raw water resulted in road salt run-off and drought conditions in winter [14].

3.2. Performance of ozonation

Fig. 3(a) and (b) shows the performance results of batch pre- and post-ozonation experiments on aldehydes formation. When both pre- and post-ozone dosage were increased to 0.5, 1.0, 1.5, 2.0, 3.0, 4.0 and 5.0 mg/L, acetaldehyde and formaldehyde concentrations increased to 2 mg/L of pre- and post-ozone dosages and at more than 3 mg/L of ozone dosages, there was no significant increase in forming aldehydes. This result was similar to the previous results that reported that TA concentration increases with increasing ozone dosages up to a maximum at moderate ozone doses of 2–5 mg/L and thereafter decreases because of excess ozone oxidation of DBP precursors [15,16].

The distribution of aldehydes speciation in pre- and post-ozonated waters were in the order of acetaldehyde > formaldehyde > methylglyoxal > glyoxal > propanal > butanal > pentanal. The percentages of acetaldehyde and formaldehyde formation as dominant compounds were 67.4 and 27.0% in pre-ozonation and 89.0 and 8.7% in post-ozonation, respectively, whereas the percentages of methylglyoxal, glyoxal, propanal, butanal and pentanal formation were approximately less than 1.0% [16].

Table 2 presents the performance results of batch pre- and post-ozonation experiments on bromate (BrO_3^-) formation in bromide-containing water. As shown in Fig. 2, Br$^-$ levels in the Nakdong river water during the monitoring period of one year were insignificant to produce BrO_3^- during ozonation. So, bromide concentration in both raw water and filtrated water was increased to 0.1, 0.2 and 0.5 mg/L by dosing bromide solution (AccuStandard, USA). As a result, when both pre- and post-ozone dosage were increased to 0.5, 1.0, 2.0 and 4.0 mg/L in the bromide-containing waters (Br$^-$ 0.1–0.5 mg/L), the percentages of BrO_3^- formation at an ozone dosage of 1.0–4.0 mg/L were 64.8 ± 24.1% for pre-ozonation and 67.6 ± 20.8% for post-ozonation higher than that at an ozone dosage of 0.5 mg/L. On the other hand, the percentages of BrO_3^- formation at Br$^-$ concentration of 0.2–0.5 mg/L were 48.0 ± 17.1% for pre-ozonation and 67.9 ± 9.0% for post-ozonation higher than that at Br$^-$ concentration of 0.1 mg/L. The data in Table 2 showed that the BrO_3^- formation rate increased steadily with the increase in both ozone dosages and Br$^-$

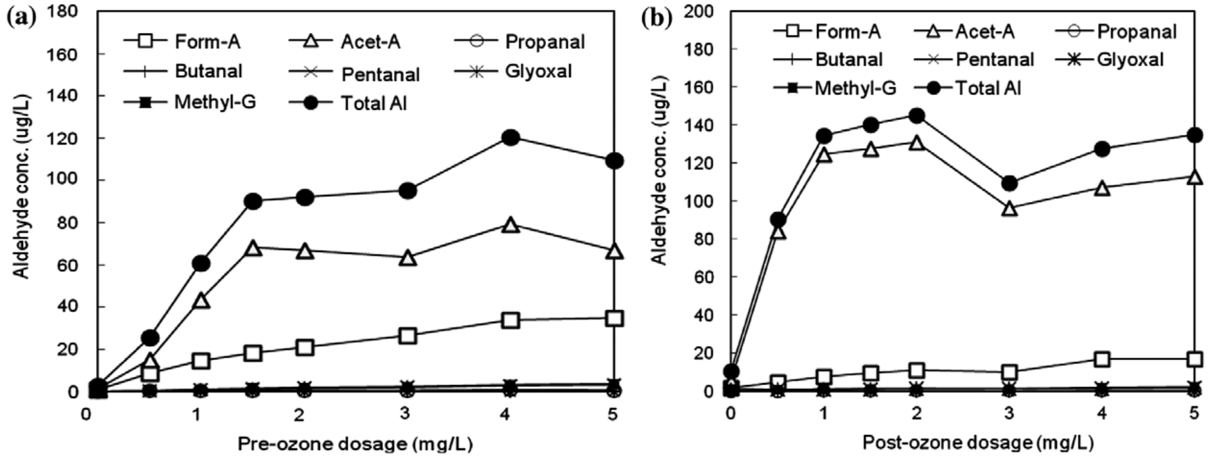

Fig. 3. Distribution and formation of aldehydes during pre- and post-ozonation.

Table 2
Charateristics of bromate formation with ozone dosage and bromide concentration

Ozone dosage (mg/L)	BrO_3^- concentration (ug/L) in pre-ozonated water			BrO_3^- concentration (ug/L) in post-ozonated water		
	Br^- concentration in raw water (mg/L)			Br^- concentration in filtrated water (mg/L)		
	0.1	0.2	0.5	0.1	0.2	0.5
0.5	4.0	5.0	7.0	2.0	4.3	6.2
1.0	5.2	8.1	15.0	3.2	8.3	12.1
2.0	10.2	18.4	34.3	6.2	16.0	26.5
4.0	21.2	37.3	55.4	10.5	35.0	51.0

concentration in the raw and filtrated water during ozonation [17].

Fig. 4 presents the effect of pH value on DBPs formation in bromide-containing water. In the batch experiments, the applied ozone dosage and contact times were 1.5 mg/L and 10 min for the raw water with Br^- concentration of 0.2 mg/L. When pH value was increased from 6 to 9, the percentages of TA and BrO_3^- formation at pH 7–9 were 28.8 ± 10.7 and 29.0 ± 4.9% higher than that at pH 6. This shows that the increase in DBPs concentration coincides with increasing pH. These results also indicate that the optimum pH value for pre- and post-ozonation is in the near-neutral range with regard to a cost-effective reduction of DBPs formation [16,18].

3.3. Performance of coagulation/sedimentation

Fig. 5 shows the profile of turbidity reduction and pH variation with coagulant dosage. The jar test was conducted to investigate the effects of different Al-based coagulants (PACl, PASCl and PASOM) on the removal of the DBPs formed during pre-ozonation (ozone dose: 1.5 mg/L, contact time: 4 min). When Al_2O_3 concentration was increased to 1.0, 2.0, 3.0, 4.0 and 5.0 mg/L corresponding to 9–45 mg/L as PACl dosage, 12–59 mg/L as PASCl dosage and 14–68 mg/L as PASOM dosage, the turbidity showed a gradual decrease to about 3.0 mg/L of coagulants dosages. After 4.0 mg/L, there was a little decrease corresponding to removal efficiency of <5%. In particular, the high removal of turbidity at 3.0 mg/L of coagulant dosage was achieved in the order of PASOM > PASCl > PACl. At more than 4.0 mg/L, three coagulant treatments resulted in a similar decrease trend in reducing turbidity. The low pH level with different coagulants was also achieved in the order of PASOM > PASCl > PACl. The high cost of coagulant depending on the percentage of Al_2O_3 in coagulant were in the order of PACl > PASCl > PASOM. The above results indicate that PASOM is more suitable than PASCl and PACl with regard to cost-effectiveness, headloss control in SF and pH control when considering the high pH value in the Nakdong river water [19].

Fig. 6 shows the profile of DBPs distribution in length of the sedimentation basin. In the coagulation process, the pre-ozonated waters (ozone dose: 1.5 mg/

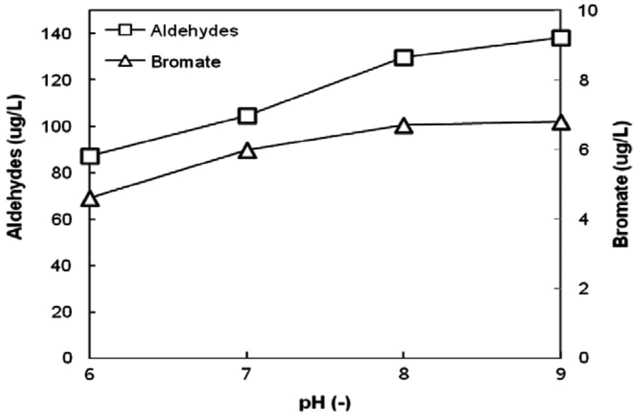

Fig. 4. Effect of pH value on DBPs reduction in the pre-ozonated water.

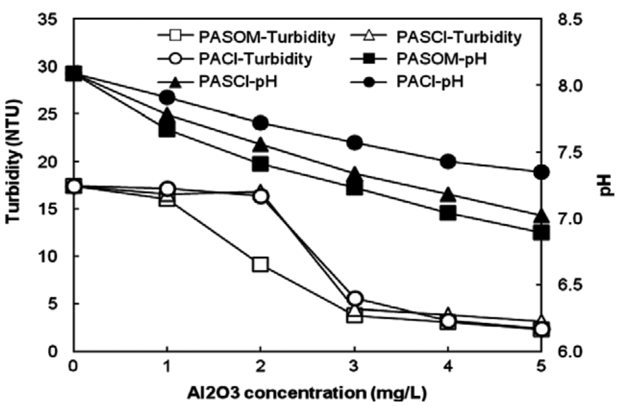

Fig. 5. Profiles of turbidity reduction and pH variation with coagulant dosage as Al_2O_3.

L, contact time: 4 min) used as raw water were stirred rapidly at 200 r/min for 1.3 ± 0.1 min during coagulant addition, followed by slow stirring at 40 r/min for 34 ± 1.0 min and then cross-flow settling for 190 ± 2.0 min. The coagulants used were PASOM and PASCl determined in the jar test.

When the optimum Al_2O_3 concentration of 3.0 mg/L determined in the batch experiment was added in coagulation process, the percentages of TA and BrO_3^- removal was found to be 34.4 ± 1.5 and 3.6 ± 0.3% for PASOM coagulation and 45.5 ± 1.7 and 4.9 ± 0.4% for PASCl coagulation at total HRT, determined as 190 min in the sedimentation process. This result showed that PASOM coagulation resulted in slightly less DBPs removal than PASCl coagulation. As shown in Fig. 6, the DBPs concentration showed a gradual decrease between the inlet and 2 m in the basin corresponding to HRT for 76 min. After 2 m from the inlet of the basin, there was a little decrease corresponding to a removal efficiency of 1.2–10.3% for PASOM coagulation and 0.6–10.7% for PASCl coagulation. The above results indicate that both PASOM and PASCl coagulation is significant for TA reduction but insignificant for BrO_3^- reduction.

3.4. Performance of sand/GAC filtration

Fig. 7 shows the profile of DBPs distribution in depth of the SF. In sand filtration experiment using the settled water, applied filtration velocity was 130 ± 3 m/d. When the mean concentrations of influent DBPs were 10.7 ug/L (10.5–11.0 ug/L) for TA and 7.0 ug/L (6.8–7.1 ug/L) for BrO_3^-, the percentages of DBPs removal in the filter were 50.1 ± 2.1% for TA and 6.9 ± 0.7% for BrO_3^-. Both TA and BrO_3^- concentration showed a relatively rapid decrease between the top and 15 cm in the filter bed. After 15 cm from the top of the filter, the TA reduction rate showed a small decrease corresponding to a removal efficiency of <6% for TA and <2% for BrO_3^-. This resulted in DBPs biodegradation by the biomass developed on the top of the SF. In particular, the removal efficiency of BrO_3^- was about 43% lower than that of TA. This result showed that the adsorption and biodegradation ability of BrO_3^- were poorer than TA.

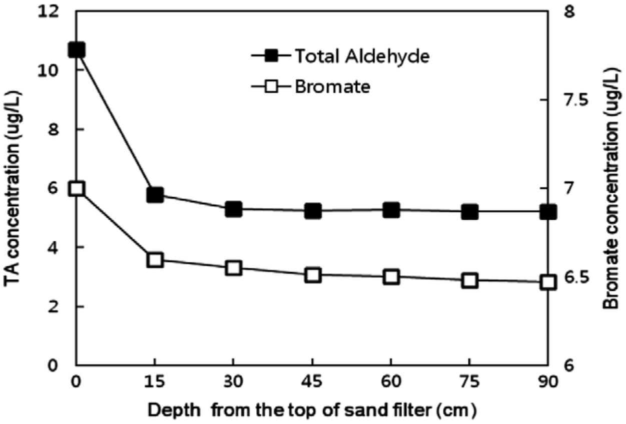

Fig. 7. Profiles of DBPs reduction in depth of SF.

Fig. 8 shows the profile of DBPs distribution in depth of the BAC filters. In the GAC filtration experiment using the filtrated water, three-month-old coal-based GAC (BAC1), thirty-month-old coal-based GAC (BAC2) and thirty-month-old coconut-based GAC (BAC3) were examined to clarify TA and BrO_3^- behaviour in BAC filters. The bed volumes of BAC were 4,440 for BAC1 and 133,200 for BAC2 and BAC3. The applied EBCT for each filter was 15 ± 0.5 min. When the mean concentrations of influent DBPs were 22.5 ug/L (21.3–23.6 ug/L) for TA and 12.5 ug/L (12.1–12.8 ug/L) for BrO_3^-, the removal of TA and bromate were 89.2 and 60.7% for BAC1, 83.5 and 14.7% for BAC2, and 79.2 and

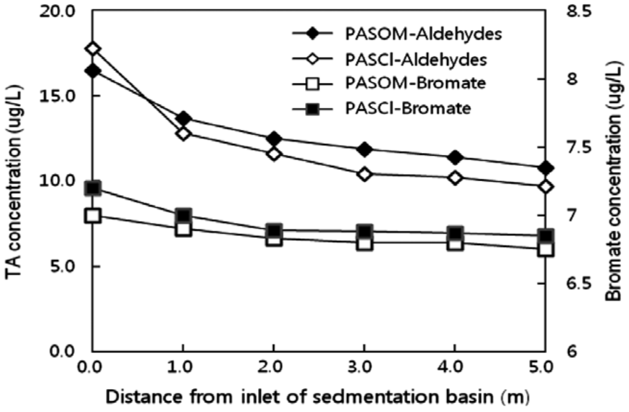

Fig. 6. Profiles of DBPs reduction in distance of sedimentation basin.

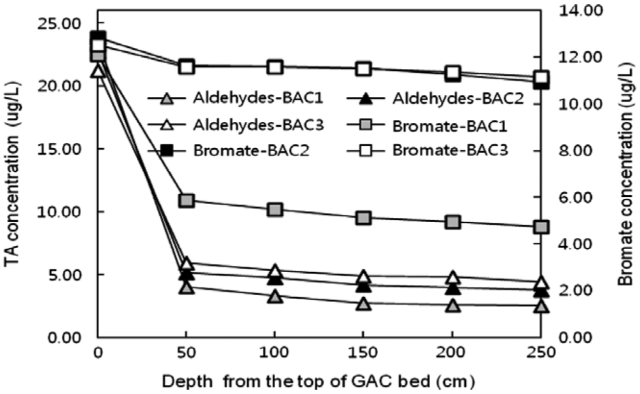

Fig. 8. Profiles of DBPs reduction in depth of BAC bed.

11.0% for BAC3, respectively. The DBPs concentration showed a relatively rapid decrease between the top and 50 cm in the filter bed. After 50 cm, DBPs reduction showed a slow decrease trends corresponding to removal efficiency of 5–6% for TA and 4–7% for BrO_3^-. The high removal of DBPs with different BAC filters was achieved in the order of BAC1 > BAC2 > BAC3. This result showed that coal-based carbon was somewhat more effective than coconut-based carbon in reducing DBPs. The BAC were also effective beds within three months for both TA and BrO_3^- reductions, whereas the BAC bed was effective within thirty months for only TA reduction. These results were similar to the previous results that reported that there was no significant change in the amount of BrO_3^- in two-year-old BAC, compared with the results for new GAC that no BrO_3^- was detected in the effluent from new GAC filter [20]. Both TA and BrO_3^- were removed efficiently by BAC filters. The above result suggests that the use of less than three-month-old BAC is efficient in reducing DBPs.

3.5. Performance of DWTP

Fig. 9 shows the performance results of real DWT plant for DBPs control during summer (August) and winter (November–February). Experiments were carried out at an efficient operating condition for the real plant. In the real DWT plant (600,000 m^3/d), the pre-chlorination (chlorine dosage 4 ± 0.5 mg/L, contact time 2.5 ± 0.5 h) prior to the pre-ozonation process was introduced to control both ammonia nitrogen and biomass growth (algae). The applied ozone dosages and contact times of pre- and post-ozonation were 1.5 and 0.7 mg/L and 4 and 7 min. The PASOM as a coagulant was used in the real DWT plant with regard to cost-effectiveness and pH control and its dosage was 40 mg/L corresponding to 4 mg/L of optimum Al_2O_3 concentration. The BAC filter used was three months old. As a result, pre-chlorination in the real DWT plant resulted in a slight increase in TA concentration, but there was no BrO_3^- formation. The mean concentrations of TA and BrO_3^- during pre-ozonation increased from 52.6 ug/L (42.0–69.9 ug/L) to 119.0 ug/L (73.0–143.9 ug/L) and from 0 to 1.8 ug/L (0–2.8 ug/L), respectively, while they were reduced to about 70.1 ± 13.4 and 5.0 ± 5.2% by coagulation/sedimentation (C–S) and SF processes. In the post-ozonated water, the mean concentrations of the TA and BrO_3^- increased from 32.2 ug/L (26.0–38.1 ug/L) to 67.9 ug/L (56.4–70.9 ug/L) and from 3.1 ug/L (0–4.8 ug/L) to 6.8 ug/L (2.7–9.5 ug/L), respectively, while they were reduced to about 58.0 ± 23.1 and 33.4 ± 2.8% by BAC bed. In pre-ozonated water, the TA and BrO_3^- concentrations in winter were 2.0 and 5.0 times greater than in summer, whereas in post-ozonated water, the TA and bromate concentrations in summer were 1.3 and 3.3 times greater than in winter. This result indicates that the concentration of DBPs formation in both the pre-ozonation and winter was relatively greater than that in both post-ozonation and summer.

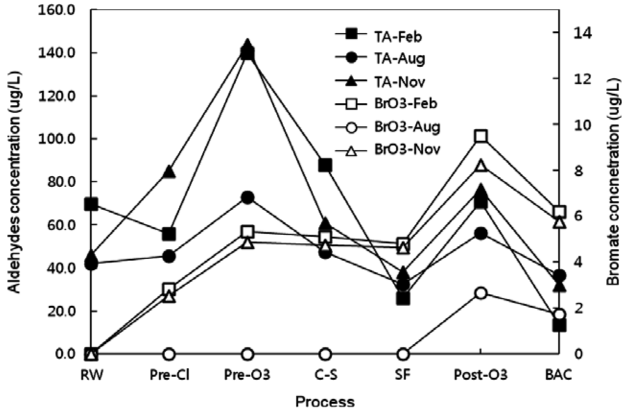

Fig. 9. Seasonal variation of DBPs formation and reduction in processes of real DWT.

4. Conclusion

Both TA and bromide concentration in NDR water increased with the decreasing water flow rate and decreasing water temperature. In ozonation, TA concentrations increased with the increasing ozone dosages of pre- and post-ozonation, whereas bromate concentration increased with increasing bromide concentration, ozone dosage and pH value. In particular, the concentration of DBPs formation in both pre-ozonation and winter was relatively higher than that in both post-ozonation and summer. In coagulation and sedimentation, both PASOM and PASCl coagulation was significant for TA reduction but insignificant for bromate reduction. On the other hand, TA was significantly reduced at the top of the SF and the BAC filters within thirty months, whereas bromate was effectively reduced in a three-month-old BAC filter. The above results reveal that parameters such as ozone concentration, contact time, water temperature, pH and precursor concentration influence the concentration of DBPs.

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