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Evaluation of a pilot scale dual media biological activated carbon process for drinking water

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Abstract−This study was carried out to optimize a dual media BAC (biological activated carbon) process for DOC removal and DBPs (disinfection by-products) control. Pilot scale tests were coducted at the Tukdo water treatment plant in Seoul, Korea. The dual media BAC process is highly efficient in the removal of DOC and THMFP, and is more capable of sustaining microorganisms than a single layer filter. The bottom layer of the sand filter functioned as a screen for turbidity, microorganisms, and other biological material. Total DOC removal efficiency was 13 to 25%, and the corresponding THMFP (trihalomethane formation potential) removal efficiency was approximately 20 to 33%. Turbidity and DOC leakage continued for 40 min after backwashing in all reactors. Breakthrough occurred from 24-72 hours in each reactor. Ten species of microorganisms were identified in the activated carbon filled in BAC reactors. The predominant species were *Clavibactor* and *Corynebacterium* and microbial species were simple at the lower end.

Key words: BAC, Dual Media, DOC, THMFP, Biomass

INTRODUCTION

An increase in biodegradable organic carbon leads to bacterial growth in drinking water distribution systems [1]. The growth of microorganisms in a distribution system promotes water quality deterioration through increased corrosion, produces taste and odor-causing compounds, and increases the amount of disinfectant demand to maintain residuals in the distribution system.

Biologically activated filters have been effective in removing significant amounts of biodegradable organic matter [2-5]. Dual media filtration with sand-BAC filters has been shown to be more efficient than conventional biologically activated carbon (BAC) filters for the removal of biodegradable organic carbon and ammonia [6]. Granular activated carbon, a highly porous material capable of supporting a large bacterial population, is placed above a supporting layer of sand in a dual media filter. When macroporous activated carbon is used, a large number of attachment sites, protected from the abrasive action of backwashing, are present for biomass fixation. Such an abundance of biomass allows for sustained nitrification and stable biodegradable dissolved organic matter removal in cold water [7].

Currently, the BAC process is a common practice for drinking water treatment and considered an alternative method for dissolved organic carbon (DOC) removal and disinfection by-products (DBPs) control. Most studies to date have not considered operation parameters such as EBCT, HLR, backwash cycles related to particles, DOC leakage, behavior of microorganisms, microbial species, and the removal of trihalomethanes (THMs) species regarding empty bed contact time (EBCT) and hydraulic loading rate (HLR), systematically. Carlson reported that DOC removal was not sensitive to HLR but sensitive to EBCT from 5-8 min. Wang and Summers [8] reported that the removal efficiency of ozonated DOC increased over an EBCT range of 3-33 min at a constant HLR. LeChevallier et al. [9] reported that total organic carbon (TOC) removal during biofiltration increased to 29, 33, 42, and 51% at 5, 10, 15, and 20 min EBCT when water was pre-ozonated with 0.8 mg O_3/mg of TOC.

The objective of this study was to understand and optimize a dual media BAC process for DOC removal and DBP control through examining DOC removal efficiency as a function of EBCT and HLR. Furthermore, this study attempted to estimate biomass concentration and identify microorganisms attached to activated carbon in a BAC reactor.

MATERIALS AND METHODS

1. Experimental Design

A pilot plant was installed at the Tukdo water treatment plant in Seoul. It consisted of an ozone generator, ozone contactor, reservoir for ozonated water, and three activated carbon reactors with different HLR and EBCT. The schematic diagram for the Tukdo pilot scale plant is shown in Fig. 1, and the design parameters for the BAC reactor process are represented in Table 1. A 20 cm sand layer followed by Picabiol activated carbon at a depth of 2 m was packed in each dual media BAC reactor. Raw water for this research was taken from the point of a filter inlet and then flowed into an ozone contactor, and was ozonated by direct feeding mode for 8 min at a dose of 0.75 mg O_3/mg of DOC and drained to a reservoir. The ozonated water was fed to the top of BAC reactors and excess water was drained through a by-pass line. A flow meter was installed at the bottom of each reactor to control the filtration rate. Eight sampling ports at intervals of 25 cm, which were connected to a pie-

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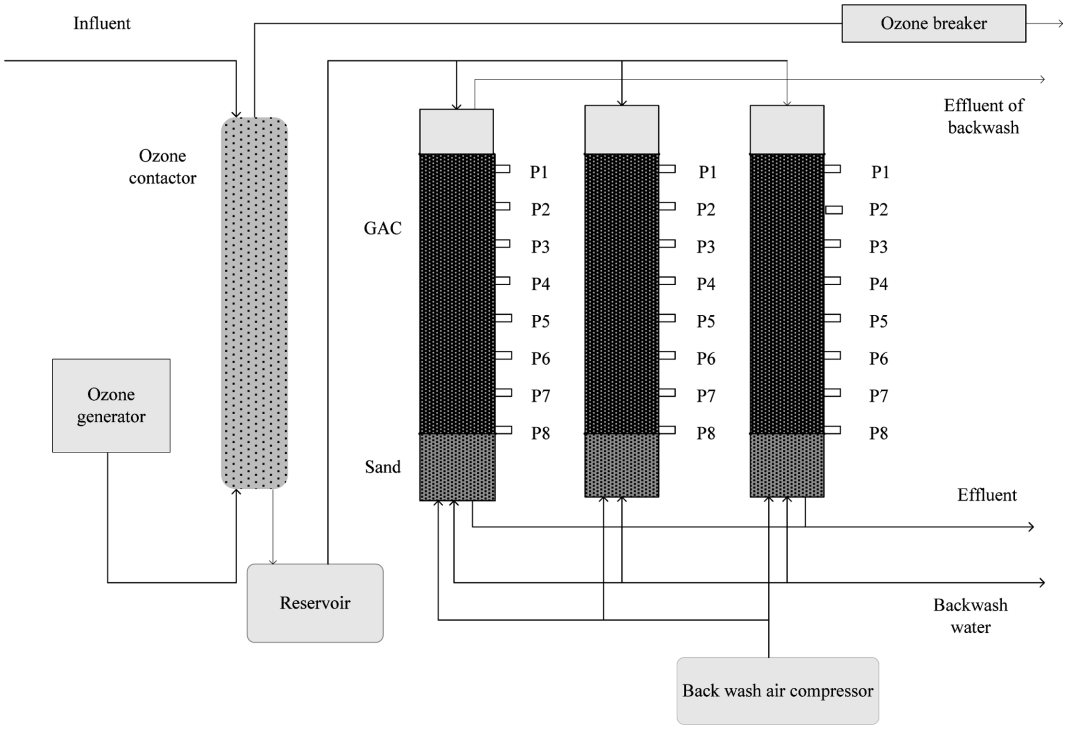

Fig. 1. Pilot plant schematic.

Table 1. Design parameters for BAC reactors

	R1	R2	R3
Diameter (cm)	5	5	5
Height (m)	3.5	3.5	3.5
AC bed height (m)	2	2	2
Sand layer (cm)	20	20	20
HLR (m/hr)	24	12	6
EBCT (min)	5	10	20

Table 2. Water quality conditions

Parameters	Average	Minimum	Maximum
Temperature (°C)	22.9	18.0	28.3
Turbidity (NTU)	1.72	0.22	3.28
pH	6.99	6.62	7.67
Alkalinity (mg/L)	35.6	16.5	54.5
Hardness (mg/L)	58	38	75
UV_{254} (cm^{-1})	0.021	0.010	0.026
DOC (mg/L)	2.32	1.28	4.11

zometer for measuring headloss, were installed on the wall of each reactor. The settled water was fed to the reactor directly to inoculate the microorganisms on the activated carbon surface and continued until a steady state was achieved. A steady state was attained at 7 months from initial instillation.

Table 2 represents the settled water quality conditions. Backwashing was conducted every three days, and flow rate for backwashing was 30 m/hr. Bed expansion during backwash was 20% and a water backwashing method was used for this research. Main analytical items were conducted on pH, temperature, DOC, UV_{254}, turbidity, heterotrophic plate count (HPC), biomass concentration, and THM.

2. Material and Analytical Procedures

2-1. THM Analysis

Four kinds of THM (Chloroform, Bromodichloromethane, Dibromo-chloromethane, and Bromoform) were analyzed by using the EPA modification method 551. The THM species were extracted from the aqueous phase to a solvent phase, which was analyzed by a gas chromatograph using HPLC grade methyl-tert-butyl ether (MTBE). 3 mL of HPLC grade methyl-tert-butyl ether (MTBE) and 4.0 g of sodium chloride (NaCl) were added to the sample vial and the vial was shaken for 30 sec and allowed to set for approximately 10 min. 1.5 mL of THM containing MTBE extract was transferred to a 2 mL vial for analysis. A Hewlett Packard 5890 Series II Gas Chromatograph equipped with a Nickel electron capture detector (ECD) and HP-5 column was used to analyze the MTBE extract. GC carrier gas was ultra high purity nitrogen with a flow rate of 1 mL/min. Injection volume was 1 μL, and the temperature of the injector and detector was 120 and 250 °C, respectively.

2-2. Biomass Determination

The biomass attached to the media was quantified by a phospholipids assay, reported by Findlay et al. [10]. Approximately 1 g of media with attached biofilm was removed from the biofilters and rinsed with deionized water. The phospholipids were removed from the biofilm through a liquid-liquid extraction procedure. Chloroform, methanol, and water were added at a ratio of 1 : 2 : 0.8. Then, the media was added at a weight ratio of 7 : 1 for chloroform and media. After 2 hours, chloroform and water were added at a ratio of 1 : 1, respectively. The mixture was left for 24 hrs at which point the aqueous layer was removed with a pipette. Chloroform was passed through a Whatman 2 V filter to remove media and insoluble cell

matter. Lipids containing chloroform phase were evaporated under a stream of nitrogen. The dried phospholipids were digested by adding a saturated potassium persulfate solution and heating the ampoules for 2 hrs at 100 °C.

Ammonium molybdate (0.1 mL; 2.5 % $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ in 5.72 N H_2SO_4) was added to the liberated phosphate and the mixture was allowed to stand for 10 min. The phosphomolybdate was complexed with malachite green (0.45 mL; 0.111% polyvinyl alcohol dissolved in water at 80 °C was cooled down and 0.011% malachite green was added) and left for 30 min. Absorbance at 610 nm for each sample was measured with a UV-visible spectrophotometer (UV-1601, Shimadzu). Glycerol phosphate standards ranging from 5 to 50 nm of phosphate were prepared. This bioassay was recorded as n moles phosphate/gram of media. These values were converted to biomass values by correlation equations that were determined by Findlay et al. [10] using an enrichment culture to enumerate cell number, dry weight, total carbon, and total nitrogen.

2-3. Microbial Identification

Heterotrophic bacteria from activated carbon filled in a pilot scale column were inoculated on R2A agar plates by using a spread method. Heterotrophic bacteria were detached from the activated carbon by violently shaking a sterilized bottle and the supernatant in the bottle was diluted to inoculate 10^3-10^4 times. The inoculated sample plates were incubated at 20±0.5 °C for 72±3 hrs, and then the produced colonies were counted.

To identify bacteria, an isolated colony was streaked on a tryptic soy-broth agar (TSBA) plate in a quadrant streak pattern with an inoculation loop sterilized in a flame and incubated at 20±0.5 °C for 72±3 hrs. Colonies were harvested from the most dilute quadrant exhibiting confluent growth along the streaking axis. Colonies were harvested by using the quadrant 3 method, which yielded the most stable fatty acid compositions since the inoculums had been adequately diluted to result in an abundant growth of colonies without a limiting nutrient supply. The fatty acid from the harvested pure colonies was extracted by procedures such as saponification, methylation, extraction, and base wash. The 2 μL of extracted fatty acid was taken and injected into a GC/FID (HP 6890 Series, USA) with a microbial identification system (MIDI). Then the resulting fatty acid compositions were compared to MIS (microbial identification system) standard libraries.

2-4. Other analyses

Turbidity, temperature, and pH were measured from samples taken during the pilot plant experimentation. UV_{254} was measured at a wavelength of 254 nm with a UV-visible spectrophotometer (Shimadzu, USA) with a quartz cell having a path length of 1 cm. All UV_{254} samples were 0.45 μm filtered prior to analysis. Turbidity was measured by using a calibrated Hach 2100P turbidity meter (Hach, USA). HPC was measured with an R2A agar plate according to Standard Methods APHA et al. [11]. Samples were inoculated in a clean bench with a sterilized 10 mL vial to prevent contamination. HPC plates were counted after 7 days of incubation at 20±1 °C and were represented as a colony-forming unit (CFU/mL).

RESULTS AND DISCUSSION

1. Dual Media BAC Process Performance

This study investigated the performance of a sand layer in a dual media BAC process for the removal efficiency of particles, DOC, and THM and the extent of microbial detainment. Fig. 2 shows removal efficiency of turbidity in effluent for R1, R2, and R3 compared with port 8. The hydraulic loading rates in each reactor were 24 m/hr for R1, 12 m/hr for R2, and 6 m/hr for R3, respectively. EBCT was 5 min for R1, 10 min for R2, and 20 min for R3, respectively.

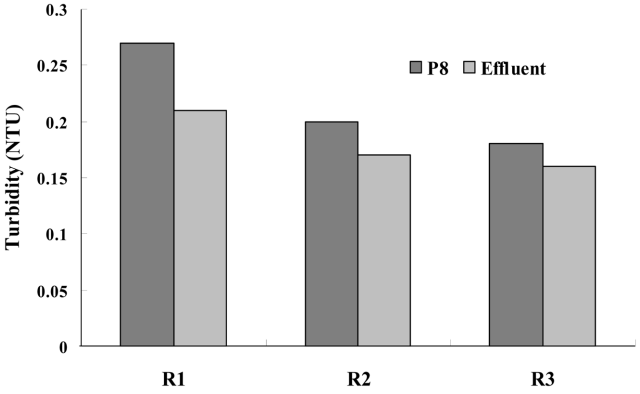

Fig. 2. Turbidity removal through a sand layer in a dual media BAC process.

It appeared that the turbidity at port 8 and effluent in each reactor was affected by HLR. In other words, higher HLR levels resulted in higher turbidity. The removal efficiency of particles was higher at high HLR levels than at low HLR levels. The largest difference between port 8 and effluent occurred in R1 and the value was 0.06 NTU. The smallest difference occurred in R3 and the value was 0.02 NTU. From this result, it was concluded that turbidity with approximately 0.02-0.06 NTU could be removed from a sand layer in a dual media BAC process.

Fig. 3 shows the HPC values in water at port 8 and effluent. The tendency of HPC values was similar to turbidity. This clearly showed that a sand layer restricts HPC leakage from GAC to effluent. This phenomenon was consistent with a study conducted by Prevost et al. [12]. The extent of restriction of HPC in filter media was larger at high HLR levels than low HLR levels. The ratio for HPC at effluent to port 8 in both R1 and R2 was approximately 0.1, respectively. However, the ratio for HPC of effluent to port 8 in R3 was 0.47.

Fig. 4 represents DOC removal efficiency in the sand layer in

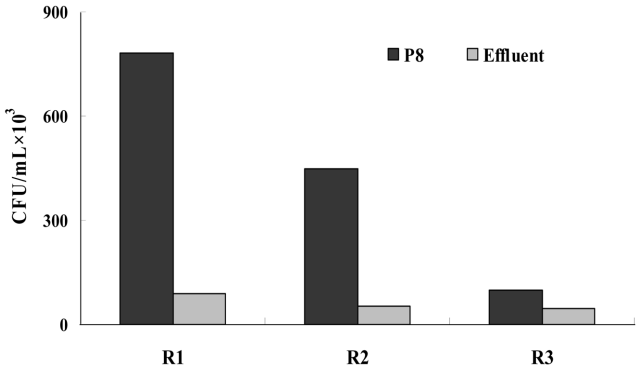

Fig. 3. The removal of HPC through a sand layer in a dual media BAC process.

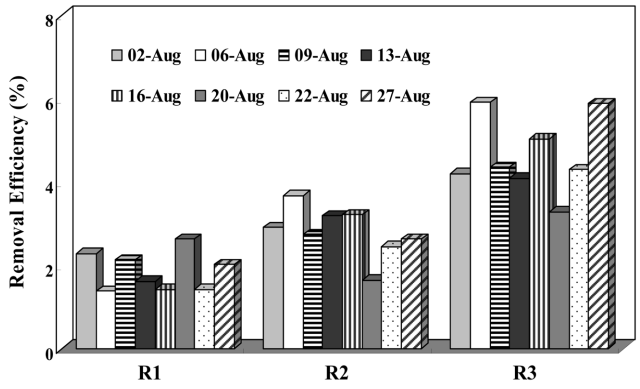

Fig. 4. DOC removal through a sand layer in a dual media BAC process.

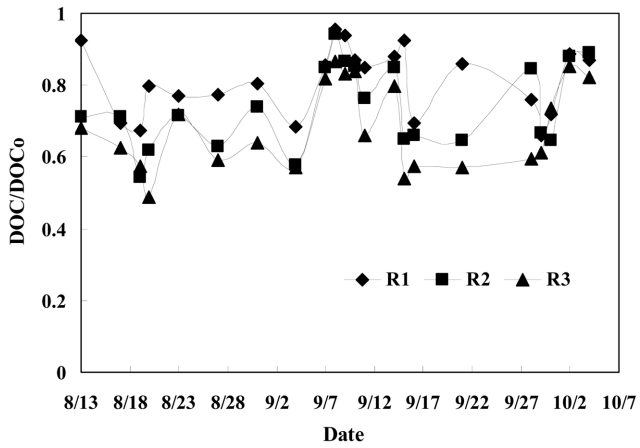

Fig. 5. Variation of DOC removal from each reactor during experimental period.

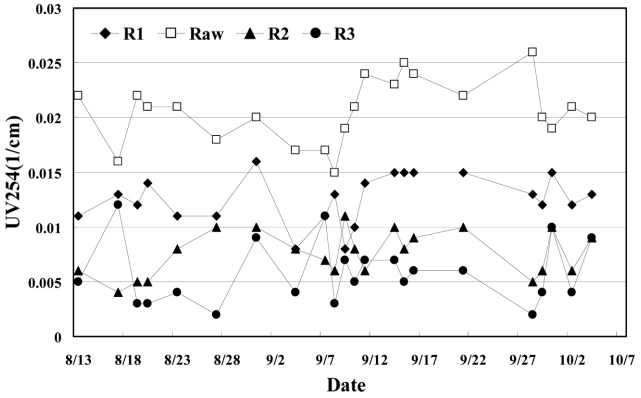

Fig. 6. Variation of UV254 from each reactor during experimental period.

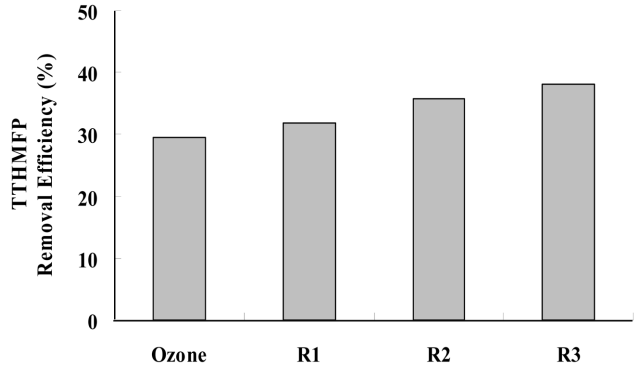

Fig. 7. TTHMFP removal efficiency in ozonated raw water and effluent from each reactor.

effluent compared to port 8. Removal efficiency increased with a decrease in HLR. The mean value of removal efficiency in each reactor was 1.87, 2.81, and 4.64% for R1, R2, and R3, respectively. Based on the above results, the presence of a fine sand layer underneath a carbon layer enhanced particle removal in this experiment.

DOC removal versus HLR during this experimental period is plotted in Fig. 5. These experiments were conducted with fixed conditions for HLR and EBCT values in each reactor. The most efficient removal of DOC was observed in the lowest HLR values, and the most inefficient removal of DOC was in the highest HLR. The average ratio of DOC/DOC_0 in each reactor during this experiment period was approximately 81, 74, and 69% for R1, R2, R3, respectively. This proved that higher EBCT showed to be more effective in DOC removal.

Fig. 6 represents a comparison between UV_{254} in the effluent from each reactor and raw water during the experimental period. The UV_{254} value of raw water ranged from 0.015 to 0.026 cm^{-1} which implies that the water did not include refractory degradable organics that are not easily degraded by microorganisms. The trends in variation of UV_{254} in each reactor were higher at a low HLR, consistent with the value of raw water and reduction of UV absorbance. However, curves sometimes intermediately humped in treated water when the UV_{254} value of raw water was low. This may be caused by UV_{254} that was detected by a leakage of microbial by-products.

When water containing dissolved organic carbon was chlorinated, chlorinated compounds such as THM were produced. This experiment was conducted to determine THM removal efficiency in ozonated raw water and effluent from each reactor during the operation of a BAC process. Fig. 7 compares TTHMFP removal efficiency between ozonated raw water and effluent from each reactor during biofiltration. Removal efficiency in the effluent from biofiltration reactors was approximately 2 to 9% higher than that of ozonated raw water and increased with an increase in EBCT. R3, which has considerable biomass based on organic mass, should be advantageous in the removal of a biodegradable reaction site.

Fig. 8 represents the removal efficiency of the THM species in each reactor. According to the results, chloroform ($CHCl_3$) and dichlorobromomethane ($CHBrCl_2$) were removed regardless of EBCT values. However, the results of dibromochloromethane ($CHBr_2Cl$) and bromoform ($CHBr_3$) clearly showed that removal efficiency was increased with increasing of EBCT.

2. Biomass on Activated Carbon

Successful performance depended on biomass and microbial activity during the operation of the BAC process. Fig. 9 shows biomass concentration determined as phospholipids in a steady state for R3. Biomass concentration was determined after 3 and 7 months from the induction of this experiment.

Biomass concentration on an activated carbon surface taken after 7

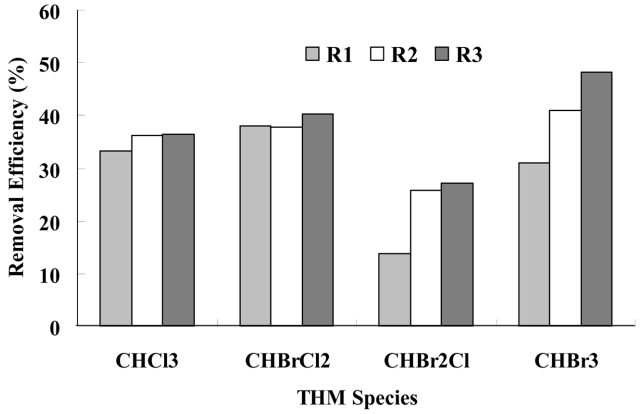

Fig. 8. Removal efficiency of THM species in each reactor.

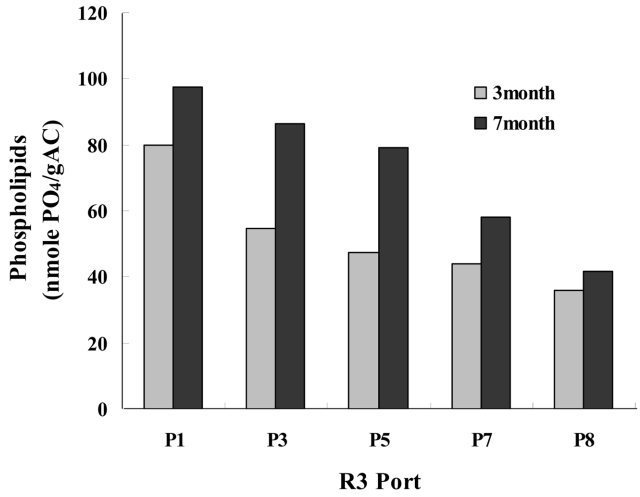

Fig. 9. Biomass concentration after 3 and 7 months according to reactor depth in R3 measured as phospholipids.

months was higher at a minimum of 15.6% to a maximum of 66.4% compared to 3 months. Biomass concentration decreased with increasing of bed depth. However, biomass at port 3 was greatly reduced in the third month sample. Furthermore, it showed an additional 25 n moles compared to port 1, which suggested that microorganisms could not disperse to a low bed depth.

Biomass concentration at the bottom port was maintained at over 50% compared to the top layer in all reactors. Prevost et al. [12] observed biological activity at the interface of sand-GAC packed in a dual media filter. However, Carlson et al. [13] reported that the biomass profile decreased according to media depth exponentially and the biomass concentration on the bottom layer was calculated as near zero in the biological single anthracite filter. It is believed that these differing results were due to the bed configuration between single and dual media filters and microorganisms detaching from the upper layer media and escaping from the bed to effluent in single layer filters. Yet, dual media filters could detain the microorganisms at the sand-GAC interface. According to these results, dual media filters were more effective for sustaining microorganisms than single layer filters.

Fig. 10 represents biomass concentration measured as phospholipids at reactor depths with different EBCT and HLR. Biomass concentration decreased with the decrease in HLR under 12 m/h of HLR. Microorganisms could not grow due to the short contact time between microorganisms and organic matter at a high HLR (over 12 m/h). Biomass decrease on bed depth was clear for R1 and R3 but the biomass difference between each port was not significant for R2. Biomass concentration in R2 was higher than R1 except port 1. The lowest value was recorded at port 8 in R3 registering 41.7 n moles of PO_4^-/g of activated carbon.

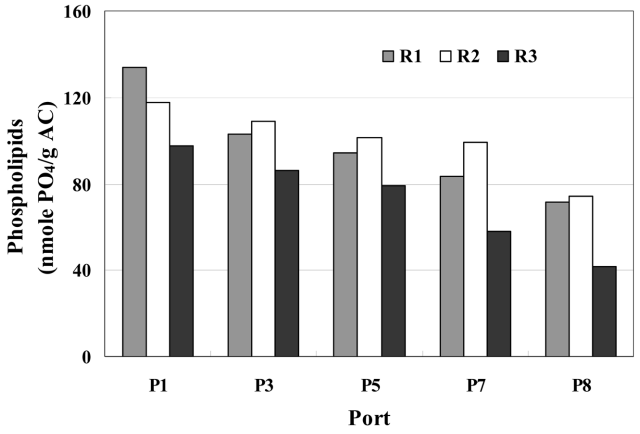

Fig. 10. Biomass concentration measured as phospholipids according to reactor depth in R1, R2, and R3.

3. Identification of Microorganisms on Activated Carbon Surface

This research identified the predominant species attached to the surface of activated carbon and evaluated microbial distribution in this dual-media BAC system. Table 3 represents the identified species and counts. There were 10 types of species identified with *Clavibacter* and *Corynebacterium* as predominant species. *Acidovorax*, *Sphingomonas*, *Flavobacterium*, *Cellulomonas*, *Serratia*, *Ancyclobacter*, *Rhodococcus*, and *Brevundimonas* were also present in these samples.

Table 4 represents the distribution of microorganisms identified at each port in R1, R2, and R3. The differences in microbial distribution were distinct at the upper and lower parts in each reactor. Various types of bacteria were found at port 1 in reactors R2 and R3, but the number of units was low except for *Clavibacter*. At port 3, the predominant species were *Clavibacter* and *Corynebactrium*. At port 5, the predominant species were *Clavibacter* and *Corynebactrium*. One unit of *Ancyclobacter* and *Cellulomonas* was found at port 8 in R1 and R3.

It was thought that HLR affected the variability of microorganisms. Seven types of microorganisms were identified in R3, which had the lowest HLR. However, four types of microorganisms were identified in R1 and R2. It was assumed that low HLR offered microorganisms the chance to attach easily to activated carbon. Most of the identified microorganisms from the activated carbon surface in this experiment are similar to the result of Yoon [14], which identified heterotrophic bacteria in the distribution system in the Seoul metropolitan area except *Clavibacter*, *Rhodococcus*, and *Serratia*. The species of *Corynebacterium*, *Serratia* were identified in distribution water by Geldreich et al. [15].

Table 3. Species and bacteria counts identified in activated carbon in a dual media BAC process

Species	Bacteria counts
Corynebacterium	7
Clavibacter	16
Acidovorax	2
Cellulomonas	1
Ancyclobacter	1
Rhodococcus	1
Sphingomonas	2
Serratia	1
Brevundimonas	1
Flavobacterium	2

Table 4. Distribution of microorganisms in R1, R2, and R3

	R1	R2	R3
Port 1	Not matching with library	*Flavobacterium*(1) *Sphingomonas*(1)	*Brevundimonas*(1) *Rhodococcus*(1) *Sphingomonas*(1) *Serratia*(1) *Clavibacter*(3)
Port 3	*Corynebactrium*(2) *Acidovorax*(2)	*Flavobacterium*(1) *Clavibacter*(2)	*Clavibacter*(2)
Port 5	*Clavibacter*(4)	*Clavibacter*(1) *Corynebactrium*(1)	*Clavibacter*(2) *Corynebactrium*(2)
Port 8	*Coynebacterium*(1) *Ancyclobacter*(1)	*Clavibacter*(1) *Corynebactrium*(1)	*Cellulomonas*(1) *Clavibacter*(1)

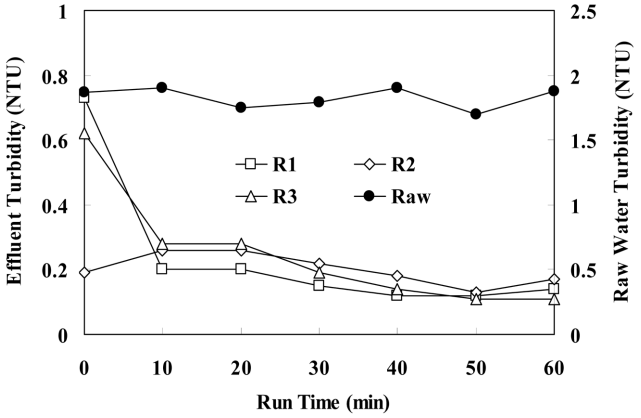

Fig. 11. Initial turbidity breakthrough after backwashing in each reactor.

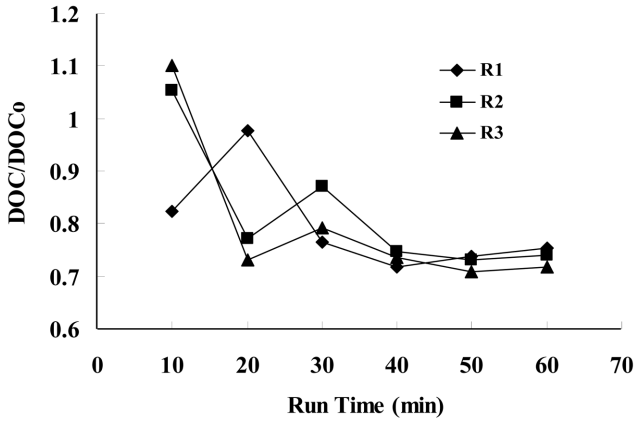

Fig. 12. Initial DOC breakthrough after backwashing in each reactor.

4. Backwashing and Head Loss

Drinking water treatment filters generally have two objectives: particle removal and DOC reduction. Filter beds must be cleaned regularly in order to maintain flow rates within an acceptable range and prevent particle and organic breakthrough. These experiments were conducted to consider backwash cycles, determine an appropriate filter stabilization time after backwashing, and measure the removal of turbidity, DOC, and the change in headloss during biofiltration with three HLRs of 24, 12, and 6 m/hr.

Fig. 11 represents the recovery time required to establish a stable condition for the removal of turbidity after backwashing. The rapid recovery and stabilization in the bed is an important factor in the removal of particles and DOC during backwashing. Initial turbidity breakthrough in all reactors was recovered within 10 min after backwashing. Turbidity in R1 and R2 with the largest HLR drastically decreased with recovering bed stabilization. However, R3 with the lowest HLR was more insensitive than R1 and R2 during backwashing. Compared with the annual filtered water turbidity in 1997 of nine water treatment plants in Seoul city, which ranged from 0.08-0.27 NTU to 0.08-0.14 NTU for the Tukdo water treatment plant, the turbidity of Tukdo plant was lower. If 0.2 NTU was considered the critical value for filtered water, the time required to reach the critical value was approximately 40 min.

Fig. 12 represents initial DOC breakthrough after backwashing in each reactor. The stable removal of DOC was accomplished after approximately 40 min of backwashing and then stability was maintained. It was discovered that DOC removal in R1 and R2 after backwashing was significantly lower, in which average operational ratio of DOC/DOCo was approximately 0.80 and 0.77, respectively.

Fig. 13 shows turbidity breakthrough during filter runs in each reactor. After initial breakthrough, turbidity remained stable under a critical value of 0.2 NTU. The breakthrough in R1 occurred at 24 hours; increase in turbidity began at 48 hours in R2 and exceeded the critical value at 72 hours after backwashing. Turbidity in R3 with the lowest HLR of 6 m/hr did not change significantly at 48 hours after backwashing. Turbidity gradually increased to 0.22 NTU, which was slightly higher than the critical value after 48 hours, and the value remained constant for up to 96 hours after backwashing.

Fig. 14 shows DOC breakthrough during filter runs in each reactor. DOC breakthrough was different from that of turbidity. The difference in value for DOC/DOCo for R1 with operation time was higher than that of R2 and R3. Rapid DOC breakthrough occurred at 24 hours after backwashing for R1. The longest DOC breakthrough time occurred in R3. The variation of DOC/DOCo was not significant until breakthrough. Breakthrough occurred abruptly for R2 and R3 and the time to breakthrough was 48 and 96 hours after backwashing, respectively.

Fig. 15 represents the variation of head loss during filter runs in each reactor. Head loss represented the reduction of water pressure

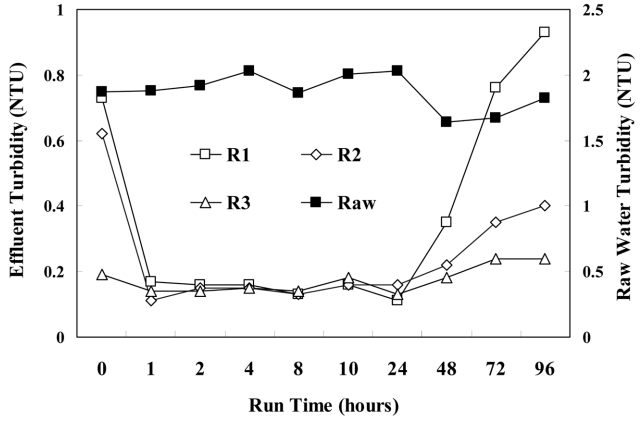

Fig. 13. Turbidity breakthrough during filter runs in each reactor.

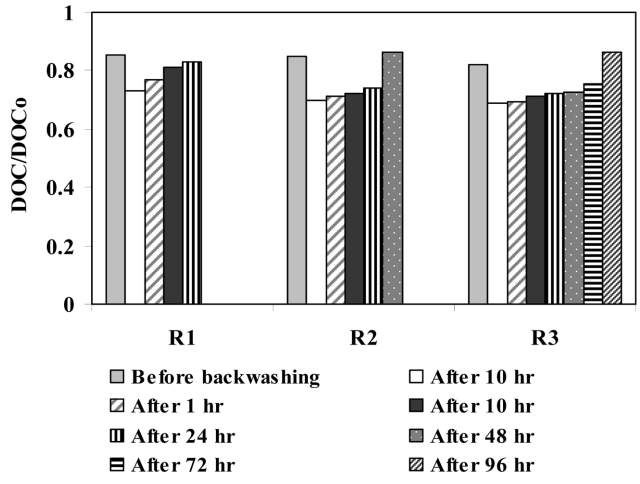

Fig. 14. DOC breakthrough during filter runs in each reactor.

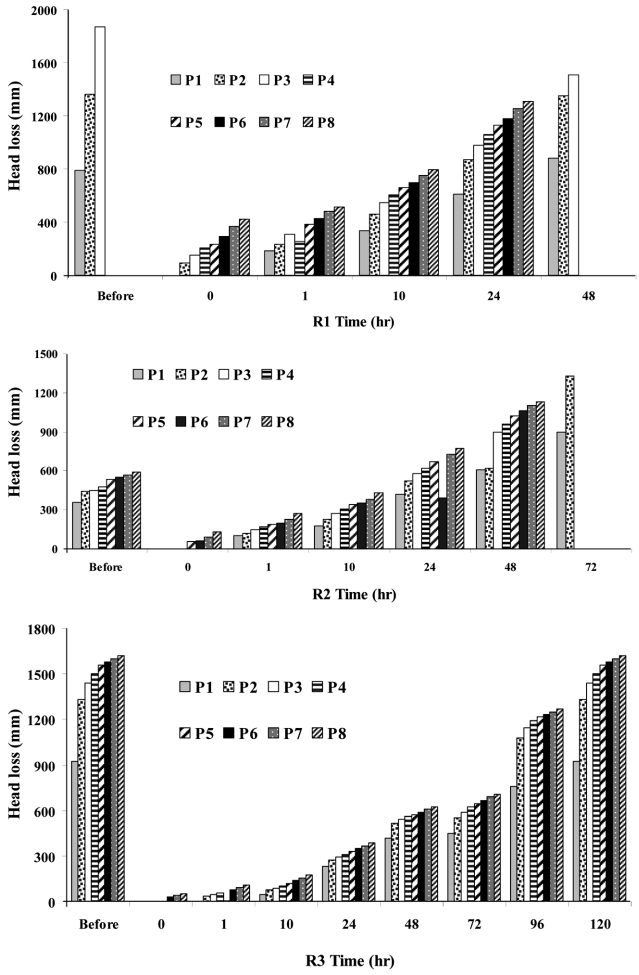

Fig. 15. Variation of head loss during filter runs in each reactor.

acting in the bed and occurred when a bed pore was blocked by particles. A water level of 1.3 m from the top of the bed surface was maintained during all experimental periods. The accumulation of head loss gradually increased with the increase in filter run time. The initial increase in head loss occurred at R1 and the longest was recorded in R3. The piezometer water level in R3 was maintained for 120 hours after backwashing. The levels in R1 and R2 were maintained for up to 24 and 48 hours after backwashing, respectively. Furthermore, head loss could not be measured because the water levels of the piezometer were not consistent. Head loss at 96 hours after backwashing significantly increased compared to 72 hours at R3. The water level of the final piezometer reading was 1,310, 1,130, and 1,270 mm for R1, R2, and R3, respectively. Breakthrough of turbidity and DOC occurred when the highest accumulated head loss value and water level were below 1,100 mm. Therefore, the backwashing cycle in this study was configured to 24, 48, and 72 hrs for R1, R2, and R3, respectively.

CONCLUSIONS

This study was carried out to estimate and optimize a dual media BAC process for DOC removal and DBP control. Pilot scale tests were carried out at the Tukdo water treatment plant in Seoul, Korea. The dual media BAC process proved to be superior in removal efficiency of DOC and THMFP and sustained capacity of microorganisms more efficiently than a single layer filter. The bottom layer of the sand filter used in this study functioned as a screen for turbidity, microorganisms, and another biological media to remove traceable DOC. Detainment of turbidity and HPC was larger at high HLR levels than low HLR levels. DOC removal was higher with a long EBCT than a short EBCT. TTHMFP of effluent from each reactor decreased approximately 2-9% more than that of ozonated raw water. When estimating the removal efficiency of THM species, brominated THM was found to decrease significantly with the increase of EBCT. The leakage of turbidity and DOC continued for 40 min after backwashing in all reactors. A backwashing cycle was performed at 24, 48, and 72 hrs for R1, R2, and R3, respectively. Biomass concentration decreased with the decrease of HLR under 12m/h of HLR. The predominant species present were *Clavibactor* and *Corynebacterium* from activated carbon filled in BAC reactors.

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