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Effects of phosphate and hydrogen peroxide on the performance of a biological activated carbon filter for enhanced biofiltration

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1 **Abstract**

2 Biofilm formation on biofilters can influence their hydraulic performance, thereby leading to head loss
3 and an increase in energy use and costs for water utilities. The effects of a range of factors, including
4 hydrogen peroxide and phosphate, on the performance of biological activated carbon (BAC) and biofilm
5 formation were investigated using laboratory-scale columns. Head loss, total carbohydrates, and
6 proteins were reduced in the nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced
7 BAC filters. However, there were no changes in the removal of dissolved organic matter,
8 trihalomethane formation potential, or selected trace organic contaminants. The biofilm formation on
9 polyvinyl chloride and stainless steel coupons using the laboratory biofilm reactor system was lower
10 when the effluent from a nutrient-enhanced column was used, which indicated that there was less
11 biofilm formation in the distribution systems. This may have been because the effluent from the
12 nutrient-enhanced column was more biologically stable. Therefore, enhanced biofiltration could be used
13 not only to reduce head loss in biofilters, but also to delay biofilm formation in distribution systems.

14

15 *Keywords:* biological activated carbon; biofilm; hydrogen peroxide; phosphate

16

17 **1. Introduction**

18 Biological activated carbon (BAC) is a cost-effective drinking water treatment process that removes
19 contaminants mainly via biodegradation, which is mediated by indigenous microorganisms. BAC
20 processes have several advantages in water treatment, including the removal of diverse contaminants
21 [1–3] and reduction of dissolved organic carbon (DOC), which includes precursors of disinfection
22 byproducts. The hydraulic performance of BAC filters is integral to meeting the supply-demand
23 requirements of water treatment works and can be substantially limited by biofilm development.
24 Excessive biofilm accumulation within BAC filters leads to significant head loss and more frequent

backwashing, which influences the production of drinking water and the operation cost. Therefore, it is important to understand and control the process of biofilm formation within BAC filters.

According to Kirisits et al. [4], extracellular polymeric substances (EPS) production is significantly reduced in phosphorus-supplemented biofilters compared with that in phosphorus-deficient biofilters. Similarly, nutrient starvation promoted EPS production, within the context of microalgae cultivation [5]. It has been shown that EPS production is enhanced in response to phosphorus limitation and that this might serve as a protective mechanism (bacterial adhesion, adsorbing and storing nutrients) [6]. Additionally, phosphorus limitation can impact biofilm physical structure and morphology, thus playing an important role in increasing head loss in BAC [7]. Low phosphorus concentrations in BAC influent can promote EPS production, which can increase the head loss in BAC filters and reduce the process efficiency. Generally, phosphate removal occurs with the addition of coagulants in coagulation-sedimentation-filtration processes, which results in phosphorus-deficient water at a concentration of 0.01 mg/L [8]. However, the impact of phosphorus on BAC biofilms, particularly the EPS characteristics, has not yet been fully explored. According to a previous study, most biofilm extracellular matrixes account for over 90% of the dry mass, whereas microorganisms account for less than 10% [9]. Peroxide enhancement reduces terminal head loss by up to 60% compared with that of control biofilters without deterioration in water quality performance [10]. Moreover, the results from laboratory studies, such as batch reactors with sand and water, have revealed that hydrogen peroxide concentrations between 3 mg/L and 5 mg/L increase the DOC removal while reducing microbial activity [11]. However, Stoddart and Gagnon reported that such oxidant and nutrient enhancement strategies do not show any improvement in water quality performance with respect to total organic carbon (TOC), DOC, specific ultraviolet absorbance (SUVA), and disinfection byproduct formation potential [12]. While biofiltration enhancement via nutrients and/or oxidants is effective in DOC removal, some studies showed that small changes in the magnitude of the mean difference have no practical operational importance [12]. Therefore, more investigations are necessary to determine the effects of nutrient and/or oxidant enhancement on the removal of DOC.

There have been no in-depth studies on the impacts of these parameters on EPS characteristics and their interactions with biofilter hydraulic performance, which includes any subsequent downstream impacts, such as on drinking water distribution systems (DWDSs). This study aimed to assess the impact of enhancing nutrients (phosphate) and/or oxidants (hydrogen peroxide) in BAC filters on the quantity and composition of EPS and filter performance. The performance of water quality improvement via BAC was determined by DOC, trihalomethane formation potential (THMFP), and trace organic contaminants (TrOCs). The biofilm formation in effluent discharged from the enhanced BAC filters was also investigated to assess the subsequent impacts on the DWDS.

2. Materials and methods

Detailed information on the batch experiments is explained in Text S1.

2.1 Columns and biofilm reactors

Four BAC columns were set up to assess the effects of phosphate and hydrogen peroxide enhancement on the performance of BAC filters and their biofilm characteristics, which included i) columns that did not have any phosphate or hydrogen peroxide added, ii) nutrient-enhanced columns using phosphate, iii) oxidant-enhanced columns using hydrogen peroxide, and iv) nutrient + oxidant-enhanced columns using phosphate and hydrogen peroxide. Each column was a glass cylinder with a diameter of 150 mm and height of 800 mm. All columns were filled with BAC collected from a full-scale drinking water treatment plant, and they were kept in a dark room (Fig. S1). The empty bed contact time of each column was 20 min (flow velocity of 1.2 m/h), and backwashes were conducted with air and water every 10 d. Feed water was collected after coagulation/flocculation-sedimentation followed by rapid sand filtration in a full-scale drinking water treatment plant (Seoul Metropolitan Waterworks, Seoul, Republic of Korea) (Table S1). Then, it was fed to all the BAC columns after ozone (O_3) treatment at a concentration of 0.8 mg O_3/mg DOC.

The nutrient-enhanced column was continuously fed with a C:P ratio of 100:10 in the influent, which was based on the results from the preliminary tests. The oxidant-enhanced columns were continuously fed with 1 mg/L of hydrogen peroxide determined based on the results of a previous study [10]. The nutrient + oxidant-enhanced column was injected with both phosphate and hydrogen peroxide at the same concentrations used for the nutrient-enhanced and oxidant-enhanced columns. Moreover, selected TrOCs, which included gemfibrozil, ibuprofen, pentoxifylline, naproxen, phenacetine, bezafibrate, diclofenac, ketoprofen, fenoprofen, caffeine, and carbamazepine, were introduced into the BAC columns at a concentration of about 1 μg/L (Table S2). Centre for Disease Control (CDC) biofilm reactors (CBR, Biosurface Technologies Bozeman, MT, USA) were fed with BAC filtered water to determine the biofilm formation potential of the post-filtered water (Table S3). The CDC reactors consisted of polyvinyl chloride (PVC) and stainless steel coupons, and the effluent from the BAC columns was used as feed water for the CDC biofilm reactors. The *p* values of all the experimental results were calculated through an analysis of variance test, which used a level of significance of 5%, and the analysis was performed using SPSS 18 (SPSS Inc., Chicago, IL, USA).

2.2 Extraction and analysis of the extracellular polymeric substances

In this study, sonication was used in order to recover biofilm. First, 1 g of each BAC sample was diluted in a 100 mL glass flask with 50 mL of deionized water. Then, samples were detached by vortexing for 1 min and sonicating using an ultrasonicator (Cole-Parmer 8890, Vernon Hills, IL, USA) at 190 W for 3 min [13]. EPS were extracted from the biofilm samples using the formaldehyde combined with heating protocol described by Evans [14], which separates the total carbohydrates and proteins. In brief, 2 g of wet BAC, 10 mL of 0.01 M phosphate buffer solution (pH of 7), and 60 μL of 35% formaldehyde (Desung, Republic of Korea) were combined in a 50 mL sterile centrifuge tube [15]. After shaking at 400 rpm for 1 h at 4 °C, the tube was heated in a water bath at 80 °C (WB-6, DAIHAN, Republic of Korea) for 10 min. Finally, the sample was centrifuged (MF300, Hanil, Republic of Korea) at 5000 g for 10 min. The carbohydrate content of the extracted EPS was determined using a phenol-sulfuric acid

assay, and glucose was used as the standard [16, 17]. A Bradford assay was used for the determination of proteins using Bradford reagent (Bradford Reagent 5x, SERVA Electrophoresis GmbH, Germany) [16]. Absorbance was measured using a UV-Vis spectrophotometer (DR5000, Hach, USA).

2.3 Dissolved organic matter characteristics and assimilable organic carbon

BAC effluents were passed through a 0.45 μm filter (Whatman, USA). DOC and UV_{254} were analyzed using a TOC analyzer (TOC-V CPN, Shimadzu, Japan) and spectrophotometer (DR5000, Hach, USA). A liquid chromatography-organic carbon detector (LC-OCD, Model 8, DOC Labor, Germany) was used to separate dissolved organic matter into high molecular weight (HMW) fractions, intermediate molecular weight (IMW) fractions, and low molecular weight (LMW) fractions [18]. A fluorescence excitation-emission matrix (EEM) was used to analyze the characteristics of fluorescent dissolved organic matter. The fluorescence emission was measured using a spectrofluorometer (RF-5301, Shidmazu, Japan) with an arc lamp as a light source. The details have been described in our previous study [19]. An assimilable organic carbon (AOC) analysis was performed using flow cytometry (FCM), as described by Eawag (Swiss Federal Institute of Aquatic Science and Technology), to evaluate the removal of AOC via BAC columns. Details of the survey method are reported elsewhere [20–22]. The AOC was used to determine the biological stability which determine the inability of drinking water to support microbial regrowth.

2.4 Total cell counts, heterotrophic plate counts, and adenosine triphosphate

Total cell counts (TCC) in a sample were enumerated using FCM (Cube 6, Partec, Germany) following a protocol described in Park et al. [23]. Heterotrophic plate count (HPC) analysis was conducted to evaluate the growth potential of bacteria. In brief, 1 mL of the samples collected from the BAC columns was diluted and streaked on an R2A agar (Difco, USA) plate medium. To evaluate cell activity, adenosine triphosphate (ATP) measurements were calculated using BacTiter-Glo™ reagent (Promega, USA).

2.5 Trihalomethane formation potential and trace organic contaminants

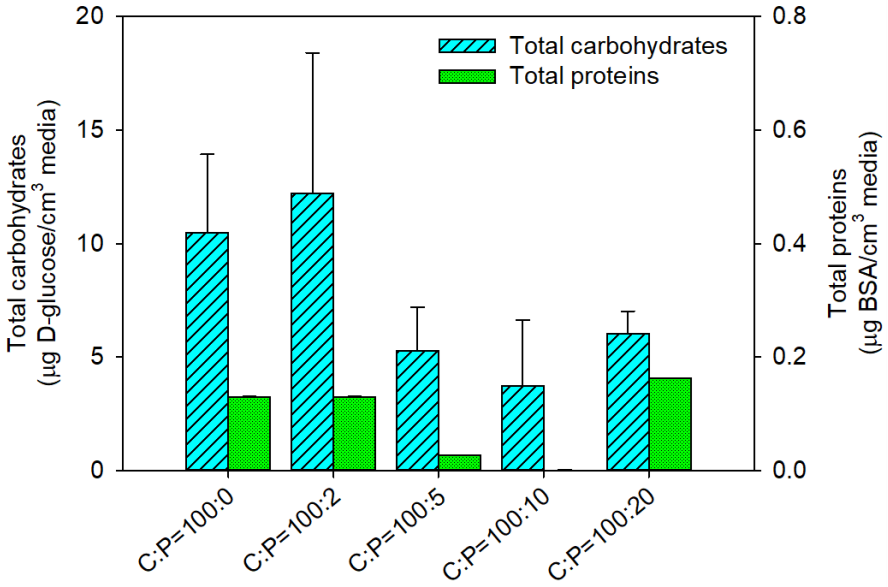
BAC effluent samples were adjusted to a pH of 8 and buffered with a borate buffer solution. The effluent samples were chlorinated with sodium hypochlorite at a dosage of 3 mg Cl_2/mg C, and were incubated at 20 °C for 48 h in headspace-free 300 mL amber bottles. After, the free chlorine remaining after the incubation was quenched with sodium sulfite prior to determining the trihalomethane concentrations using gas chromatography - electron capture detection (GC-ECD, Agilent 6890N, USA). The details have been described in our previous work [19]. The selected TrOCs were prepared in a stock solution of 1 g/L in high performance liquid chromatography (HPLC)-grade methanol (JT Baker, USA) and introduced at a concentration of 1 µg/L into the BAC columns. Selected TrOCs were preconcentrated using the column switching method with a concentration column (Hypersil Gold aQ; Thermo Fisher Scientific) and an analytical column (Hypersil Gold C18). Mass spectrometry measurements were performed on a high-resolution full scan Orbitrap Exactive mass spectrometer (Thermo Fisher Scientific, Bremen, Germany) [24].

3. Results and discussion

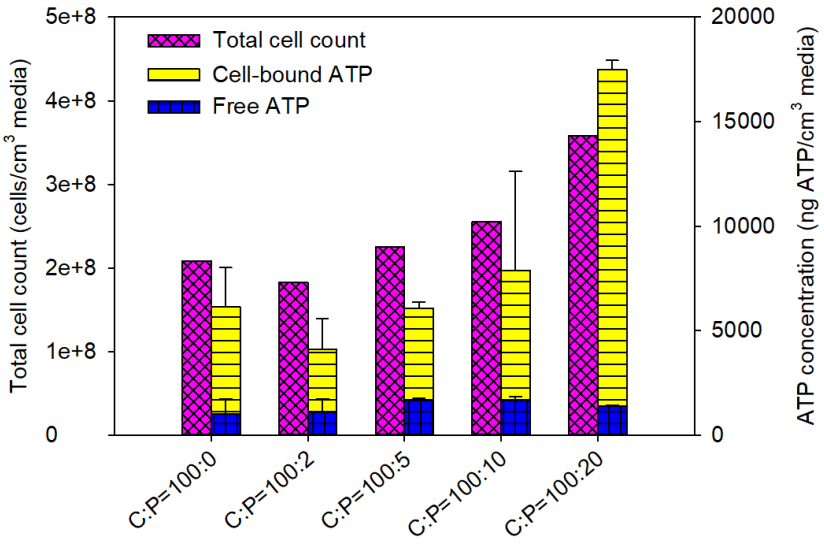
3.1 Impact of phosphate concentration on biological activated carbon biofilms

Comparison of the BAC biofilms developed under different C:P ratios using batch reactors showed that the highest concentration of carbohydrates and proteins, which were used as an estimate of EPS, was observed in a C:P ratio of 100:0, as shown in Fig. 1a. The EPS concentrations were 10.3, 9.0, 5.3, 3.7, and 6.0 µg/cm^3 BAC for C:P ratios of 100:0, 100:2, 100:5, 100:10, and 100:20, respectively (Fig. 1a). The concentration of EPS formed at the C:P ratio of 100:10 was 64% lower than that at the C:P ratio of 100:0, and the TCC gradually increased as the C:P ratio decreased. The data collected indicated that EPS production was reduced with phosphate addition. However, the total EPS and protein concentration increased between the C:P ratios 100:10 and 100:20. This trend has not previously been reported and requires further exploration, which was beyond the scope of the current study. However, there may be

a phosphate threshold that favors changes in the EPS characteristics when a different microbial community is selected. Alternatively, the excess phosphate caused a substantial increase in cell activity and promoted EPS production. The total ATP in BAC increased with phosphate addition and showed a similar trend to that of the TCC (Fig. 1b). The C:P ratio of 100:10 was selected and used in the nutrient-enhanced columns because the lowest formation of EPS was observed in the batch study.



(a)

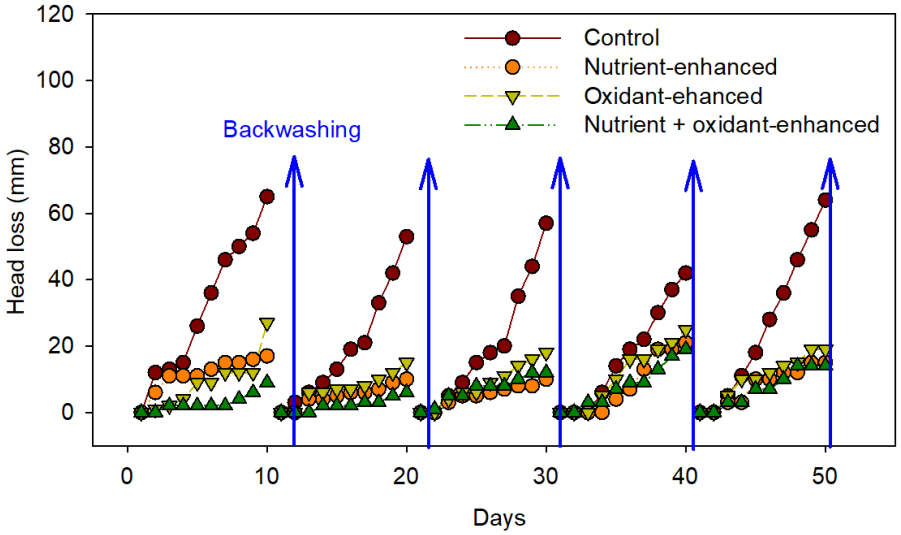


(b)

Fig. 1. (a) Total carbohydrates and total proteins associated with biological activated carbon (BAC) and (b) total cell counts and adenosine triphosphate (ATP) associated with BAC (n = 6).

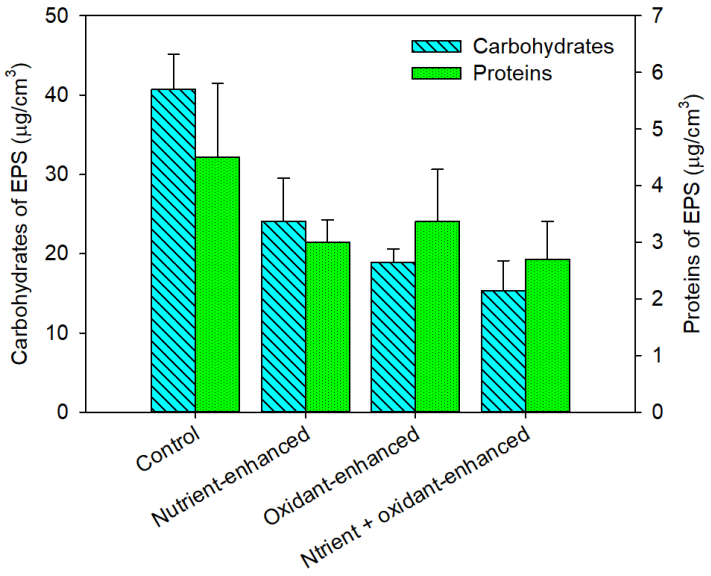
3.2 Head loss and extracellular polymeric substances characteristics in enhanced biological activated carbon filters

Fig. 2 shows the changes in head loss in the BAC columns as a function of backwashes that were conducted every 10 d (p<0.05) for 50 d of operation. The maximum head losses were 56, 15, 21, and 12 mm for control, nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced columns, respectively. In the oxidant-enhanced columns, the head loss resulted in a 63% reduction. Nutrient-enhanced and nutrient + oxidant-enhanced columns reduced the head loss by 74% and 79%, respectively. The differences in the average head loss between the enhanced BAC columns were not significant, which suggested that any enhancement reduced head loss. Fig. 3 shows the composition and concentration of EPS associated with each of the four BAC filters. The potential formation of EPS, which was estimated as protein and carbohydrates, is known as a potential fouling agent of biological filters [10]. Therefore, the higher production of EPS in the control BAC filter (C:P ratio of 100:0) may have accelerated the head loss as a result of filter clogging. The observation of the greatest head loss in the control column has previously been attributed to greater EPS production due to a community of long-term phosphorus-deficient bacteria [25], and it is possible that this also occurred in the BAC filters of the current study.



1

2 **Fig. 2.** Head loss profiles of enhanced biofilters.



3

4 **Fig. 3.** Extracellular polymeric substances (EPS) composition in terms of total carbohydrates and total
5 proteins in the biological activated carbon collected from the top of the column (0–50 mm) (n = 6;
6 p<0.05).

The concentrations of carbohydrates and proteins from biomass associated with BAC along the columns at different depths are presented in Fig. S2. The concentration of carbohydrates, which is the main component of EPS, was significantly high at the top of the control column. The EPS determined by summing the total carbohydrates and total protein concentrations was 41 μg/cm^3 BAC in the top layer (50 mm) of the control. For the nutrient-enhanced column, the EPS concentration was 24 μg/cm^3 BAC, which was 41% less than that at the top of the control. In the case of the oxidant-enhanced column, the total EPS concentration was 19 μg/cm^3 BAC, which was 54% lower than that of the control. For the nutrient + oxidant-enhanced column, the total EPS concentration was 15 μg/cm^3 BAC, which was 63% lower than that at the top of the control. The enhancement of BAC filter columns via phosphate and/or hydrogen peroxide reduced the EPS from 41% to 63%.

EPS are composed of protein-like substances, and fluorescence EEM spectroscopy has been extensively utilized to determine changes in protein-like substances [26]. Fluorescence EEM spectroscopy was performed to determine the fluorescent dissolved organic matter in EPS extracted from the top of each column (0–50 mm) where the highest EPS was observed (Fig. 4). The fluorescence intensities of the protein like-species T1 and T2 were not significantly different between the control and enhanced BAC columns. As shown in Fig. 1a and Fig. 3, the concentration of proteins was also not different between the control and enhanced BAC columns. A high-intensity fulvic-like substances peak was observed in the control, which indicated that more decomposed organic matter, such as fulvic acid, was associated with the control BAC. Therefore, the main fraction in the EPS associated with the BAC was humic, such as fulvic-like substances. The fluorescence EEM can only determine organic matter that has fluorescence characteristics, such as proteins, but not polysaccharides. However, an LC-OCD can detect both proteins and polysaccharides using an OCD detector. Therefore, this analysis was also applied to the EPS extracted from the top of the BAC columns (Fig. 5). The control column showed the highest concentration of the HMW fraction (565 μg/cm^3 BAC). The HMW fractions in the nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced columns were 53%, 60%, and 69% lower than those of the control, respectively (p<0.05). The concentration of carbohydrates, which was in the HMW

fraction, in the enhanced BAC columns showed a good correlation with the HMW fraction determined by the LC-OCD ($R^2 = 0.98$).

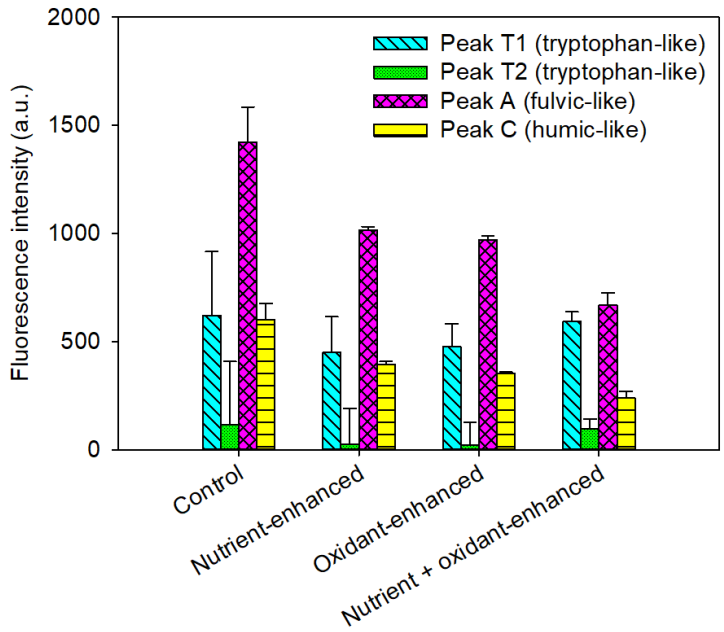


Fig. 4. Fluorescent dissolved organic matter characteristics in extracellular polymeric substances collected from the top of the biological activated carbon columns (0–50 mm) (n = 6).

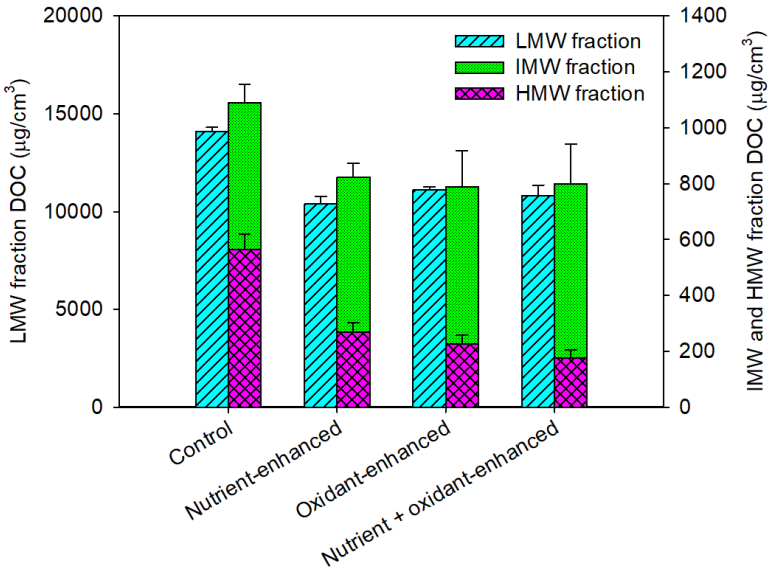


Fig. 5. Extracellular polymeric substances constituents separated into high molecular weight (HMW) fractions, intermediate molecular weight (IMW) fractions, and low molecular weight (LMW) fractions (0–50 mm) (n = 6).

3.3 Changes in dissolved organic matter characteristics in enhanced biological activated carbon filters

Changes in DOC and SUVA were observed in all four BAC columns between the influent and effluent (Fig. S3a). Across all the columns, the DOC removal rates were between 13% and 17%, and the differences were not significant between any of the BAC filters (p<0.05). According to previous studies, the DOC removal rate of the enhanced BAC columns was between 11.6% and 12.1% [10] and showed similar removal rates. Enhanced O_3/biofiltration has been reported to result in lower organic carbon by adding phosphate to the feed water [27], but there was no improvement in the removal of DOC in the filter fed with phosphate in our study. The SUVA of the column effluent increased between 9% and 18% compared with that of the influent (Fig. S3a), and again there were no significant differences between the filters. The difference in DOC removal could be confirmed by the difference in biodegradability of DOC according to the feed water characteristics and oxidizing power of O_3. The increase in DOC removal confirmed that HMW organic matter could be converted into LMW organic matter by O_3 oxidation, which increases biodegradable DOC [10, 27]. However, biofiltration without the O_3 process as a pretreatment did not increase the removal of DOC [12]. The O_3 dose used from the previous study [27] was 2.18 mg O_3/mg DOC, which was relatively higher than the dose of 0.8 mg O_3/mg DOC in this study. In our study, the biodegradable organic matter of the feed water was relatively low; therefore, there was no difference in the DOC removal rate with the addition of phosphate.

An AOC analysis was conducted to assess the biological stability in the BAC filtrates by evaluating the bacterial growth potential (Fig. S3b). It is important to investigate the fate of AOC in BAC, which is effective in the removal of biodegradable organic matter in drinking water treatment processes. The AOC concentration in the influent was 54 μg/L on average, and the removal rate was between 57% and 62% in the BAC columns. The AOC is often related to LMW dissolved organic matter, and this

corresponded with the LC-OCD result of the preferential removal of LMW organic matter [28]. As with DOC and SUVA, no difference in the removal of AOC was observed between the control and enhanced BAC columns, and no significant improvement was observed in the enhanced columns for AOC or DOC.

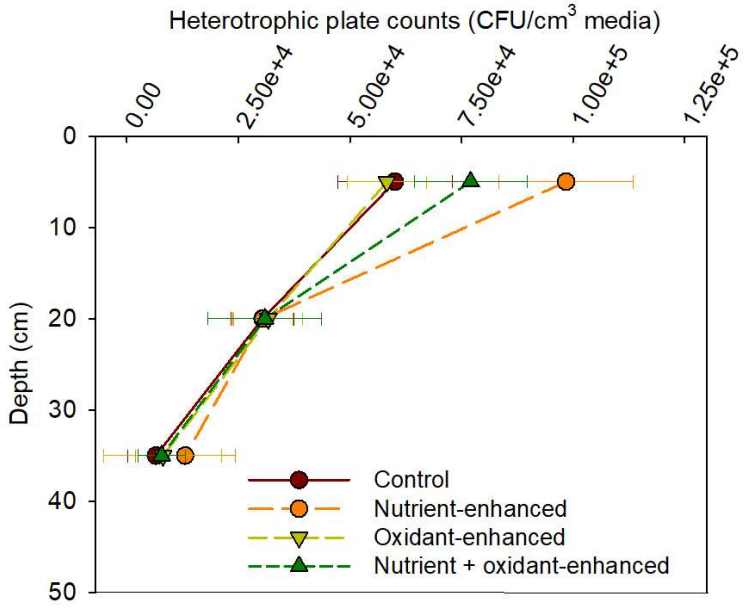
The LC-OCD analysis results of O_3-treated water (influent) and the four column effluents are shown in Fig. S4. LMW neutrals (>71%) and acids (>98%) were effectively removed. The removal of the LMW fraction was greatest in the nutrient-enhanced column (80%), but the difference between the BAC filters was not significant. LMW organic matter, such as LMW acids and neutral compounds, were preferentially removed in relation to humic substances, which have inert organic matter characteristics. The reduction of the LMW fraction was due to biodegradation processes [20, 29], which suggested that LMW biodegradation rates were unaffected by BAC enhancements of any kind in the current study.

The fluorescence EEM results for the rapid sand filtrate, influent (O_3-treated rapid sand filtrate), and BAC effluent were plotted according to T1, T2, A, and C peak regions (Fig. S5). The fluorescence intensities observed in the rapid sand filtrate were considerably reduced (61–90%) in selected peak regions after ozonation. This suggested that organic matter structures changed to leave fewer fluorophores. The florescence EEM also showed an increase in both humic-like and tryptophan-like substance regions after the BAC columns, which agreed with the increased SUVA shown in Fig. S3a. According to Lohwacharin et al. [30], there was an increase in humic-like substance regions after 6 y of BAC treatment. During biofiltration via the BAC columns, there were changes in organic matter characteristics, which indicated that the biodegradation led to more fluorophores, and these changes were similar between the four BAC filters.

3.4 Microbial characterization

In order to analyze the microbial behavior in the BAC columns, HPC and TCC associated with BAC were performed (Figs. 6a and 6b). The HPC and TCC were the greatest at the top of all the columns. Ozonation of the influent likely enhanced the biodegradability in the feed water by oxidation, and thus

increased the AOC concentration [31–33]. Therefore, HPC and TCC associated with BAC likely decreased with distance through the BAC filters because the amount of AOC and the dissolved oxygen concentration would have been depleted toward the lower end of the columns. The biomass at the top of the column was higher in the nutrient-enhanced and nutrient + oxidant-enhanced columns compared with that of the control and oxidant-enhanced columns (Figs. 6a and 6b). At the top of the column, the nutrient-enhanced column had higher HPC associated with BAC than those of the control column (Fig. 6a), but the EPS in the nutrient-enhanced column were lower than those in the control column. It was confirmed that the enhancement in phosphate was effective in reducing EPS formation. The cell-bound ATP associated with BAC along with column depth were analyzed, and are shown in Fig. S6. Similar to HPC and TCC, the columns fed with phosphate showed relatively higher microbial activities with respect to ATP than those of the control. The limiting nutrients in the BAC columns could lower the microbial activity in the control.



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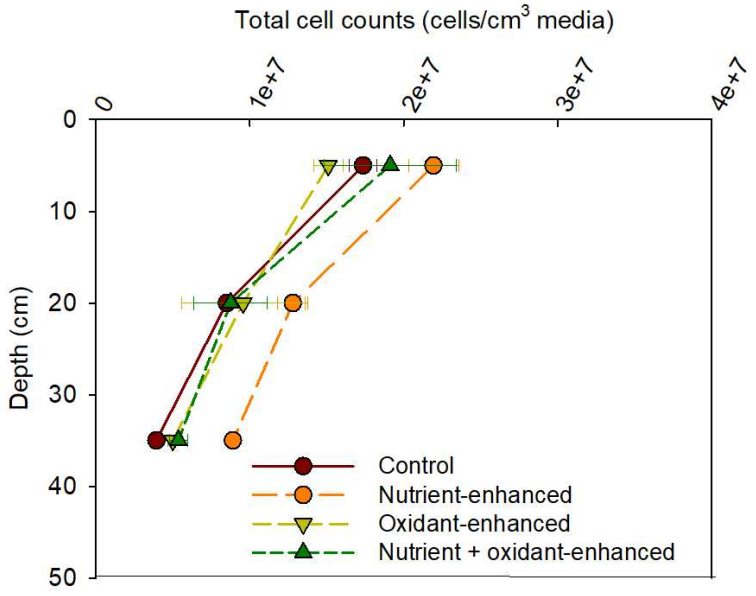
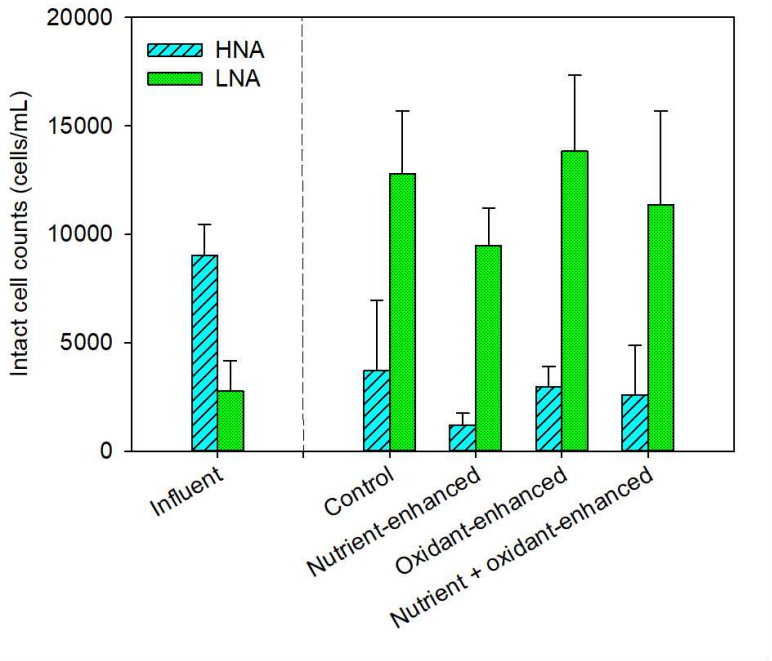


(b)

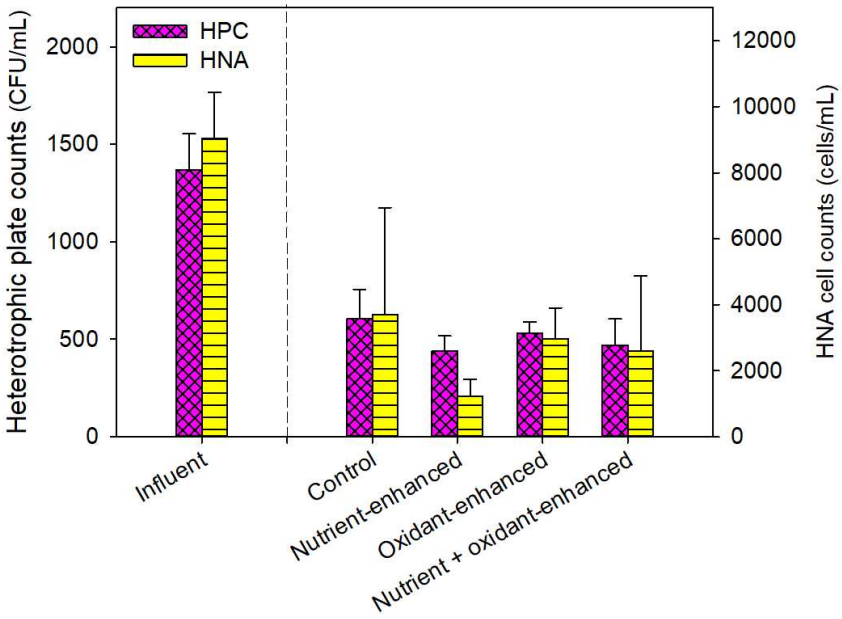
Fig. 6. Distribution of heterotrophic plate count bacteria (a) and total cell counts (b) at different depths (n = 3).

The average intact cell count (ICC) in the influent was 1.2×10^4 cells/mL, and this appeared to be dominated by the high nucleic acid (HNA) group (77%; Fig. 7a.). In contrast, the ICC increased in the column effluent compared with that in the influent. The ratio of HNA group decreased because the number of cells assigned to the low nucleic acid (LNA) group was higher in the BAC effluents. HNA and cell-bound ATP showed some correlation ($R^2 = 0.96$). According to Gasol and Del Giorgio, these results indicated that the HNA group could be an active group of heterotrophic bacteria [34]. In previous studies, the HPC was found to be a better indicator of microbial regrowth than TCC [35]. Herein, the average HPC of the influent water was 1368 CFU/mL, and in the BAC columns, 56%, 68%, 61%, and 66% of the HPCs was removed in the control, nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced columns, respectively. HNA and HPC in the BAC columns showed a good correlation ($R^2 = 0.84$), which suggested that the cells classed as HNA were more likely to be culturable. It takes much less time to measure HNA compared with HPC, which requires incubation time; this is advantageous for evaluating biofilm formation. Owing to the presence of microbial cells (as indicated

by ICC), specifically heterotrophic bacteria (as indicated by HPC) in effluents from the control and enhanced BAC columns, it was necessary to investigate the effects of enhanced BAC filters on the formation of biofilm in the distribution systems as a further parameter of assessing their performance.



(a)



(b)

Fig. 7. (a) High nucleic acid and low nucleic acid bacteria and (b) heterotrophics plate count in the biological activated carbon columns (n = 6).

3.5 Evaluation of biofilm formation potential using Centers for Disease Control and Prevention biofilm reactors

CDC biofilm reactors were used to evaluate the biofilm formation potential of the effluents from BAC columns. Subsequently, the performance data was used to determine which BAC columns would be preferred to limit any downstream bacterial regrowth or seeding in the DWDS. The attached HPC levels of PVC and stainless steel coupons from CDC reactors were determined (Fig. 8). The differences in DOC and AOC in the feed water used for the CDC biofilm reactors were negligible, but the HPC was found to be different, as shown in Fig. 8. The attached HPC levels of the coupons from the CDC reactor fed with effluent from the control column showed $3.3\times10^5 \pm 3.1\times10^3$ cells/cm^2 and $2.4\times10^5 \pm 2.7\times10^3$ cells/cm^2 for PVC and stainless steel coupons, respectively. The HPC level in the nutrient-enhanced BAC columns was 72% lower than that of the control ($9.10\times10^3 \pm 2.80\times10^3$ cells/cm^2) and 83% ($4.00\times10^3 \pm 1.44\times10^3$ cells/cm^2) lower than that of PVC and stainless steel coupons, respectively.

Overall, the highest HPC level detected in a CDC biofilm reactor was related to the control column. The biofilm reactor fed with the control effluent also showed the highest HPC. Conversely, CDC reactors fed with effluents from the nutrient-enhanced column and nutrient + oxidant-enhanced column exhibited a relatively smaller biofilm formation on the coupons. Therefore, the distribution systems could also benefit from the use of nutrient-enhanced BAC, but further work to determine the community composition of the BAC effluent would be beneficial to determine the downstream impact on the DWDS.

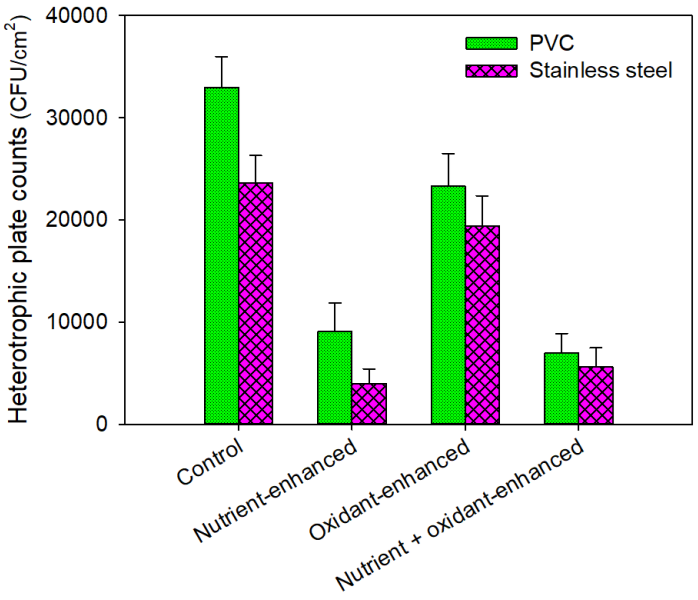


Fig. 8. Effects of nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced biological activated carbon filtration on polyvinyl chloride (PVC) and stainless steel coupons using heterotrophic plate counts (n = 3).

3.6 Trihalomethane formation potential and trace organic contaminants

The THMFP was analyzed to evaluate the potential for formation of disinfection byproducts in the effluent from the BAC columns (Fig. S9). The total THMFP of the influent was 103 µg/L, and the chloroform formation potential ($CHCl_3FP$) accounted for 76% of the THMFP. Therefore, the THMFP

was governed by the extent of the reduction of $CHCl_3$, $CHCl_2Br$, $CHClBr_2$, and $CHBr_3$ produced by the bromination reaction, which accounted for 14%, 1%, and 9% of the total THMFP, respectively. The total THMFP for the control, nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced columns were 70, 68, 74, and 76 µg/L, respectively. Fig. S4 shows that more than 98% of the LMW acid fraction was effectively removed from the feed water in the BAC columns. The acidic organic matter contributed to the formation of THM [19, 36, 37]. Therefore, it was considered that the reduction of the LMW acidic fraction in the column contributed to the removal of THMFP. The $CHCl_3$ concentrations in all column effluents was between 56 µg/L and 61 µg/L, which was reduced by about 22–29% compared with that in the feed water. The significant reduction of $CHBr_3$ was confirmed in all the columns. Therefore, it was also confirmed that the reduced THMFP did not differ from that in the control in the enhanced BAC columns. An earlier study also reported that biofilter enhancement did not improve water quality regarding the THMFP [12].

The removal of selected TrOCs was not significant between the enhanced BAC columns. Table S4 shows the removal performance of selected TrOCs in the BAC columns. In order to remove the effect of ozonation, selected TrOCs were introduced without ozonation into the enhanced BAC columns and more than 78% were attenuated, except for carbamazepine (18–33%). Carbamazepine, which is an antiepileptic drug, is a recalcitrant compound that is highly stable in soil and sewage treatment plant effluents. Moreover, carbamazepine was proposed as an anthropogenic marker to determine the water quality owing to its persistent characteristics in the aquatic environment [38]. Table S5 shows the removal of TrOCs via ozonation followed by BAC. Selected TrOCs further attenuated the ozonation followed by BAC (>99%; O_3+BAC). Therefore, hybrid systems combine one or more treatments in the removal of persistent compounds. However, there was no difference in the removal of selected TrOCs in the enhanced BAC columns compared with that of the control.

4. Conclusions

Significant reduction with respect to head loss, namely 63–79%, was observed in the nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced BAC columns compared with that of the control column. EPS formation, which included proteins and carbohydrates, tended to be reduced in the enhanced BAC columns when compared with that of the control. Moreover, the HMW organic fractions in nutrient-enhanced, oxidant-enhanced, and nutrient + oxidant-enhanced columns were significantly reduced. The enhancement of BAC filters using phosphate was successful in promoting bacterial growth but decreased the EPS production. HNA bacteria were predominant in the influent but decreased remarkably in the effluent. CDC biofilm reactor studies showed that the microbial abundance on coupons connected to the enhanced BAC columns was relatively low compared with that of the control. Enhanced BAC filters via phosphate or hydrogen peroxide had no effect on water quality improvement with respect to the removal of DOC, THMFP, and selected TrOCs. The enhanced BAC-treated water reduced the head loss and formed less biofilm on selected coupons in this study, which indicated that there was less biofilm formation in the distribution systems.

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