

A Study on the Removal of Trace Organic Matter in Water Treatment Procedures using Powder-activated Carbon Biofilm

Rou-Han Lee, Jie-Chung Lou, Huang-Ming Fang

Abstract—This study uses natural water and the surface properties of powdered activated carbon to acclimatize organics, forming biofilms on the surface of powdered activated carbon. To investigate the influence of different hydraulic retention times on the removal efficacy of trace organics in raw water, and to determine the optimal hydraulic retention time of a biological powdered activated carbon system, this study selects ozone-treated water processed by Feng-shan Advanced Water Purification Plant in southern Taiwan for the experiment. The evaluation indicators include assimilable organic carbon, dissolved organic carbon, and total organic carbon. The results of this study can improve the quality of drinking water treated using advanced water purification procedures.

Keywords—Water Purification Procedures, Biological Powdered Activated Carbon System, Assimilable Organic Carbon

I. INTRODUCTION

TO guarantee that drinking water is safe and sanitary, standards for drinking water in Taiwan have become gradually more rigorous. In addition to traditional water treatment procedures (including coagulation, gelation, sedimentation, filtration, and chlorination), advanced water treatment procedures (including ozone, biological activated carbon, and film treatment) can also cope with issues of water source degradation due to pollution. [1]

According to the literature, the deterioration of water quality in water distribution pipes primarily arises from the growth and reproduction of abnormal microbes within the pipes. These microbes significantly and negatively affect the quality of the water distribution system and the water supply. The reproduction of microbes accelerates corrosion of the pipeline [2]; the metabolism and biofilm shedding of the microbe produces discoloration and a foul stench [3]; and the consumption of large volumes of chlorine causes pathogen reproduction. [4] Currently, the most effective method for controlling after growth is believed to be the control of the nutrients that the microbes need for growth. Therefore, a reduction in the concentration of organic carbon in the water can restrain microbial growth. This water is considered drinking water with a high biological stability. [5]

Rou-Han Lee is with the *Ph. D. Candidate, Institute of Environmental Engineering, National Sun Yat-Sen University, Kaohsiung City, Taiwan 804.* (e-mail: d993030003@student.nsysu.edu.tw).

Jie-Chung Lou is with the Institute of Environmental Engineering, National Sun Yat-Sen University, Kaohsiung City 80424, Taiwan. (phone: +886-7-5254410; fax: +886-7-5254411; e-mail: loujc@mail.nsysu.edu.tw).

Huang-Ming Fang is with the *U-TECH Technology Corporation, Vice-Managing Director, Kaohsiung City 802, Taiwan.* (e-mail: hming@email.green99.com.tw).

A survey of water quality in the water originating from the greater Kaohsiung region revealed that traditional water treatment combined with membrane filtration is nearly ineffective in removing assimilable organic carbon (AOC). [6] While biologically activated carbon filters (BACF) effectively remove organic matter, [7] they occupy a large area, and the filters need to be changed regularly. Additionally, BACF often is installed at the end of water treatment. If it is operated inappropriately or the water quality undergoes a significant change, pieces of biofilm and organic matter will enter the clean water, directly causing the growth of micro-organic matter in the water distribution network.

Therefore, this study used natural bodies of water and the surface properties of powder activated carbon to cultivate microbes. After the surface of the powder activated carbon was formed into a biological film, ozonated water from the Fengshan Advanced Water Purification Plant in southern Taiwan was selected as the experimental water source. This study examined the removal of trace organic matter in water at different hydraulic retention times (items evaluated: AOC and DOC), and found the impact of the water retention time on the biological powder activated carbon system.

II. MATERIALS AND METHODS

A. Powder Activated Carbon Biofilm Cultivation

Microbes were cultivated using 40 g of biological powder activated carbon as the carrier. Domestic water treated in a rapid sand filter was poured into a 4 L reactor at the Fengshan Advanced Water Purification Plant in southern Taiwan. The microbes were attached to the powder activated carbon's porous surface, forming biofilm.

The literature indicates that microbes in BACF require two to three months for cultivation. [8] At this stage, organic matter will primarily decompose biologically, preventing experimental errors in biological treatment efficiency caused by powder activated carbon adsorption. To allow the biofilm to grow completely on the powder activated carbon, this experiment used dissolved organic carbon (DOC) and assimilable organic carbon (AOC) to indicate the amount of powder activated carbon cultivated.

B. Biological Assimilable Organic Carbon

AOC is analyzed using the improved method developed by Van der Kooij. [8] *Pseudomonas fluorescens strain* (P17) and *Spirillum species strain* (NOX) were each implanted in a bacteria liquid bottle and preserved at 2 to 6 °C.

After chlorine extraction, the test water sample was pasteurized (60 °C water bath for 30 min). Once the water sample was cooled, it was embedded into the two types of pure bacteria - P17 and NOX - and immediately smeared onto a petri dish. Besides being immediately smeared onto NB (LLA) agar to analyze the amount of bacteria, the water sample was cultivated below 15 °C, and the amount of bacteria was sampled and analyzed every 1 to 2 days. The amount of bacteria in the petri dish after culturing at 25 °C for 3 days depicts the growth curve until the stationary phase. The average value for the maximum number of P17 and NOX bacteria grown in the water sample (N_{max}) multiplied by each bacterial strain's yield value provides the values for AOC-P17 and AOC-NOX, which added together equals AOC-Total (μg acetate eq-C/L). The calculation method is as follows:

$$AOC (\mu\text{g acetate} - C / L) = \frac{N_{max} (CFU / mL) \times 1000 (mL)}{Yield (CFU / \mu\text{g acetate} - C)} \quad (1)$$

$$AOC_{total} = AOC_{P17} + AOC_{NOX} \quad (2)$$

C. Total Organic Carbon and Dissolved Organic Carbon

This study used a 1030 W Wet Oxidation TOC Analyzer (OIA) to analyze the total organic carbon and standard Potassium Hydrogen Phthalate products ($\text{KHC}_8\text{H}_4\text{O}_4$, KHP, Nacalai Tesque, Japan) for calibration. The water sample was led into a digestion reactor that could be heated to 95 to 100 °C, and mixed with per sulfate solution and acid solution. The organic and inorganic carbons in the water sample were oxidized, converting them into carbon dioxide. A non-dispersive infrared analyzer with high purity carried nitrogen that is able to absorb the specific wave length of carbon dioxide could then immediately test for the concentration of total organic carbon in the water. When analyzing a solution that had passed through a 0.45 μm filter, the above procedure can be used to test for the concentration of DOC.

D. Microscope Equipment Analysis

Through secondary electron imaging, a scanning electron microscope (SEM) can be used to observe the surface appearance, diameter, and length of the nano-structure. Specific X-rays can be detected with the energy dispersive spectrometer (EDS) to conduct component analysis for elements.

III. RESULTS AND DISCUSSION

A. Powder Activated Carbon Biofilm Cultivation Process

This study used change in DOC concentration to determine microbial quantity, and used SEM and EDS microscopic equipment to analyze microbial properties, primarily to determine whether microbes had attached to the pores of the powder activated carbon.

During the initial phase of this study, 40 g of powder activated carbon was embedded in the 4 L incubator, and the activated carbon concentration was 10 g/L. After 50 days of cultivation, the average DOC concentration was 704.3 ± 116.1 mg/L. During the middle phase of this study, an additional 40 g

of powder activated carbon was placed in the incubator, and the activated carbon concentration was 20 g/L. After an additional 50 days of cultivation, the average DOC concentration was 593.9 ± 90.0 mg/L. During the final phase of this study, an additional 120 g of powder activated carbon was placed in the incubator, and the activated carbon concentration in the incubator was 50 g/L. After an additional 50 days of cultivation, the average DOC concentration was 316.0 ± 77.9 mg/L. The change in microbial DOC cultivated on powder activated carbon is displayed in Fig. 1. The results of the experiment show that an increase in the concentration of powder activated carbon corresponds to an increase in carriers on which microbes can grow, thereby improving DOC removal.

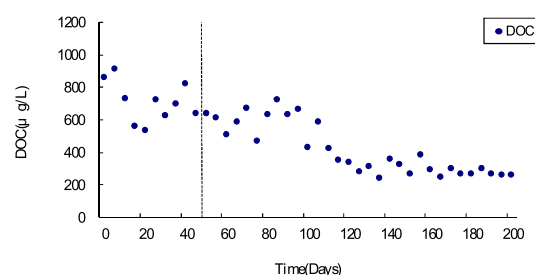


Fig. 1 Change in concentration over time of DOC for powder activated carbon-cultivated microbes

The 5000x magnified SEM pictures showing powder activated carbon before and after cultivation are displayed in Fig. 2. The Fig. 2 (a) shows that prior to cultivation, no biofilms were attached to the porous surface of the powder activated carbon. Fig. 2 (b) shows that after 50 days of cultivation, the pores of the powder activated carbon were surrounded by biofilm. Table 1 and Fig. 3 both show the EDS spectral analysis for powder activated carbon before and after cultivation. The analysis reveals that after cultivation, powder activated carbon was comprised of 0.47 % calcium. This is perhaps due to the prevalence of limestone in southern Taiwan. Therefore, partial calcium elements will settle in the pores when ozonated water from the Fengshan Advanced Water Purification Plant is used to cultivate powder activated carbon, and 8.52 % more oxygen exists in the powder activated carbon after cultivation than before.

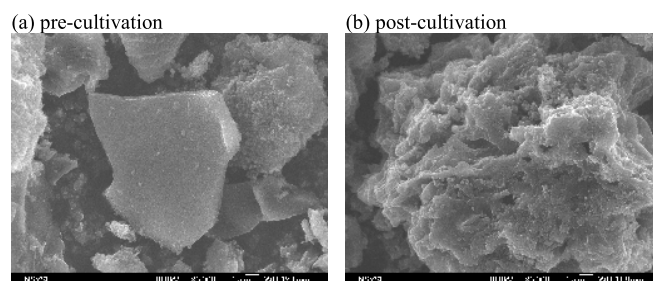


Fig. 2 5000x magnified SEM structure of powder activated carbon -cultivated microbes; (a) pre-cultivation; (b) post-cultivation

B. Basic Properties of Water Quality

This study used microbes to degrade trace organic matter, researching the Fengshan Advanced Water Purification Plant in southern Taiwan. To avoid disrupting the change in water quality in the prior batch of water, each time the water was changed; the water quality was only analyzed after 1 to 2 days of water quality stabilization. Table 2 provides the water quality data for this experiment: average DOC was 0.70 ± 0.14 mg/L and average AOC was 50.16 ± 14.41 μ g acetate-C/L; where AOC-P17 and AOC-NOX comprised 46 % and 54 % of total AOC.

TABLE I
 EDS ELEMENTS FOR POWDER ACTIVATED CARBON BEFORE AND AFTER CULTIVATION

Element	PAC Before Cultivation		PAC Before Cultivation	
	Weight%	Atomic%	Weight%	Atomic%
C	93.88	95.33	84.89	88.41
O	6.12	4.67	14.64	11.45
Ca	-	-	0.47	0.15
Totals	100.00	100.00	100.00	100.00

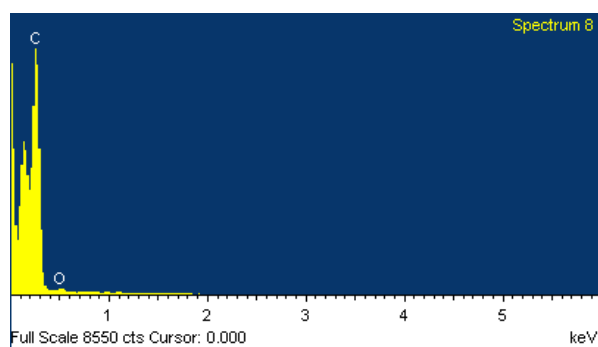
TABLE I
 EDS ELEMENTS FOR POWDER ACTIVATED CARBON BEFORE AND AFTER CULTIVATION

Element	PAC Before Cultivation		PAC Before Cultivation	
	Weight%	Atomic%	Weight%	Atomic%
C	93.88	95.33	84.89	88.41
O	6.12	4.67	14.64	11.45
Ca	-	-	0.47	0.15
Totals	100.00	100.00	100.00	100.00

TABLE II
 OUTFLOW AND INFLOW WATER PROPERTIES FOR POWDER ACTIVATED CARBON SYSTEM

Parameters	Unit	Influent	Effluent	Effluent (I)	Effluent (II)	Effluent (III)
HRT	hr	-	-	1	3	6
Temperature	°C	23.0±3.10	25.2±1.02	25.1±1.06	25.4±0.55	25.2±1.33
pH	-	8.3±0.11	8.3±0.08	8.3±0.07	8.3±0.07	8.4±0.08
Conductivity	μ mho/cm	506±16.7	467±111.0	507±20.6	502±15.5	523±11.4
TDS	mg/L	325±26.6	329±13.4	309±3.0	326±9.8	340±7.4
DOC	mg/L	0.70±0.14	0.49±0.96	0.59±0.76	0.47±0.64	0.42±0.46
AOC	μ g acetate-C/L	50.3±14.4	16.1±5.4	21.2±4.3	15.5±3.3	11.2±3.0
NH ₃ -N	mg/L	0.03±0.02	0.02±0.01	0.02±0.01	0.02±0.01	0.02±0.01
NO ₃ -N	mg/L	1.09±0.39	1.62±0.69	2.11±1.08	1.33±0.33	1.51±0.20
Free Residual Chlorine	mg/L	3.11±0.31	3.66±1.24	3.97±0.52	3.40±1.10	3.63±1.73
DOC load	mg DOC/hr	HRT 1hr: 0.65±0.09, HRT 3hrs: 0.21±0.02, HRT 6hrs: 0.13±0.04				

(a) pre-cultivation



(b) post-cultivation

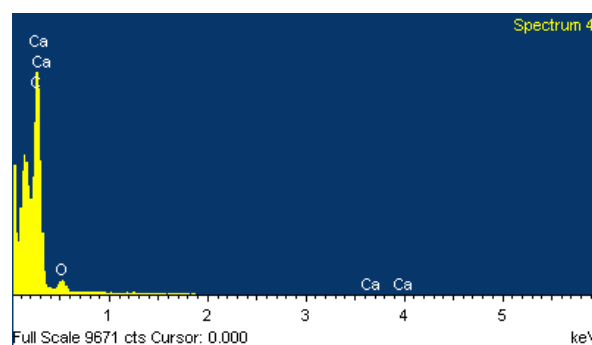


Fig. 3 EDS spectral element analysis of powder activated carbon-cultivated microbes; (a) pre-cultivation; (b) post-cultivation

C. AOC Removal Rate of Biological Powder Activated Carbon Treatment System

Using different hydraulic retention times and a constant activated carbon concentration of 50 g/L, this study found that the AOC removal rate for a hydraulic retention time of 1 hr was 57 %. Additionally, total AOC was primarily composed of AOC-P17, at over 73 %. NOX primarily used a carboxylic acid substrate, especially ozone oxidized products such as formate, oxalate, glyoxylate, and glycolate substrates. [5] When the hydraulic retention time is extended to 6 hrs, the AOC removal rate can increase to 77 %. This increase demonstrates that the biological powder activated carbon treatment system can effectively remove ozone oxidized products. According to van der Kooij's research, [9], [10] total heterotrophic colony growth can be controlled even if no chlorine is added to the water. The correlation between hydraulic retention time and AOC is displayed in Fig. 4.

D. DOC Removal Rate of Biological Powder Activated Carbon Treatment System

Fig. 5 indicates that the change to DOC outflow and inflow concentration was insignificant. When hydraulic retention time was 1 hr, the DOC removal rate was approximately 24 %. When the hydraulic retention time was extended to 6 hrs, the DOC removal rate was approximately 33 %. These results show that powder activated carbon can effectively remove DOC.

IV. CONCLUSION

The observations of the surface properties and other elements of powder activated carbon before and after cultivation using SEM and EDS showed that a layer of biofilm appeared on the porous surface of powder activated carbon after cultivation, which is corroborated by the increase in oxygen discovered using EDS. Therefore, using water quality from an actual facility can feasibly cultivate microbes. Using rapid-filtered water from a water treatment plant with powder activated carbon to directly cultivate microbes that can be used to remove trace organic matter in the water (such as AOC and DOC) showed that biologically activated carbon adsorption stabilizes after a long period of cultivation. Once stabilized, biodegradation is emphasized in the removal of organic matter. Under a constant concentration of powder activated carbon, AOC and DOC removal efficiency will increase with hydraulic retention time.

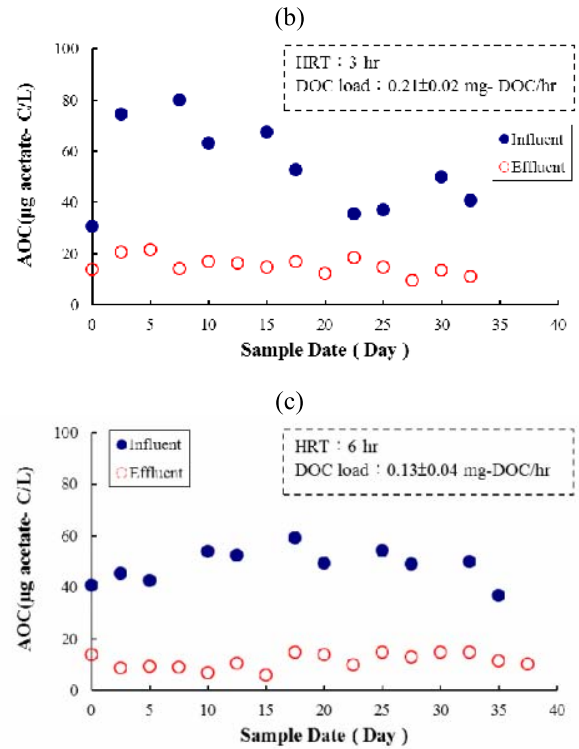
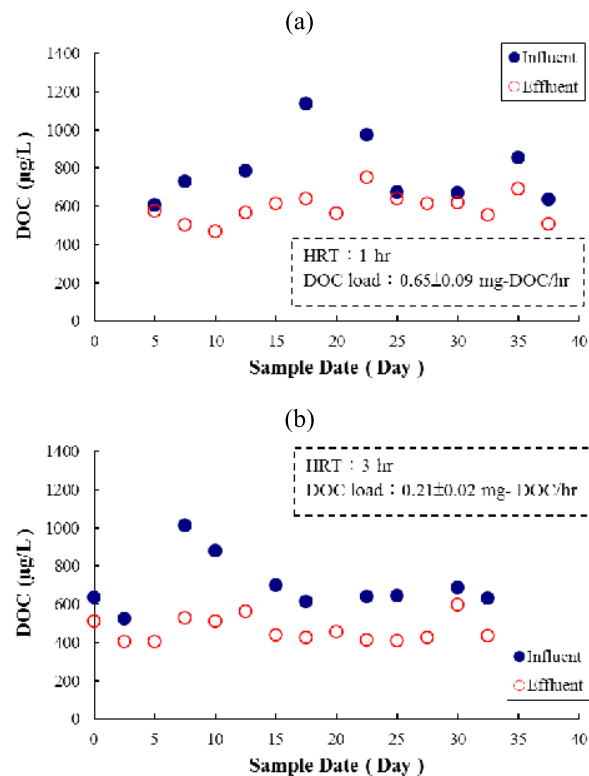
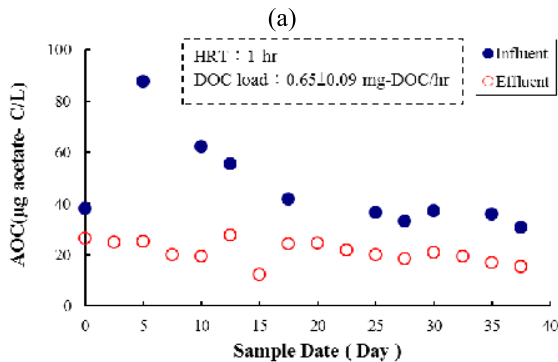


Fig. 4 Change in AOC over time for different hydraulic retention times in the biological powder activated carbon system



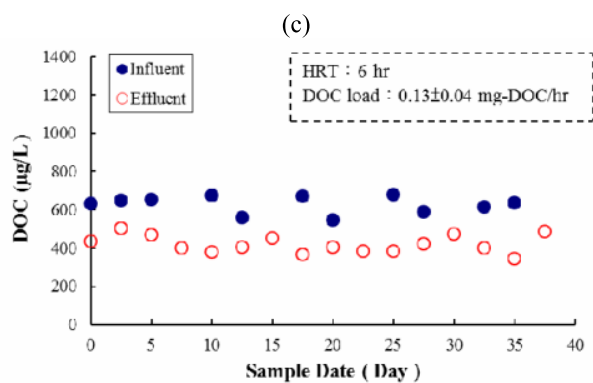


Fig. 5 Change in DOC over time for different hydraulic retention times in the biological powder activated carbon system

REFERENCES

- [1] Krasner S. W., J. P. Croue, J. Buffle, and E. M. Perdue, "Three approaches for characterizing NOM", *AWWA.*, vol. 88, no. 6, pp. 66-79, 1996.
- [2] Lee, S.H., O'Connor, J.T. and Banerji, S.K., "Biologically mediated corrosion and its effects on water quality in distribution systems", *Jour. AWWA.*, vol. 72, no. 11, pp. 636-645, 1980.
- [3] Levy, R.V., Hart, F.L. and Cheetham, R.D., "Occurrence and public health significance of invertebrates in drinking water systems", *Jour. AWWA.*, vol. 78, no. 9, pp. 105-110, 1986.
- [4] Geldreich, E.E., "Biofilms in water distribution systems." *Microbial Quality of Water Supply in Distribution System*, Lewis publisher, Boca Raton, Florida, 1996.
- [5] Van Der Kooij, D. and Veenendaal, H.R., "Determination of the concentration of easily assimilable organic carbon (AOC) in drinking water with growth measurements using pure bacterial cultures." *SWE 95.002, KIWA, Nieuwegein, Netherlands*, 1995.
- [6] Zhang, M. M., Li, C., Benjamin, M. M. and Chang, Y. J., "Fouling and natural organic matter removal in adsorbent/membrane systems for drinking water treatment", *Environmental Science and Technology*, vol. 37, no. 8, pp. 1663-1669, 2003.
- [7] Su, D. J. and Gao, N. Y., "A study of removing organic matters in slightly-polluted water by an ozone-active carbon combined process", *Ind. Water Waste-water*, vol. 32, pp. 26-28, 2005.
- [8] Servais, P., Laurent, P. and Randon, G., "Comparison of the bacterial dynamics in various french distribution system", *J. Water SRT-Aqua.*, vol. 44, no. 1, pp. 10-17, 1995.
- [9] Van Der Kooij, D., "Assimilable organic carbon (AOC) in drinking water", in *Drinking Water Microbiology*, G. A. Mcfeter, ed., Springer-Verlag, New York, 1990.
- [10] Van der Kooij, D., Hijnen, W.A.M. and Kruithof, J.C., "The effect of ozonation, biological filtration and distribution on the concentration of easily assimilable organic carbon (AOC) in drinking water", *Ozone: Science and Engineering*, vol. 11, pp. 297-311, 1989.