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Evaluation of chemical washing for granular activated carbon during drinking water treatment

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ABSTRACT

Contaminants are retained on the internal pore surfaces of granular activated carbon (GAC) and periodic regeneration and/or replacement of GAC is mandatory for robust water treatment. Alternatively, the chemical washing can be acceptable for adsorptivity recovery of short-term fouled GAC. In this study, Fenton's reagent was employed to restore adsorptivity of saturated GAC, focusing on the removal of organic components from surface water after cleaning in comparison with fresh GAC. The chemical washing with Fenton's reagent was carried out for 10 min, which was identical to that applied for the hydraulic backwash of GAC. Four different GAC beds were prepared, that is, fresh, saturated, saturated/hydraulically washed, and saturated/chemically cleaned GACs, and their performance was compared during the treatment of surface water. An innovative suite of analytical tools was applied to better elucidate the changes in compositional and functional properties of organic matter during GAC filtration. In addition to the characterization of organic matter, this study also investigated the fate of biomass-derived organic matter during GAC filtration. The results indicate that the use of in situ chemical washing is a highly feasible technology to effectively remove extracellular polymeric substances that are retained on activated carbon materials and to accelerate GAC saturation.

Keywords: Granular activated carbon; Adsorptivity recovery; Hydraulic backwash; Chemical washing; Fenton's reagent

1. Introduction

Granular activated carbon (GAC) plays an important role in the adsorption of organic compounds, and taste and odor compounds for drinking water treatment. When GAC is exhausted due to particle saturation, bulk organic matter, and biomass in the bed, the regeneration and replacement of GAC are essential methods for the recovery of GAC absorptivity. On the other hand, hydraulic backwashing is a commonly used technique to remove saturated particles of the fixed bed filters and to prevent saturation of the media. However, the effect of hydraulic backwashing was usually limited to reduction of head loss by the removal of inorganic particles. Organic adsorbate of the extracellular polymeric substance (EPS) will not be easily removed by hydraulic backwashing [1]. Expensive thermal regeneration of carbon and media replacement in the bed is recommended methods for the GAC enhancement. The most commonly used thermal regeneration is not only a major part of operating cost but also requires considering extra treatment of exhaust gas and a 5%–15% loss of carbon during the regeneration process. Saturated GAC is heated to high temperature

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(650°C–900°C), baked, dried, and gasified in multiple hearth furnaces [2]. Much research has focused on the development and optimization of GAC regeneration methods in various ways such as physical, chemical, biological, and mechanical treatments: ultrasound technique, electrochemical process, hydrothermal, ozone, and Fenton, to name a few. Among them, Fenton's reagent has been verified to be effective as an in situ chemical oxidation (ISCO) and ISCO GAC regeneration technology [3]. ISCO is an environmental remediation technology for groundwater or soil contaminants using an advanced oxidation process, which introduces a chemical oxidant into the subsurface of groundwater or soil contaminants to reduce the concentrations of contaminants [4].

In this study, Fenton chemical washing was considered for GAC enhancement through the addition of chemicals into the readily available backwash pipelines. The chemical backwashing is conducted very short periods of time (10–15 min) during the GAC operation. The chemical injection line speed applies to the removal of physical and chemical using the 30% of expansion rate. This chemical process can be implemented to both absorption bed and filtration bed of advanced water treatment process excluding biological activated carbon (BAC) process, as the chemical process may impact the BAC process. In contrast, it would be necessary to repeat the regeneration method for every 4 months to 4 years depending on the bed condition.

This paper proposes the use of Fenton as a chemical reagent to backwash the GAC bed to enhance operation and maintenance. The use of Fenton reagent provides an opportunity to overcome the limitations of traditional hydraulic backwashing and provide the effectiveness associated with saturated GAC. The application of a column test using actual field-spent GAC is provided to demonstrate the benefits of Fenton chemical backwashing in comparison with conventional hydraulic backwashing.

2. Experimental methods

2.1. GAC column experimental setup

GAC columns were deployed to assess the adsorptivity recovery of fouled GAC after cleaning, focusing on the removal of organic components from surface water in comparison with fresh GAC. Four glass columns with an inner diameter of 1.44 cm and a length of 40 cm were used. Among the columns, one was packed with the fresh GAC and the others were packed with the saturated GAC. Saturated GAC was obtained from a local waterworks plant and transported to the laboratory and stored in a refrigerator at 4°C before use. Meanwhile, fresh GAC (8 × 30 mesh) was thoroughly washed with deionized water (Milli-Q Advantage A10, Millipore, MA, USA), dried in an oven at 105°C for a day, and stored in a desiccator.

Two different cleaning methods were used for the saturated GAC: (1) hydraulic backwash and (2) chemical washing with Fenton's reagents (10,000 ppm). Preliminary tests were conducted to optimize the concentration of Fenton's reagents for chemical washing and to verify the consistent recovery of the GAC adsorptivity during multi-cycle chemical cleanings (Figs. S1 and S2 in supporting information). The conductivity of Fenton's regents was 600 ± 10 µs/cm and the solution pH was adjusted to 3. An electrolyte was prepared with deionized water and KCl for hydraulic backwash. Both hydraulic backwash and chemical washing were carried out at 30% GAC bed expansion for 10 min.

Surface water collected from the Tancheon River (Seoul, Korea) was used as influent for four different GAC columns (i.e., fresh, saturated, saturated/hydraulically washed, and saturated/chemically cleaned GAC). Prior to the column experiment, the adsorption of iodine and methylene blue was measured using the prepared GAC. Organic matter and EPS retained on the activated carbon materials were also extracted by sonicating the prepared GAC. All the columns were put into operation at an empty bed contact time (EBCT) of 10 min at ambient temperature (16°C–22°C). The average concentration of dissolved organic carbon (DOC) in the influents prepared for the columns was 3.7 mg/L. The fate of organic matter and bacteria through the GAC columns was investigated.

2.2. Characterization of dissolved organic matter

Molecular weight (MW) distribution of organic matter was determined by liquid chromatography with online organic carbon detection (LC-OCD) (DOC-Labor, Germany). For this purpose, size-exclusion chromatography was coupled with online equipment for the real-time measurement of both UV254 and organic carbon concentration of the effluent passing through the exclusion column. Detailed information regarding the LC-OCD system was provided in an earlier report [5]. Fluorescence spectra were collected using a Perkin-Elmer LS-50B luminescence spectrometer (PerkinElmer, Waltham, MA), which uses a 450 W xenon lamp source. Spectroscopic analyses were carried out as previously described [6]. Three-dimensional spectra were obtained by repeatedly measuring the emission (Em) spectra within a range of 280–600 nm, with excitation (Ex) wavelengths from 200 to 400 nm, spaced at 10 nm intervals in the excitation domain. Spectra were then concatenated into an excitation–emission matrix (EEM). The EEM components determined in this study were T1 (Ex/Em 220–240/330–360 nm), T2 (Ex/Em 270–280/330–360 nm), A (Ex/Em 230–260/400–450 nm), and C (Ex/Em 300–340/400–450 nm) [7].

2.3. Cell counting and ATP measurement

We described in our previous report the microbial assay procedures in detail [6]. All water samples were immediately stained with a mixture of SYBR® Green I and propidium iodide in order to differentiate cells with intact cell membranes from the disrupted microbial cells. Subsequent cytometric analysis was carried out using a Partec CyFlow® Cube6 flow cytometer (Partec GmbH, Görlitz, Germany) equipped with a 20 mW blue diode pumped solid-state laser emitting at 488 nm. The flow cytometer (FCM) operated to monitor the cell density using volumetric counting hardware measuring the number of fluorescent particles in 200 µL of the sample. All collected fluorescence data were processed using the FCS Express 4 Cytometry software (De Novo Software, Glendale, CA); the electronic gating application of the software was used to isolate positive signals from the cells and to exclude noise from the non-microbial particles or from the instrument itself. The total and soluble cell-bound adenosine triphosphate (ATP) contents were determined as described in our previous work by Kim et al. [6] using the BacTiter-GloTM reagent (Promega Corporation, Fitchburg, WI) and a luminometer (GloMax® 20/20; Promega, Madison, WI). The soluble ATP was generally referred to as free-ATP (i.e., extracellular ATP) for the sample filtered using 0.2 µm membranes, according to the operational definition. The cell-bound ATP was calculated by subtracting the free ATP from the total ATP using the whole sample.

2.4. Analytical methods

Non-purgeable organic carbon is generally referred to as total organic carbon (TOC) for the whole sample (or DOC for a sample filtered with a 0.22 µm membrane) according to the operational definition. Therefore, TOC and DOC are identical unless microfiltration is required for the sample prior to analysis. Organic carbon in the liquid sample was also measured (TOC-V CPN, Shimadzu, Japan). A DR/5000 spectrophotometer (Hach, USA) was used to determine UV254 absorptivity after passage through 0.22 µm membrane filters. The specific UV absorbance (SUVA) value was calculated from the UV254 transmittance divided by the amount of dissolved organic carbon in the water sample. The pH was measured using a pH meter (Orion 3 STAR, Thermo Scientific, USA). Each measurement was carried out in triplicate and average values were reported.

3. Results and discussions

3.1. Chemical washing with Fenton's reagent

The adsorption capacity of iodine and methylene blue toward fresh and saturated GAC was determined and compared with those observed after chemical washing and hydraulic backwash of the saturated GAC (Fig. 1). Fenton-cleaned GAC showed the iodine adsorptivity of 498 ± 32 mg/g, which had a very high efficiency compared with saturated GAC (342 ± 21 mg/g) and was less than fresh GAC (567 ± 21 mg/g). It showed that promising results in terms of time-saving and simplicity. For the methylene blue dye reduction test, fresh GAC has the adsorptivity of 40.3 ± 2.5 mL/g. Fenton-cleaned

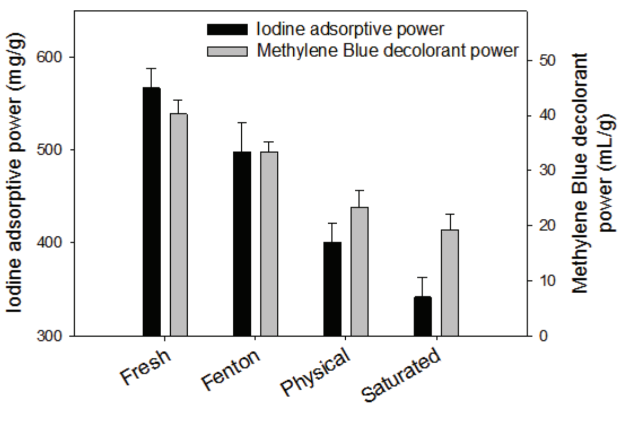

Fig. 1. Effects of chemical cleaning and hydraulic backwash on the adsorption capacity of iodine and methylene blue toward different GAC prior to the filtration runs. Unground GAC was used for the adsorption tests.

GAC showed very high efficiency of 82.8% compared with the methylene blue adsorptivity observed for fresh GAC. Contrarily, hydraulic backwashing increased the methylene blue adsorptivity from 19.3 ± 2.8 to 23.4 ± 3.1 mL/g. Fenton chemical washing shows high efficiency for NOM saturated GAC in lab scale tests (section 3.1).

Preliminary tests were conducted to optimize the concentration of Fenton's reagents for chemical washing and to verify the consistency of the adsorptivity recovery during multi-cycle chemical washing. Different levels of dosage concentration were applied to identify the effectiveness of Fenton chemical washing as compared with hydraulic backwash (Fig. S1 in supporting information). In general, the higher the concentration of Fenton's reagent used, the greater adsorption rate was observed; for example, when 1,000 mg/L of Fenton is applied, the phenol adsorption rate was 44% improved compared with fresh GAC showing 13% enhancement over hydraulic washing. As 10,000 mg/L of Fenton was applied, the phenol adsorption rate showed 78% recovery compared with fresh GAC, indicating 47% enhancement to physical washing. For humic acid adsorption, likewise, when the dosage concentration increases, the recovery of adsorption rate is increased. High concentration (10,000 mg/L) of chemical washing helps the GAC recovering the adsorptivity better than hydraulic washing. Both phenol and humic acid showed insufficient oxidation reaction of Fenton's reagent in low concentrations [8]. The efficiency increased when it goes to high concentrations. High-concentration chemical washing is necessary to get the expected efficiency in chemical washing using Fenton's reagent. The previous research on GAC in situ regeneration using Fenton's reagent showed the efficiency of Fenton's reagent chemical washing (equivalent to 10,000 mg/L of H_2O_2 concentration) of 92% for phenol break through GAC [9]. In this study, the recovery of chemical washing for phenol break through GAC showed 78%, indicating less absorptivity than the regeneration. However, the regeneration consumes more chemicals and takes a longer time than chemical washing.

The adsorptivity recovery during multi-cycle hydraulic and chemical washing of saturated GAC was determined in comparison with that observed for fresh GAC (Fig. S2 in supporting information). For the phenol break through GAC, the adsorption recovery rate of hydraulic washing ranged between 26% and 29% when measured at initial 30 min after washing. Whereas, the adsorption recovery rate of Fenton chemical washing showed 77% ± 1% at initial 30 min after washing. For humic acid, the adsorption recovery rate of hydraulic washing ranged between 50% and 55% at initial 30 min after the washing conducted, while Fenton chemical washing resulted in adsorption recovery rate of 78% ± 3% at initial 30 min after washing. This indicates the reproducibility of hydraulic backwash and Fenton chemical washing for both phenol and humic acid break through GAC. The physical treatment did not encounter any loss or damage, breakage of the micropore, or breakage of micropore for Fenton chemical washing. The chemical washing technique has a minor loss rate of GAC and little breakage of the micropore as the process was conducted inside GAC bed as in in situ chemical regeneration method [3]. A short injection time with a high-concentration oxidant removes the adsorbate on GAC with less harm to the physical and chemical effect to

GAC. The thermal regeneration technique may destroy the structure of micropore and occur a loss of surface area with an increase of pore size. Moreover, the repetition of thermal regeneration accelerates the destruction process of the structure, resulting in a decrease of the absorptivity of GAC. Chemical washing can prevent destruction of micropores and reduce loss rate with excellent reproducibility.

3.2. Fate of dissolved organic matter during the filtration through GAC columns

Fig. 2 shows the effects of chemical washing and hydraulic backwash on the removal of organic matter in surface water passing through different GAC columns. The DOC of surface water was 3.7 mg/L and decreased to 1.1 mg/L through Fenton washed GAC column. The efficiency of absorptivity for Fenton chemical washing shows 87% recovery compared with fresh GAC. SUVA lowered after surface water passed through all the GAC columns, indicating that GAC selectively adsorbed non-biodegradable organic compounds (e.g., humic-like substances) from surface water. Fenton chemical washing resulted in the adsorptivity recovery of saturated GAC toward UV-causing materials; thus the effluent SUVA for chemically washed (0.830 L/mg m) GAC was lower than that observed for saturated (0.946 L/mg m) and hydraulically washed (0.935 L/mg m) GAC. Hydraulic washing physically removed organic matter retained onto the saturated GAC, but a greater extent of organic removal was accomplished with Fenton chemical washing (Fig. S3 in supporting information). For the case of hydraulic and chemical washing, SUVA of organic matter retained on the GAC bed was higher than that observed for saturated GAC. In particular, Fenton chemical washing removes more aliphatic organics than aromatic ones. The diffusion velocity of aliphatic organic is faster than aromatic organic material as they have less MW [10].

Fig. 3 shows the effects of chemical washing and hydraulic backwash on the fate of five different organic components (i.e., biopolymer, humic substances, building blocks, low molecular weight (LMW) neutrals, and LMW humics) in surface water passing through different GAC columns. When compared with influent surface water, hydraulically washed GAC and saturated GAC shows 30% removal of LMW neutrals, 30% reduction of building blocks, and 5%–6% reduction of humic materials. Whereas, Fenton-cleaned GAC shows 79% removal of humic-like substances, 54% removal of building blocks, and 61% removal of LMW neutrals, indicating that Fenton chemical washing improved organic adsorptivity drastically. Saturated GAC could not remove adsorbate in GAC even after the application of hydraulic backwash, while Fenton could remove organic adsorbate matter in GAC, which resulted in an increased absorptivity of GAC that reacts to both LMW and polymeric materials such as humic-like substances [10]. Fenton chemical washing substantially removed biopolymer and LMW neutral from the saturated GAC bed, while the removal of humic substances was relatively less and an insignificant change was found for building blocks (Fig. S4 in supporting information). As shown in SUVA, Fenton chemical washing showed a better removal of LMW aliphatic organic matter compared with aromatic

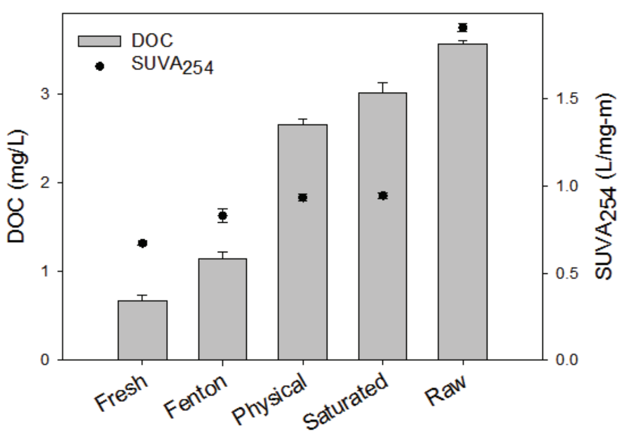

Fig. 2. Effects of chemical cleaning and hydraulic backwash on the removal of organic matter in surface water passing through different GAC columns.

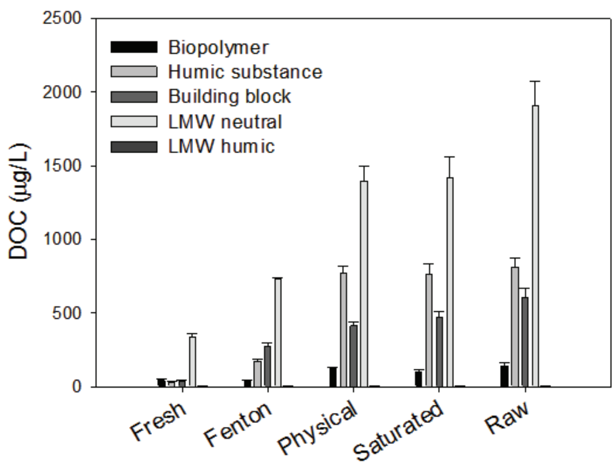

Fig. 3. Effects of chemical cleaning and hydraulic backwash on the fate of organic components in surface water passing through different GAC columns.

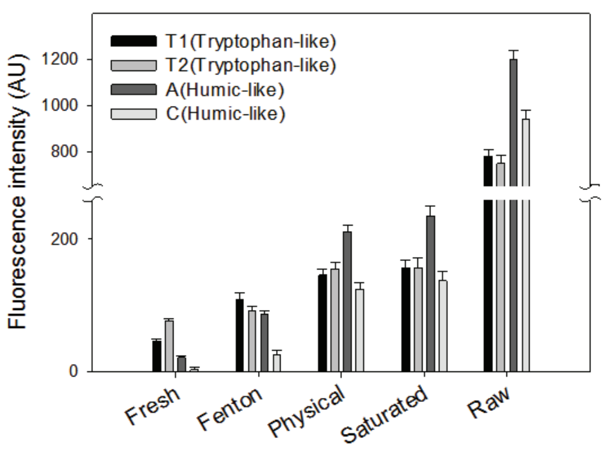

Fig. 4. Effects of chemical cleaning and hydraulic backwash on the fate of fluorescent organic components in surface water passing through different GAC columns.

organic matter among the organic fractions retained onto the saturated GAC. Hydraulic backwashing shows a limitation in the removal of dissolved organic matter adsorbed onto GAC. Fig. 4 shows the effects of chemical washing and hydraulic backwash on the fate of fluorescent organic components in surface water passing through different GAC columns. The fluorescent components can be classified into two major groups, that is, protein-like substances (T1 and T2 regions) and humic-like substances (A and C regions). Fluorescent intensity for surface water has T1 (781 ± 30), T2 (750 ± 36), A (1,199 ± 39), and C (942 ± 38) arbitrary unit. Hydraulically washed GAC and saturated GAC show similar results, indicating that the adsorptivity recovery of saturated GAC was negligible after hydraulic backwash. Fresh GAC and Fenton-cleaned GAC show a less fluorescent intensity for all regions, suggesting a significant restoration in the adsorptivity of saturated GAC toward the fluorescent dissolved organic matter. Fenton-cleaned GAC shows 63% peak reduction in A and 80% reduction in C, which implies that Fenton chemical washing increased the retention of humic materials onto the GAC beds. This result coincided with the LC-OCD result (Fig. 3) showing an increase of the adsorptivity for humic substances when Fenton chemical washing applied. Likewise, Fenton chemical washing was more effective at taking off the fluorescent dissolved organic matter from the saturated GAC bed compared with when using hydraulic backwash (Fig. S5 in supporting information).

3.3. Effects of chemical washing on the bacterial characteristics

Table 1 shows the effects of chemical washing and hydraulic backwash on the variations in bacterial cell concentration of surface water passing through different GAC columns. The intact cell count of surface water was 2.6E+06 and an insignificant change was found after the filtration of surface water through different GAC columns (i.e., fresh, chemically cleaned, hydraulically washed, and saturated GAC). Among the examined GAC columns, saturated GAC shows a slight increase in the total cell count after the filtration run. It has been reported that GAC increases the number of microbes due to the bacterial growth on the GAC bed during water treatment [11,12]. The increased bacterial number was associated with the release of microbes grown on the saturated GAC bed. Fresh GAC shows decreased number of microbes due to the adsorption compared with influent water. Fenton-cleaned GAC and hydraulically washed GAC also showed a decreased number of microbes because of the impact of washing. In water treatment facilities, the number of microbes introduced to the GAC process is meager because it normally measured after an ozone oxidation process. However, the result did not give a clear variation of cell count number as reported in the literature. One possible explanation is that the cell count of the influent is very high. In the FCM analysis of biomass detached from GAC (Table 2), Fenton-cleaned GAC shows a low number of intact cells compared with both saturated GAC and hydraulically washed GAC. Fenton-cleaned GAC also shows a high distribution of damaged cells (76%) as intact cell/damaged. In contrast, hydraulically washed GAC shows a greater number of intact cell than damaged cell. Saturated GAC also shows the higher distribution of intact cell as the proportion of intact cell/damaged cell is 145%. Fenton chemical washing uses oxidation reactions which can directly affect the bacteria retained by the GAC bed.

Fig. 5 shows the effects of chemical washing and hydraulic backwash on the variations in total and cell-bound ATP in surface water passing through different GAC columns. Cell-bound ATP of the influent sample was 92 ± 21 ng-ATP/L, which was very close to that of fresh GAC and Fenton-cleaned GAC. This implies that either absorption or release of microbes was not found for hydraulically washed GAC or saturated GAC. In contrast, the cell-bound ATP increased

Table 1
Effects of chemical cleaning and hydraulic backwash on the variations in bacterial cell concentration of surface water passing through different GAC columns

Sample		Intact cell (cell/mL)	Damaged cell (cell/mL)	Total cell (cell/mL)	Intact/damaged (%)
Influent and Effluent	Fresh	2.3E+06	6.4E+05	2.94E+06	368
	Fenton	2.5E+06	4.7E+05	2.97E+06	532
	Physical	2.5E+06	5.7E+05	3.07E+06	444
	Saturated	2.7E+06	5.7E+05	3.27E+06	465
	Raw	2.6E+06	6.0E+05	3.20E+06	443

Table 2
Fenton chemical cleaning and physical backwash of saturated GAC and their impacts on the concentration of bacterial cells retained onto the GAC

Sample		Intact cell (cell/mL)	Damaged cell (cell/mL)	Total cell (cell/mL)	Intact/damaged (%)
Adsorbate (biomass)	Fresh	7.6E+05	1.3E+06	2.06E+06	60
	Fenton	2.5E+06	3.3E+06	5.80E+06	76
	Physical	4.0E+06	3.5E+06	7.50E+06	116
	Saturated	4.6E+06	3.1E+06	7.70E+06	145

Note: Results shown in comparison with that observed for fresh GAC.

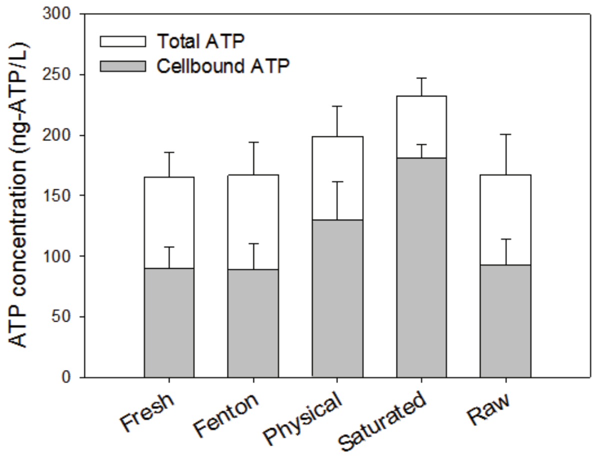

Fig. 5. Effects of chemical cleaning and hydraulic backwash on the variations in total and cell-bound ATP in surface water passing through different GAC columns.

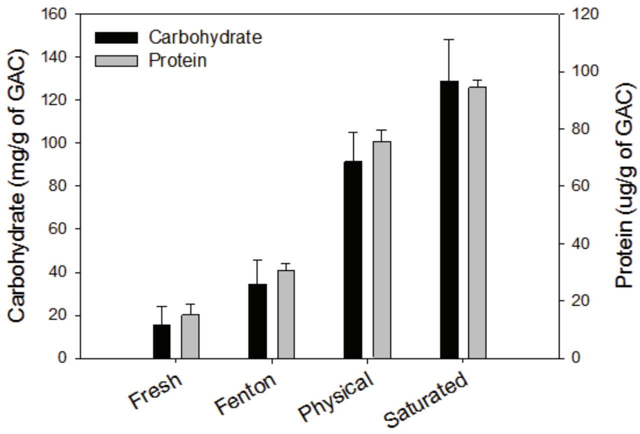

Fig. 6. Effects of chemical cleaning and hydraulic backwash on the composition of EPS extracted from different GAC prior to the filtration runs.

after the filtration of surface water through saturated GAC (180 ± 11 ng-ATP/L), and even after hydraulic backwash (130 ± 31 ng-ATP/L) was conducted. An increased number of cell-bound ATP of hydraulically washed GAC and saturated GAC is due to the release of bacteria retained onto the GAC column beds, which coincided with the FCM analysis (Table 1). Hydraulically washed GAC shows decreased cell-bound ATP compared with saturated GAC cell-bound ATP, but it shows a less decrease number than Fenton-cleaned GAC as EPS of biofilm limits the microbial release [13]. Consistent results were also shown in the ATP measurement of biomass detached from the GAC (Fig. S6 in supporting information). Fenton-cleaned GAC shows lower activity of retained bacteria compared with that observed for hydraulically washed GAC and saturated GAC, as shown in the results of intact cell count using FCM (Table 2).

Excessive EPS will make GAC operation difficult in water treatment facility due to the loss of absorptivity and increase of head loss [14]. This study measured EPS of biomass detached from GAC. Fig. 6 shows the effects of chemical washing and hydraulic backwash on the composition of EPS extracted from different GAC prior to the filtration runs using Tancheon River water. Saturated GAC has 129 mg/g of GAC for carbohydrate and 92 μg/g of GAC for protein. Both EPS components were removed to some extent after hydraulic backwash (91 mg/g of GAC for carbohydrate and 75 μg/g of GAC for protein), while a substantial removal was achieved with Fenton chemical washing (35 mg/g of GAC for carbohydrate and 30 μg/g of GAC for protein). Physical treatment can hardly remove EPS as they are strongly bonded into the surface area. Fenton chemical washing was effective at removing adsorbed EPS onto GAC. The removal of adsorbed organic matter can be compared with the EPS results. Biopolymer was removed by Fenton chemical washing to a greater extent than by hydraulic backwash (Fig. S4 in supporting information). The removal of EPS takes part in the reduction of biopolymer. A similar trend was also found in the result from the fluorescence measurements (Fig. S5 in supporting information).

4. Conclusions

In this study, we employed Fenton's reagent to restore the adsorptivity of field-spent GAC and estimated the fate of organic components during the filtration of surface water through the restored GAC. In contrast to a thermal regeneration of GAC and media replacement in the bed for GAC enhancement, Fenton chemical washing through the addition of chemicals into the readily available backwash pipelines is considered as a cost-effective and time-saving strategy not only to overcome the limitations of traditional hydraulic backwashing, but also to provide the effectiveness associated with adsorbents. Our results indicate that the use of in situ chemical washing is a highly feasible technological procedure which effectively removes EPS that are retained on carbon materials and accelerate GAC breakthrough. Fenton chemical washing uses oxidation reactions which can directly affect bacteria and organic matter retained onto the GAC bed, which improved organic adsorptivity drastically. Contrarily, hydraulic backwash alone could hardly remove EPS adsorbed onto the carbon materials as they were strongly bonded to the surface or the pore walls of GAC. Long-term studies should be performed using techniques analogous to those used in this paper to evaluate process robustness and determine the long-term effects of Fenton chemical washing which may affect the structure of micropores and subsequently cause a loss of surface area with an increase of pore size. More research and pilot studies should also be conducted to generate a cost comparison of traditional regeneration and Fenton chemical washing.

Acknowledgments

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Supporting information

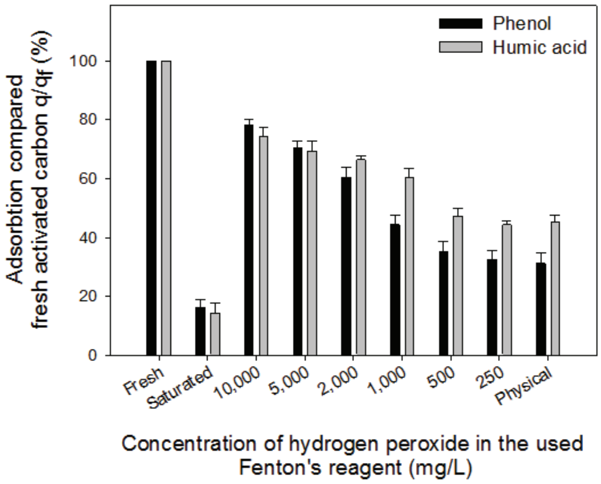

Fig. S1. Adsorptivity recovery after chemical washing of saturated GAC as a function of hydrogen peroxide level. Phenol (1,000 mg/L as TOC) and purified humic acid (50 mg/L as TOC) were used to saturate fresh GAC beds. Aldrich humic acid was purified to remove inorganic impurities using the method previously proposed by Hur and Schlautman [15]. Both chemicals were also used as influents for columns to determine the adsorption recovery after hydraulic backwash and chemical cleaning of the saturated GAC beds. All columns were operated at an EBCT of 10 min at ambient temperature (16°C–22°C).

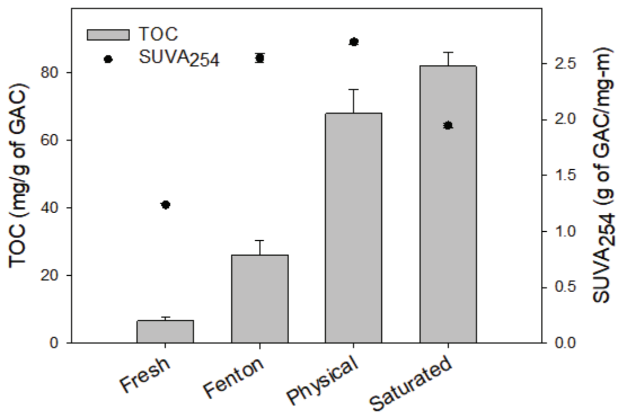

Fig. S3. Fenton chemical cleaning and physical backwash of saturated GAC and their impacts on the concentration and characteristics of organic matter retained onto the GAC. Results shown in comparison with that observed for fresh GAC.

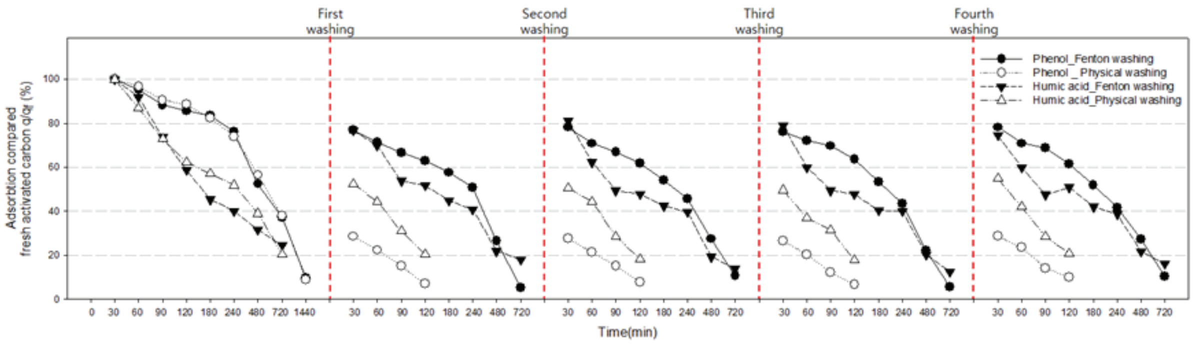

Fig. S2. Multi-cycle cleaning of saturated GAC beds. Hydraulic backwash and chemical washing were conducted using deionized water and Fenton's reagent (10,000 ppm), respectively. GAC columns were saturated with either phenol (1,000 mg/L as TOC) or purified humic acid (50 mg/L as TOC).

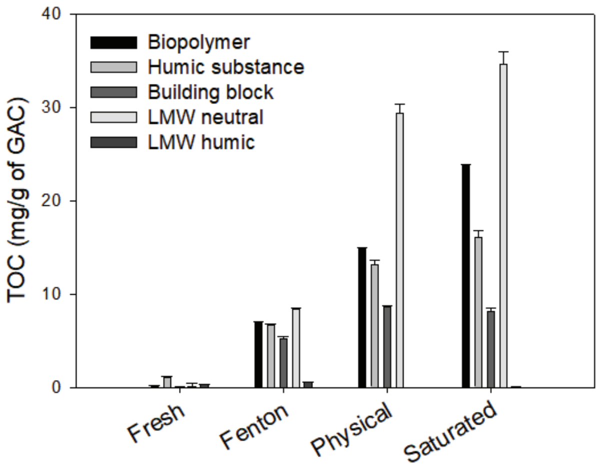

Fig. S4. Fenton chemical cleaning and physical backwash of saturated GAC and their impacts on the compositional fractions of organic matter retained onto the GAC. Results shown in comparison with that observed for fresh GAC.

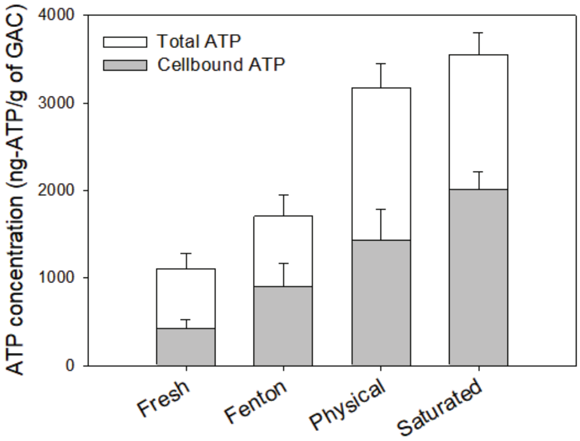

Fig. S6. Fenton chemical cleaning and physical backwash of saturated GAC and their impacts on the ATP levels of microbes retained onto the GAC. Results shown in comparison with that observed for fresh GAC.

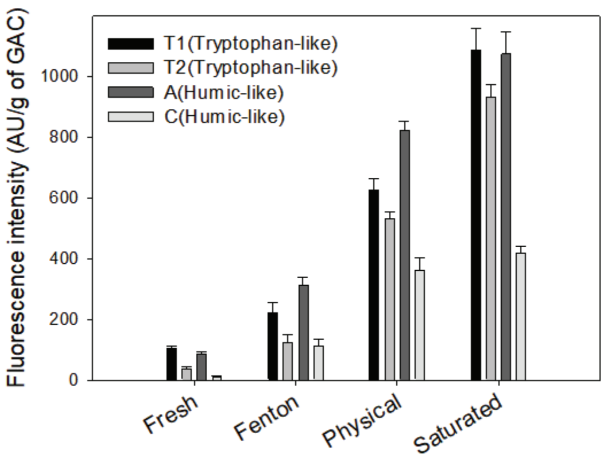

Fig. S5. Fenton chemical cleaning and physical backwash of saturated GAC and their impacts on the fluorescent fractions of organic matter retained onto the GAC. Results shown in comparison with that observed for fresh GAC.