

Comparison of the water purification efficiency of two different flocculation-sediment-filtration (FSF) processes in Chaohu Lake in China

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ABSTRACT

A novel biofilter (BF) with the potential to improve pollutant removal when used as a substitute for sand filters in water purification processes is presented. To investigate BF's pollutant removal performance, a flocculation-sediment-biofiltration (FSBF) process was compared with a flocculation-sediment-sand filtration (FSSF) process on a lab scale, with water collected from Chaohu Lake. The results showed that, during the stable period, the average removal efficiencies of COD_{Mn} , $\text{NH}_3\text{-N}$, $\text{NO}_2\text{-N}$ and UV_{254} of the FSSF process were 31.7, 29.9, 33.3, and 21.1%, respectively, while 42.7, 92.7, 90.3, and 27.8% of COD_{Mn} , $\text{NH}_3\text{-N}$, $\text{NO}_2\text{-N}$, and UV_{254} were removed by the FSBF process, respectively. In the BF, microorganisms in the biofilm on granular activated carbon appeared to play an important role in pollutants removal. Moreover, the FSBF process was more efficient at removing compounds from each organic content molecular weight fraction than the FSSF process, particularly for the removal of fractions with a molecular weight less than 10 KD, and especially for fractions less than 3 KD, which predominated the organic contents in raw water. Gas chromatography-mass spectrometry (GC-MS) indicated that the FSBF process could reduce more species than the FSSF and tended to produce or induce more kinds of new organic chemical simultaneously. Ultimately, the total organic species in the effluent of the FSBF process was less than that in the effluent of the FSSF process.

Key words | biofiltration, Chaohu Lake, drinking water, GC-MS, molecular weight distribution, water treatment

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INTRODUCTION

When rivers, lakes, and reservoirs are polluted, the quality of drinking water supplied by them is likely to be under threat. Moreover, some pollutants and by-products from the disinfection process may be carcinogenic, mutagenic, and/or teratogenic (Sagbo *et al.* 2008; Wu *et al.* 2009; Zhang *et al.* 2010). Furthermore, it is very difficult for traditional water treatment processes to remove these types of pollutants which has created new challenges in water treatment design over the last decade (Guzzella *et al.* 2002; Cho *et al.* 2003).

Chaohu Lake, which is one of the biggest lakes in China, is an important water supply. Despite its importance, some areas of the lake are currently being polluted

(Chinese Environment Bulletin 2015). As in many other Chinese regions, the flocculation-sedimentation-sand filtration-disinfection process is a common drinking water treatment operation used in the areas supplied by Chaohu Lake; however, this process has a low efficiency at removing dissolved organic materials such as ammonia and nitrite (Frederik & Guntena 2006; Song *et al.* 2015). Thus, new types of water purification processes are urgently needed. Numerous new drinking water treatment processes have recently emerged, such as membrane separation, advanced oxidation, photo catalytic oxidation, and biological processes (Seredyńska-Sobecka *et al.* 2006; Nandy *et al.* 2007; Toor & Mohseni 2007; Huang *et al.*

2008). Although membrane separation, advanced oxidation, and photo catalytic oxidation provide satisfactory pollutant removal, they are all costly and will add complexity when used in water purification processes. Furthermore, advanced oxidation and photo catalytic oxidation have not yet been applied to large scale drinking water treatment processes. Additionally, many researchers favor biofiltration over these processes because it is easy to operate. A typical biofilter (hereafter referred to as BF) has two filtering media layers, and good potential to be applied to drinking water treatment processes in China and other developing countries. In a BF, the upper media comprises materials that promote microbial adhesion, and the substrate usually consists of sand, which is used to filter the insoluble suspended substances. Additionally, the BF can remove some pollutants through biodegradation if it is operated properly. Moreover, a sand filter can be transformed into a BF by simply replacing a certain thickness of sand with materials that are favorable to the retention of microorganisms. As no additional structure is needed, upgrading to a BF in a conventional water treatment operation represents a significant advantage in regions in which space or funds are not sufficient to support waterworks improvements.

Even though there have been some reports of BFs or similar techniques, these studies have commonly used synthetic polluted water, while few studies have focused on the direct treatment of actual raw polluted water. Since the composition and characteristics of the contaminants of raw polluted water can be complex, existing reports may not be complete. In this study, we collected raw water from Chaohu Lake, China, then compared the flocculation-sediment-sand filtration (FSSF) with the flocculation-sediment-biofiltration (FSBF) process on the capacity of removing pollutants. COD_{Mn} , $\text{NH}_3\text{-N}$, $\text{NO}_2\text{-N}$, and UV_{254} were detected to evaluate the water quality. By analyzing the molecular weight distribution and the components of the organic contents in the raw water and the treated water respectively, the elimination or migration of these pollutants in the two processes could be identified further. The results of this study can provide useful information about BF for its application to drinking water treatment processes used in areas supplied by polluted water, which are similar to Chaohu Lake, China.

MATERIALS AND METHODS

Experimental design

Raw water was collected from a site in Chaohu Lake, which is near the inlet of a water treatment facility. As shown in Figure 1, the raw water was pumped into an overhead storage tank, from which it flowed into a flocculation and sedimentation (FS) tank by gravity. The FS tank was a sequencing batch reactor made of stainless steel, and its running cycle included a filling phase (1 h), a flocculation phase (25 min) and settling phase (1.5 h). The flocculant used in this experiment was polyaluminum chloride (PAC) at a concentration of 30 mg/L^{-1} and polyacrylamide (PAM) at a concentration of 0.2 mg/L^{-1} . In the flocculation phase, the water was first stirred at 300 rpm for 5 min, then at 100 rpm for 10 min. The operation parameters were determined by a set of preliminary coagulation tests.

After the settlement phase, the supernatant was decanted into two filters (80 mm in diameter, 2.5 m in height) as shown in Figure 1 at the same flow rate (20 L r^{-1}). The sand filter was filled with a 1-m thick layer of sand supported by a 0.4-m thick layer of pebbles underneath, and the BF was filled with a 0.4-m thick supporting layer of pebbles, followed by a 0.4-m thick layer of sand (particle size 0.5–1.2 mm), and finally a 0.6-m thick layer of granular activated carbon (GAC) (particle size 0.5–1.5 mm). The GAC were taken from a laboratory BF apparatus that had been treating artificial influent water for one year. Moreover, the GAC was adhered by developing a biofilm onto biologically activated carbon (BAC). The biofilm

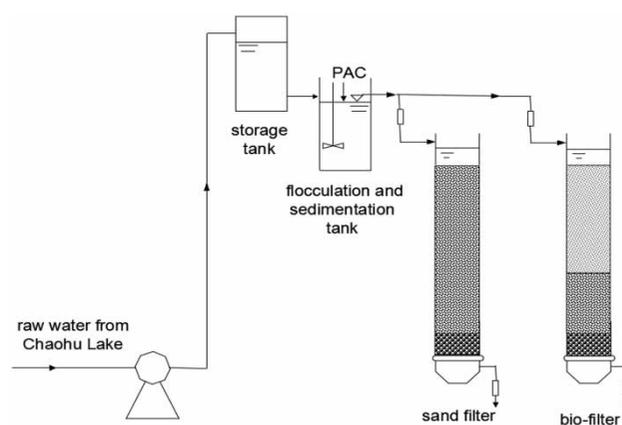


Figure 1 | Process diagram.

microorganisms on the BAC were allowed to acclimate to the water collected from Chaohu Lake until the FSBF process showed stable pollutant removal, after which the two filters were run for 30 days continually (defined as stable operation period in this paper). During these stable periods, the raw water temperature ranged from 28 to 31 °C, the flow rates of the two filters were both 4 m³/h, and both empty bed residence times were 15 min.

The two filters were backwashed every 3–4 days, which included air backwashing for 3 min at 10 L/m⁻²/s⁻¹, followed by 4 min of backwashing with air for 10 L/m⁻²/s⁻¹ and water for 4 L/m⁻²/s⁻¹, respectively, with a final step of backwashing with water for 4 min at 8 L/m⁻²/s⁻¹.

Indicators and analytical methods

COD_{Mn}, NH₃-N, NO₂⁻-N, and UV₂₅₄ were detected to evaluate water quality. As a comprehensive index of organic and reducing inorganic materials, COD_{Mn} is usually an important quality indicator for drinking water and source water. In this study, COD_{Mn} was determined according to the method specified in the ISO standards (ISO8467-1993). Ammonia, which can account for the extra consumption of disinfectant in waterworks, and nitrite, which is harmful to human health and usually present in polluted resource water, were both selected as water quality indicators for this study and analyzed according to *Standard Methods (APHA/AWWA 1998)*. UV₂₅₄ represents absorbance at a wavelength of 254, reflecting the humus and aromatic compounds, which contain a C=C double bond or a C=O double bond. This value was chosen as an auxiliary comprehensive indicator of organic pollution in this study, and quantified using a Shimadzu UV-1800 ultraviolet spectrophotometer.

The molecular weight distributions of the organic contents in raw water and treated water were analyzed by a centrifugal ultrafiltration method (Chow *et al.* 2009). In this method, a set of water samples of equal volume were first passed through a series of Millipore filters blocking the passage of different molecular weights, which was achieved by centrifugal action in a high-speed (Z206A) centrifuge, and total organic carbon (TOC) was determined by measuring each filtered liquor through a TOC Analyzer (Shimadzu TOC-6-WP). Finally, the molecular weight

distributions were clarified by calculating the differences among the TOC values in different filter liquors.

The compositions of organic chemicals in the raw water and treated water on the 30th day of the experiment were determined by gas chromatography–mass spectrometry (GC-MS) (Thermo Scientific ITQ 1100TM). When the FSBF process showed stable efficiency for treating raw water from Chaohu Lake, the biomass activity of the biofilm in the BF was measured according to Urfer & Huck (2001). Bioactivity was evaluated by specific oxygen uptake rate (SOUR), which is defined as oxygen uptake rate per unit biomass. The biomass was represented by the phosphatide amount in terms of nanomoles of phosphorus (nmol Ph) in this study (Liu *et al.* 2001).

RESULTS AND DISCUSSION

COD_{Mn}, NH₃-N, NO₂⁻-N and UV₂₅₄ removal by the two processes

We collected 15 sets of water quality data during the stable period and determined the average and range of the concentrations of each pollutant in raw water. The results for the influent of the two filters, as well as the water treated by the two processes (FSSF and FSBF), are shown in Table 1.

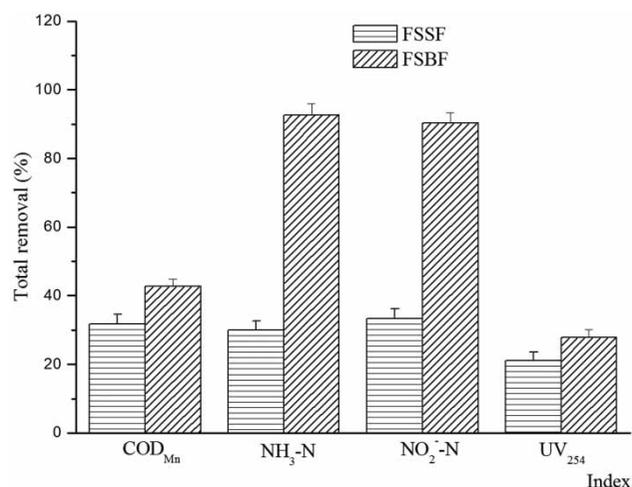
All pollutant removal data using the water quality indicators was represented as removal efficiency, which is presented in Figure 2, in the form of running averages.

The removal efficiencies of COD_{Mn}, NH₃-N, NO₂⁻-N and UV₂₅₄ of the FSSF process over 30 days were 31.7, 29.9, 33.3 and 21.1%, respectively. Additionally, 42.7, 92.7, 90.3, and 27.8% of the COD_{Mn}, NH₃-N, NO₂⁻-N, and UV₂₅₄, respectively, were removed by the FSBF process, demonstrating that the FSBF process was more efficient at reducing all pollution indicators than the FSSF process.

As shown in Table 1, the values of COD_{Mn}, NH₃-N, NO₂⁻-N and UV₂₅₄ in the effluent of the sand filter were nearly equal to those in the inflow of the sand filter after FS. These findings indicate that the initial stages of the treatment were responsible for pollutant removal, which suggests that sand filtration made almost no contribution in this regard. Furthermore, it can be concluded that the BAC in the BF might play an important role in the efficient removal of these pollutants.

Table 1 | Water qualities of each step of the two processes during flocculation-sediment-filtration

| Water samples | COD _{Mn} (mg/L) | | NH ₃ -N (mg/L) | | NO ₂ -N (mg/L) | | UV ₂₅₄ (cm ⁻¹) | |
|-----------------------------------------------------------------|--------------------------|------|---------------------------|-------|---------------------------|-------|---------------------------------------|-------|
| | Range | Mean | Range | Mean | Range | Mean | Range | Mean |
| Raw water | 3.33–3.97 | 3.72 | 0.361–0.493 | 0.417 | 0.141–0.171 | 0.158 | 0.061–0.070 | 0.066 |
| Influent of the two different filters | 2.17–2.18 | 2.53 | 0.257–0.358 | 0.294 | 0.093–0.130 | 0.107 | 0.040–0.055 | 0.050 |
| Effluent of the sand filter (water treated by the FSSF process) | 2.14–2.77 | 2.54 | 0.255–0.352 | 0.292 | 0.096–0.119 | 0.105 | 0.048–0.055 | 0.052 |
| Effluent of the BF (treated water of the FSBF process) | 1.89–2.39 | 2.13 | 0.014–0.055 | 0.031 | 0.011–0.023 | 0.015 | 0.044–0.052 | 0.048 |

**Figure 2** | COD_{Mn}, NH₃-N, NO₂⁻-N, and UV₂₅₄ removal by the two processes.

Specifically, the COD_{Mn} removal efficiency of the FSBF process was 11% higher than that of the FSSF process, suggesting that BAC in the BF had a significant effect on organic content removal. Liao *et al.* (2013) indicated that BAC could remove organic materials through the biodegradation activity of the biofilm-associated microorganisms on the surface of the GAC, as well as the adsorption of activated carbon, and the biodegradation of this organic content is mainly achieved through the metabolic activity of these microorganisms and the hydrolytic activity of extracellular enzymes. In this study, the average SOUR of heterotrophic biofilm-associated bacteria along the GAC layer in the BF changed from 0.2257 to 0.3405 μg O₂/(nmol Ph) during the stable operation period, suggesting that the activated carbon was colonized by a certain amount of heterotrophic bacteria, and these microorganisms likely contributed to the removal of organics. However, the relative contributions of biodegradation and

adsorption by activated carbon to pollutant removal could not be clarified entirely by the results of this study.

The FSBF process showed considerably higher removal efficiencies for NH₃-N and NO₂⁻-N than the FSSF process, with both NH₃-N and NO₂⁻-N removal efficiency in the FSBF process reaching over 90%. Furthermore, tests in this study showed that activated carbon used in the BF had a very low efficiency (less than 4% after adsorption for 1 h) at removing NH₃-N and NO₂⁻-N from the raw water collected from Chaohu Lake (data not published). Taken together, these findings indicate that NH₃-N and NO₂⁻-N were primarily removed by the bacteria in BF. Over the stable running period, the average SOUR of nitrobacteria and Nitrococcus present in the carbon layer ranged from 0.0227 to 0.0693 μg O₂/(nmol Ph) and from 0.0414 to 0.1003 μg O₂/(nmol Ph), respectively, indicating that a significant amount of Nitrococcus and nitrobacteria might have grown on the surface of the GAC in the BF.

During this experiment, nitrite in the effluent of BF decreased, indicating that the nitrification removed more nitrite than the nitrosation produced. Furthermore, the BF is not likely to cause increases of nitrite in the effluent.

Molecular weight distribution of the organic materials in the raw and treated water

As shown in Figure 3, molecular weights were divided into six intervals following the distribution of molecular weights fractionated by Millipore filtration, with organic contents correspondingly divided into six parts as shown in Table 2.

The molecular weight distribution data of raw water, water treated by the FSSF process, and water treated by the FSBF process were estimated by quantifying the organic

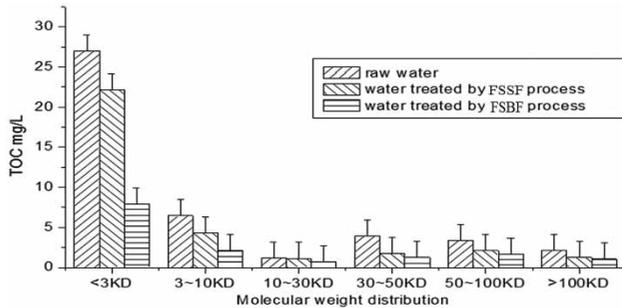


Figure 3 | Molecular weight distribution of the organic contents in untreated water, FSSF-treated water, and FSBF-treated water.

matter contents, which were evaluated by TOC (Figure 3). The average proportion of each organic matter fraction in the treated or untreated water is shown in Table 3, and the average extent of removal of each part is shown in Table 4.

During the treatment processes, the change in concentration across all organic content fractions may be caused by degradation of the original organic content in the raw water, or the production or introduction of new chemicals, as the process details are not completely understood at this time. In this study, the removal of organic materials was not restricted to the removal of original organics in the water, but the results of multiple processes including degradation of the original organic contents, as well as the production or introduction of new chemicals.

As shown in Figure 3 and Table 4, the FSBF process could remove each fraction at higher efficiency than the FSSF process, and the FSBF process was significantly better than the FSSF process at removing organic materials with molecular weights less than 3 KD. It should also be noted that Fractions

I and II were the major constituents of total organic contents in untreated water from Chaohu Lake, with Fraction I accounting for most of the organic materials in the raw water. As shown in Table 4, Fraction I was removed by the FSSF process at a lower efficiency. Based on these findings, we concluded that FS was the main contributor to the removal of organic materials in the FSSF, which suggests that the FS process was not suitable for the removal of organic matter with molecular weights under 3 KD. These findings are consistent with other studies (Volk & Lechevallier 2002), and explain why the FSSF process is not efficient at removing total organic contents.

The FSBF process removed Fractions I and II more effectively in this experiment, with average removal efficiencies of 70.4 and 67.4%, respectively. Because we concluded that the organic fractions with molecular weights less than 3 KD were not efficiently removed by the FS process, the removal of this organic content fraction can be attributed to the activity of the BF. Moreover, the finding that the majority of fractions (Fractions I and II) were removed by the FSBF process in a superior fashion can help explain the high efficiency of FSBF on reducing total organic contents in source water.

Components of organic contents in raw and treated water

The composition of organics in drinking water are likely to influence the quality of the water. Therefore, the organic chemicals in the raw water, the water treated by the FSSF process and the water treated by the FSBF process on the

Table 2 | Organic content fractionation according to molecular weight

| Organic content fraction | Fraction I | Fraction II | Fraction III | Fraction IV | Fraction V | Fraction VI |
|--------------------------|------------|-------------|--------------|-------------|------------|-------------|
| Molecular weight | <3 KD | 3–10 KD | 10–30 KD | 30–50 KD | 50–100 KD | >100 KD |

Table 3 | Average proportions of the organic contents with different molecular weight

| Water samples | Proportion of each organic matter fraction (%) | | | | | |
|--------------------------------------|------------------------------------------------|-------------|--------------|-------------|------------|-------------|
| | Fraction I | Fraction II | Fraction III | Fraction IV | Fraction V | Fraction VI |
| Raw water collected from Chaohu Lake | 61.3 | 14.6 | 2.7 | 9.0 | 7.6 | 4.8 |
| Water treated by the FSSF process | 67.7 | 13.2 | 3.5 | 5.4 | 6.4 | 3.8 |
| Water treated by the FSBF process | 53.7 | 14.2 | 4.9 | 8.5 | 11.5 | 7.1 |

Table 4 | Average removal of organic contents with different molecular weights

| Water sample | Extent of removal of each organic matter fraction (%) | | | | | |
|-------------------------------|-------------------------------------------------------|-------------|--------------|-------------|------------|-------------|
| | Fraction I | Fraction II | Fraction III | Fraction IV | Fraction V | Fraction VI |
| Water treated by FSSF process | 18.0 | 33.0 | 4.2 | 55.2 | 37.4 | 42.3 |
| Water treated by FSBF process | 70.7 | 67.4 | 38.1 | 68.3 | 49.4 | 50.7 |

30th day of the stable running period of the two processes were examined by GC-MS (Table 5).

Overall, 71 organic chemicals were detected in source water collected from Chaohu lake, including two priority pollutants identified by the EPA (dichlorobenzene and 2,4-DNT), as well as one group of endocrine disruptors (i.e. phthalates). The organic chemicals were classified into 13 categories, which are listed in Table 5.

Moreover, the GC-MS results showed that the FSSF process could eliminate 19 organic chemicals, and produce or induce four new chemicals. There were some organic chemicals eliminated in most of the categories, and the esters and the heterocyclic compounds showed more removal than other species. The four new chemicals belonged to the amine or arenes groups, while more chemicals were

eliminated in both categories at the same time. Moreover, the GC-MS results revealed that, although elimination of heterocyclic compounds and esters was incomplete, the levels of these chemicals were significantly reduced. This differed from other non-eliminated organic chemicals, which showed little reduction. Taken together, these findings indicate that the organics were removed almost completely by the FS process. These results suggest that the new chemicals produced in the FSSF process originate from coagulant impurities, which might be created during the production of PAM reagent or when PAMs are hydrolyzed.

In the FSBF process, 45 kinds of organic chemicals disappeared from the treated water, which indicated that this process could eliminate more types of organic chemicals than the FSSF process. However, 19 new chemicals

Table 5 | Results of component analysis of the organic contents

| Categories | Raw water | | Water treated by FSSF | | Water treated by FSBF | |
|------------------------|---------------------------|-------------------|---------------------------|-------------------|---------------------------|-------------------|
| | Number of organic species | Total peak area | Number of organic species | Total peak area | Number of organic species | Total peak area |
| Alcohol | 8 | 5×10^9 | 7 | 4.2×10^9 | 5 | 3.2×10^9 |
| Ketones | 6 | 2×10^9 | 5 | 1.2×10^9 | 4 | 0.9×10^9 |
| Esters | 13 | 3×10^9 | 7 | 2.3×10^9 | 5 | 1.7×10^9 |
| Ethers | 3 | 0.6×10^9 | 3 | 0.3×10^9 | 2 | 0.2×10^9 |
| Amine | 15 | 8×10^9 | 14 | 5×10^9 | 10 | 4×10^9 |
| Nitriles | 1 | 0.3×10^9 | – | – | – | – |
| Phenols | 2 | 0.6×10^9 | 2 | 0.5×10^9 | 2 | 0.3×10^9 |
| Arenes | 5 | 2.9×10^9 | 5 | 2×10^9 | 4 | 1.7×10^9 |
| Halohydrocarbons | 1 | 0.4×10^9 | 1 | 0.2×10^9 | – | – |
| Nitrobenzene | 2 | 1×10^9 | 2 | 0.7×10^9 | 2 | 0.4×10^9 |
| Heterocyclic compounds | 12 | 6×10^9 | 7 | 4×10^9 | 7 | 3.3×10^9 |
| Alkane | 2 | 2×10^9 | 2 | 1.5×10^9 | 2 | 1.1×10^9 |
| Others | 1 | 0.2×10^9 | 1 | 0.1×10^9 | 1 | 0.1×10^9 |
| Total | 71 | 32×10^9 | 56 | 22×10^9 | 45 | 17×10^9 |

appeared in the water treated by the FSBF process, while only four new chemicals appeared during the FSSF process, indicating that a number of new chemicals were produced in the BF during the biodegradation of these organic chemicals. The organic chemicals removed from the water by the FSBF process included organic chemicals of each category, while the new chemicals mainly belonged to alkane, amine, and alcohol groups. Overall, the total amount of organic chemicals eliminated by the FSBF process increased relative to FSSF.

As shown in Table 4, 56 organic chemicals were detected in the effluent of the FSSF process eventually, while 45 were present in water treated by the FSBF process. Moreover, there were more species eliminated by the FSBF process than the FSSF process for each category of chemical. Additionally, it seems that the FSBF process can reduce more organic species than the FSSF process. However, it should be acknowledged that new organic species were produced or induced in the FSBF, indicating that the safety of the treated water of the FSBF process still needs to be studied further.

CONCLUSIONS

In this study, the FSBF process was compared to the FSSF process to clarify the effect of replacing a sand filter with a BF in a water purification process when treating the water collected from Chaohu Lake. The FSBF process was more efficient than the FSSF process at removing all pollutants detected in our experiment, indicating the superiority of the BFs over the sand filters for purifying slightly polluted water. Additionally, the results of this study indicated that microorganisms in the biofilm along the GAC contributed to pollutants removal. Moreover, the $\text{NH}_3\text{-N}$ and $\text{NO}_2\text{-N}$ removal was mainly attributed to microorganisms along the GAC, while the contributions of adsorption of GAC and biodegradation to organic contents removal could not be determined at this point. Furthermore, The FSBF was significantly more efficient at removing the organic content fraction for each molecular weight than the FSSF process, in particular for the removal of fractions with a molecular weight less than 10 KD, and especially for fractions less than 3 KD, which accounted for most of the organic

contents in the raw water. Our findings also showed that the FSBF process could reduce more species than the FSSF and tended to produce or induce more kinds of new organic chemicals simultaneously. Ultimately, water treated by the FSBF process contained fewer total organic species than that treated by the FSSF process. In summary, a FSBF process significantly improved pollutant removal; therefore, it is recommended that such a system be used in place of existing systems with BFs in sand filters that are utilized for water purification. However, the safety of water treated by the FSBF process still needs to be studied further.

ACKNOWLEDGEMENTS

The authors thank the National High Technology Research and Development Program of China (2012AA062605) and the Natural Science Foundation of Anhui province of China (1508085ME91) for the partial support of this study.

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First received 23 June 2016; accepted in revised form 8 December 2016. Available online 10 February 2017