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Removal of dissolved organic matter from algae-polluted surface water by coagulation

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ABSTRACT

Dissolved organic matter (DOM) is ubiquitous in the algae-polluted surface water and it possesses a potential threat to drinking water safety. It is important to investigate the removal efficiency of DOM by coagulation. Polyaluminum chloride a widely used coagulant is applied in the treatment of DOM derived from Minzhu Lake in Chongqing of China. Based on the analysis of dissolved organic carbon, UV_{254}, specific ultraviolet absorbance, and fluorescence excitation-emission matrix spectroscopy, the results indicate that the small proportion of DOM, such as aromatic-like organic substances, is removed with algae and other colloid particles via coagulation, but most of DOM, such as he tryptophan-like and fulvic-like organic substances, is remained in the treated water. Co-flocculation, bridging, adsorption, and entrapment would be the main removal mechanisms of DOM. DOM removal will be slightly improved through optimizing pH of surface water and using a composite coagulant.

Keywords: Dissolved organic matter; Polyaluminum chloride; Algae; Coagulation; Surface water

1. Introduction

Algae blooms have grown up to be a challenge for drinking water treatment in numerous countries all over the world [1,2]. They often cause water quality deterioration including hypoxia, odors, bad taste, and scum formation [2]. Although algae are properly treated by coagulation, filtration, and adsorption [3–5], these water treatment technologies often lead to algae cell lysis and the release of intracellular organic matter which results in secondary pollution of aquatic environments [2]. Besides, organic matter such as microcystin yielded by algae has a detrimental effect on human health [6]. And dissolved organic matter (DOM) is often hard to be removed from water [7]. DOM also easily transforms to disinfect byproducts (DBPs) in chlorination and reduces the membrane flux sharply in the drinking water treatment [7,8].

Coagulation as a basic and essential water treatment technology has been widely applied in drinking

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water treatment [9,10]. The small suspended and colloidal particles in the water become the bigger ones after coagulation, and the aggregated particles are easily removed by gravity settlement or filtration [11]. However, it is controversial whether dissolved organic carbon (DOC) is able to be effectively removed by a coagulant [9]. And little information is available on the DOM removal from the algae-polluted surface water by coagulation.

Polyaluminum chloride (PAC) coagulant is extensively applied in drinking water treatment for its high efficiency and low cost [9,11]. In this study, the algae-polluted surface water was collected from Minzhu Lake and treated by the self-prepared PAC coagulant. The removal efficiencies of algae and DOM were investigated via the analysis of turbidity, DOC, UV_{254}, specific ultraviolet absorbance (SUVA), and fluorescence excitation-emission matrix (FEEM) spectroscopy. And coagulation mechanisms in the treatment were discussed. Moreover, enhanced coagulation was taken into account to improve the DOM removal rate.

2. Materials and methods

2.1. Surface water source

The surface water was gathered from Minzhu Lake that is located in Chongqing of China. Minzhu Lake was the scenic lake, and its total area was about 10,000 m^2. The main water source of Minzhu Lake was rain, and there were no potential contaminants discharged from domestic sewage and industrial wastewater. But the excessive reproduction of algae in Minzhu Lake was often observed in summer. The characteristics of water, such as turbidity, algae cell concentration, DOC, UV_{254}, temperature, and pH, were measured [12].

2.2. Chemicals and coagulants

All the chemical reagents were analytical grade chemicals. De-ionized water was used to make all the solutions of the reagents. In this work, PAC was prepared using fresh aluminum hydroxide gels [11]. And products were denoted as $PAC_{2.5}$, $PAC_{2.0}$, and PAC_0 according to the basicity values (the ratio of OH/Al) of 2.5, 2.0, and 0. Aluminum species distributions of the self-prepared PAC were measured by Ferron assay [11,13].

2.3. Coagulation tests

The surface water was collected from Minzhu Lake in summer after the algal bloom. Glass container was soaked and washed by chromic acid lotion before use. To avoid more and more intracellular organic matter releasing for the algae cell lysis in the storing, the collected surface water was directly treated by the self-prepared PAC under the natural temperature and pH value. And commercial PAC with the basicity value of 2.0 (Lanjie Water Purification Material Co., Ltd, China) was performed as a comparison in the same condition. A ZR4–6 six-paddle gang stirrer (Shenzhen Zhongrun Water Industry Technology and Development Co., Ltd, China) was used in the coagulation. After adding the coagulant, the raw water of 1 L was mixed at a high speed of 300 rpm for 1 min and at a low speed of 40 rpm for 10 min, and then it was allowed to settle for 30 min [11]. The treated water and raw water were immediately analyzed by a 2100P turbidity meter (HACH, USA) and a HQ11 pH meter (HACH, USA). Zeta potential was measured by a ZS90 Malvern potential analyzer (Malvern, UK) [11]. To enhance the coagulation performance of PAC in the algae-polluted surface water treatment, increasing the dosage and optimizing the pH of raw water was applied. And two composite coagulants of PAC-poly dimethyl diallyl ammonium chloride (PDMDAAC) and PAC-polyacrylamide (PAM) were also used.

2.4. Analysis methods

2.4.1. DOC, UV_{254}, and SUVA

The raw water and treated water were immediately filtered through 0.45 μm membrane filter after coagulation. Their DOC concentrations were measured by a Liqui TDC II total organic carbon analyzer (Elementar, Germany). Reproducibility of DOC was acceptable with deviations of less than 5% in duplicate. UV_{254} was measured by a TU-1900 ultraviolet/visible spectrophotometer (Purkinje General Instrument Co., Ltd, China) [14]. And the SUVA was calculated via the values of DOC and UV_{254} [15].

2.4.2. Fluorescence excitation-emission matrix spectroscopy

FEEM spectra of raw water and treated water were recorded on a F-7000 fluorescence spectrometer (Hitachi High Technologies, Tokyo). The operational parameters were set as the previous study [16,17]. Emission spectra were scanned from 200 nm to 550 nm with 1 nm increment and excitation spectra were scanned from 200 nm to 450 nm with 5 nm increments. The scan was performed at a speed of 12,000 nm/min. The slits of emission and excitation were 5 nm and PMT voltage was set at 725 V [16,17]. The intensity of the Raman line of de-ionized water was measured at 350 nm to monitor the stability of instrument [16].

3. Results and discussion

3.1. Characterization of the prepared polyaluminum chloride

Aluminum species distributions of PAC prepared using fresh aluminum hydroxide gels are represented in Table S1. Aluminum species are broadly divided into three categories, monomeric aluminum species (Al_a), small/middle polymeric aluminum species (Al_b), and high/insoluble polymeric aluminum species (Al_c) [11,13]. It has been reported that Al_b would be the most efficient species for the DOM removal [9]. And Al_b content increases with the increase of basicity value [13]. However, in this study, the highest Al_b content of 72.38% is observed in $PAC_{2.0}$ solution. Al_b content in $PAC_{2.5}$ solution abnormally decreases due to the presence of fresh aluminum hydroxide gels. The self-prepared PAC is applied in the algae-polluted surface water treatment.

3.2. Algae-polluted surface water treatment

3.2.1. Characterization of surface water

Algae blooms are often observed in Minzhu Lake during summer. Although water circulation system and wetland are implemented to control the excessive reproduction of algae, the algae (cell count of 1.1 ± 0.3 × 10^6 cells/mL) still exists in the water. The concentration of DOM in the algae-polluted Minzhu Lake is about 6.2 ± 0.5 mg DOC/L (Table 1). DOM might originate from the rainwater runoff, and it is also largely produced via the metabolism of algae cells and microorganisms especially in the case of algae blooms [18]. The SUVA value of surface water is only 2.145 L/m mg. Organic matter in natural water is mainly hydrophilic, low molecular weight and low removability, when the SUVA value is less than three [12,14]. The water quality of Minzhu Lake does not significantly change in the experimental period.

3.2.2. Turbidity removal

The acceptable performances of PAC in the turbidity removal are presented in Fig. 1 and Fig. S1. In optimal dosage range of PAC, the turbidity removal rates of algae-polluted surface water reach 90% and the residual turbidity in the treated water is below one NTU. However, the optimal dosage of PAC is high compared with other similar studies [11]. It suggests that turbidity derived from algae and DOM might decrease the coagulation efficiency. Besides, coagulation efficiencies of four PAC are different. The self-prepared $PAC_{2.0}$ has the highest coagulation efficiency. The coagulation efficiency of PAC_0 is slightly low. And the excessive PAC_0 easily causes the restabilization of colloid particles due to the charge reversal, and the performance becomes worse. Fresh aluminum hydroxide gels in $PAC_{2.5}$ lead to the lower coagulation efficiency of $PAC_{2.5}$ for the weaker capacity of charge neutralization and bridging.

3.2.3. DOM removal

UV_{254} and DOC as the important indexes are often used to assess organic matter concentration level in the water. And when the value of UV_{254} is higher, organic matter is more easily transformed to DBPs [19]. UV_{254} and DOC concentration of DOM in the algae-polluted surface water is measured before and after coagulation. And UV_{254} and DOC concentration in the raw water is about 0.133 cm^{-1} and 6.2 mg/L, respectively (Table 1). It has been noted that DOM is rarely removed by coagulation [20]. In the research, the values of UV_{254} decrease to 0.082 cm^{-1} and DOC concentration decreases to 4.28 mg/L under the optimal coagulation condition (Figs. 2 and 3). But overdosing $PAC_{2.0}$ and PAC_0 will lead to the increase in DOC in the treated water. It is represented that the DOM in Minzhu Lake is partly removed by PAC. Moreover, DOM removal is positively correlated with turbidity removal. For instance, the best removal rates of DOM and turbidity are obtained by $PAC_{2.0}$ at the dosage of 20 mg Al/L, and their removal rates decrease after overdosing $PAC_{2.0}$. Thus, DOM removal would be attributed to its co-flocculation with algae and other colloid particles in the water. However, the residual

Table 1
Water quality of Minzhu Lake in the experimental period

Turbidity (NTU)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/m mg)	pH	Temperature (℃)
9.32 ± 0.3	6.2 ± 0.5	0.133 ± 0.005	2.145 ± 0.28	8.25 ± 0.2	21.2 ± 0.2 (at the surface of Minzhu Lake); 19.7 ± 0.2 (in the laboratory)

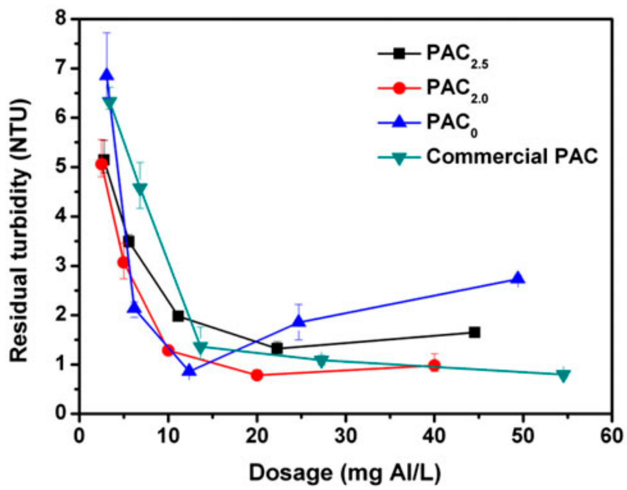

Fig. 1. Residual turbidity of surface water after being treated by PAC.

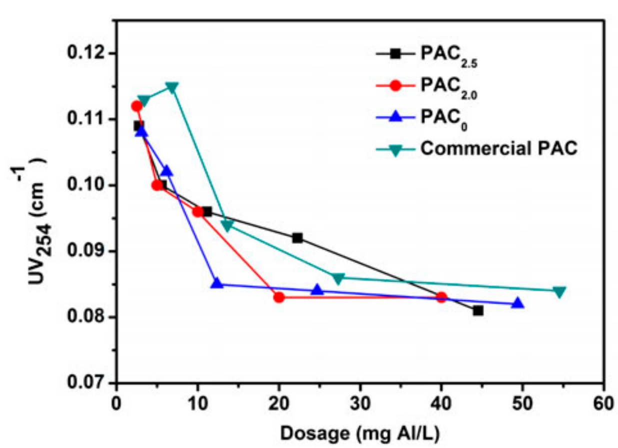

Fig. 2. UV_{254} of surface water after being treated by PAC.

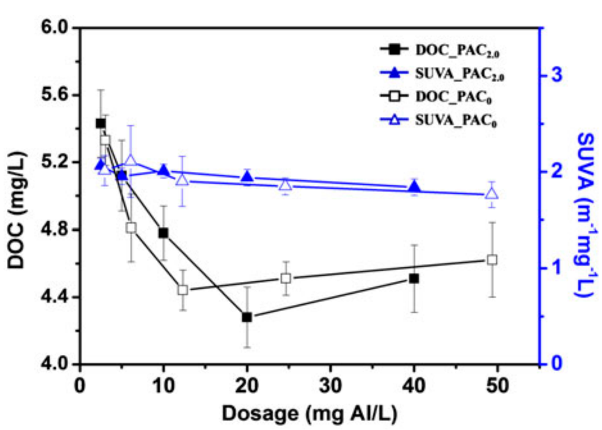

Fig. 3. DOC concentration and SUVA value of surface water after being treated by $PAC_{2.0}$ and PAC_0.

DOM is tough and fails to be removed by coagulation. SUVA values of treated water slightly decrease after coagulation (Fig. 3). It indicates a positive correlation between the removal of DOM and hydrophobic organic compounds [9].

In FEEM spectra, five typical peaks, named A, B, C, T_1, and T_2, are commonly observed in the surface water. Peak A appearing at Ex/Em of 320–350 nm/400–500 nm is related to humic-like substances. Peak B presented at Ex/Em of 225–237 nm/309–321 nm is associated with tyrosine-like (protein-like) substances. Peak C locating at 230–260 nm/400–500 nm is ascribed to fulvic-like substances. Peak T_1 and T_2 occurring at Ex/Em of 270–280 nm/305–335 nm and Ex/Em of 220–230 nm/325–340 nm belong to tryptophan-like (protein-like) substances and aromatic-like substances, respectively [7,21]. In the Fig. 4, three peaks are discovered, and peak T_2 is the most obvious fluorescence peak. Its fluorescence intensity decreases under the optimal dosage of $PAC_{2.0}$ (20.0 mg Al/L). It implies that some aromatic-like organic substances are removed, which is also correlated with the decrease in DOC concentration and UV_{254} (Figs. 2 and 3). On the contrary, peak T_1 and C are weaker in the FEEM spectra. And the fluorescence intensity of peak T_1 and C is almost unchanged after coagulation. Thus, these tryptophan-like and fulvic-like organic substances are retained in the water.

Zeta potential is an important indicator of the stability of colloidal dispersions. And colloids with high zeta potential possess electrically stabilized, while colloids with low zeta potentials tend to coagulate in water due to Brownian motion and turbulence of water [22]. Positively charged PAC is available to neutralize the negative charge on the surface of algae cell and colloid particles, which improves the aggregation and sedimentation of pollutants in the algae-polluted surface water [23]. Charge neutralization could be detected by the zeta potential. In the Fig. 5 and Table S2, the zeta potential of surface water slowly increases with the increase in the dosage of PAC. Although the dosage increases to 40 mg Al/L, the zeta potential is still negative in the case of $PAC_{2.5}$ and $PAC_{2.0}$. The results represent that some DOM might react with hydroxy-aluminum to form the soluble complex, which impedes the charge neutralization and increases the demand of dosage. And it also implies that charge neutralization is not the only coagulation mechanism. The bridge of Al_b improves the removal of algae and colloid particles in the algae-polluted surface water, and the adsorption of $Al(OH)_{3(am)}$ and Al_b contribute to DOM removal. Besides, entrapment of flocs is also indispensable in the treatment. Conversely, the increase rate of zeta potential is high in

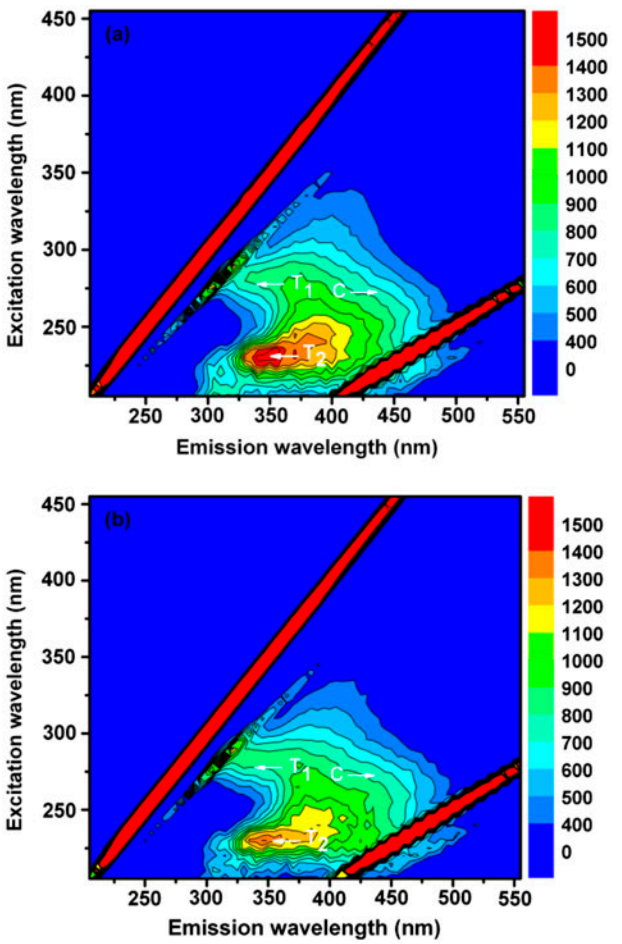

Fig. 4. FEEM spectra of surface water: (a) before and (b) after being treated by moderate $PAC_{2.0}$.

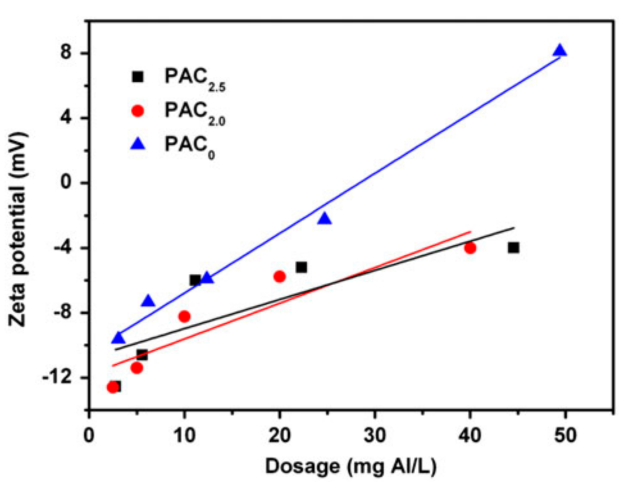

Fig. 5. Zeta potential of surface water after being treated by PAC.

the case of PAC_0 since Al_b content in PAC_0 is much less than that in $PAC_{2.0}$ and pH of PAC_0 is lower. The performance deterioration caused by charge reversal is conspicuous after overdosing PAC_0, which is represented in Figs. 1 and 3.

3.3. Enhanced coagulation

Coagulation is the highly efficient technology in the water treatment, but the removal rate of organic matter by coagulation is markedly low in some studies [24,25]. Thus, enhanced coagulation is considered to be used. The principal methods of enhanced coagulation include the choice of the suitable coagulant, the increase of coagulant dosage, the optimization of coagulation processes, and the control of pH of raw water [26,27]. In this study, adding more PAC will not enhance the removal rate of DOM and even result in the worse turbidity removal for the restabilization of algae and colloidal particles (Figs. 1 and 3). Thus, other enhanced coagulation methods, such as adjusting pH of algae-polluted surface water and using composite coagulant, were applied to increase the removal efficiency of DOM.

3.3.1. Effect of pH

In the water, pH is an important factor that will influence the aluminum hydrolysis and the characteristics of pollutants [10,28]. Normally, PAC will perform better at the optimal pH range [29]. In the study, the effects of pH of algae-polluted surface water on the algae and DOM removal are investigated under the optimal dosage of $PAC_{2.0}$ (20.0 mg Al/L) (Fig. 6). The lowest residual turbidity is observed at pH 7.0 and the optimal pH range for turbidity removal is wide, while the best removal rate of DOM is discovered at pH 9.0 and the optimal pH range for DOM removal is narrow. In weakly alkaline surface water, Al_a will tend to hydrolyze and form Al_b and Al_c, which enhances the bridge and adsorption of hydroxy-aluminum. However, DOC concentration and UV_{254} only reduce to 4.16 mg/L and 0.075 cm^{-1} at the optimal pH 9.0. It is not economically feasible in the field application.

3.3.2. The performance of composite coagulants

Two composite coagulants, PAC-PDMDAAC and PAC-PAM, have also been applied to improve the treatment of DOM at the optimal pH 9.0. But until then, PDMDAAC and PAM as the comparison are

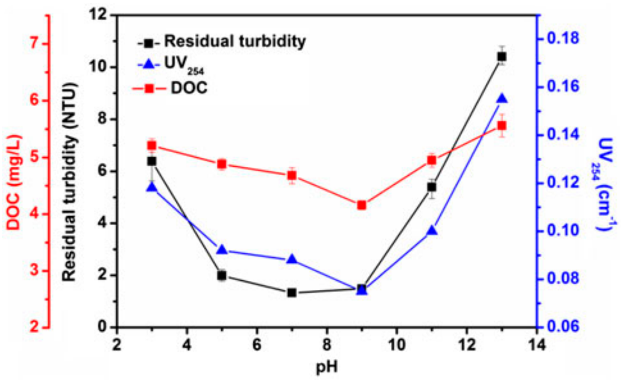

Fig. 6. Residual turbidity, UV_{254}, and DOC concentration of surface water after being treated by moderate $PAC_{2.0}$ under the varied pH.

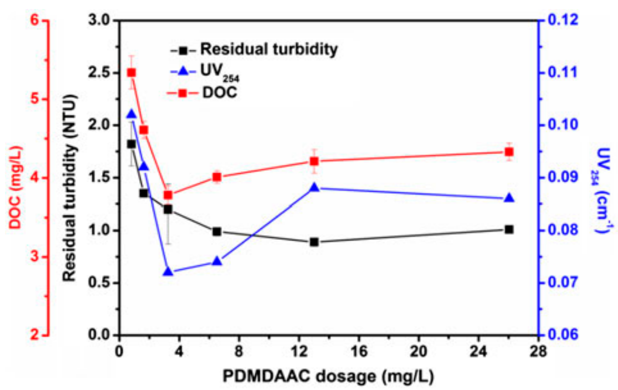

Fig. 7. Residual turbidity, UV_{254}, and DOC concentration of surface water after being treated by a composite coagulant of PAC-PDMDAAC at pH 9.0.

separately used to treat the algae-polluted surface water, and both of them result in the lower removal rates of DOM than $PAC_{2.0}$ (Table S3). It confirms that the adsorption of $Al(OH)_{3(am)}/Al_b$ contributes to DOM removal and the important role of co-flocculation in DOM treatment according to the turbidity removal rate. Besides, PAC-PAM also fails to improve DOM removal (Table S3). Only PAC-PDMDAAC makes better performance (Fig. 7). In this case, the dosage of PAC is always 40 mg Al/L. Residual turbidity, DOC concentration, and UV_{254} in the treated water decrease firstly and then increase with the increase in PDMDAAC dosage. The best DOM removal rate is observed at the PDMDAAC dosage of 3.25 mg/L. The possible removal mechanism of DOM is speculated that overdosing PAC forms large amount of $Al(OH)_{3(am)}$ which adsorbs and mesh DOM on the surface. And PDMDAAC neutralizes the surface charge of colloid particles and bridges these colloid particles to form the large flocs. DOM is removed with flocs. Although DOC concentration and UV_{254} reduce to 3.78 mg/L and 0.072, this method is of low efficiency for a large amount of PAC is added in the treatment. Therefore, the more efficient treatment method of DOM should be studied in future.

4. Conclusions

DOM is discovered in the algae-polluted Minzhu Lake. And PAC prepared by fresh aluminum hydroxide gels is applied in the surface water treatment. Algae and colloid particles are easily removed by PAC. The turbidity removal rate is able to reach 90% and the residual turbidity is below one NTU in most cases. However, the removal rate of DOM is always low. The lowest DOC concentration in the surface water is 4.28 mg/L after being treated by $PAC_{2.0}$. DOM is comprised of aromatic-like, tryptophan-like, and fulvic-like organic substances. Some aromatic-like organic substances are removed via coagulation, while the tryptophan-like and fulvic-like organic substances are still found in the treated water. There is a positive correlation between the removal of DOM and hydrophobic organic compounds. Co-flocculation of DOM with algae and other colloid particles should play a key role in the DOM treatment. And the bridging, adsorption, and entrapment of Al_b, $Al(OH)_{3(am)}$ and flocs are indispensable. To enhance DOM removal, pH of surface water is adjusted to 9.0 and a composite coagulant of PAC-PDMDAAC is used. Although the removal rates of DOM are slightly increased, the methods are not cost-effective.

Supplementary material

The supplementary material for this paper is available online at http://dx.doi.org.10.1080/19443994.2016.1152510.

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Abbreviations

DOM	—	dissolved organic matter
DBPs	—	disinfection byproducts
PAC	—	polyaluminum chloride
Al_a	—	mononuclear aluminum
Al_b	—	small/middle polymeric aluminum
Al_c	—	high polymeric or colloidal aluminum
DOC	—	dissolved organic carbon
UV	—	ultraviolet
SUVA	—	specific ultraviolet absorbance
FEEM	—	fluorescence excitation-emission matrix
PDMDAAC	—	poly dimethyl diallyl ammonium chloride
PAM	—	polyacrylamide

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