

57 (2016) 18619–18629 August



# Multiple views of biological stability and optimized coagulation in the control of biostability in traditional water treatment processes: a pilot test

### Jin Wang, Huaqiang Chu\*, Bingzhi Dong

State Key Laboratory of Pollution Control and Resource Reuse, Tongji University, Shanghai 200092, China, Tel. +86 21 65982691; email: jinwang0126@hotmail.com (J. Wang), Tel./Fax: +86 21 65985811; emails: chuhuaqiang@tongji.edu.cn (H. Chu), dbz77@tongji.edu.cn (B. Dong)

Received 6 September 2014; Accepted 2 September 2015

#### ABSTRACT

This study investigated biological stability in a pilot test of traditional water treatment processes in eastern China. The variations of assimilable organic carbon, biodegradable dissolved organic carbon, bacterial regrowth potential, and total phosphorus were analyzed, and the relationships among them were studied. High-purity aluminum sulfate was added as a coagulant, and the corresponding removal efficiencies of the coagulation–sedimentation unit and the sand filtration unit were evaluated. The experimental results indicate that the removal efficiencies of the dissolved organic carbon (DOC) and  $UV_{254}$  both increased as the alum dosages increased in the coagulation and sedimentation process. The removal efficiencies of the filtration treatment. The bacterial regrowth potential was correlated with DOC concentration, and the introduction of total dissolved phosphorus into the evaluation of biostability is recommended to help confirm the C:P ratio and to select the appropriate strategy for controlling bacterial regrowth. An explanation for the changes in biostability indicators is provided.

*Keywords:* Biostability; Assimilable organic carbon; Biodegradable dissolved organic carbon; Bacterial regrowth potential; Total phosphorus

#### 1. Introduction

Bacterial regrowth in water distribution systems is commonly recognized as a cause of a wide range of problems, such as deterioration of water quality, increased risk of pipeline leakage, and endangerment of human health. To address these hazards, the biological stability of treated drinking water has been researched in the field of water treatment globally.

The extensive research in this field, which has been conducted at multiple levels, has provided a

significant amount of insight into the causes, enabling conditions, and indicators of bacterial regrowth. The concentration of assimilable organic carbon (AOC) is considered as the main indicator of the regrowth of micro-organisms. Meanwhile, biodegradable dissolved organic carbon (BDOC), which is mineralized (to CO<sub>2</sub>) and assimilated (into biomass), has been identified as an indicator for addressing reductions in chlorine demand or for the formation of disinfection byproducts [1,2]. Instead of focusing on organic carbon, several other indicators, such as bacterial regrowth potential (BRP), have been introduced based on the

<sup>\*</sup>Corresponding author.

<sup>1944-3994/1944-3986 © 2015</sup> Balaban Desalination Publications. All rights reserved.

presumption that inorganic nutrients might be a limiting factor for bacterial regrowth [3].

Natural organic matter (NOM), which is defined as a complex matrix of organic material that is present in natural water, is composed of two fractions: biodegradable matter and refractory matter. Humic substances are generally defined as refractory dissolved organic carbon (RDOC). Although the coagulation and sedimentation (C and S) process is generally designed for particle and turbidity treatment, it can be used to remove organic matter in treatment plants that must treat highly colored water [4]. Several studies have reported contradictory removal efficiencies of biodegradable organics by different coagulants. Three coagulants, poly aluminum chloride (PACl), aluminum potassium disulfate dodecahydrate (alum), and ferric chloride (FeCl<sub>3</sub>), were used to treat water samples which achieved mean BDOC removal rates of 33, 31, and 30%, respectively [4]. Similarly, Edzwald [5] reported that PACl had the same efficiency as alum and FeCl<sub>3</sub> in removing total organic carbon. In contrast, Croue et al. [6] documented a higher removal rate of BDOC when applying ferric chloride compared to alum. Amy [7] found that enhanced coagulation was able to achieve 50% dissolved organic carbon (DOC) removal. In comparison, enhanced and optimized coagulation resulted in an additional BDOC reduction of 8%, which is not a significant improvement. Filtration (FLTR) has also been used to remove impurities, and its effectiveness is a strong function of biological removal, especially in slowly flowing water that is subject to growth of biofilms [8].

The removal of AOC has been reported to be correlated with decreases in the concentrations of other drinking water indicators [9]. Escobar and Randall [1] found that AOC causes the improvement in the bacterial regrowth potential achieved by nanofiltration to be underestimated; however, BDOC caused the improvement to be overestimated. Hence, the collection of complementary information by measuring both AOC and BDOC is advised. The vast majority of previous studies have focused on biologically available

Table 1 Raw water quality in pilot-scale tests

organic carbon (AOC and BDOC), and a few studies have focused on inorganic nutrients. However, a comprehensive examination of their combined influence on the control of biological stability that integrates the roles of both organic and inorganic nutrients using various indicators has not been performed.

The objective of this study is to develop a comprehensive understanding of the control of biological stability in traditional treatment processes and to investigate the interrelationships between various indicators. We also attempt to optimize the alum dosage to control bacterial regrowth.

#### 2. Materials and methods

#### 2.1. Raw water quality

Raw water was transported approximately 27 km from the Lake Tai and was fed into the pilot test and into the water plant. The quality of the raw water is summarized in Table 1. Peak A (Ex = 230 nm, Em = 340 nm) and Peak B (Ex = 280 nm, Em = 320 nm) can be easily identified in the three-dimensional fluorescence spectrum of the raw water. Hence, the raw water was rich in proteins with aromatic structures and soluble microbial products, including several low molecular weight amino acids that can be effectively used by bacteria (strains P17 and NOX) for AOC analyses. In addition, several types of humic-like components are evident from the broad response areas in other regions.

#### 2.2. Pilot setup

A continuous, pilot-scale test, including coagulation, sedimentation, and sand filtration, was conducted at the Xiang Cheng water plant in Suzhou, China from 2009 to 2013. A comprehensive assessment was performed in July and August 2013 after the pilot system had achieved a steady operation. The pilot system was operated at a design flow of  $3 \text{ m}^3$ /h. The main operational parameters are summarized in

Coag dose (mg/l (ppm))	Turb (NTU)	T (°C)	pН	Alkalinity (mg/l)	NH <sub>3</sub> -N (mg/l)	COD <sub>Mn</sub> (mg/l)	DOC (mg/l)	UV <sub>254</sub> (cm <sup>-1</sup> )
10	$9.5 \pm 0.2$	$26.8 \pm 0.1$	$8.51 \pm 0.09$	57 ± 3	$0.07 \pm 0.01$	$2.98 \pm 0.02$	$3.62 \pm 0.04$	$0.066 \pm 0.002$
30 50	$5.7 \pm 0.1$ $5.0 \pm 0.2$	$26.9 \pm 0.1$ $26.9 \pm 0.1$	$8.58 \pm 0.07$ $8.77 \pm 0.05$	$49 \pm 4$ $49 \pm 3$	$0.03 \pm 0.00$ $0.03 \pm 0.00$	$2.94 \pm 0.01$ $2.90 \pm 0.01$	$3.63 \pm 0.02$ $3.61 \pm 0.03$	$0.067 \pm 0.002$ $0.064 \pm 0.001$
70	$3.0 \pm 0.2$ $2.7 \pm 0.1$	$20.9 \pm 0.1$ $27.5 \pm 0.2$	$8.38 \pm 0.13$	$49 \pm 3$ $48 \pm 2$	< 0.02	$2.90 \pm 0.01$ $2.98 \pm 0.01$	$3.50 \pm 0.03$	$0.064 \pm 0.001$ $0.064 \pm 0.001$
90	$2.6\pm0.1$	$27.5\pm0.2$	$8.36 \pm 0.04$	$48 \pm 3$	< 0.02	$2.98 \pm 0.01$	$3.46\pm0.03$	$0.064 \pm 0.001$

Table 2	
Process	parameters

Process	Parameter	Note
Coagulation	Coagulant: Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> 10–90 mg/l	Size: $L \times B \times H = 800 \text{ mm} \times 1,200 \text{ mm} \times 1,200 \text{ mm}$ Retention time: 23 min
Sedimentation	Tube settler, clean water area 0.4 m <sup>2</sup>	Clean water region plan size: $L \times B = 500 \text{ mm} \times 825 \text{ mm}$ Heights: clean water region = 1.2 m, tube area = 0.866 m, sludge discharge region = 1.6 m
Filtration	Sand media Speed = 8 m/h, period = 24 h	Size: $D \times H = 700 \text{ mm} \times 3,250 \text{ mm}$ Sand: $d_{10} = 0.94 \text{ mm}$ , $d_{60} = 1.26 \text{ mm}$ , $K_{80} = 1.51$ , $\rho = 2.62 \text{ g/cm}^3$ , $H = 1,250 \text{ mm}$

Table 2. The alum dosages were 10, 30, 50, 70, and 90 ppm. Each dosage was run for one week, and samples of the raw water, the coagulating sedimentation process, and the sand filtration process were collected five times. All of the biostability indicators (e.g. AOC, BDOC, BRP) were tested immediately after the samples were collected. The pilot-scale test system was backwashed every two days.

#### 2.3. Coagulant parameters

High-purity alum used as the coagulant was manufactured by the Light Industry Auxiliary Factory (Jiangsu Province, China). The characteristics of the alum were measured twice; the detailed parameters as well as the related Chinese standards [10] are listed in Table 3.

#### 2.4. Analytical methods

#### 2.4.1. Water sample collection

As shown in Fig. 1, C and S and FLTR water samples were collected one hour after the pilot system had reached the stable operation. Samples of raw water and effluent were collected for direct measurements of temperature, pH, total phosphorus (TP), turbidity, and UV<sub>254</sub>. The samples were filtered

through a 0.45-µm membrane prior to AOC, BDOC, BRP, and DOC analyses.

#### 2.4.2. AOC measurement

AOC was measured using the method of Van der Kooij. In this approach, two strains of bacteria, Pseudomonas fluorescens strain P17 (ATCC 49642) and Spirillum species strain NOX (ATCC 49643), were used for biometric testing. First, the water samples were sterilized at 70°C for 30 min. After the samples had cooled to room temperature, they were inoculated with approximately 10<sup>4</sup> CFU/ml P17 and cultivated at 25°C for 3 d. The samples were then extracted and spread onto the appropriate medium to measure the number of bacterial colonies. The culture medium was plate count agar because of its applicability in enumerating bacteria. Then, the same samples were sterilized again and inoculated with approximately 10<sup>4</sup> CFU/ml NOX. After cultivation at 25°C for 4 d, the numbers of colony-forming units were measured. Because the traditional method of simultaneously inoculating with P17 and NOX may result in interference when the colonyforming units are counted, a successive inoculation method was applied in this study. In addition, extra days were included in the cultivation period because the AOC concentration is a product of the largest

Table 3

Main characteristics of the alum and Chemical Industry Standard of the People's Republic of China (water treatment chemicals—aluminum sulfate)

Parameter		Alum-test 1	Alum-test 2	Standard [10] (Class I: liquid)
$\overline{\text{Al}_2\text{O}_3}^a$	≥	10.06	8.29	7.8
pH (1% aqueous solution)	≥	4.08	3.57	3.0
Fe <sup>a</sup>	$\leq$	0.05	< 0.01	0.25
$\rho (g/cm^3)$		1.30	1.32	b

<sup>a</sup>Mass fraction in %.

<sup>b</sup>Not given.

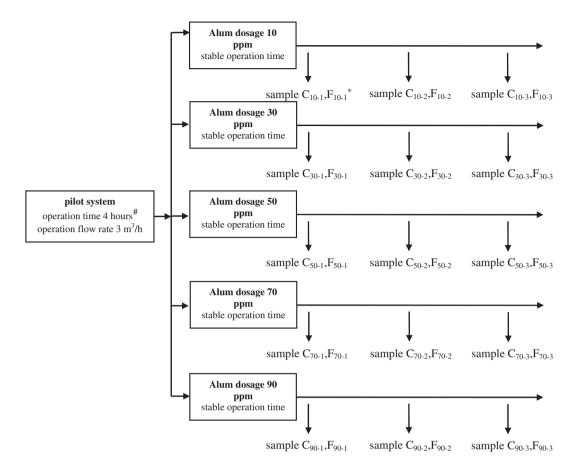


Fig. 1. Process schematic for water sample collection.

Note: "The pilot system is considered to reach the stable operation when operating for 4 h. From then on, the extra operation time is counted as stable operation time;  $*C_{10-1}$  and  $F_{10-1}$  stand for sample 1 at 10 ppm dosage of C and S process and FLTR process, respectively.

colony-forming units. The AOC concentration was calculated; the units are micrograms acetate-C per liter [11]. The yield factors for P17 and NOX are  $6.33 \times 10^6$  and  $2.21 \times 10^6$  CFU/µg acetate-C, respectively.

#### 2.4.3. BDOC measurement

The BDOC analysis used the indigenous bacteria in the raw water as the inoculum. The inoculum was collected by filtering the raw water through a 2-µm membrane. First, the water samples were filtered through a 0.45-µm membrane. Then, a portion of the filtered water was subsampled to measure the DOC (DOC<sub>0</sub>) concentration. The remaining filtered water was inoculated at a volume ratio of 100:1 and then cultivated at 20°C for 28 d. After cultivation, the remaining water samples were filtered and the DOC concentration (DOC<sub>28</sub>) was measured. The BDOC concentration was calculated as the loss of DOC (BDOC = DOC<sub>0</sub> – DOC<sub>28</sub>). Thus, the BDOC represents the fraction of DOC that is either mineralized or assimilated by the heterotrophic flora.

#### 2.4.4. BRP measurement

BRP measurements were performed using the method reported by Sathasvian and Ohgaki [3] with a minor modification. In this approach, the indigenous bacteria were used as the inoculum for the BRP test. The indigenous bacteria were obtained by filtering raw water through a 2-µm membrane and then cultivating the samples at 20°C for 5 d to allow for adjustment to the oligotrophic environment. After sterilization at 70°C for 30 min, a 40-ml water sample was mixed with 400 µl of inoculum (volume ratio of 100:1). After a seven-day incubation at 20°C using R2A agar as the culture medium, the number of colony-forming units was counted. The culture time on the R2A agar was seven days at 25°C to ensure an integrated count. The growth potential was calculated from the direct count without subtracting the number of bacteria in the inoculum because of the inoculum size [12].

#### 2.4.5. Other analyses

The DOC concentrations were measured using a wet oxidation TOC analyzer (O.I. Analytical Aurora 1030). The  $UV_{254}$  was determined using a UV2100

spectrophotometer (Unico (Shanghai) Instrument). The SUVA<sub>254</sub> values, which indicate the proportion of humic material, were calculated by determining the ratio of UV254 to DOC.  $COD_{Mn}$ , NH<sub>3</sub>, and TP were measured according to the Chinese State Environmental Protection Agency (SEPA) Standard Methods (Chinese SEPA, 2002). The turbidities of the influent and effluent were measured using a turbidity meter (2100 N, Hach, USA).

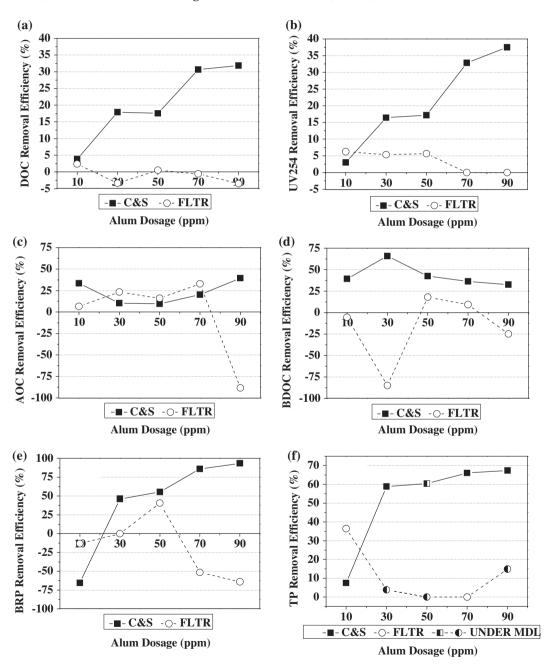


Fig. 2. Effect of high-purity aluminum sulfate on the removal efficiencies of the biological stability indices (C&S stands for coagulation and sedimentation process, FLTR stands for filtration process, UNDER MDL stands for under method detection limit).

#### 3. Results and discussion

#### 3.1. Organic matter removal

Fig. 2 shows the mean removal efficiencies of biological stability-related indices for five different alum dosages in the conventional treatment process in July 2013. Three water samples were taken and tested at different processes of different alum dosages. The removal efficiencies of DOC and UV<sub>254</sub> increased as the alum dosages increased during the C and S process. The DOC and UV<sub>254</sub> increased from approximately 3 to 17% as the alum dosages increased from 10 to 30 ppm, reached a plateau at 50 ppm, then increased to slightly greater than 30% at a dosage of 70 ppm followed by a modest increase at 90 ppm. The DOC removal efficiency was closely related to that of UV<sub>254</sub> in the C and S process. This relationship indicates that the aluminum sulfate did not exhibit any specific selectivity in terms of removing dissolved organics and humic-like components with high molecular weights, including various aromatic compounds.

As shown in Fig. 2, FLTR process had little effect on DOC and  $UV_{254}$  removal. Under certain circumstances, the removal of DOC was less than 0%, which is most likely attributed to the accumulation of organics in the sand filter. Similarly, the  $UV_{254}$  removal remained at 5% when the dosages were less than 50 ppm. Compared with the increased removal efficiencies of the C and S process, the removal of  $UV_{254}$ in the FLTR process decreased to 0% (70 and 90 ppm). Hence, the FLTR process provides only a limited contribution to the removal of organic matters, though it plays an important role in controlling the turbidity (almost all of the samples were under 0.3 NTU after FLTR) of the effluent.

## 3.2. Controls of biological stability indices and internal relationships

The AOC is an important parameter that measures the biological stability in a distribution system. It refers to the most readily degradable fraction, which tends to be composed of low molecular weight compounds that generally contribute a relatively small proportion of the BDOC [1,4]. Because the AOC assay involved two strains of bacteria, P17 and NOX, the concentration of AOC contains two fractions (AOC-P17 and AOC-NOX). AOC-P17 consists of several organic components, including carboxylic acids, amino acids, alcohols, and various carbohydrates. In contrast, AOC–NOX contains formic acid, glyoxylic acid, oxalic acid, various carboxylic acids, and a few amino acids. The removals of AOC for each alum dosage are shown in Table 4.

Fig. 2 shows that alum sulfate was effective for AOC removal during the C and S process. The mean AOC concentrations of the raw water and the sand filtration effluent for different dosages of alum were 75 and 57 µg/L acetate-C, respectively. The removal rate increased gradually from 10% at 30 ppm to 39% at 90 ppm. The finding that the removal remained constant between 30 and 50 ppm may indicate that a higher dosage was needed for a higher efficiency. Moreover, the removal of AOC-NOX in the coagulation-sedimentation process is generally less efficient than the removal of AOC-P17 [13]. A similar result was observed in this study; the removal of AOC-P17 exceeded that of AOC-NOX at high dosages  $(\geq 50 \text{ ppm})$  and the high removal of AOC was mainly attributed to the elimination of AOC-P17. Overall, the mean AOC removal efficiency during the C and S process was 23%, which is slightly greater than that reported by Owen et al. [14] for alum coagulation (16%). Treatment with alum could also be more effective than nanofiltration [1]. Several researchers have reported that ferric chloride has a high removal efficiency (38%) for AOC control [15]. However, because the raw water quality varies by location, this suggested treatment method requires further investigation. A generally upward trend of AOC removal was also observed for the FLTR process except for the 90 ppm case. The decomposition of organic matter by micro-organisms in the biofilm as the water slowly passes through the sand filter bed causes the large reduction in AOC [8].

The BDOC concentration represents the fraction of DOC that is assimilated or mineralized by the heterotrophic flora [16]. The BDOC is calculated as the difference between the initial DOC value and the minimum DOC value that is observed during the incubation period [17]. Volk et al. [17] suggested that BDOC concentrations of 0.15 mg/L at 20°C and 0.30 mg/L at 15°C are required for biostability. The method detection limit (MDL) for BDOC testing was determined by the TOC measurement technique to be 0.15 mg/L [18]. The mean BDOC/DOC ratio for the raw water samples in this study was 12%, which is consistent with the results of Joret et al. [19] that BDOC values represent 10-30% of the total dissolved organic carbon content in drinking water. The DOC that remains after biodegradation is defined as the refractory (non-biodegradable) dissolved organic carbon (RDOC). The mean BDOC concentrations for the raw water, the C and S effluent, and the FLTR effluent were 0.41, 0.24, and 0.27 mg/L, respectively. The mean BDOC removal rate for the C and S process by the addition of high-purity aluminum sulfate was 43%, while the FLTR process had a -18% reduction

Parameter		Parameter AOC Fraction percentage Remo			Fraction percentage	ercentage	Removal	Removal efficiency		Other factors		
Alum dosage (ppm)	Unit	P17 (μg acetate-C/L)	$ \begin{array}{lll} P17 \ (\mu g \\ acetate-C/L) \end{array} \begin{array}{ll} NOX \ (\mu g \\ acetate-C/L) \end{array} $	Total (μg acetate-C/L)	P17/ NOX/ AOC (%) AOC (%) P17 (%) NOX (%) AOC (%)	NOX/ AOC (%)	P17 (%)	(%) XON		SUVA (L/mg·m)	Turb (NTU) pH (-)	(-) Hq
10	RAW	$28.06 \pm 5.29$	$22.21 \pm 0.64$	$50.27 \pm 5.93$	$56 \pm 4$	$44 \pm 4$	I	1	I	1.82	9.5	8.51
	C and S	$24.26 \pm 6.65$	$9.16 \pm 2.27$	$33.42 \pm 8.92$	$73 \pm 1$	$27 \pm 1$	13.55	58.76	33.53	1.84	8.7	7.97
	FLTR	$25.00 \pm 12.56$	6.23 ±	$31.23 \pm 13.36$		$21 \pm 7$		31.96	6.54	1.77	4.5	7.90
30	RAW	$66.85 \pm 13.40$ 14.05 $\pm$	$14.05 \pm 1.29$	$80.90 \pm 14.68$	82 ± 2	18 ± 2	I	I	I	1.80	5.7	8.58
	C and S	$61.42 \pm 5.72$	$11.11 \pm 0.32$	$72.53 \pm 5.40$	$85 \pm 2$	$15 \pm 2$	8.12	20.95	10.35	1.83	1.0	7.87
	FLTR	$46.77 \pm 12.99$	$8.84 \pm 3.74$	$55.61 \pm 16.73$	$84 \pm 2$	$16 \pm 2$	23.85	20.41	23.32	1.67	0.3	7.72
50	RAW	$75.08 \pm 11.69$	$10.77 \pm 2.72$	$85.84 \pm 8.97$	+ 5	$13 \pm 5$	I	I	I	1.82	5.0	8.77
	C and S	$57.00 \pm 7.67$	$20.55 \pm 1.14$	$77.55 \pm 6.18$	$73 \pm 4$	± 4	24.08	-90.89	9.66	1.83	0.5	7.64
	FLTR	$44.51 \pm 9.71$	$20.63 \pm 1.28$	$65.14 \pm 8.43$	68 ± 6	32 ± 6	21.91	-0.36	16.01	1.74	0.2	7.55
70	RAW	$64.94 \pm 39.16$	$8.93 \pm 0.67$	$73.87 \pm 39.83$	86 ± 7	$14 \pm 7$	I	I	I	1.83	2.7	8.33
	C and S	$36.13 \pm 0.47$	$22.78 \pm 10.10$	$58.91 \pm 9.63$	$62 \pm 11$	$38 \pm 11$	44.36	-154.97	20.26	1.77	0.3	7.11
	FLTR	$24.91 \pm 2.75$	$14.66 \pm 9.83$	$39.56 \pm 7.08$	$65 \pm 19$	35 ± 19	31.06	35.66	32.84	1.76	0.3	7.07
06	RAW	74.22 ± 38.70 8.39 ±	$8.39 \pm 0.96$	82.60 ± 37.74	88 ± 6	12 ± 6	I	I	I	1.85	2.6	8.36
	C and S	$39.68 \pm 0.47$	$10.31 \pm 0.48$	$49.99 \pm 0.96$	$79 \pm 1$	$21 \pm 1$	46.54	-22.97	39.48	1.70	0.5	7.11
	FLTR	$36.81 \pm 1.06$	$57.34 \pm 5.45$	$94.16 \pm 4.39$	$39 \pm 3$	$61 \pm 3$	7.21	-456.04	–88.36 <sup>a</sup>	1.64	0.2	7.08
Notes: RAW = 1 <sup>a</sup> The high negat	aw water, C ai ive removal m	Notes: RAW = raw water, C and S = coagulation and sedimentation process, FLTR = sand filtration process, and $-$ = no removal efficiency. <sup>a</sup> The high negative removal might be attributed to the accumulation of organic matter in the sand filter.	n and sedimenta to the accumula	tion process, FL ation of organic 1	TR = sand filt matter in the	ration proce sand filter.	ss, and – =	no removal	efficiency.			

Table 4 Removal of AOC and its fractions with different alum dosages in the traditional treatment process

18625

(Table 4). Several studies have examined the efficiency of BDOC removal during the coagulation process when using various coagulants. Large variations in BDOC removal (between 0 and 74%) have been observed between different water sources [4].

The BDOC removal ranged from 33 to 66% (Table 5). In comparison, the reduction in BDOC at the St. Rose water treatment plant (Canada) ranged from 50 to 86% when alum was used. These conflicting results may be attributed to the specific physicochemical characteristics of the different raw water samples, the test conditions and the running status of the units that treated the water samples. As shown in Fig. 3, the ratio of BDOC removed (mg) to DOC removed (mg) decreased as the alum dosage increased. This relationship indicates that RDOC was easier to eliminate by coagulation than BDOC [4], which is consistent with previous studies in which humic substances were found to be preferentially targeted by the coagulation process [5,7]. Specifically, because the initial DOC removal was entirely attributed to BDOC removal with a moderate level of removal at a low alum dosage, which represents conditions under which a significant amount of AOC could also be removed (34% at 10 ppm), this scenario may be conducive to the development of a new method of pretreatment that could reduce the biological stability load using a low dosage of coagulant.

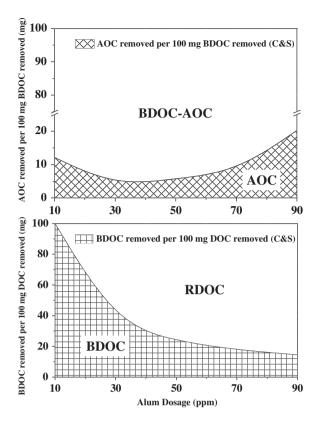


Fig. 3. Removal of AOC and BDOC during the C and S process with different alum dosages.

Table 5
Removal efficiencies for various biological stability indices in the traditional treatment process

Parameter Alum dosage (ppm)	Unit	BRP (cfu/ml)	BRP (removal %)	X (C:P = 100:X) <sup>a</sup> (-)
10	RAW	82,500	_	10.05
	C and S	136,500	-65	15.30
	FLTR	154,500	-13	9.22
30	RAW	108,500	_	7.18
	C and S	58,500	46	8.67
	FLTR	58,583	0	4.50
50	RAW	86,750	_	7.45
	C and S	38,750	55	5.13
	FLTR	23,000	41	6.25
70	RAW	117,500	_	5.86
	C and S	16,500	86	3.12
	FLTR	25,000	-52	3.44
90	RAW	379,000	_	7.30
	C and S	25,000	93	3.53
	FLTR	41,000	-64	2.40

<sup>a</sup>C:P stands for the bacterial requirement and is described as BDOC:TP in this study. X is equal to 100 P/C.

Obviously, additional studies are required, and other raw water sources should be tested. The results shown in Fig. 3 indicate that the removed AOC only contributed a small fraction of the total BDOC removed, which is consistent with the results of Tryby et al. [20] that the reduction rates of hydrophobic compounds surpassed those of hydrophilic compounds.

The removal of BDOC during the FLTR process ranged between -85 and 18% (Fig. 2(d)). The mean BDOC/DOC ratio in the raw water was 12%, which is consistent with the BDOC/DOC ratios in the range of 10-30% that were reported by Joret et al. [19]. The C and S process achieved high BDOC removal (32.59-65.91%) and irregular fluctuations occurred as the amount of added coagulant was increased. The highest BDOC removal efficiency (65.91%) was obtained with the coagulant addition of 30 mg/L coagulant. The BDOC removal by the sand filtration process ranged from -85 to 17.95%; the large variation contrasted with that observed for the C and S process (Fig. 2). The amount of BDOC removal by the sand filtration process was closely related to the BDOC concentration that was treated by the sedimentation process, which facilitates the stable quality of treated water after the entire treatment process. In this experiment, BDOC removal by the typical treatment process with the addition of Al<sub>2</sub>(SO4)<sub>3</sub> coagulant ranged from 15.79 to 52.80% and the mean removal efficiency was 36.74%.

As one of the most essential inorganic elements for bacterial growth, low concentrations of phosphorus can restrict microbial growth when organic matter supplies are sufficient [3,21]. Hence, inorganic nutrients such as phosphorus should also be considered when measuring the level of biological stability. The BRP method was reported by Sathasivan and Ohgaki [3]. This method considers the nutrients (including those from autoclaved cells) that can be utilized for bacterial regrowth in the original water sample [3]. Fig. 4 shows the relationship between DOC and BRP in the raw water and C and S units; the two parameters are correlated, while there is a relatively weak association ( $R^2 = 0.6874$ ) between TP and BRP. Hence, in addition to the previous conclusion, the modest difference may indicate that the ratio of concentrations of DOC and phosphorus is similar to the bacterial requirement (C:P = 100:1.7-2). However, it should also be emphasized that the actual concentration of phosphorus that is available for microbial growth is lower than the TP value. Therefore, total dissolved phosphorus (TDP) (measured after filtering the water samples through a 0.45-µm membrane before performing the normal TP measurement procedure) should be considered when calculating the C:P ratio to obtain more relevant measurements. Fig. 2 shows that the reduction

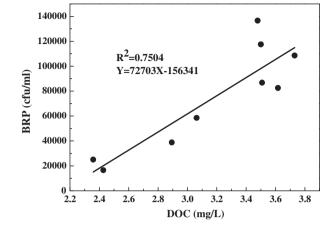


Fig. 4. Association between DOC and BRP in the RAW and C and S units.

in AOC was accompanied by an increase in the BRP at the 10 ppm alum dosage. Similarly, Ryu et al. [22] and Thayanukul et al. reported that the microbial regrowth (an increase in the HPC) that was observed in their studied distribution systems occurred in parallel with AOC reductions.

The reduction in BRP was as high as 93% for the 90 ppm dosage at the highest DOC removal and TP removal conditions. The FLTR process had unstable removal efficiency for BRP elimination. In contrast, the TP removal by the FLTR process was relatively stable and was almost always less than 15% because the C and S process had already removed the majority of the phosphorus (Fig. 2).

The results shown in Fig. 2 and Table 5 demonstrate that the sand filtration process could disturb the removal of organic matter, mainly biodegradable organic matter such as hydrophilic substances, and cause increases in the DOC, BDOC, and AOC concentrations. However, it is notable that no negative reductions in the  $UV_{254}$  and TP removal values were observed, which indicates that the sand filtration process did not introduce additional humic or phosphorous contaminants into the effluent.

### 3.3. Optimization of the alum sulfate dosage to control bacterial regrowth

Based on the results for the five dosage levels and ten different indices, the effect of adding alum sulfate to the C and S process to maintain the biological stability of the effluent has been accurately estimated. Additionally, the removal efficiencies of the organic compounds and nutrients that are required for bacterial regrowth have been calculated. Although the representativeness of the calculated removal efficiencies is limited by the raw water quality and the operational conditions of the units, these data provide a specific model (based on the low DOC, low SUVA, and low-alkalinity raw water in this pilot test) of the changes in the alum sulfate coagulation removal efficiencies of biostability indicators and also explain the mechanisms of interaction between the biostability parameters. It is clear that the subsequent increase in the removal efficiency of DOC, UV<sub>254</sub>, and TP at dosages above 70 ppm was limited (Fig. 2). Thus, the increase in the BRP (which synthetically measures the influence of various organics and nutrients on overall microbial regrowth) reduction was also limited. The AOC removal efficiency increased continuously at dosages above 50 ppm. However, a corresponding decrease in the BDOC removal efficiency was observed at the same dosages. Because a portion of the BDOC also represents the AOC-formation potential [1], an additional increase in the dosage might eventually lead to a limited increase or even a modest decline in AOC removal. The results presented in Table 4 demonstrate that almost all of the filtration effluents except the 10 ppm dosage satisfy the turbidity removal criteria. In conclusion, the 70 ppm dosage of high-purity alum (used as a coagulant) is believed to have reached the point of diminishing returns in terms of the functioning of the C and S process during this pilot test.

#### 4. Conclusions

This study observed variations in the removal of natural organic matter and inorganic elements that are available for microbial regrowth that could be attributable to their preferential removal by alum sulfate, the water quality matrices, and the operational status of the treatment unit. As the alum dosages in the C and S process increased, the removal efficiencies of DOC and  $UV_{254}$  increased. Generally, the C and S process is much more effective than the FLTR process at controlling the biostability indices, including DOC, BDOC, AOC, BRP, and TP.

Overall, the traditional treatment process was shown to be beneficial for enhancing the biological stability. It provides a potential model, which is associated with a specific raw water quality, to describe the changes in the reductions in various biostability indices. The results also indicate that the optimization of the concentration of alum coagulant is closely related to the raw water quality. Therefore, the optimal control of biological stability during the C and S and filtration processes should allow for the control of the AOC index as well as the BDOC and bacterial regrowth potential indices.

#### Acknowledgment

This work was supported by the National Water Pollution Control and Treatment Key Technologies R&D Program of China (2011ZX07410-002-1).

#### References

- I.C. Escobar, A.A. Randall, Assimilable organic carbon (AOC) and biodegradable dissolved organic carbon (BDOC), Water Res. 35 (2001) 4444–4454.
- [2] L.A. Kaplan, D.J. Reasoner, E.W. Rice, A survey of bom in United States drinking waters, J. Am. Water Works Assoc. 86 (1994) 121–132.
- [3] A. Sathasivan, S. Ohgaki, Application of new bacterial regrowth potential method for water distribution system—A clear evidence of phosphorus limitation, Water Res. 33 (1999) 137–144.
- [4] C. Volk, K. Bell, E. Ibrahim, D. Verges, G. Amy, M. Lechevallier, Impact of enhanced and optimized coagulation on removal of organic matter and its biodegradable fraction in drinking water, Water Res. 34 (2000) 3247–3257.
- [5] J.K. Edzwald, Coagulation concepts for removal of TOC, in: AWWA WQTC Conf., San Francisco, 1994.
- [6] J.P. Croue, B. Martin Mousset, E. Lefebvre, A. Deguin, B. Legube, Evolution of the humic/non humic natural organic matter distribution and its biodegradable fraction during drinking water treatments, Revue. Des. Sci. De L'Eau. 10 (1997) 63–81.
- [7] G.L. Amy, Using NOM characterization for the evaluation of treatment, in: Workshop on NOM in Drinking Water, Chamonix, France, 1993.
- [8] D. Van Der Kooij, H.R. Veenendaal, Determination of the concentration of easily assimil able organic carbon (AOC) in drinking water with growth measurements using pure bacterial cultures, in: SWE 95.002, KIWA, Nieuwegein, Netherlands, 1995.
- [9] M.W. LeChevallier, Coliform Regrowth in Drinking Water: A Review, J. Am. Water Works Assn. 82 (1990) 74–86.
- [10] The National Development and Reform Commission of the People's Republic of China, Chemical Industry Standard: Water Treatment Chemicals-aluminum Sulfate, PRC, China, 2004.
- [11] D. Van der Kooij, Development and application of methods for assess ing the biostability of drinking water and materials in contact with drinking water, in: The 5th International Workshop on Drinking Water Quality Management and Treatment Technology, Taipei, Taiwan, 1999.
- [12] Y. Yang, S. Takizawa, H. Sakai, M. Murakami, N. Watanabe, Removal of organic matter and phosphate using ferrihydrite for reduction of microbial regrowth potential, Water Sci. Technol. 66 (2012) 1348–1353.
- [13] P.M. Huck, P.M. Fedorak, W.B. Anderson, Formation and removal of assimilable organic carbon during bio-

logical treatment, J. Am. Water Works Assn. 83 (1991) 69-80.

- [14] D.M. Owen, G.L. Amy, Z.K. Chowdhury, Characterization of natural organic matter and its relationship to treatability, AWWAR, Denver, CO, 1993.
- [15] C. Chen, X.J. Zhang, W.J. He, W. Lu, H.D. Han, Comparison of seven kinds of drinking water treatment processes to enhance organic material removal: A pilot test, Sci. Total Environ. 382 (2007) 93–102.
- [16] P.M. Huck, Measurement of biodegradable organic matter and bacterial growth potential in drinking water, J. Am. Water Works Assn. 82 (1990) 78–86.
- [17] C. Volk, C. Renner, C. Robert, J.C. Joret, Comparison of two techniques for measuring biodegradable dissolved organic carbon in water, Environ. Technol. 15 (1994) 545–556.
- [18] I.N. Najm, L. Boulos, M. LeChevallier, C. Norton, C. Volk, A. Randall, I. Escobar, L. Kiene, C. Campos, Case Studies of the Impacts of Treatment Changes on

Biostability in Full Scale Distribution Systems, American Water Works Research Foundation, Denver, CO, 2000.

- [19] J.C. Joret, Y. Levi, C. Volk, Biodegradable dissolved organic carbon (BDOC) content of drinking water and potential regrowth of bacteria, Water Sci. Technol. 24 (1991) 95–101.
- [20] M.E. Tryby, R.J. Miltner, S.R. Scott, TOC removal as a predictor of DBP control with enhanced coagulation, in: AWWA WQTC Conf., Miami, FL, 1993.
- [21] J.S. Vrouwenvelder, F. Beyer, K. Dahmani, N. Hasan, G. Galjaard, J.C. Kruithof, M.C.M. Van Loosdrecht, Phosphate limitation to control biofouling, Water Res. 44 (2010) 3454–3466.
- [22] H. Ryu, A. Alum, M. Abbaszadegan, Microbial characterization and population changes in nonpotable reclaimed water distribution systems, Environ. Sci. Technol. 39 (2005) 8600–8605.