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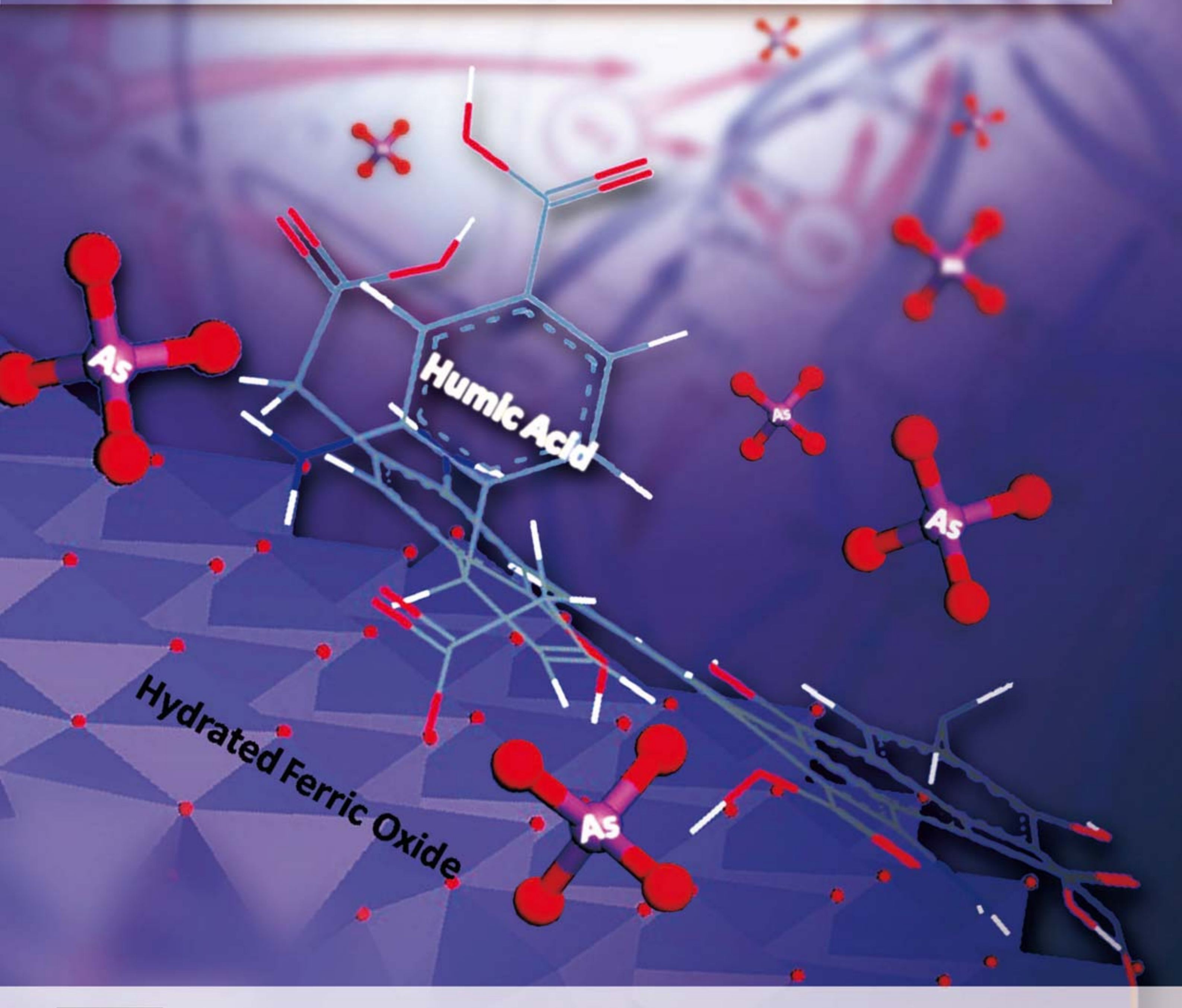

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CONTENTS

Aquatic environment

Atmospheric environment

Environmental health and toxicology

Environmental catalysis and materials

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Biostability in distribution systems in one city in southern China: Characteristics, modeling and control strategy

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ABSTRACT

This study investigated the bacterial regrowth in drinking water distribution systems receiving finished water from an advanced drinking water treatment plant in one city in southern China. Thirteen nodes in two water supply zones with different aged pipelines were selected to monitor water temperature, dissolved oxygen (DO), chloramine residual, assimilable organic carbon (AOC), and heterotrophic plate counts (HPC). Regression and principal component analyses indicated that HPC had a strong correlation with chloramine residual. Based on Chick-Watson's Law and the Monod equation, biostability curves under different conditions were developed to achieve the goal of HPC $\leqslant$ 100 CFU/mL. The biostability curves could interpret the scenario under various AOC concentrations and predict the required chloramine residual concentration under the condition of high AOC level. The simulation was also carried out to predict the scenario with a stricter HPC goal ($\leqslant$ 50 CFU/mL) and determine the required chloramine residual. The biological regrowth control strategy was assessed using biostability curve analysis. The results indicated that maintaining high chloramine residual concentration was the most practical way to achieve the goal of HPC $\leqslant$ 100 CFU/mL. Biostability curves could be a very useful tool for biostability control in distribution systems. This work could provide some new insights towards biostability control in real distribution systems.

Introduction

The qualified finished water from water treatment plants can experience complex chemical, physical and biological changes during transportation in the distribution systems before it reaches the taps. The problems of water deterioration in the networks include chemical instability effects, such as elevated turbidity, color, taste, odor and iron concentration (Rigal and Danjou, 1999; Lehtola et al., 2004; Niu et al., 2006; Husband and Boxall, 2011), as well as biological instability effects, such as bacterial regrowth, nitrification and propagation of protozoa (Sibilie et al., 1998; Lipponen et al., 2002; Chowdhury, 2012; Lu et al., 2013).

The regrowth of microorganisms, especially pathogens, can have large impacts on public health. Non-pathogens can also lead to bio-corrosion and off-flavor. High priority has been given to the control of bacterial regrowth in the USA. and Europe (Ashbolt, 2004; McGuire, 2006). Heterotrophic plate count (HPC) is widely used as an index to evaluate the bacterial regrowth in drinking water distribution systems. Different criteria of HPC have been set, ranging from 100 to 500 CFU/mL (Pavlov et al., 2004).

Generally, there are two approaches available to con-

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trol the bacterial regrowth in distribution systems. First, maintain an adequate disinfectant concentration for better inactivation efficiency. Second, lower the substrate concentration to cut down the food supply of microbes. However, the measures to control bacterial regrowth are quite site-specific. It is hard to design a universal control strategy for all distribution systems. Van der kooij (1992) found a significant correlation between assimilable organic carbon (AOC) and the geometric mean of heterotrophic bacteria in one distribution system in the Netherlands where the chlorine residual in waters was less than 0.1 mg/L. The requirement of HPC < 100 CFU/mL could be satisfied with AOC < 10 μg/L. Lu (2005) revealed that HPC had a good relationship with chloramine residual (r = –0.59, $P < 0.001$) and AOC (r = 0.39, P = 0.002) in one distribution system in China when chloramine residual was 0.05–1.0 mg/L. Zhang et al. (2002) pointed out that AOC was not the limiting factor for bacterial regrowth in distribution systems. High residual chloramine (> 2 mg/L) could effectively repress microbial activity in waters even at high AOC levels. HPC had a significant correlation with chlorine (r = –0.74, P = 0.0001) but a relatively poor correlation with AOC (r = –0.21, P = 0.028) (Zhang and DiGiano, 2002). Therefore, the HPC may be influenced by AOC and disinfectant residual simultaneously. The increase of HPC may be due to high AOC feeding, mainly in distribution systems with low disinfectant concentration, while the HPC density can be limited by a high disinfectant residual concentration even at a relatively high AOC concentration.

Biostability analysis in real distribution systems has mainly been carried out with regression models. Some mechanistic models, such as SANCHO, PICCOBIO and BAM, were developed with the pipe loop reactor or biological annular reactor. There are few reports about the assessment of these models in real distribution systems (DiGiano and Zhang, 2004; Zhang et al., 2004). The biostability in real distribution systems in large cities remains poorly understood. Moreover, there is no information available on control strategy based on systemic analysis of whole water supply networks (from water treatment plant to distribution systems). The aim of this study was to systemically investigate the factors that influenced HPC levels in real drinking water distribution systems in one large city in Southern China and find a cost-effective strategy to control bacterial regrowth. Mathematical tools were applied to identify the most important parameters and evaluate their impacts on HPC concentration. Different water treatment processes and disinfection technologies were also evaluated to establish a feasible strategy for control of bacterial regrowth.

1 Materials and methods

1.1 Profile of water treatment plant, distribution system and sampling points

Raw water and the effluents of a horizontal sedimentation tank, sand filter, O_3 contact tank, biological activated carbon (BAC) filter, and pumping station were selected as the sampling points (**Fig. 1**). Water quality monitoring was performed in sub-district D and sub-district Z, both of which were served by the N water treatment plant (**Table 1**). Field surveys were conducted in summer (August 2007; water temperature at approximately 30°C) and winter (November 2007, January 2008 and March 2008; water temperature at approximately 20°C), respectively. Water temperature, dissolved oxygen (DO), HPC, AOC, and chloramine residual were determined.

1.2 Water quality analysis

Water temperature, DO and chloramine residual were measured using a mercury thermometer, YSI 550A Handheld Dissolved Oxygen Instrument (YSI, USA) and Pocket Colorimeter™II (Hach, USA), respectively. HPC was detected according to the Pour Plating Method with R2A media (22°C, 7 days) (APHA, 1995). AOC was measured according to the literature (Liu et al., 2002).

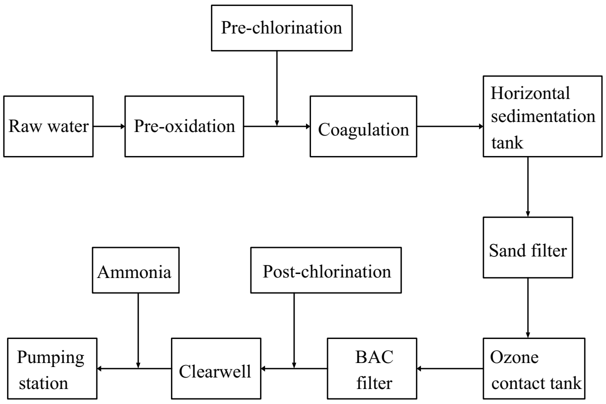

Fig. 1 Flowchart of the studied water treatment plant. BAC: biological activated carbon.

Table 1 Distribution systems and sampling points

Water plant	Distribution systems	Pipe material	Service year of pipe before investigation	Number of sampling points	Number of samples
N water treatment plant	Sub-district D	Cast iron	4	8	35
	Sub-district Z	Cast iron	8	5	18

1.3 Principal component analysis

Principal component analysis (PCA) was performed using IBM SPSS Statistics (19th version, IBM, USA) on the water temperature, chloramine residual, DO, HPC, and AOC of the distribution systems in sub-district D and sub-district Z ($n = 53$).

1.4 Biostability curve

A biostability curve was drawn according to the method developed by Srinivasan and Harrington (2007), assuming that the Monod Equation can be used to describe the growth of a bacterium, which can be expressed by Eq. (1), and the Chick-Watson Equation can be applied to calculate the inactivation by disinfectants, which can be expressed by Eq. (2)

$$\mu_H = \mu_{max} \frac{S_S}{K_S + S_S} \tag{1}$$

where, μ_H (min^{-1}) is the specific growth rate constant of the bacterium; μ_{max} (min^{-1}) is the maximum specific growth rate of the bacterium; S_S (μg/L) is the concentration of limiting substrate for growth, which can be assumed as AOC; and K_S (μg/L) is the half-maximum substrate concentration.

$$\mu_A = -k \times C_D \tag{2}$$

where, μ_A (min^{-1}) is the specific inactivation rate of the bacterium; C_D (mg/L) is the chloramine residual concentration; and k (L/(min·mg)) is the inactivation rate constant of the bacterium.

When the specific growth rate of a bacterium equals its inactivation rate (i.e., μ_H equals μ_A), Eq. (3) can be deduced

$$C_D = R \frac{S_S}{K_S + S_S} \tag{3}$$

$$R = \frac{\mu_{max}}{k} \tag{4}$$

where, R (mg/L) is the ratio of maximum specific growth rate to specific inactivation rate.

Based on Eq. (3), a curve can be plotted with C_D against S_S, which is the so-called biostability curve. Each individual bacterial species has an individual biostability curve. In a distribution system with multiple bacterial species, for any specific HPC control objective (for instance, HPC ⩽ 100 CFU/mL), a conservative biostability curve can be developed. When the operation point of the system is above the curve, the control objective is surely satisfied. On the other hand, when the operation point of the system lies on or is below the curve, the control objective may be not met. In other words, for any sampling point where the control objective is achieved, Eq. (5) should be satisfied:

$$C_D \geqslant R \frac{S_S}{K_S + S_S} \tag{5}$$

In a real drinking water distribution system, when the HPC goal is determined and the conservative biostability curve is plotted, two sampling points (A and B), which lie exactly on the curve, can be selected. Applying measured C_D and S_S values of point A and B to Eq. (3), R value and K_S value of this system can be calculated by Eqs. (6) and (7), respectively.

$$R = \frac{C_{DA} \times C_{DB} \times (S_{SA} - S_{SB})}{S_{SA} \times C_{DB} - S_{SB} \times C_{DA}} \tag{6}$$

$$K_S = \frac{S_{SA} \times S_{SB} \times (C_{DB} - C_{DA})}{S_{SB} \times C_{DA} - S_{SA} \times C_{DB}} \tag{7}$$

where, C_{DA} (mg/L) and C_{DB} (mg/L) are the residual chlorine concentrations at sampling points A and B, respectively; S_{SA} (mg/L) and S_{SB} (mg/L) are the concentrations of limiting substrate for growth (assumed as AOC) at sampling points A and B, respectively.

This study set the control objectives of HPC ⩽ 100 CFU/mL and HPC ⩽ 50 CFU/mL, respectively. Based on the water quality monitoring database of sub-district D and sub-district Z, R values and K_S values for different distribution systems under various water temperatures were obtained.

2 Results and discussion

2.1 Bacterial regrowth in the distribution systems

2.1.1 Comparison of biostability indicators in the two sub-distribution systems

The comparison of basic biostability indicators in the two sub-distribution systems showed obvious differences. As mentioned above, the two sub-distribution systems received the same finished water from a water treatment plant. The HPC in sub-district D were overall higher than in sub-district Z (**Fig. 2a**). Although the pipe ages in sub-district D (4 years) were younger than in sub-district Z (8 years), the higher chloramines (22%) and lower AOC (10%) accounted for the lower HPC concentration (27%) in the latter one (**Table 2**). The fairly high DO level in the sub-district Z also indicated the depression of bacterial metabolism (Liu et al., 2005).

Although the aging of pipe usually brings more chlorine decay and hence stimulates the bacterial regrowth in the distribution system (Al-Jasser, 2007), the situation can

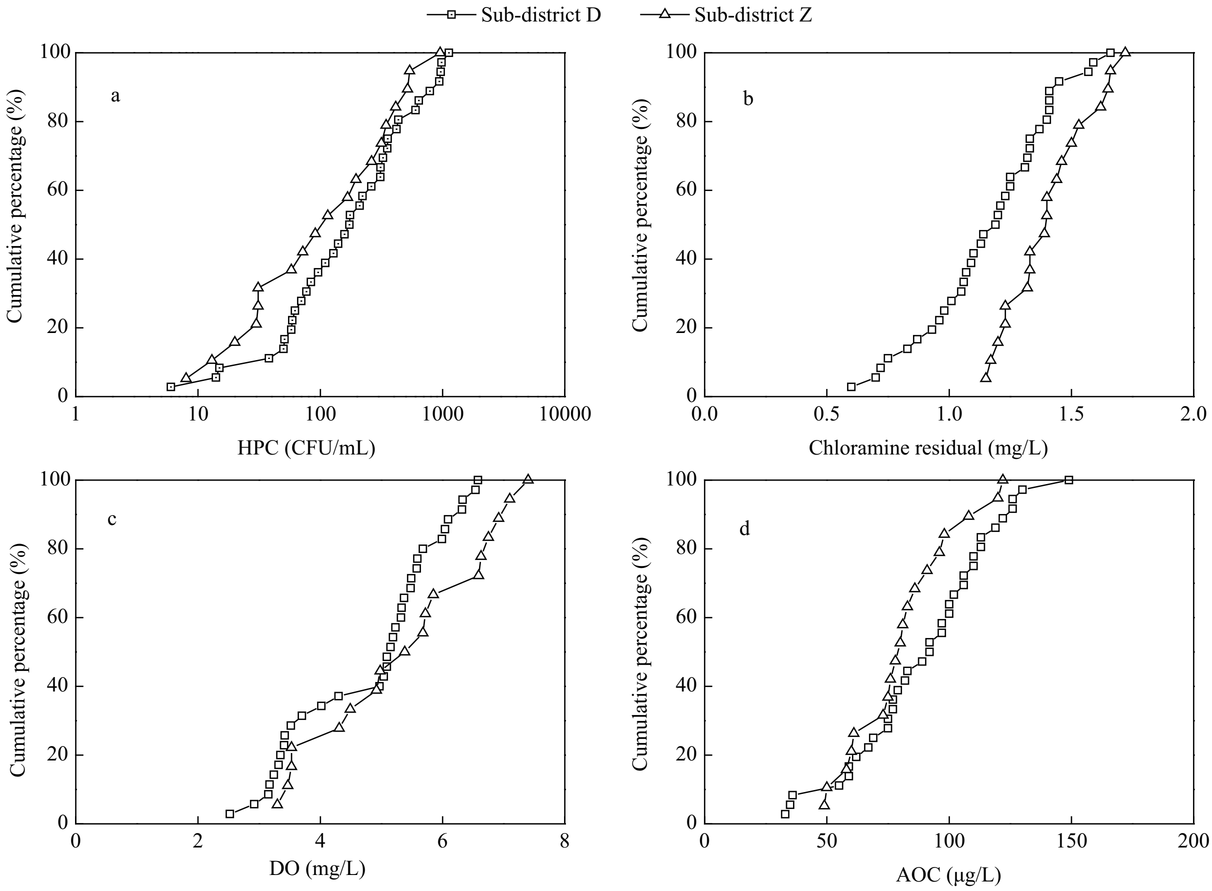

Fig. 2 Accumulative curve of basic biostability indicators. (a) HPC, (b) chloramine residual, (c) DO, and (d) AOC.

Table 2 Comparison of basic biostability indicators in the two sub-distribution systems

Distribution systems	DO (mg/L)	AOC (μg/L)	Chloramine (mg/L)	HPC (CFU/mL)	HPC > 500 CFU/mL
Sub-district D	4.79 ± 1.19	90 ± 28	1.16 ± 0.26	300 ± 314	19%
	(2.52–6.58)	(33–149)	(0.60–1.66)	(6–1130)	(*n* = 36)
Sub-district Z	5.36 ± 1.37	81 ± 21	1.41 ± 0.17	220 ± 248	16%
	(3.30–7.40)	(49–122)	(1.15–1.72)	(8–955)	(*n* = 19)

usually be controlled by maintaining higher chloramine residual, as evidenced in this case. The result obtained in this study also indicated the necessity to maintain relatively high disinfectant residual and low AOC concentration for biostability control.

2.1.2 Principal component analysis

The PCA method is a good tool to reduce the dimension of space formed by multiple variables. By this means, the correlationship with each variable can be described more easily and clearly. The coefficients of the original five parameters on the extracted components are shown in **Fig. 3**. The correlations between HPC and water temperature, chloramine residual, DO or AOC were calculated (**Table 3**). As shown in **Fig. 3** and **Table 3**, it is notable that HPC had strongly negative correlation with chloramine residual, weakly positive correlation with temperature, weakly negative correlation with DO, and no obvious relationship with AOC, which is quite different from the results in previous studies (Van der kooij, 1992; Lu, 2005). The PCA method allows us to reduce the space dimension of five variables, i.e. water temperature, DO, HPC, AOC and chloramine residual, into three components, which are the linear combination of the previous five parameters. The new space can explain 89.8% of the variety of the original five parameters.

The concentration of chloramine residual in Tianjin was 0.05–1 mg/L in water mains and ends (usually less than 0.5 mg/L) (Lu, 2005). However, in this study, the concentration of chloramine residual was 0.5–1.5 mg/L in water mains, and usually more than 1 mg/L in sub-district D and sub-district Z (**Fig. 2b**). The fairly high chloramine

Table 3 Correlation between HPC and other parameters

Coefficient	Dependent variable	Water temperature	Chloramine residual	DO	AOC
r	HPC	0.43	–0.47	–0.38	0.02
P	HPC	0.001	< 0.001	0.002	0.448

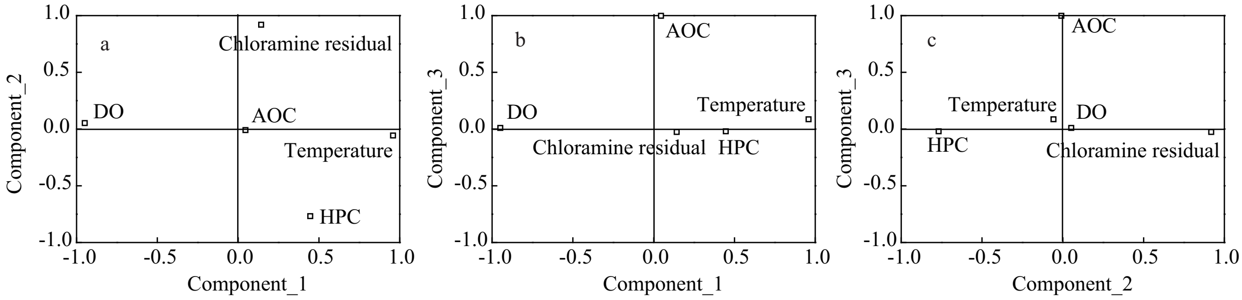

Fig. 3 Component plot in rotated space. Extraction method: principal component analysis; Rotation method: Varimax with Kaiser Normalization.

residual in this studied city repressed the bacterial regrowth in waters even with sufficient organic substrate.

The AOC concentration had weak relationship with chloramine residual in this study. This was possibly due to the weak oxidation potential of chloramines, which cannot increase the AOC level as much as free chlorine (Wu, 2000). The AOC concentration was also influenced by consumption by microorganisms, transformation by disinfectant, re-suspension of pipe sediment, and the contribution from dead bacteria, which complicated its origin (Srinivasan et al., 2008; Huck and Gagnon, 2004).

2.2 Biostability curves in distribution systems

2.2.1 Biostability curves with the goal of HPC ⩽ 100 CFU/mL

The biostability results were classified into four groups: I. sub-district D, summer (water temperature was about 30°C); II. sub-district D, winter (water temperature was about 20°C); III. sub-district Z, summer (water temperature was about 30°C); and IV. sub-district Z, winter (water temperature was about 20°C). Thus, the influences of water temperature and sampling zones could be visualized in the biostability curves.

The US Environmental Protection Agency believes that HPC in any distribution system could reach 100 CFU/mL or lower. For distribution systems with HPC of 100–500 CFU/mL, pipe-flushing is recommended. HPC higher than 500 CFU/mL indicates that the distribution system has poor microbial quality (Bartram et al., 2003). Therefore, the objective of HPC ⩽ 100 CFU/mL was set for the sub-districts.

The chloramine residual (R) and half-maximum substrate concentration (K_S) of biostability curves have been previously used for the control of bacterial regrowth in simulated distribution systems (Srinivasan and Harrington, 2007). However, to the authors' knowledge, this is the first study to use biostability curves for biostability control in real distribution systems. The biostability curves represented the variation of R and K_S value with season in the same sub-networks. As shown in **Fig. 4a, b**, in sub-district D, the R value was higher in summer than in winter, suggesting that higher chloramine residual was needed to control the bacterial regrowth and achieve the goal of HPC ⩽ 100 CFU/mL. The higher K_S value in the summer also showed evidence of a more active metabolism needing more substrate. The biostablity curves in the sub-district Z also demonstrated the same pattern.

The disinfectant residual (C_D, as chloramine concentration) had a closely linear relationship with substrate (C_S, as AOC concentration) if AOC was below K_S (**Fig. 4a**). However, the real AOC concentration (36–149 μg/L, shown in **Fig. 2d**) in sub-district D was higher than K_S. Thus, the residual chloramine, C_D, substrate, and C_S had no close correlation with each other in real situations, which was also observed in the PCA results. If a water plant wants to lower the disinfectant dosage in order to avoid a high yield of disinfection byproducts without impairing the biostablity, much lower AOC should be of practical importance. For example, when AOC below K_S = 26 μg/L in summer in sub-district D was satisfied, the chloramine residual could be decreased to half of R (R = 1.91 mg/L), i.e. 0.95 mg/L.

2.2.2 Biostability curves with different HPC goals

The biostability curves could also be used to determine the water quality requirement with different HPC goals. As shown in **Fig. 5**, if a stricter biostability goal, HPC ⩽ 50 CFU/mL, is set in sub-district D, the R value will be as high as 2.33 mg/L under 30°C and 1.67 mg/L under 20°C, respectively. The stricter the HPC goal is, the higher the R value needed. The higher temperature favors bacterial regrowth in the distribution system, so a higher chloramine residual is necessary to satisfy the HPC goal.

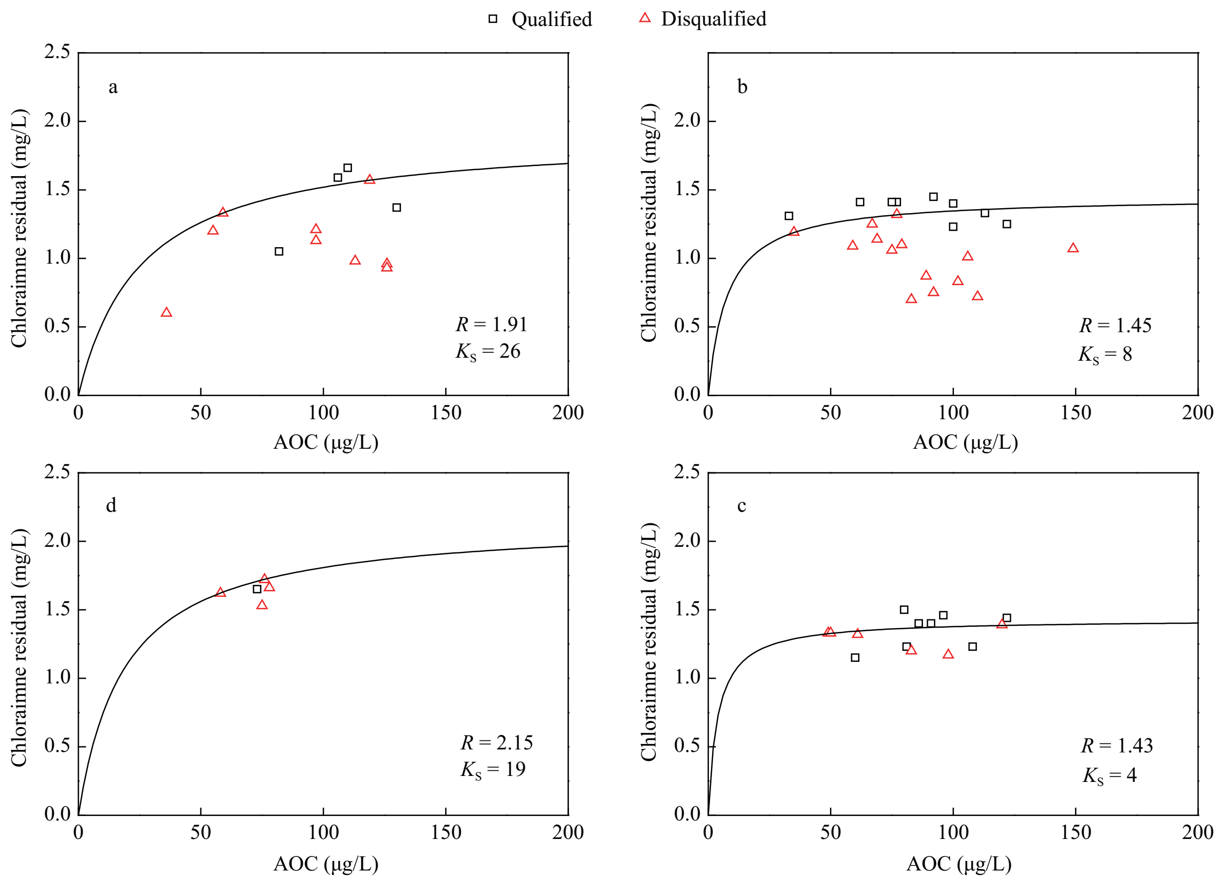

Fig. 4 Biostability curves with data qualified or not judged by HPC goal ⩽ 100 CFU/mL. (a) in the sub-district D at 30°C, (b) in the sub-district D at 20°C, (c) in the sub-district Z at 30°C, and (d) in the sub-district Z at 20°C.

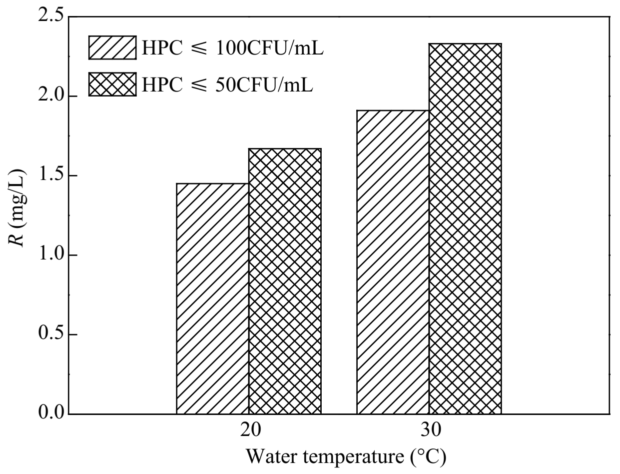

Fig. 5 *R* of Biostability curves for HPC ⩽ 100 CFU/mL and HPC ⩽ 50 CFU/mL in sub-district D.

2.3 Efficiency of AOC removal by water treatment process

The efficiency of AOC removal by different water treatment processes in the real water treatment plant was evaluated to address the bacteria regrowth in the distribution system in this city. As illustrated in **Fig. 6**, the conventional coagulation-sedimentation-sand filtration process could remove the AOC by –28%~60%, which was also observed by other researchers (Chen et al., 2007b; Lou et al., 2012). The removal rate of coagulation-sedimentation could vary with the organic molecular weight and hydrophobicity in the source water (Volk and Lechevallier, 2002; Klimenko et al., 2012). The removal in the sand filter was attributed to biodegradation since the biomass in the sand filter was still high in the subtropical region (Magic-Knezev and Van der kooij, 2004; Feng et al., 2012; Liao et al., 2013). Moreover, the active biodegradation in the sand filter could be proven by the change of AOC removal with different pre-chlorination dosages. When a high pre-chlorination dosage was applied, the AOC concentration increased by 30%, with 0.65 mg/L of chlorine residual in the sand filter effluent; however, when low pre-chlorination dosage was applied, the AOC concentration decreased by 45%, with 0.21 mg/L of the chlorine residual in the sand filter effluent.

The ozonation and BAC process was used to further lower the AOC concentration. The AOC concentration decreased by 35% or 46%, both below 50 μg/L for two sampling dates, and reached the accepted biostable level (Lechevallier, 1990; Han et al., 2012). However, the low AOC concentration after the O_3-BAC process could be

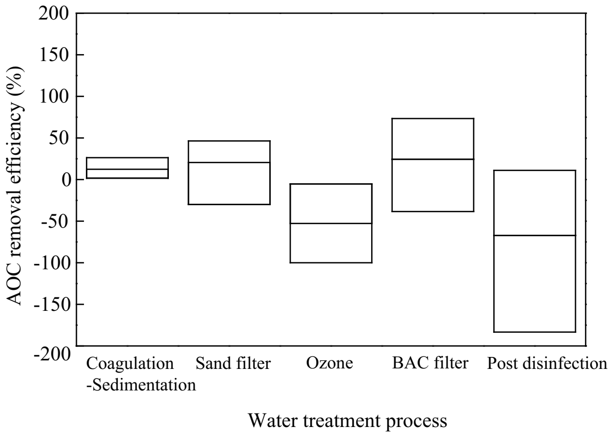

Fig. 6 AOC removal in the N water treatment plant. The upper, middle, and lower edges of the boxes show the maximum, average, and minimum, respectively.

elevated by post-chlorination, which was also reported by previous researchers (Volk et al., 2002; Chen et al., 2007). With the combined treatment of the conventional and advanced treatment processes, the AOC concentration in the finished water was 62–80 μg/L.

Hence, the required chloramine residual in the different sub-distribution systems at different temperatures could be obtained using the aforementioned biostability curves (**Table 4**). This goal could be satisfied easily since the detected chloramine residuals in the finished water were above these levels (1.73 mg/L in August 2007; 1.54 mg/L in November 2007). Therefore, the AOC removal by the water treatment process and the applied high chloramine dosage could jointly satisfy the goal of controlling the HPC concentration below 100 CFU/mL.

Elevating disinfectant dosage is a much easier way to control bacterial regrowth in the distribution system, compared with AOC control by conventional and advanced treatment processes. For example, the detected AOC level in the network was in the range of 33–149 μg/L. If AOC = 149 μg/L was taken into the biostability curve (20°C in sub-district D), the C_D value of 1.38 mg/L could be obtained; if AOC = 33 μg/L was taken into the curve (20°C in the sub-district D), the C_D value of 1.17 mg/L could be obtained. The difference of chloramine dosages between two scenarios was just 0.21 mg/L. However, it was quite difficult to maintain a low AOC level in the finished water (below 50 μg/L). Therefore, the high dosage of chloramine was a practical option for this subtropical city.

Table 4 Calculated chloramine values for the control of HPC $\leqslant$ 100 CFU/mL

Systems	Water temperature (°C)	Chloramine residual needed (mg/L)
Sub-district D	30	1.35–1.45
	20	1.29–1.32
Sub-district Z	30	1.65–1.74
	20	1.35–1.36

Application of chloramine as post-disinfectant can reduce the formation of halogenated disinfection by-products (DBPs), such as THMs and HAAs (Chen et al., 2007a; Cromeans et al., 2010). However, chloramination will favor the formation of N-nitrosamines and other nitrogenous DBPs (Krasner, 2009; Kristiana et al., 2009; Wang et al. 2012b). The yield of N-nitrosamines or other nitrogenous DBPs was determined by the concentration and characteristic of DBP precursors, chloramine dosage, pH, and temperature (Krasner, 2009; Nawrocki and Andrzejewski, 2011; Wang et al., 2013). Therefore, the elimination of nitrosamine precursors will be an effective strategy with the conventional and advanced treatment processes at hand to control the nitrosamine formation in networks (Wang et al., 2012a) without decreasing the chloramination dosage. Moreover, further study should be conducted to obtain the optimized dosage that can well balance bacterial regrowth and DBP formation.

3 Conclusions

The correlation analysis indicated that HPC had the closest relationship with chloramine residual, fairly strong with water temperature or DO, but quite weak with AOC. The biostability curve based on Chick-Watson's law and the Monod Equation was developed to depict the sampling results. By this means, the chloramine residual (R) and half-maximum substrate concentration (K_S) could be determined for two different biostability goals (HPC $\leqslant$ 100 CFU/mL; HPC $\leqslant$ 50 CFU/mL). The stricter the HPC goal was, the higher the R value required. Biostability curves could be a very useful tool for biostability control in real distribution systems. The AOC concentration could only be decreased to 62–80 μg/L even with the combined conventional and advanced treatment processes, and cannot be the limiting factor for bacterial regrowth in the distribution system. The addition of a high dosage of disinfectant is much more cost-effective than increasing the substrate removal by water treatment processing to meet the biostability goal of HPC $\leqslant$ 100 CFU/mL.

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