Inactivation of particle-associated *Escherichia coli* with chlorine dioxide

Tao Lin, Bingwei Hou, Zhe Wang and Wei Chen

ABSTRACT

In this paper, the inactivation of both free *Escherichia coli* (FE) and particle-associated *E. coli* (PAE) with chlorine dioxide (ClO_2) were investigated using granular activated carbon effluent water samples. The inactivation rate of FE was higher than that of PAE and the reactivation ratio of PAE was higher than that of FE, indicating the threat of particle-associated bacteria. Response surface methodology (RSM) was applied to determine the factors influencing the disinfection efficiency of ClO_2 in inactivating PAE. The experimental results indicated that particle concentration was a principal factor influencing the PAE inactivation efficiency, presenting a negative correlation, while exposure time and ClO_2 dosage revealed a positive correlation. The inactivation kinetics of PAE using ClO_2 was also investigated and the results demonstrated that PAE inactivation with ClO_2 fitted the Chick–Watson kinetic model. The inactivation rate constants of PAE were found to follow the Arrhenius expression with an activation energy of 107.5 kJ/mol, indicating a relatively strong temperature dependence. However, there are minor effects of pH and initial ClO_2 dosage on PAE inactivation rate constant.

Key words | chlorine dioxide, disinfection, kinetics, particle-associated *E. coli*, response surface methodology

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INTRODUCTION

Granular activated carbon (GAC) filtration/adsorption is an advanced water-treatment technology which has been widely used in drinking water facilities (Zhang 2009). Owing to its porosity and weak polarity, GAC possesses high adsorptive capabilities in removing undesirable dissolved organic carbon (DOC) fractions (Velten *et al.* 2007). In addition, GAC may remove pathogens; for instance, a 0.1–1.1 log reduction of *Escherichia coli* could be achieved as a result of attachment of the bacteria to GAC (Hijnen *et al.* 2010). Thus, GAC may act as a microbial carrier in water purification, with bacteria being attached to its surface or inner pores. The GAC filtering layer may accumulate a large number of biological and non-biological particles that have the ability to penetrate and break through the GAC filter and reach the effluent (Castaldelli *et al.* 2005). The effluent bacteria may be attached to particles, forming particle-associated bacteria (PAB). The inactivation of pathogens which attach to particles has been regarded as a key process to attenuate the concentration of viable pathogens in potable water (Pedley *et al.* 2006; Tufenkji & Emelko 2011). However, PAB have been proven to be more resistant to disinfection using chlorine, chloramines or ultraviolet irradiation than their planktonic counterparts (Hess-Erga *et al.* 2008; Lynch *et al.* 2014). As a result, re-growth of the PAB that has survived disinfection is enhanced in the water distribution system (Hallam *et al.* 2001). Therefore, a method to effectively inactivate attached bacteria and generate fewer by-products is needed in the field of drinking water disinfection.

Disinfectants are important in controlling opportunistic bacteria and their residuals are the primary sources for limiting microbial re-growth in drinking water distribution systems (Hong *et al.* 2013). Chlorine dioxide (ClO_2), as an alternative disinfectant, has attracted considerable attention since it has been proven to minimize trihalomethane

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(THM) formation and has a better biocidal efficiency than free chlorine over a wide pH range (Sutton *et al.* 2002; Navalon *et al.* 2009; Zhang *et al.* 2015). Previous research has found that the disinfection efficacy of ClO_2 is higher than that of free chlorine for waterborne human rotavirus (Xue *et al.* 2013). A previous study revealed that the resistance of PAB to chlorination could mainly be attributed to the incomplete penetration of disinfectants into the particles (Berman *et al.* 1988). The inactivation of microorganisms depended largely on the permeability of ClO_2 into the bacterial cell (Li *et al.* 2004). Although the disinfection efficiency of ClO_2 has been widely investigated (Gagnon *et al.* 2005; Kim *et al.* 2009), there is still less research on its inactivation effect on PAB. In addition, very few reports have been involved in the influence of particles concentration on the disinfection efficiency of PAB with ClO_2, and the kinetic model of PAB disinfection using ClO_2 is inadequate (Liu *et al.* 2014). Normally, conventional microorganism culture and enumeration methods can hardly reflect the actual amount of PAB and can also mislead the actual inactivation performance (Liu *et al.* 2013).

ClO_2 disinfection efficiency is influenced by many factors such as disinfectant dose, water quality and temperature (Barbeau *et al.* 2005; Ayyildiz *et al.* 2009; López-Velasco *et al.* 2012). The response surface methodology (RSM) is a statistical experimental protocol used in mathematical modeling (Gong *et al.* 2012). This method reduces measurements, while improving statistical interpretation and indicating the interaction between variables (Yim *et al.* 2012). In this study, the inactivation performance of PAB via ClO_2 was investigated. During experiments, the GAC filter effluent was used as water sample, in which *E. coli* was the targeted organism. The influences of ClO_2 dosage, particle concentration, and particle size on inactivation rate were investigated using RSM. Furthermore, the PAB disinfection kinetics were discussed.

METHODS AND MATERIALS

Water samples

Water samples were taken from the effluent of a pilot GAC facility in Nanjing, China. The characteristics of the raw water are shown in Table S1 (in the Supplementary Material, available with the online version of this paper). In the treatment processes, a GAC filter column was installed immediately following sand filtration and prior to final disinfection. The experimental schematic is illustrated in Figure 1.

The design parameters of the GAC filter are given in Table 1. The GAC type used in the pilot facility is broken charcoal. The backwashing procedure comprised an initial air–water backwash step followed by a water backwash.

Water samples were simultaneously obtained from taps located at the side of the GAC filter. These samples were collected in sterile glassware which had been sterilized under high temperatures in advance and rinsed using deionized water and air-dried before use.

Particle preparation

An autoclaved gauze filter with a pore size of 2.0 μm (Millipore Corp., USA) was used to trap the particles from the GAC effluent which was prepared as previously described (Camper *et al.* 1985). The water samples were first filtered through the gauze filter, and then the filter was aseptically cut in half and placed in a vessel containing 300 mL of cold, sterile, reagent-grade water. Each vessel was shaken vigorously for 2 h to dislodge

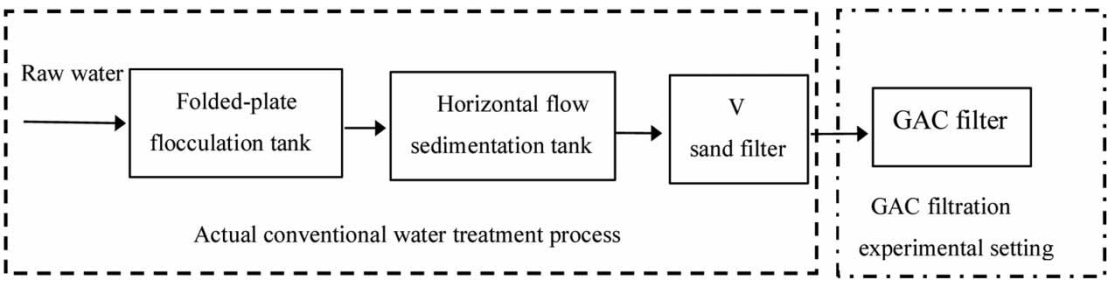

Figure 1 | Schematic diagram of the conventional treatment process and GAC filter.

Table 1 | The design parameters of the GAC filter

			Air-water backwashing rate (L/(m²*s))		Single water backwashing		
Empty bed contact time (min)	Diameter (mm)	Depth (mm)	Air	Water	Time (min)	Rate (L/(m²*s))	Time (min)
20	200	1,800	10	3	3	4	6

the particles from the filter. Subsequently, the gauze was removed from the vessel and the autoclaved GAC-filtrate, without particles, was added to the vessel in order to attain particle concentrations of approximately 2×103 counts/mL as determined by a particle counter instrument (WHB1-IBR-B1, Interbasic Resources, USA).

To investigate the effects of particle size on disinfection, autoclaved gauze filters with different pore sizes (Millipore Corp., USA) were used to separate the particles in the GAC effluent into three size distributions: 2–5, 5–8, and >8 μm.

To investigate the effects of particle concentration on the disinfection efficiency, three concentrations of particles, for the 5–8 μm size range, were prepared by serial dilutions at 2×10^3, 1.25×10^3 and 5×10^2 counts/mL.

Bacteria culture and PAB inoculation

A broth culture (100 mL) of an *E. coli* K-12 (ATCC 10798) strain was inoculated in 100 mL of Luria Bertani (LB) nutrient broth (Sigma Aldrich, USA) and incubated on a rotary shaker (KS501, IKA, Germany) at 37 °C overnight. The cells were then harvested after 5 h in the exponential growth stage. Then, an aliquot of 1.0 mL of the *E. coli* suspension was transferred into another 100 mL fresh LB broth and incubated on the rotary shaker at 37 °C for another 5 h. To prepare the reaction suspensions, 50 mL of the harvested *E. coli* suspension (concentration around 10^8 CFU/mL) was centrifuged at 13,500 rpm at ambient temperature for 2 min and then washed and re-suspended in 0.9% NaCl solution three times to remove any impurities present in the solution. The supernatant was discarded and the pellet was re-suspended and then transferred to the water sample (Angela-Guiovana & Cesar 2004).

Prior to the inoculation of the *E. coli* pellet, prepared water samples with particles were vigorously vibrated and autoclaved for 24 h at 121 °C to eliminate any potential interference of other bacteria in the water sample. After transfer, the inoculated water sample was transferred to the autoclaved glass beaker (autoclaved at 121 °C for 15 min). Then the beaker was fixed again on a rotary shaker at 37 °C overnight to promote effective attachment of *E. coli* to particles in the water sample, forming particle-associated *E. coli* (PAE).

Generation of ClO_2 and disinfection experiments

ClO_2 was freshly prepared no more than 1 day in advance for each experiment, using ClO_2 solution-generating equipment (Ecosia Co., Seoul, Korea) with Nalgene tubing for all generator gas lines. All of the chlorine dioxide was bubbled through a diffuser into a clean amber bottle containing chilled distilled deionized (DDI) water and stored in a dark room at 4 °C until used.

The concentration of the ClO_2 stock solution, approximately 100 mg ClO_2/L, was measured using a direct spectrophotometric method after a 50-fold dilution with DDI water. Absorbances were measured at 360 nm with a UV-1601 Shimadzu spectrophotometer using a quartz cuvette with a light path length of 1 cm. ClO_2 concentrations used in all experiments, <5.0 mg ClO_2/L, were within the linear range of the spectrophotometric method and thus were measured without dilution.

All disinfection experiments were carried out in 1 L flasks with 500 mL intermixture of water samples and disinfectant, fixed in thermostatic water bath oscillators. The ClO_2 residual and *E. coli* counts were then investigated.

Separation of PAE from particles

During the inoculation process, not all *E. coli* cells are able to attach to particles and form PAE. Therefore, some steps were required prior to PAE enumeration in order to eliminate free *E. coli* (FE). The water sample containing the particles was chlorinated using 0.3 mg/L sodium hypochlorite for 30 min at 4 °C

(pH = 7.0) in the dark, which effectively eliminated the residual free-living bacteria, but had no inactivation effect on the attached bacteria (Camper *et al.* 1986). Then, particles in water samples (attaining particle concentration about 2×10^3 counts/mL) were intercepted and rinsed in new vessels. A homogenization technique was used to quantitatively desorb microorganisms from particles as previously described (Camper *et al.* 1985).

E. coli enumeration

Before enumeration of bacteria, the water samples were first shaken for 45 s to break apart clumped bacteria (Kerim & Banu 2012). The *E. coli* counts were measured according to Method 1604 (USEPA 2002). The inactivation efficiency was calculated using the following equation:

$$\text{Inactivation efficiency} = \frac{(N_0 - N_t)}{N_0} \times 100\%$$

where N_0 and N_t represent the initial number of *E. coli* and those at the sampling point during the process, respectively.

Reactivation of *E. coli*

After inactivation, treated water samples were placed in a closed sterilized tube which prevented sample contamination with bacteria in the air. After 24 h of incubation at ambient temperature the samples in enclosed tubes were withdrawn for bacteria count. The reactivation ratio of *E. coli* was estimated based on the bacterial count before and after cultivation. The reactivation ratio was calculated using the following equation:

$$\text{Reactivation efficiency} = \frac{(N_{ac} - N_t)}{(N_0 - N_t)} \times 100\%$$

where N_{ac} represents the number of *E. coli* which were cultured after exposure to ClO_2.

Response surface design of PAE inactivation

RSM is a statistical processing and analysis technology based on an experimental design for modeling and analysis of multivariable problems (Liu & Wang 2005). In the present study, RSM was used to determine the sensitivity of different factors influencing PAB inactivation. The independent variables were denoted as A, B, and C, representing ClO_2 dosage, exposure time, and particle concentration, respectively. Furthermore, in this response surface design, a minimum or low range (denoted as −1), an average or medium range (denoted as 0), and a high or maximum range (denoted as +1) were defined for each experimental factor (Table 2).

The software packages Design Expert 7.1.3 and Statistics Analysis System (SAS 8.2) were used to analyze the effects of the variables on the disinfection rate.

Kinetics of PAE inactivation model

The water sample containing particles at a concentration of approximately 2.0×10^3 counts/mL was used to estimate the kinetics of the PAE inactivation model. Different initial ClO_2 concentrations were prepared as described previously, and different phosphate buffers with varied pH (6.0, 7.4 and 10.0) were prepared by mixing various doses of sodium phosphates ($NaH_2PO_4 \cdot 2H_2O$ and $Na_2HPO_4 \cdot 12H_2O$).

Analytical methods

To ensure the reliability of the results, each experiment was performed in triplicate and the average value was determined ($p < 0.05$, p expresses the statistical test value).

RESULTS AND DISCUSSION

Inactivation and reactivation of PAE

As shown in Figure 2, the inactivation efficiency of both FE and PAE increased with the increasing ClO_2 dosage. The inactivation efficiency of FE reached 88% when the ClO_2

Table 2 | Independent variables and levels for the response surface design

Independent variables	Variable factor	Variable range		
		−1	0	+1
ClO_2 dosage (mg/L)	A	1.0	1.5	2.0
Exposure time (min)	B	10	20	30
Particle concentration (counts/mL)	C	500	1,250	2,000

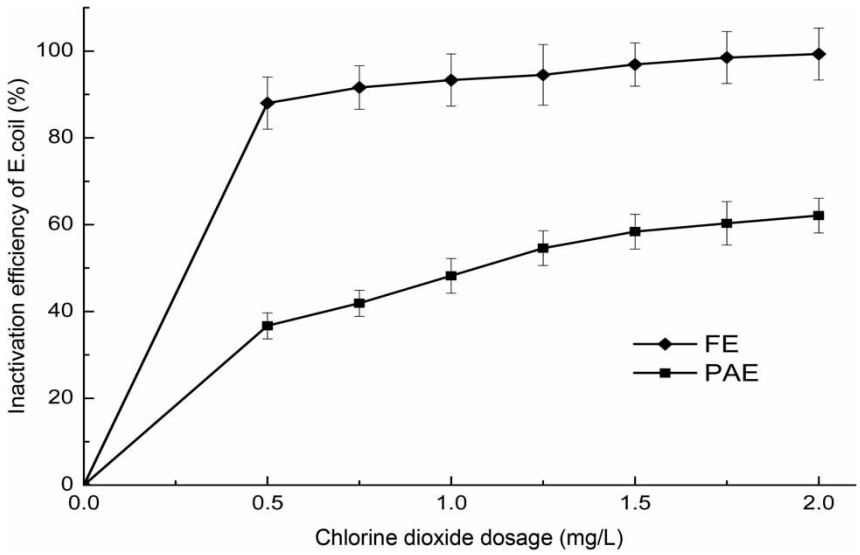

Figure 2 | ClO_2 inactivation of FE and PAE (10^8 CFU/mL) for 30 min at ambient temperatures with particle concentration of 2×10^3 counts/mL.

dosage was 0.5 mg × min/L and it increased to 99.3% at 2.0 mg × min/L of ClO_2. However, the inactivation efficiency of PAE was much lower than that of FE. PAE is considered to be resistant to disinfection because *E. coli* located in the cracks, crevices, and pores of the particles may not come into contact with the ClO_2 molecule. The particles possess a relatively coarse specific surface and their abundant pore structures provide good protection to the adsorbed *E. coli* (Wojcicka *et al.* 2008). The hydrophobic inner surface minimizes the adsorption of hydrophilic molecules, therefore ClO_2 cannot sufficiently access the inner structure of the particles. Furthermore, reducible organic compounds adsorbed onto the particles may react with ClO_2, thus preventing disinfection of PAE (Inoue *et al.* 2004; Mavrocordatos *et al.* 2004).

The reactivation rate of *E. coli* after different disinfection processes are shown in Figure 3. It is clear that PAE have a higher reactivation efficiency (11–43%) than those of FE (4–23%). ClO_2 can inactivate microorganisms by disrupting protein synthesis or increasing the permeability of the outer membrane due to its reaction with the membrane protein and lipids (Mahmoud *et al.* 2007). As depicted in Figure 3, increasing exposure time or ClO_2 dosage increases the extent of damage to microorganisms and decreases the reactivation efficiency. However, particles provide protection for PAE, leading to relatively higher reactivation efficiency. PAE may also accumulate in the distribution systems as loose

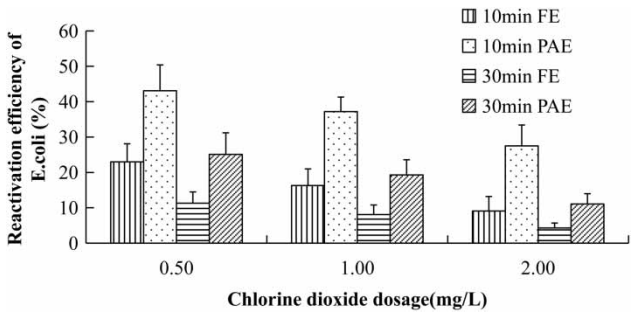

Figure 3 | Reactivation efficiency of FE and PAE after inactivation for 10 and 30 min of exposure to ClO_2.

deposit and may re-grow in an environment with low or even depleted disinfectant residues (Jjemba *et al.* 2010; Thayanukul *et al.* 2013). ClO_2 dosage and exposure time are both important factors in controlling reactivation of *E. coli*.

RSM of inactivation efficiency on variables

The effect of independent variables on the disinfection efficiency (Table S2, available with the online version of this paper) was assessed by developing an experimental matrix in which the treatment groups were designed using the software package Design Expert. The experimental results were used for the evaluation of the reliability of response to variables. Analysis of variance (ANOVA) was used to evaluate the significance of the model equation, as shown in Table 3.

Table 3 | ANOVA for independent variables

Source	Sum of squares	df	Mean square	F-value	*p*-value prob > F
Model	0.21	9	0.023	9.04	0.0010
A	0.011	1	0.011	4.46	<0.0001
B	0.018	1	0.018	7.21	0.0087
C	0.15	1	0.15	58.27	<0.001
Residual	198.45	10	19.84	–	–
LOF	173.91	5	34.78	7.09	0.0254
Pure error	24.54	5	4.91	–	–

As shown in Table 3, the lack of fit (LOF) *p*-value of 0.025 implied that the LOF was significant relative to the pure error. The matrix had a *p*-value of 0.001, indicating that the response to the variables was reliable. The *p*-values for ClO_2 dosage (A) and particle concentration (C) were less than 0.001, meaning that the two variables play a key role in influencing the sensitivity of PAB to ClO_2 disinfection.

Three-dimensional response surface curves were generated as a function of the interaction of any two variables by holding the other at a significant level, and the results are shown in Figure 4. The plots illustrate a similar relationship for the effects of ClO_2 concentration and exposure time, whereas the effect of particle concentration was adverse. As shown in Figure 4(a), the PAE inactivation rate increased with the increasing ClO_2 dose. The tendency of inactivation response gradually stabilized at a ClO_2 dosage higher than 1.7 mg/L. Previous studies have reported that bacterial inactivation could be achieved within several minutes when ClO_2 comes into contact with the bacterial cells, and ClO_2 can increase the permeability of the outer membrane by reacting with the membrane protein and lipids, then, the leakage of intracellular substance can react with ClO_2 (Mahmoud *et al.* 2007). With regard to PAE inactivation, ClO_2 should essentially penetrate into the particles and then reach the bacterial cells. High concentration is preferred in order to improve ClO_2 penetration, which may attain a steady rate

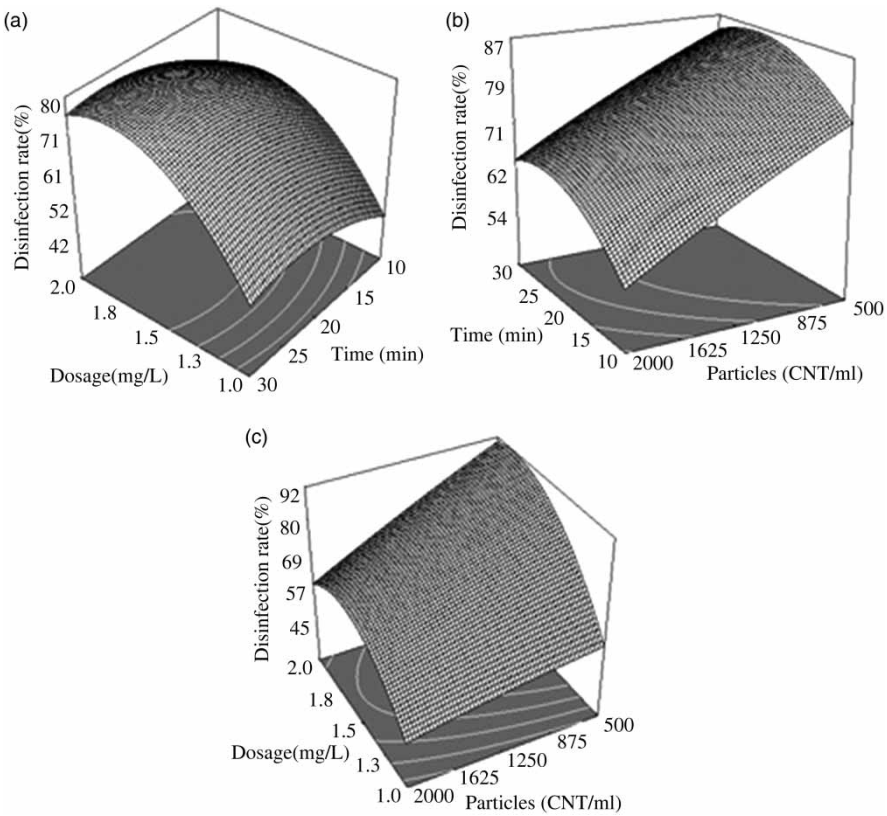

Figure 4 | Response surface plots for PAB inactivation as an interaction function of independent variables (temperature: 25 °C; pH: 7.4). (a) Particle concentration: approximately 1,250 counts/mL; (b) ClO_2 dosage: 1.5 mg/L; (c): exposure time: 20 min.

when the ClO_2 dose reaches a certain concentration, and a higher concentration may produce stagnation in the disinfection efficiency (Liu *et al.* 2014). With respect to the exposure time (Figure 4(b)), the inactivation rate was improved with the increase in the exposure time. However, when the exposure time reached a certain duration, about 25 min, the inactivation rate gradually stabilized. In contrast to common free bacteria, PAE is protected by the particles from disinfectant. Hence, it is essential for the ClO_2 molecules to penetrate into the particles to come in contact with the attached *E. coli*. Figure 4(c) shows that a lowered particle concentration favored an increase in disinfection efficiency. When the particle concentration decreased from 2,000 to 500 counts/mL, the disinfection efficiency increased from 46 to 67%. The number of PAE was increased, resulting in a higher probability of residual surviving *E. coli* after disinfection. Therefore, the presence of a greater number of particles may cause a loss of disinfectant in bulk concentration, which decreases the PAE inactivation efficiency.

Effects of particle size distribution on inactivation performance

It can be clearly noted from Figure 5 that increasing the particle size decreases the inactivation rate of PAE, especially when the particle size was larger than 8 μm and the inactivation rate is lower than 40%. In a previous study, it was demonstrated that particles larger than 7 μm were mainly responsible for shielding the coliforms from chlorination (Berman *et al.* 1988). It is considered that larger particles have more complicated inner structures with more inner caves and paths (Chen *et al.* 2009), which may provide more interspaces for attachment of PAE. This complicated structure may also affect the transport efficiency of ClO_2. Therefore, it is necessary to control large size particles in effluent, especially those larger than 8 μm.

Inactivation kinetics of PAE with ClO_2

Kinetic models

The residual disinfectant concentrations C_t for each experiment were fit separately to a first-order rate equation:

$$C_t = C_0 \exp(-k_1 t) \tag{1}$$

where C_t is the free available residual ClO_2 concentration at the sample point (mg/L), C_0 is the initial free available residual ClO_2 concentration (mg/L), t is the exposure time (min), and k_1 is the ClO_2 decay rate constant (min^{-1}).

PAE inactivation with ClO_2 was described by a simple Chick–Watson kinetics:

$$\ln\left(\frac{N_t}{N_0}\right) = -k'CT \tag{2}$$

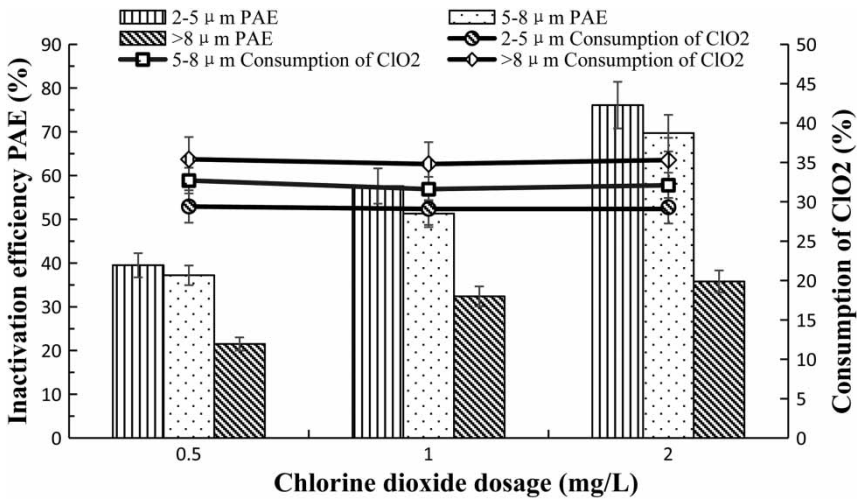

Figure 5 | Influence of particle distribution on PAE inactivation rate at ambient temperature with particle concentration of 2×10^3 counts/mL for 30 min.

where N_t/N_0 is the fraction of viable *E. coli* cells after time t of exposure to the disinfectant, k' is the PAE inactivation rate constant, and CT is the integrated exposure to the disinfectant over time in mg·min/L and can be calculated in Equation (3):

$$CT = \int_0^t C_\lambda d\lambda \tag{3}$$

where C_λ is the disinfectant concentration at time λ ($0 \leq \lambda \leq t$).

As illustrated in Figure 6, the ClO_2 residual gradually decreased when the exposure time increased. Furthermore, it can be observed that a linear relationship existed between $\ln(C_0/C_t)$ and exposure time. The ClO_2 decay rate constant k_1 was 0.0825, 0.1029 and 0.1194 min^{-1} respectively when the initial ClO_2 dosages were 1.0, 1.5 and 2.0 mg/L, also the correlation coefficient (R^2) of the respective line was greater than 0.98.

Inactivation kinetics of PAE with ClO_2 are shown in Figure 7. It is clear that a single curve was obtained revealing that the disinfectant concentration had no effect on the inactivation kinetics of PAE with ClO_2, which is consistent with former research (Vicuña-Reyes *et al.* 2008). The average PAE inactivation rate constant was 0.0692 L/(min/mg), which is much lower than that of free bacteria (Vicuña-Reyes *et al.* 2008; Li *et al.* 2011), indicating that particles can shelter *E. coli* attached to or wrapped in particles. Figure 7 also demonstrates that when exposure time exceeds 20 min, the inactivation curves of different initial ClO_2 dosage had a slight uptrend, indicating that long contact time plays an important role in improving the inactivation rate of PAE.

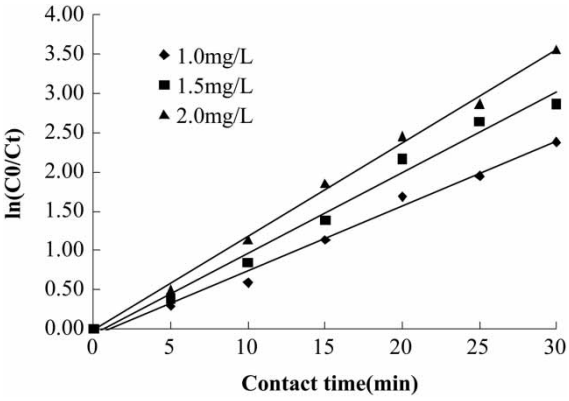

Figure 6 | First-order kinetic plot for the decay of ClO_2.

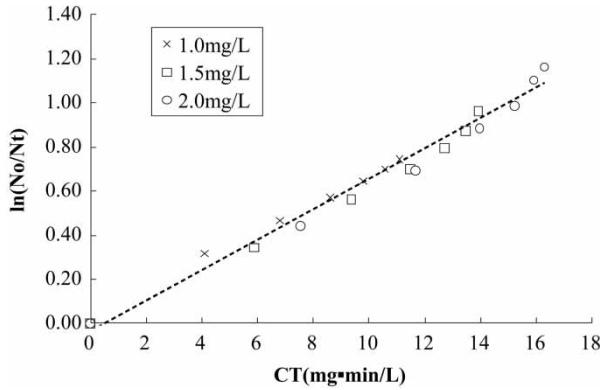

Figure 7 | Inactivation kinetics of PAE with ClO_2 at pH 7.4 and 25 °C.

Impact of temperature and pH

It may be considered that the inactivation rate constant for *Salmonella* spp. is dependent on temperature and pH. An empirical formula for the PAE inactivation rate constant (k') could be obtained according to the Arrhenius equation as follows (Kohpaei & Sathasivan 2011):

$$k' = A \exp\left(-\frac{E_a}{RT}\right) \tag{4}$$

where T is the absolute temperature in K, R is the gas constant (8.314 J/(mol·K)), E_a is the reaction activation energy in J/mole, and A is the collision frequency factor in L/(mg·min).

As shown in Figure 8, the inactivation of PAE with ClO_2 has a certain temperature dependence while pH showed no observable effect on k', which is consistent with previous research (Mahmoud *et al.* 2007; Sun *et al.* 2007).

The parameters obtained by least-squares fitting to Equation (4) were $A = 4.72 \times 10^{17}$ L/(mg·min) and $E_a =$ 107.5 kJ/mol, the latter reveals that the increasing temperature can significantly increase the PAE inactivation rate. An increase in temperature can promote the dissolution of ClO_2 in water, thus increasing its oxidative action, favoring inactivation of PAE (López-Velasco *et al.* 2012). However, the activation energy is much higher than that of 74.1 kJ/mol reported for the inactivation of free *Mycobacterium avium* (Vicuña-Reyes *et al.* 2008) indicating that PAE is more resistant to disinfectant in comparison to free bacteria.

Generally, the pH may not affect the inactivation rate, whereas pH 10 results in a slight decrease of inactivation

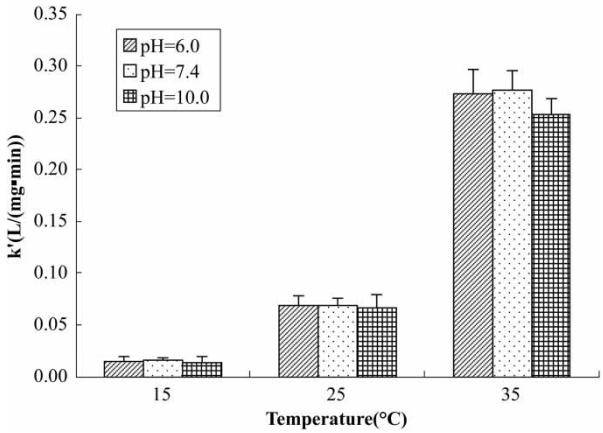

Figure 8 | Effect of temperature and pH on PAE inactivation rate constant.

rate. However, a reduction of inactivation rate constant was found when the pH was 10. It can be speculated that a disproportionate reaction of ClO_2 will occur under alkaline conditions, also high temperature can promote this disproportionate reaction thus reducing the available ClO_2 in water solutions, decreasing the inactivation rate (Sun *et al.* 2007).

CONCLUSIONS

This study has shown that PAE were more difficult to inactivate than the free bacteria due to the protection of particles. Increasing concentrations of ClO_2 and contact time result in higher rates of inactivation. The inactivation rate constants of PAE were found to follow the Arrhenius expression with activation energy of 107.5 kJ/mol, indicating a relatively strong temperature dependence. However, there are no observable effects of pH and initial ClO_2 dosage on PAE inactivation rate constant. The results demonstrated that PAE inactivation with ClO_2 fitted the Chick–Watson kinetic model. Therefore, the presence of particles should be avoided in the disinfection process in order to maximize its effectiveness to inactivate waterborne bacteria.

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