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Synergistic effect of the sequential use of UV irradiation and chlorine to disinfect reclaimed water

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ABSTRACT

The effectiveness of UV and chlorination, used individually and sequentially, was investigated in killing pathogenic microorganisms and inhibiting the formation of disinfection by-products in two different municipal wastewaters for the source water of reclaimed water, which were from a microfilter (W1) and membrane bioreactor (W2) respectively. Heterotrophic plate count (HPC), total bacteria count (TBC), and total coliform (TC) were selected to evaluate the efficiency of different disinfection processes. UV inactivation of the three bacteria followed first-order kinetics in W1 wastewater, but in W2 wastewater, the UV dose–response curve trailed beyond approximately 10 mJ/cm^2 UV. The higher number of particles in the W2 might have protected the bacteria against UV damage, as UV light alone was not effective in killing HPC in W2 wastewater with higher turbidity. However, chlorine was more effective in W2 than in W1 for the three bacteria inactivation owing to the greater formation of inorganic and organic chloramines in W1 wastewater. Complete inactivation of HPC in W1 wastewater required a chlorine dose higher than 5.5 mg/L, whereas 4.5 mg/L chlorine gave the equivalent result in W2 wastewater. In contrast, sequential UV and chlorine treatment produced a synergistic effect in both wastewater systems and was the most effective option for complete removal of all three bacteria. UV disinfection lowered the required chlorine dose in W1, but not in W2, because of the higher chlorine consumption in W2 wastewater. However, UV irradiation decreased total trihalomethane formation during chlorination in both wastewaters.

1. Introduction

Reclaimed water as an alternative water source for domestic, industrial, agricultural, and recreational purposes is providing an increasing contribution to sustainable water resources in many areas of the world. To minimize the public risks associated with exposure to reclaimed water, an adequate disinfection process is necessary. Traditionally, chlorination has been the most commonly adopted disinfection process for the treatment of both drinking and reclaimed water. However, with the appearance of *Cryptosporidium parvum* oocysts and *Giardia lamblia* cysts, which are chlorine-resistant pathogenic microorganisms, free chlorine can seldom achieve the required target goals for microbial inactivation (Korich et al., 1990; Mackenzie et al., 1994). Additionally, chlorine disinfection results in numerous by-products such as haloacetic acids and trihalomethanes, which exhibit potentially carcinogenic activity (Adin et al., 1991; Fawell et al., 1997). Achieving an appropriate balance between disinfection and disinfection by-products (DBPs) has

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been the focus in extensive investigations of alternative disinfection processes.

Ultraviolet irradiation is a promising alternative to free chlorine as a primary disinfectant, because of its ability to inactivate *C. parvum* without producing DBPs at common disinfection doses (Liu et al., 2002; Liberti et al., 2003). However, owing to a lack of residual activity and the possible repair of UV-damaged microorganisms (Hassen et al., 2000; Oguma et al., 2004), UV irradiation cannot be used as a stand-alone disinfectant. Nevertheless, using UV light as a primary disinfectant could potentially lower effective chemical doses and provide further disinfecting capabilities. The combination of UV irradiation as a primary disinfectant and free chlorine or monochloramine as a secondary disinfectant has been shown not only to prevent subsequent microbial regrowth but also to produce a synergetic disinfection (Ballester and Malley, 2004; Shang et al., 2007). However, other studies have reported mixed results for the effect of UV irradiation on DBP formation and chemical disinfectant dosages. Some researchers have found that UV disinfection lowered the possibility of harmful DBP formation (Cotton et al., 2001), whereas others have reported that UV treatment neither lowered the required chemical dosages nor significantly affected DBP formation upon subsequent chemical disinfection (Rand et al., 2007). The quality of the water to be treated may have a great influence on the role of UV in combined disinfection. All of the studies cited above were conducted as drinking water treatments. To our knowledge, little studies of the combination of UV and secondary chemical disinfection in treating reclaimed water has been carried out to determine microbial inactivation and DBP formation (Montemayor et al., 2008).

We investigated the effectiveness of UV and chlorination, alone and together, in killing pathogenic microorganisms and inhibiting DBP formation in wastewater from two different wastewater reclamation plants. Heterotrophic plate count (HPC), total bacteria count (TBC), and total coliform (TC) in the wastewater were examined to evaluate the effectiveness of different disinfection processes.

2. Materials and methods

2.1. Wastewater samples

Wastewater samples were collected from points upstream of the disinfection process at two different wastewater reclamation plants in Tianjin, China. All samples were stored at 4 °C before analysis. The wastewater treatment processes before disinfection were prechlorination, coagulation, sedimentation, and microfiltration at the first plant (W1), and fine screening, aerated grit removal, primary clarification, and membrane bioreactor application at the second (W2).

2.2. Water quality analysis

Measurement of particle size distributions was carried out using a Mastersizer 2000 (Malvern, UK). One sample was repeated five times and background measurement time of 25s and a refractive index of 1.52. The minimum and maximum detection limits were 0.2 μm and 2000 μm. Nitrate and nitrite were measured using the Dionex ICS-2000 ion chromatography system (Dionex Corp., CA), while NH_3–N was analyzed by Nesslerization method (UV–vis spectrophotometer-U-3100, Hitachi Co., Japan). Dissolved organic carbon (DOC) and total dissolved nitrogen (TDN) were measured using a Shimadzu TOC-V_{CPH} analyzer with a TNM-1 TN unit (Shimadzu, Japan). The amount of dissolved organic nitrogen (DON) was obtained by subtracting the concentration of inorganic nitrogen from the total dissolved nitrogen. In addition, total suspended solids (TSS), volatile suspended solids (VSS), pH and UV_{254nm} were determined according to standard methods for the examination of water & wastewater (APHA, 1998). The water quality parameters of both wastewaters were shown in Table 1.

2.3. Enumeration of indicator bacteria

In the experiment, heterotrophic plate count (HPC), total bacteria count (TBC), and total coliform (TC) in the wastewater were measured for evaluating the disinfection efficiency. TC was enumerated by membrane filtration followed by growth on Basic Fuchsin/sodium sulfite agar. Colony-forming units (CFUs) were counted within 48 h of incubation at 37 °C. The spread plate method was used to determine HPC and TBC. For HPC, bacteria were grown on R2A agar at 25 °C for 7 d; for TBC, bacteria were grown on nutrient agar at 37 °C for 48 h. The detection limits between the analytical methods are different. We defined the absence of microorganisms as <30 CFU/ml for the spread plate method and <10 CFU/100 ml for the membrane filtration method.

2.4. UV disinfection and photoreactivation

UV irradiation experiments were conducted using standard collimated beam tests. A low-pressure lamp (11 W; Beijing Lighting Research Institute, China) with emission primarily at 253.7 nm was mounted horizontally over a collimating tube. A Petri dish (diameter, 60 mm) containing 20 mL of wastewater was placed, with stirring, under the collimated beam. At the center of the solution surface, the incident intensity was approximately 0.125 mW/cm^2. The UV dose was calculated as described by Bolton and Karl (2003).

The photoreactivation experiments were performed under three fluorescent lamps, with the light intensity of

Table 1 – Water quality characteristics of wastewater samples used in this study.

Parameter	W1	W2
$Abs_{254\ nm}$	0.1035	0.2434
pH	7.74	8.62
DOC (mg/L)	5.243	9.911
DON (mg/L)	2.903	1.393
NH_3–N (mg/L)	0.203	0.298
Turbidity (NTU)	0.2	1.6
TSS (mg/L)	0.56	2.71
VSS (mg/L)	0.23	0.88
HPC (CFU/ml)	1.27×10^5	3.13×10^4
TBC (CFU/ml)	1.02×10^4	2.91×10^3
TC (CFU/100 ml)	9.01×10^3	4.37×10^4

0.066 mW/cm^2 at 360 nm. The percentage of photoreactivation was computed as follows (Lindenauer and Darby, 1994):

$$\text{Percentage photoreactivation (\%)} = \frac{\text{No. of cells photoreactivated}}{\text{No. of cells inactivated by UV}} \times 100\%$$

2.5. Chlorine disinfection

A 9% sodium hypochlorite solution was diluted in ultrapure water to prepare a stock solution. The chlorine concentration was measured using the N,N-diethyl-p-phenylenediamine colorimetric method, according to the standard method of Water and Wastewater Monitoring of China. Chlorine residuals in the bacteria enumeration samples were immediately neutralized with 10% sodium thiosulfate. To ensure sterility, all materials used in the experiments were autoclaved at 121 °C for 25 min. All experiments were performed at room temperature (25 ± 2 °C) and were repeated three times.

2.6. Disinfection by-product formation and analysis

The concentration of trihalomethanes (THMs) was determined using a gas chromatograph (6890 N; Agilent) with an electron capture detector and a fused silica capillary column (HP-5, 30 m, 320 μm × 0.25 μm), according to USEPA method 551.1. Chlorine in the samples was quenched with sodium sulfite, and THMs were measured using the following temperature program: hold at 35 °C for 4 min and ramp to 60 °C at 10 °C/min.

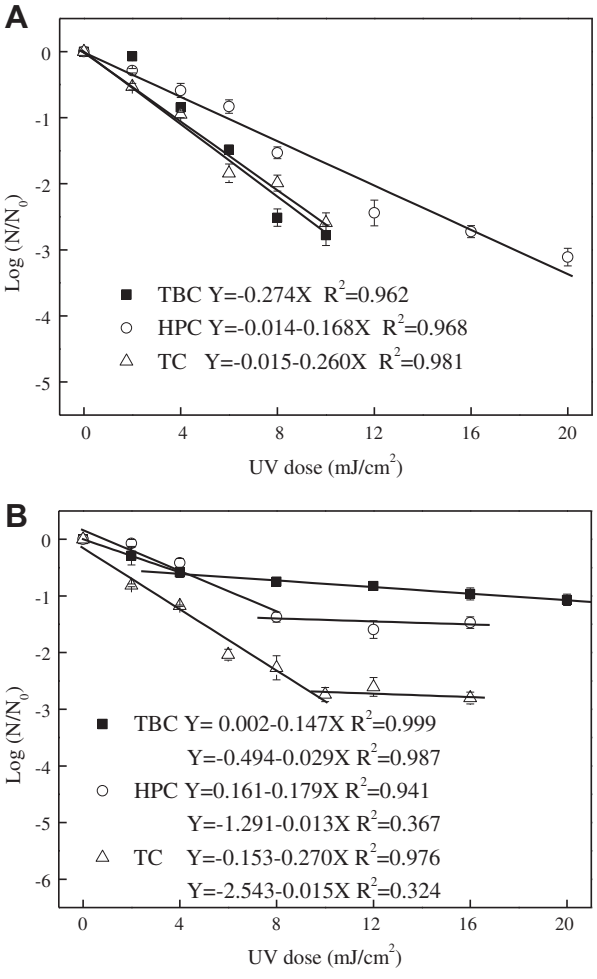

Fig. 1 – Kinetics of the tested bacteria in W1 (A) and W2 (B) response to UV irradiation.

3. Results and discussion

3.1. UV disinfection and photoreactivation in wastewater

Fig. 1 shows the inactivation kinetics of HPC, TBC, and TC in W1 and W2 wastewaters with UV irradiation. In W1 wastewater, the log survival of the three bacteria and the UV dose used for inactivation showed a strong first-order relationship ($r^2 > 0.960$). Among the three bacteria, TBC was the most sensitive to UV irradiation (k = 0.274 cm^2 m W^{-1} s^{-1}), followed by TC (k = 0.260 cm^2 m W^{-1} s^{-1}) and HPC (k = 0.168 cm^2 m W^{-1} s^{-1}). Before disinfection, the concentrations for TBC, HPC, and TC in W1 wastewater were 3.98, 4.86, and 4.29 log units, respectively. Treatment with UV at 8, 20, and 10 mJ/cm^2, respectively, reduced these values nearly to the detection limits. In contrast, in W2 wastewater, UV inactivation of the three bacterial indicators followed first-order kinetics until approximately 10 mJ/cm^2, after which significant tailing occurred with slower bacteria inactivation.

The UV inactivation kinetics of pathogenic bacteria in filtered W1 and W2 wastewaters have been reported to be the same as those in phosphate-buffered saline (Wang et al., 2011), indicating that dissolved matter in W2 wastewater had no impact on the UV inactivation of pathogenic bacteria. The turbidity and total suspended solids were 1.6 NTU and 2.71 mg/L, respectively, in W2 wastewater, compared with 0.2 NTU and 0.56 mg/L, respectively, in W1 wastewater. This suggests that particles might account for the decreased UV inactivation in W2 wastewater, as attached or embedded bacteria have shown increased resistance to UV inactivation (Winward et al., 2008). Cantwell and Hofmann (2008) reported that particles as small as 11 μm, which are naturally

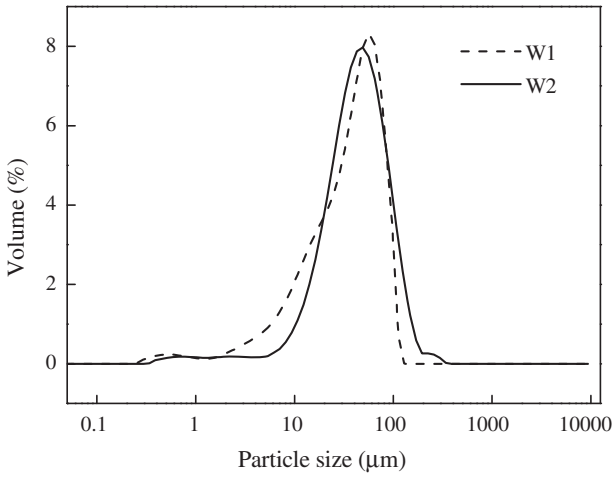

Fig. 2 – Particle size distributions of wastewater samples.

found in surface water with low turbidity (<3 NTU), can harbor indigenous coliform bacteria and offer protection from low-pressure UV light. The volume-weighted mean particle sizes were 52 μm and 41 μm and in W2 and W1 wastewater respectively (Fig. 2). The result indicated that the efficacy of UV disinfection of wastewaters was linked to both the size of the particles and the turbidity in wastewaters. However, the inactivation of the three bacteria in W2 wastewater still occurred with the increase of UV dose. TC and TBC were reduced to below the detection limits at UV doses of 20 mJ/cm^2 and 40 mJ/cm^2, respectively, whereas the HPC was 64 CFU/ml, even at a UV dose of 160 mJ/cm^2 (Supplementary Material Fig. S1). These results suggest that most of the coliform bacteria were dispersed in the W2 wastewater, while some of the other bacteria were embedded within particulate matter, where they were shielded from UV light.

Fig. 3 illustrates the photoreactivation of bacteria after exposure to different UV doses. After UV treatment with 4 mJ/cm^2, 28% of the TBC and 50% of the HPC bacteria in W1 wastewater were reactivated under a fluorescent lamp; the respective photorepair rates were 80% and 53% in W2 wastewater. Other UV doses showed a similar trend, with lower photoreactivation percentages in W1 wastewater compared with W2 wastewater. In addition, bacteria embedded in particles in the W2 wastewater might have been protected from UV-induced bacterial damage and released into solution after UV disinfection. The photoreactivation rate decreased in all three bacterial indicators as the UV dose increased, suggesting that higher UV doses should be applied to completely eliminate photoreactivation.

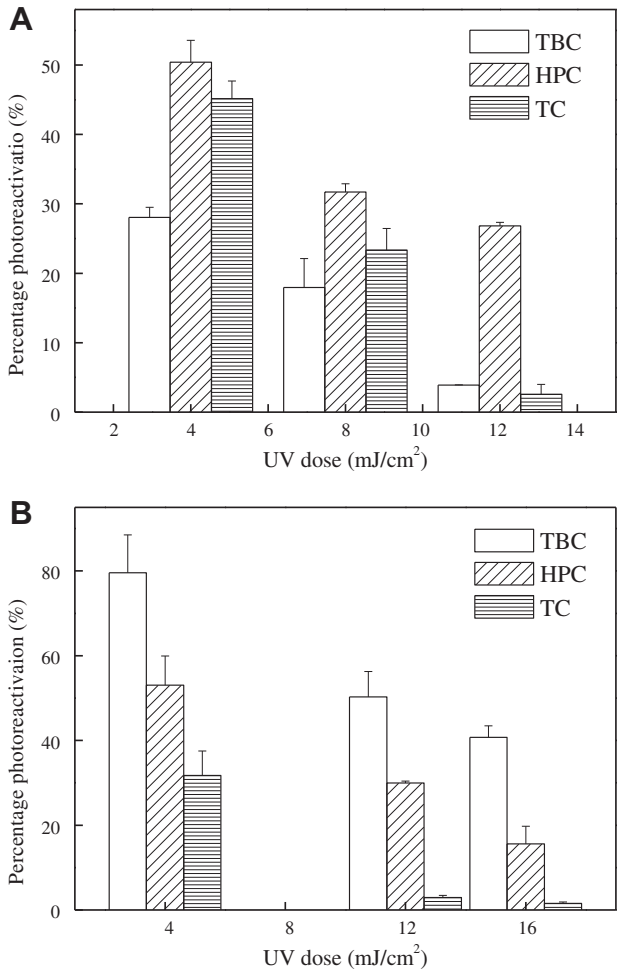

Fig. 3 – Effect of UV dose (mJ/cm^2) on photoreactivation of the tested bacteria in W1 (A) and W2 (B).

3.2. *Wastewater disinfection with chlorine*

The effect of the chlorine dose on the inactivation of bacteria in W1 and W2 wastewaters is shown in Figs. 4 and 5 respectively. At the contact time of 30 min, which is standard in wastewater treatment plants, a chlorine concentration of 3.5 mg/L reduced the TBC to an undetectable level in W1 wastewater, whereas more than 5.5 mg/L chlorine was required to decrease the HPC to an undetectable level. In W2 wastewater, both the TBC and HPC were reduced to undetectable levels by chlorine concentrations of 4 and 4.5 mg/L, respectively. Thus, suspended solids did not inhibit bacterial inactivation by chlorine, implying that chlorine is capable of

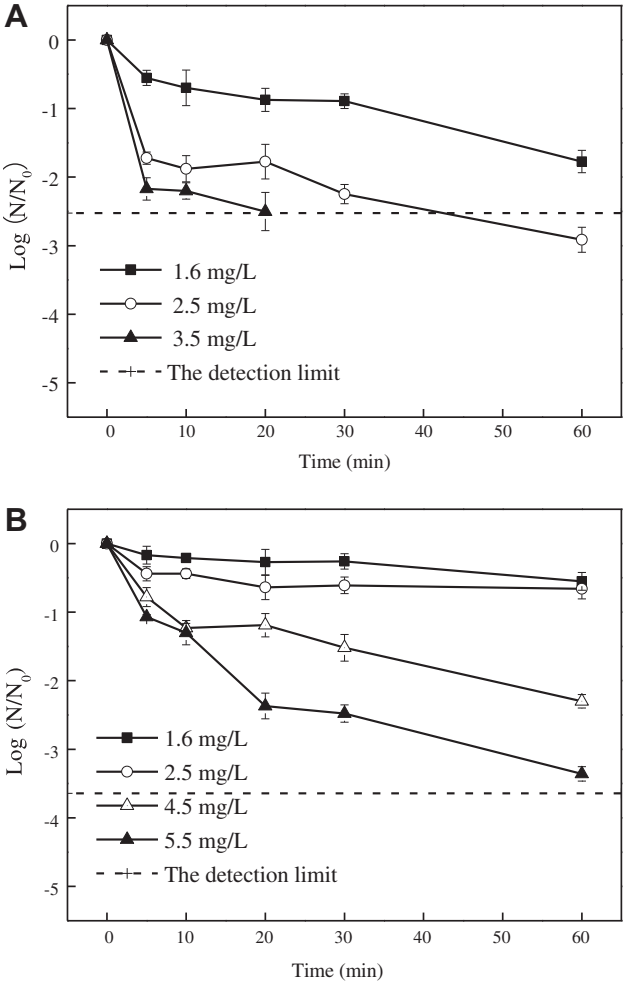

Fig. 4 – Inactivation of bacteria indicators by chlorine in W1. The symbols are means of three independent experiments and the error bars indicate standard deviation. (A) TBC (B) HPC.

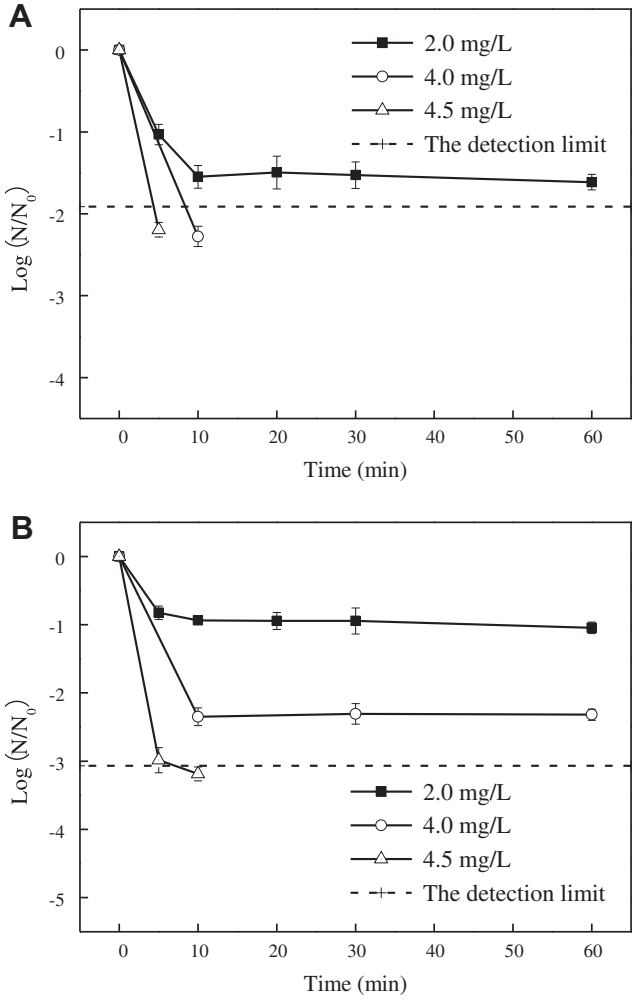

Fig. 5 – Inactivation of bacteria indicators by chlorine in W2. The symbols are means of three independent experiments and the error bars indicate standard deviation. (A) TBC and (B) HPC.

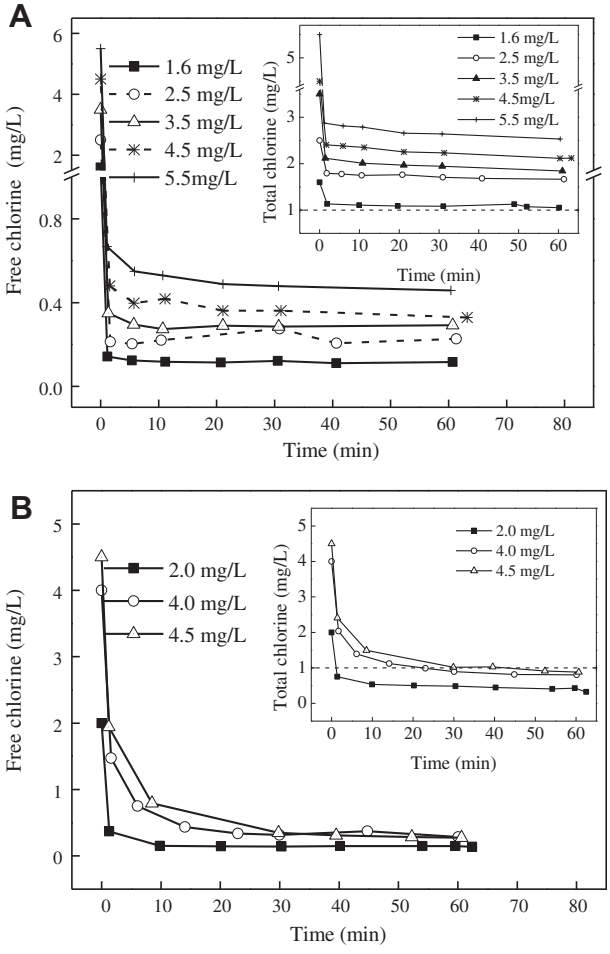

Fig. 6 – Decay of free chlorine and total chlorine as a function of disinfection time in W1 (A) and W2 (B).

penetrating macroporous network of pathways within wastewater particles (Dietrich et al., 2003).

The free and total chlorine were monitored by standard methods for the examination of water and wastewater (APHA, 1998) during the disinfection. As shown in Fig. 6, in W1 wastewater, the free chlorine was promptly consumed within initial 1.5 min, giving residual free chlorine concentrations of 0.14, 0.21, 0.35, 0.48, and 0.67 mg/L, and total chlorine concentrations of 1.13, 1.79, 2.12, 2.41, and 2.88 mg/L for initial chlorine doses of 1.60, 2.50, 3.50, 4.50, and 5.50 mg/L, respectively. Obviously, the combined chlorine was predominant in the total chlorine. Moreover, total chlorine in W1 wastewater did not decay rapidly within the reaction time. After 30 min of contact, the total residual chlorine concentrations were higher than the standard for reclaimed water (1 mg/L) in different initial chlorine doses. Since the NH_3–N and dissolved organic nitrogen concentration were 0.203 mg/L and 2.903 mg/L in W1 wastewater respectively, the amount of inorganic chloramines was smaller, the major portion was organic chloramines, which was produced by the reaction of chlorine with organic N-containing compounds in the wastewater (Shang and Blatchley, 2001). The lower total chlorine depletion was mainly contributed to the organic chloramines. In W2 wastewater, the residual free chlorine were 0.37, 1.47 and 1.94 mg/L, and the total chlorine were 0.75, 2.04 and 2.41 mg/L at the reaction time of 1.5 min for initial chlorine doses of 2.00, 4.00, 4.50 mg/L, respectively. The free chlorine was predominant in the total chlorine and total chlorine decayed more rapidly in W2 wastewater than in W1 wastewater. At the contact time 30 min, the total chlorine concentration were much more than 1 mg/L in W1 for all the tested initial chlorine doses, while those ones were near to 1 mg/L in W2 just for more than 4 mg/L initial chlorine dose. Since in W2 wastewater, the NH_3–N and dissolved organic nitrogen concentration were 0.298 mg/L and 1.393 mg/L, less than those ones in W1. Correspondingly, the combined chlorine was formed less than that one in W1, which was one reason for higher total chlorine depletion. On the other hand, the dissolved organic carbon (DOC) concentration was higher in W2 wastewater (9.911 mg/L) than that in W1 wastewater (5.243 mg/L), and this might have contributed to the faster decay rate of total chlorine in W2 wastewater. No significant difference in total chlorine consumption was observed between raw W2 and filtered W2 wastewater, indicating that

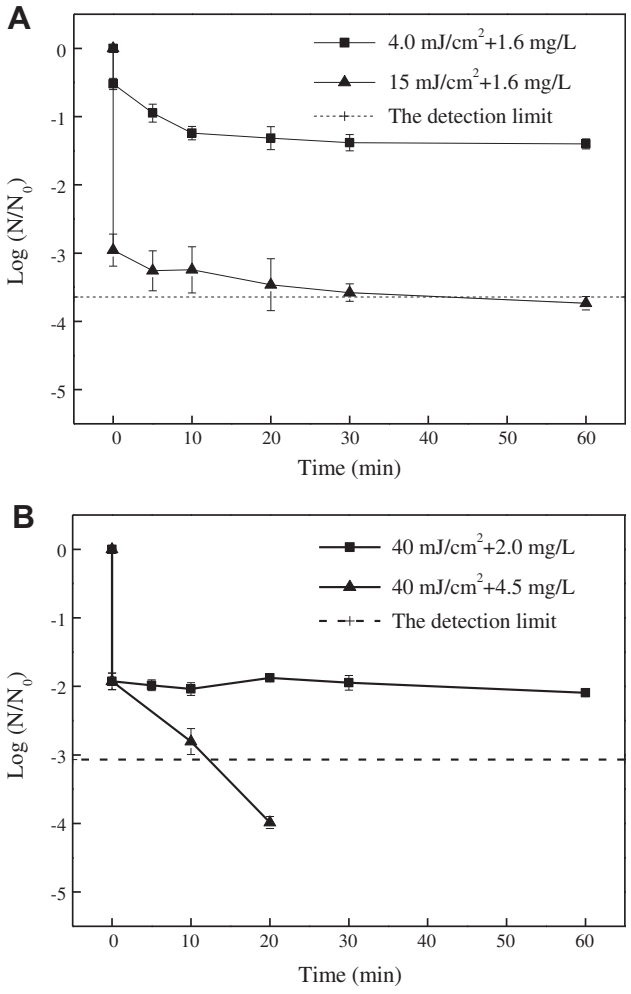

Fig. 7 – Inactivation of HPC by sequential UV and Cl_2. The symbols are means of three independent experiments and the error bars indicate standard deviation. (A) W1, (B) W2.

suspended particles in W2 wastewater did not affect the decay of chlorine (Supplementary Material Fig. S2).

3.3. *Combined UV and chlorine treatment*

Our results revealed deficiencies in the use of UV irradiation alone or chlorine alone for disinfection, including incomplete inactivation of bacteria, photoreactivation of bacteria, and increased requirements for chlorine. To address these limitations, we investigated the combined use of UV and chlorine. As shown in Fig. 7A and Supplementary Material Fig. S3, UV irradiation of 15 mJ/cm^2 almost completely reduced the TBC and HPC, by 4.01 and 5.10 log units, respectively, in W1 wastewater. A subsequent 30 min treatment with 1.6 mg/L chlorine further reduced the TBC and HPC to below the detection limits. Moreover, the free chlorine and total chlorine decay rates were almost the same as those with chlorine disinfection alone (Supplementary Material Fig. S4), with a total residual chlorine above the standard value of 1 mg/L. These results verified that the synergistic effect of UV and chlorine increased the inactivation rate of bacteria and lowered the required chlorine dose for W1 wastewater disinfection. A synergistic bactericidal effect of UV and chlorine also occurred in W2 wastewater. After UV treatment (40 mJ/cm^2) followed by chlorine treatment (2.0 mg/L), the TBC and HPC were reduced by 3.46 and 4.50 log units, respectively, with no observable photoreactivation of bacteria. Treatment with 4.5 mg/L chlorine for 30 min produced even greater reductions in the TBC and HPC, to below the detection limits, and gave total residual chlorine of 1.02 mg/L, just above the standard value for reclaimed water. Thus, UV disinfection lowered the required chlorine dose in W2 wastewater.

3.4. *DBP formation during different disinfection protocols*

The formation of total trihalomethanes (TTHMs) was determined after different disinfection protocols. As shown in Table 2, the concentration of TTHMs in W1 wastewater was 16.0 μg/L before disinfection, which resulted from the process of prechlorination, and decreased to 14.9 μg/L after irradiation by 15 mJ/cm^2 of UV alone. With chlorine treatment alone at 3.5 and 4.5 mg/L, the TTHM concentrations were 25.0 and 27.2 μg/L, respectively. However, when UV treatment at 15 mJ/cm^2 preceded treatment with 1.6 mg/L chlorine, the TTHM concentration was 19.7 μg/L, indicating that UV disinfection lowered the required chlorine dose and decreased TTHM formation in W1 wastewater. In W2 wastewater, the TTHM concentration was 8.6 μg/L before disinfection. No TTHMs were detected after exposure to UV of 40 mJ/cm^2. The TTHM concentration after treatment with 4.5 mg/L chlorine alone was 65.7 μg/L, and the concentration decreased to 60.2 μg/L when the same chlorine treatment was preceded by UV treatment at 40 mJ/cm^2. UV irradiation slightly decreased the TTHM formation in W2 wastewater, possibly due in part to UV-induced structural changes in DOM compounds. Various studies had revealed that UV irradiation at high UV doses could alter organic matter in water by reducing TOC content, color and molecular size (Corin et al., 1996). Magnuson et al. (2002) found that UV irradiation resulted in the changes of mass spectra of NOM at the range of UV dose from 20 to 140 mJ/cm^2. These changes might alter the reactivity of NOM with chlorine

Table 2 – Disinfection by-product yields (μg/L) for experimental conditions.

Water source	W1						W2			
UV (mJ/cm^2)	–	15	15	–	–	–	–	40	40	–
Cl_2 (mg/L, 1 h)	–	–	1.6	1.6	3.5	4.5	–	–	4.5	4.5
TTHMs (μg/L)	16.0	14.9	19.7	19.2	25.0	27.2	8.6	–	60.2	65.7

and then effect the formation of DPBs in subsequent chlorination, which would be studied in detail in another work.

4. Conclusions

UV irradiation alone did not effectively disinfect W2 wastewater because of its higher particle content, and chlorine alone exhibited low disinfection efficiency in W1 wastewater owing to the increased formation of inorganic and organic chloramines. However, sequential disinfection with UV and chlorine worked synergistically to effectively reduce the HPC, TBC, and TC and to inhibit TTHM formation in both W1 and W2 wastewater. The complete inactivation of HPC in W1 wastewater was accomplished by treatment with 15 mJ/cm^2 UV followed by 1.6 mg/L chlorine. This result could not be achieved with chlorine treatment alone, even with 5.5 mg/L chlorine. In W2 wastewater, TBC were completely inactivated by sequential application of 40 mJ/cm^2 UV and 2 mg/L chlorine. To produce this level of inactivation with chlorine treatment alone, a chlorine dose of 4 mg/L was required. In W1 wastewater, initial treatment with UV irradiation allowed the chlorine dose to be lowered to 1.6 mg/L, which enabled the total residual chlorine to be maintained at 1 mg/L. However, UV irradiation did not decrease the required chlorine dose of 4.5 mg/L in W2 wastewater because of the higher consumption of chlorine in W2 wastewater. In both wastewaters, prior UV irradiation inhibited TTHM formation during chlorine treatment, reducing the TTHM concentration by 7.5 μg/L in W1 wastewater and by 5.5 μg/L in W2 wastewater.

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Appendix. Supplementary data

Supplementary data related to this article can be found online at doi:10.1016/j.watres.2011.12.027.

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