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| 보 문 |

Combined Chlorite-Monochloramine Application for Controlling Nitrifying and Heterotrophic Bacteria in Drinking Water Distribution System

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상수관망에서 Chlorite-Monochloramine 소독제를 이용한 질산화 세균 및 종속영양세균의 제어

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In the present work, the reactors that harbor bacterial biofilms including ammonia-oxidizing bacteria (AOB) and heterotrophic bacteria were treated with a continuous dose of chlorite (0.66 ± 0.01 mg/L) either with or without monochloramine at 1.77 ± 0.03 mg/L. Both chlorite alone and combined chlorite-monochloramine applications effectively reduced biofilm and bulk AOB levels to near or below the detection limit (0.6 MPN/cm^2 and 0.2 MPN/ml). The combined chlorite-monochloramine application exhibited greater AOB inactivation than chlorite alone. Unlike AOB, heterotrophic plate count (HPC) was unaffected by chlorite alone. In contrast to chlorite-only application, a combination of chlorite and monochloramine resulted in a significant reduction in HPC levels with log reductions of 3.1 and 3.0 for biofilm and bulk water, respectively. The results demonstrate that the combined chlorite-monochloramine application can provide an effective treatment for the inhibition of AOB and heterotrophic bacteria in a drinking water distribution system.

***Keywords*:** biofilm, chlorite, drinking water, monochloramine

Chloramine disinfectant is formed by the combination of free chlorine with ammonia at a Cl_2/NH_3-N weight ratio of 3~5:1. The forms of chloramine are monochloramine (NH_2Cl), dichloramine ($NHCl_2$), and trichloramine (NCl_3); monochloramine is the predominant species under the typical conditions found in drinking water treatment (Vikesland *et al.*, 2001). In drinking water system, the use of chloramine is advantageous in terms of a lower concentration of disinfection by-products or a longer lasting residual (Norman *et al.*, 1980; Mitcham *et al.*, 1983; Norton and LeChevallier, 1997). However, there is a growing concern that free ammonia either from excessive dose or from chloramine decay may serve as an energy source for nitrifying bacteria and subsequently result in a nitrification incidence in the distribution system.

Nitrification is a biological process through which ammonia is oxidized to nitrite by ammonia-oxidizing bacteria (AOB) and further to nitrate by nitrite-oxidizing bacteria (NOB). In a chloraminated water distribution system, biological nitrification is the cause for concern because it can have a number of adverse effects on water quality including decrease in chloramine residual, increase in nitrite and/or nitrate, decrease in alkalinity, pH and dissolved oxygen concentration, and increase in heterotrophic bacteria (Odell *et al.*, 1996; Wilczak *et al.*, 1996; Zhang *et al.*, 2009). Particularly, nitrifying bacteria associated with biofilms on pipe walls can be more resistant to substrate limitation and also less susceptible to inactivation by disinfectants (Furumai and Rittmann, 1994; Baribeau, 2006; Ling and Liu, 2013). Due to the problems associated with nitrification in the chloraminated drinking water system, controlling nitrification is becoming one of the most pressing challenges facing water utilities that have experienced nitrification episodes.

The use of chlorite (ClO_2^-) has been proposed in an attempt to control nitrification in chloraminated water (McGuire *et al.*,

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1999; O'Connor *et al.*, 2001; McGuire *et al.*, 2006; Rahman *et al.*, 2011) and its antibacterial action is related to the inhibition of bacterial metabolism (Bichai and Barbeau, 2006). It is found in literature that the action of chlorite is bacterial species-specific and appears to be inhibitory to nitrifying bacteria (McGuire *et al.*, 1999, 2006; Gagnon *et al.*, 2005; Bichai and Barbeau, 2006; Rahman *et al.*, 2011). However, little information is provided concerning its effect on inactivating heterotrophic bacteria which are of concern because of their drinking water regulations.

A significant increase in heterotrophic bacteria has been reported in drinking water distribution systems where nitrifying bacteria are growing (Zhang *et al.*, 2009). They are either present in bulk water or in biofilms on the inner surface of the pipelines of distribution systems. Based on our experience, the presence of heterotrophic bacteria (especially in biofilm) can be a significant contributor to chloramine decay in a chloraminated distribution system before nitrification begins. When chlorite is applied, maintaining an adequate disinfectant residual is therefore necessary for the control of heterotrophic bacteria. From this point of view, the combined application of chlorite and monochloramine are worthy of interest.

This study was therefore intended to demonstrate the effectiveness of chlorite/chloramine combination on the control of bacterial colonization consisting of AOB and heterotrophic bacteria under a controlled pH condition, since the combined application of the two disinfectants would be expected to give a meaningful result.

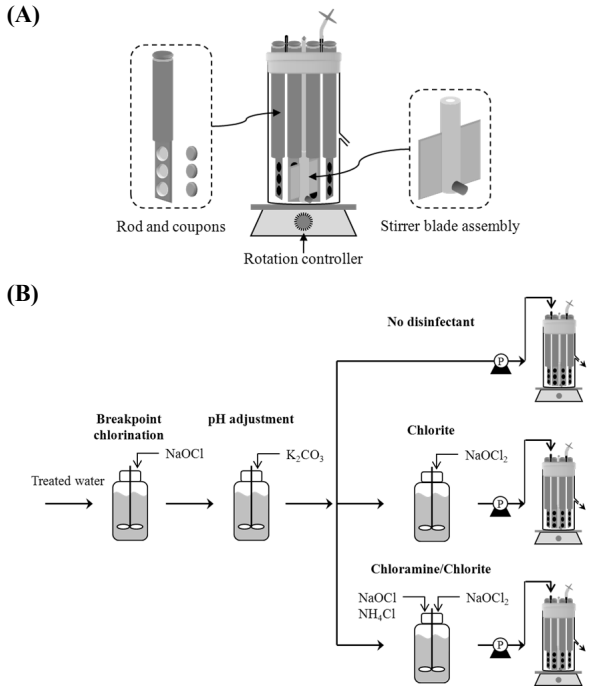

Fig. 1. Schematics of (A) biofilm reactor and (B) experimental setup.

Table 1. Characteristics of drinking water used as feed water for the experiments

Parameters	Average	Range
Total chlorine (mg/L)	0.83	0.58–1.29
NH_2Cl (mg/L)	0.78	0.58–1.16
NH_3-N (mg/L)	0.25	0.18–0.31
NO_2-N (mg/L)	0.043	0.006–0.088
NO_3-N (mg/L)	0.582	0.370–0.795
pH	7.96	7.65–8.09
NPOC[a] (mg/L)	0.72	0.65–0.78
Alkalinity (mg/L as $CaCO_3$)	39.6	38–41
AOB[b] (MPN/ml)	2.5	ND–6.2
HPC[c] (CFU/ml)	17.2	0–44

[a] Non-purgeable organic carbon
[b] Ammonia-oxidizing bacteria
[c] Heterotrophic plate counts

Materials and Methods

Model distribution system

A laboratory biofilm reactor system, CDC Biofilm Reactor (Model CBR 90-1, Biosurface Technologies Co., USA), was used for disinfection experiments (Fig. 1A). The reactor consisted of eight polypropylene coupon holders supported by an ultra-high-molecular-mass polyethylene ported lid. Each coupon holder accommodates three removable coupons which have an exposed surface area of 2.53 cm^2 (1.27 cm diameter and 0.3 cm thickness). A total of 24 polyvinyl chloride (PVC) coupons which support biofilm growth were held in the reactor. The lid with coupon holders and coupons was mounted in a 1-L Pyrex glass vessel with side-arm discharge port. The glass vessel was placed on a controlled stir plate to provide constant rotation of the baffled stir bar at a desired speed. Rotation of the baffle provides constant mixing and consistent shear to the coupon surface. The reactor has a 350 ml working volume.

Prior to the beginning of each run, the reactors were prepared as recommended by the manufacturer (Biosurface Technologies Co.); reactor components (reactor top/vessel, coupons, rods, baffled stir bar) were soaked in 2 M HCl for 2 h, rinsed thoroughly with Milli-Q water, air-dried, and then assembled. The assembled reactor was then autoclaved at 121℃ for 15 min, and air vent and tubing were attached to the reactor top.

Experimental setup

Three reactors were operated in parallel for a period of 3 months to allow bacterial colonization. For the first two-month period, drinking water (Table 1), produced from a nearby water

treatment plant that used coagulation-flocculation, sedimentation, sand filtration, and chloramination was pumped into the reactor at a hydraulic retention time (HRT) of 1 day. After the two-month accumulation period, the reactors were dosed for 1 month with AOB growth medium (per L distilled water): 0.5 g $(NH_4)_2SO_4$, 0.2 g KH_2PO_4, 0.04 g $MgSO_4 \cdot 7H_2O$, 0.04 g $CaCl_2 \cdot 2H_2O$, 1 ml 5 mM EDTA iron(III) sodium salt solution, pH 8.0. During bacterial accumulation period, the reactors were maintained at a fixed rotational speed of 50 rpm at a temperature of 26–30℃. At the end of the bacterial accumulation period, growth medium within the reactor was poured out, and the reactor was washed gently with sterile distilled water to eliminate loosely attached bacteria. Subsequently, the reactor was immediately refilled with 350 ml of fresh feed water.

Fig. 1B shows a schematic diagram of the experimental setup. Prior to the disinfection study, chlorine was added to the feed water so that the monochloramine residual and the background ammonia disappear by a breakpoint reaction. Approximately 0.6–0.7 ml of sodium hypochlorite solution (NaOCl, 0.5% as Cl_2) was added to 2-L drinking water, and then agitated for 1 h with stirring (100 rpm). This allowed monochloramine and ammonia-N to disappear to < 0.1 and < 0.01 mg/L, respectively. After completing the breakpoint reaction, sequential disinfectant dosage followed by pH adjustment was performed (Fig. 1B). Water pH was adjusted to 9.0 using 50% (w/v) K_2CO_3. Following pH adjustment, disinfection runs were carried out in a continuous dose of chlorite either with or without monochloramine. For application of chlorite disinfectant, an aliquot of chlorite stock solution (500 mg/L) prepared daily by dissolving sodium chlorite ($NaClO_2$, assay 80%, Sigma-Aldrich) in Milli-Q water was added into influent to the reactor, which provided an influent ClO_2^- concentration of 0.7 mg/L. For a combined chlorite/monochloramine application, monochloramine influent concentration of 1.8 mg/L was produced by reacting chlorine (2.0 mg/L) with NH_3-N (0.4 mg/L) under a constant stirring (100 rpm) for 1 h. Sodium hypochlorite (NaOCl, 0.5% as Cl_2) and ammonium chloride (NH_4Cl, 500 mg/L as NH_3-N) solutions were used for monochloramine production. Disinfectant-dosed influent was fed to the reactor at a flow rate of 60 ml/h to provide a HRT of 6 h throughout the course of disinfection experiments. One reactor was run without any disinfectant amendment as a control purpose.

Water quality analysis

Chlorine and chloramine were measured with DPD colorimetric method using a spectrophotometer (Model DR5000, Hach Co., USA). Ammonia was determined using Phenate method (4500-NH_3 F) of Standard Methods (APHA, 2005). Alkalinity and hardness were analyzed with a titration method using a mixed bromocresol green-methyl red indicator (2320 B) and the ethylenediaminetetraacetic acid titrimetric method (2340 C) of Standard Methods (APHA, 2005), respectively. TOC and non-purgeable organic carbon (NPOC) were measured using a TOC analyzer (TOC-V_{CSH}, Shimadzu, Japan). Cations and anions including nitrite and nitrate were analyzed with ion chromatograph (Model DX 500, USA) using CS12A cation column and AS9-HC anion column. Chlorite present in sample was preserved with addition of 50 mg/L of ethylenediamine (US EPA, 1997; Baribeau *et al.*, 2002). Chlorite was measured by ion chromatography (Model ICS-3000, Dionex) with suppresser and conductivity detector (US EPA, 1997). Detection limit of the method was 5 μg/L.

The number of AOB in water sample was enumerated by most probable number (MPN) method. The mineral medium used for AOB contained following ingredients per L distilled water: 0.5 g $(NH_4)_2SO_4$, 0.2 g KH_2PO_4, 0.04 g $MgSO_4 \cdot 7H_2O$, 0.04 g $CaCl_2 \cdot 2H_2O$, and 1 ml 5 mM EDTA iron(III) sodium salt solution. The medium pH was adjusted to 8.0 using 50% (w/v) K_2CO_3 before autoclaving 121℃ for 15 min. Four 10-fold serial dilutions were used. Each tube which contained 9 mL of medium was inoculated with 1 ml of original or serially 10-fold diluted samples. The tubes were incubated at 30℃ in the dark for three weeks and then tested for the presence of nitrite using 300 μl of an equal parts mixture of 0.6% dimethyl-α-naphthylamine and 0.8% sulfanilic acid (Wolfe *et al.*, 1990; Zhang *et al.*, 2008). Deep reddish color indicates the presence of nitrite, and the tube was scored positive for viable AOB. MPN index was calculated according to Standard Methods (APHA, 2005), which gives most probable numbers and corresponding 95% confidence limits.

Heterotrophic plate counts (HPC) in water samples were analyzed by spread plate method (9215 C) of Standard Methods (APHA, 2005) on R2A agar (Difco Laboratories, USA) and incubation at 25℃ for 7 days.

Biofilm sampling and analysis

Biofilm samples were obtained by removing coupon installed in the reactor and placing it in a tube containing 10 ml of 0.85% (w/v) NaCl solution. The coupon was scraped manually with a sterile disposable cell lifter (Fisherbrand, USA). The tube was vigorously vortexed for 3 min, sonicated for 3 min in an ultrasonic cleaning bath (Elmasonic S60, Elma Hans Schmidbauer GmbH & Co. KG, Germany), and vortexed again for 3 min. Subsequently the coupon was removed from the biofilm suspension. AOB and HPC in biofilm samples were determined by the same method used for bulk water analysis.

Scanning electron microscopy (SEM) analysis

Biofilm-attached PVC specimens were fixed in glutaraldehyde (2.5%), washed in phosphate-buffered saline and gradually dehydrated in ethanol series from 50% to 100%. They were then dried at the critical point of carbon dioxide and coated with Pt-Pd. Prepared samples were scanned with SEM (Model S-4300, Hitachi). SEM observations were documented through the acquisition of at least 20 representative microphotographs for each experiment.

Results and Discussion

Chlorite (ClO_2^-) is a known by-product of chlorine dioxide (ClO_2) (Gordon, 2001; Baribeau *et al.*, 2002). When ClO_2 is applied to disinfect drinking water, its decomposition and reaction lead to the formation of a wide range of ClO_2^- (Baribeau *et al.*, 2002). The presence of chlorite can affect the distribution system in a positive way. For example, McGuire and co-workers (1999) found in field work that the presence of chlorite which is formed from ClO_2 in the water distribution system suppresses the incidence of nitrification. They have also shown in laboratory study that even low concentration of chlorite (0.05 mg/L) can cause a significant reduction in the culturability of AOB over several hours. They had hypothesized that as chlorite could be transformed to ClO_2 in an acidic environment created by ammonia oxidation during nitrification, localized ClO_2 can thus change the permeability of the cell membrane, hinder the enzyme and protein function and destroy nucleic acids. Since the findings of McGuire *et al.* (1999), sodium chlorite ($NaClO_2$) has been directly applied to the effluent of drinking water treatment plant. Sodium chlorite is commercially available; it dissolves in water and forms chlorite ions. O'Connor *et al.* (2001) who carried out a field study on controlling nitrification by adding sodium chlorite to drinking water observed that chlorite ion markedly reduced nitrification phenomenon in a distribution system. Recent pilot tests of McGuire *et al.* (2006) showed that continuous or intermittent dosage of chlorite at 0.2–0.4 mg/L could significantly arrest AOB growth. Interestingly, in a study performed by Rahman *et al.* (2011), nitrifying bacteria were only inactivated at unrealistic dose level (20 mg/L) of chlorite, which is different from the findings of other studies (McGuire *et al.*, 1999, 2006; O'Connor *et al.*, 2001). This may be due to a difference in experimental setup conditions such as water quality.

Although commercially available sodium chlorite can be used in drinking water to form chlorite ion, its direct use needs meeting drinking water regulations with a maximum level of 1 mg/L (US EPA, 2012). Therefore, a tailored-dose strategy below the concentration of 1 mg/L is thought to be required for a practical application of chlorite. In the present work, chlorite was applied at a specified concentration (0.66 ± 0.01 mg/L) that complies with drinking water regulations. As shown in Fig. 1B, the reactors that harbor bacterial biofilms including AOB and heterotrophic bacteria were treated with a continuous dose of chlorite either with or without monochloramine. The chlorite (0.66 ± 0.01 mg/L) and monochloramine (1.77 ± 0.03 mg/L) feeds were practiced in the water adjusted with pH 9.0 to maintain stable disinfectant residuals (Karmakar, 1999; Skadsen, 2002; Wilczak *et al.*, 2003). During the course of disinfection, chlorite residuals were remained stable at 0.65 ± 0.02 and 0.61±0.01 mg/L in the absence and presence of monochloramine, respectively (Fig. 2). Chlorite can react with free chlorine to produce chlorate (ClO_3^-) and Cl^- and possibly ClO_2 (Baribeau *et al.*, 2002). In this work, free chlorine concentrations were determined and remained below 0.1 mg/L Cl_2. Although free chlorine concentration remained fairly low, its involvement in the reaction that reduces chlorite could not be ruled out under chloramination conditions.

AOB and heterotrophic bacteria in the biofilm and bulk water were monitored from the reactors that were being disinfected with chlorite alone or chlorite-monochloramine combined, and the results are shown in Fig. 3 and 4. A continuous dose of chlorite at 0.66 ± 0.01 mg/L resulted in a significant reduction in biofilm AOB levels (Fig. 3A). After the 6-day disinfection period, biofilm AOB levels were decreased to near or below the detection limit (0.6 MPN/cm^2). The antibacterial effect of chlorite on AOB is in agreement with the results of other studies (McGuire *et al.*, 1999, 2006; O'Connor *et al.*, 2001). As shown in Fig. 3A, the combined dose of

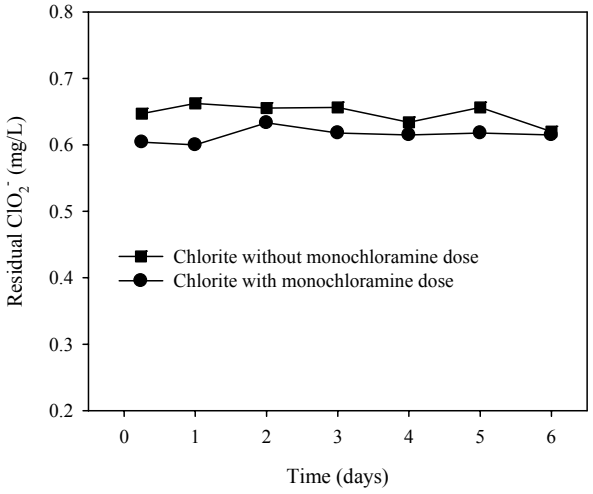

Fig. 2. Residual chlorite concentrations in the absence or presence of monochloramine.

chlorite and monochloramine disinfectants exhibited greater AOB inactivation than chlorite alone suggesting a synergistic effect in the inactivation. A possible mechanism for this synergy might be related to the ability of monochloramine to penetrate deeply into the biofilm matrix and inactivate the cells within. LeChevallier *et al.* (1990) found that monochloramine was effective in controlling biofilm microorganisms because they interacted poorly with capsular polysaccharides that protects against disinfectant. The results suggest that the combined chlorite-monochloramine application can provide an effective treatment for the control of biofilm formation in distribution systems.

It is generally recognized that the release of biofilm bacteria under a continuous flow regime can contribute to an increase of bacterial numbers in bulk water. For the reactor that was operated without disinfectant residual, bulk AOB were observed at levels greater than the detection limit (0.2 MPN/ml) throughout the experimental period (Fig. 3B). However, no bulk AOB was detected in both chlorite alone and chlorite-monochloramine combined applications, demonstrating the effectiveness of disinfection methods on bulk AOB. Distribution systems exhibit varying conditions and multiples forms of microorganisms that may influence pathogen inactivation (US EPA, 2004). For instance, in the distribution system, bacteria and viruses can be found as part of the bulk water, attached to particles, or as part of biofilms. Compared to bulk water environments biofilms can provide ecological niche that are more suited to microorganisms resulting more protection from disinfection.

Fig. 4 shows the effectiveness of chlorite alone and chlorite-monochloramine combined applications on the biofilm and bulk HPC levels. For the reactor that was operated without disinfectant residual, HPC levels were 4.1×10^5–1.9×10^6 CFU/cm^2 for biofilm and 8.0×10^4–4.4×10^5 CFU/ml for bulk water, respectively. No significant decrease in the biofilm and bulk HPC levels was found for chlorite-only application. In case of chlorite alone, the HPC levels were 6.2×10^5–3.8×10^6 CFU/cm^2 for biofilm and 1.1×10^4–3.2×10^5 CFU/ml for bulk water, respectively (Fig. 4), which were similar to the control

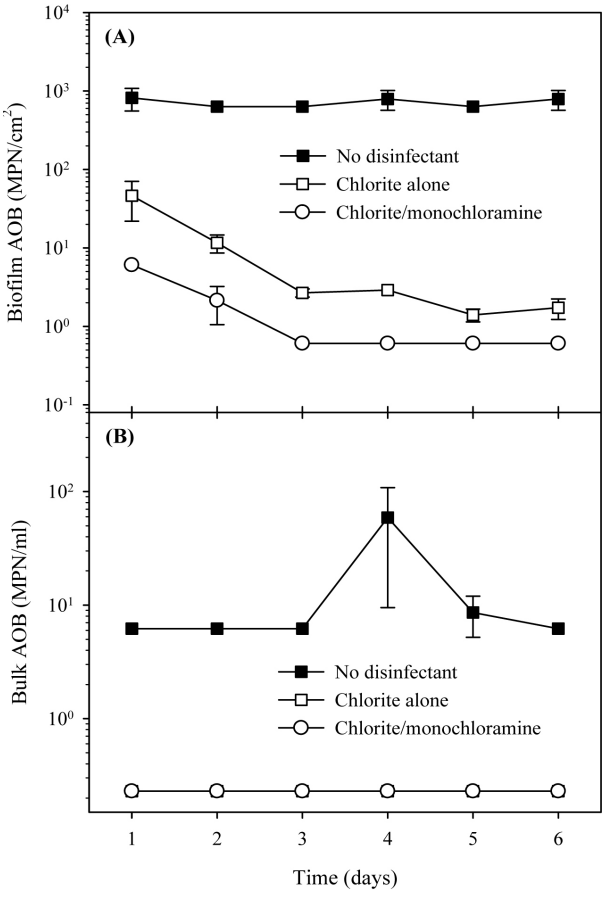

Fig. 3. Ammonia-oxidizing bacteria determined from (A) biofilm and (B) bulk water in reactors disinfected with chlorite alone or chlorite/monochloramine combined.

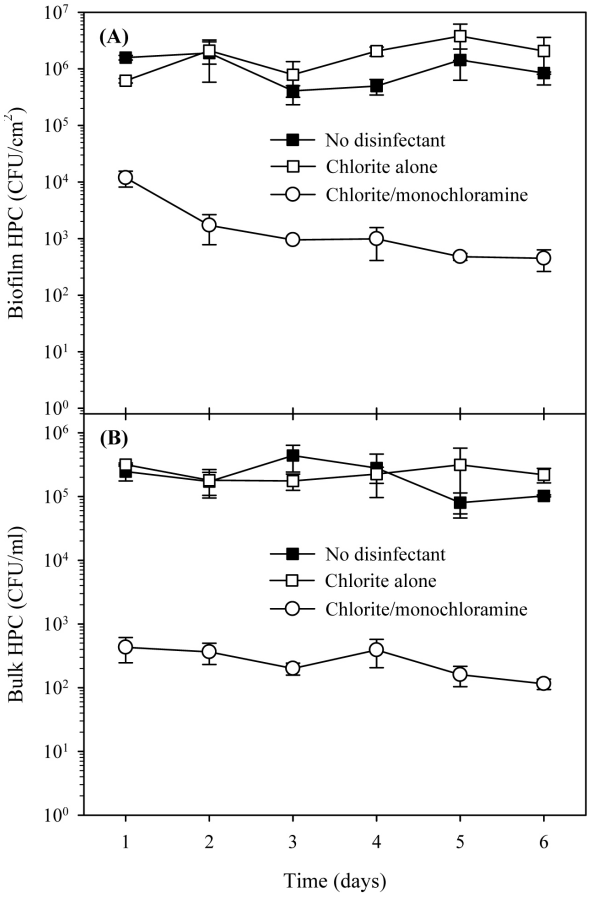

Fig. 4. Heterotrophic plate counts determined from (A) biofilm and (B) bulk water in reactors disinfected with chlorite alone or chlorite/monochloramine combined.

that received no disinfectant. These results are not contradictory to the findings of other studies (McGuire *et al.*, 1999, 2006; Gagnon *et al.*, 2005; Rahman *et al.*, 2011), where chlorite was ineffective in inhibiting heterotrophic bacteria.

In contrast to the chlorite-only application, the combination of chlorite and monochloramine resulted in a significant reduction in HPC levels both in biofilm and bulk water (Fig. 4). After the implementation of the combined dose, the HPC levels decreased to 4.5×10^2 CFU/cm^2 and 1.2×10^2 CFU/ml for biofilm and bulk water, respectively, which corresponded to log reductions of 3.1 and 3.0. In this case, a significant reduction in the HPC levels could be attributed to the antibacterial effect of monochloramine, since chlorite has no disinfection effect on them. Ling and Liu (2013) evaluated a continuous impact of monochloramine disinfection on laboratory grown biofilms through the characterization of biofilm architecture. Based on confocal laser scanning microscopy and image analysis the authors reported that chloramination could lead to 81.4–83.5% and 86.3–95.6% reduction in biofilm biomass and thickness, respectively.

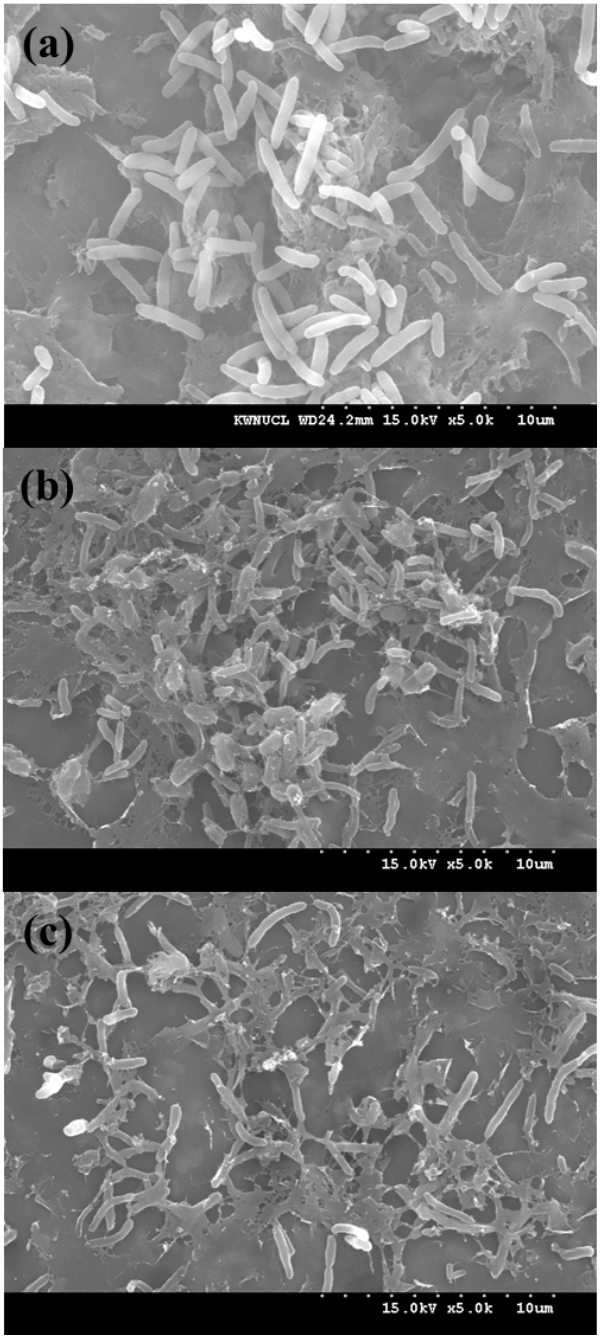

Fig. 5. Scanning electron microscopic images of biofilms formed on PVC surfaces (A) without disinfectant application, (B) after disinfection with chlorite, and (C) after disinfection with chlorite/chloramine.

SEM images were obtained from the biofilm formed on PVC slides in the absence of disinfectant and the possible damage resulting from chlorite alone and combined chlorite-monochloramine applications (Fig. 5). The observations showed structural changes in biofilms were more evident for combined addition of chlorite-monochloramine.

Consequently, we propose chlorite-monochloramine process as an effective disinfection option for controlling bacterial biofilm consisting of AOB and heterotrophic bacteria in chloraminated distribution system. Continuous dosing of chlorite in combination with monochloramine is likely to hold significant promise for controlling problems arising from the growth of nitrifying and heterotrophic bacteria in drinking water distribution system. In terms of disinfection by-product, there is evidence in the literature that the use of chlorite for nitrification control seldom form a by-product of chlorite such as chlorate and perchlorate (McGuire *et al.*, 2009). Nevertheless a further study may be needed to better understand the relevant by-product formation in the combined chlorite-monochloramine application. From a practical prospective, a tailored dose of chlorite seems to be desirable lest its concentration exceed a maximum level of 1 mg/L in drinking water distribution system, which meets drinking water regulations. In our opinion, the combined chlorite-monochloramine disinfectant can be applied to flushing process that is commonly practiced in distribution system for cleanup purposes. It would provide an effective disinfection against microorganisms inhabiting in the biofilm.

적 요

본 연구에서는 암모니아 산화세균과 종속영양세균을 포함한 세균 생물막에 대한 chlorite (0.66 ± 0.01 mg/L)의 영향을 monochloramine (1.77 ± 0.03 mg/L)의 존재 유무에 따른 조건에서 알아보았다. Chlorite 단독 또는 monochloramine과 함께 적용한 경우에서 공히 생물막과 물 시료에 존재하는 암모니아 산화

세균은 검출한계(0.6 MPN/cm^2 and 0.2 MPN/ml)에 근접한 수준으로 감소되는 것으로 나타났지만, chlorite/monochloramine으로 함께 첨가했을 때의 저해효과가 더욱 크게 나타났다. 종속영양세균의 경우 암모니아 산화세균과 달리 chlorite에 의한 저해가 거의 나타나지 않았다. 하지만 chlorite/monochloramine으로 적용한 경우 생물막과 물 시료에서 종속영양세균의 개체 수는 대조군 대비 각각 3.1 log와 3.0 log 감소하는 상당한 저해 효과를 보여주었다. 이러한 결과는 상수관망에 형성된 생물막에 존재하는 질산화 세균과 종속영양세균에 대한 효과적 제어방법 중의 하나로 chlorite와 monochloramine 혼합사용의 성공적 가능성을 제시해 준다.

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