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Effects of O_3/Cl_2 disinfection on corrosion and opportunistic pathogens growth in drinking water distribution systems

Haibo Wang[1], *Chun Hu*[1,2,3,*], *Suona Zhang*[1,3], *Lizhong Liu*[1,3], *Xueci Xing*[1,3]

1. *Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China. E-mail: hbwang@rcees.ac.cn*

2. *Research Institute of Environmental Studies at Greater Bay, School of Environmental Sciences and Engineering, Guangzhou University, Guangzhou 510006, China*

3. *University of Chinese Academy of Sciences, Beijing 100049, China*

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A B S T R A C T

The effects of O_3/Cl_2 disinfection on corrosion and the growth of opportunistic pathogens in drinking water distribution systems were studied using annular reactors (ARs). The corrosion process and most probable number (MPN) analysis indicated that the higher content of iron-oxidizing bacteria and iron-reducing bacteria in biofilms of the AR treated with O_3/Cl_2 induced higher Fe_3O_4 formation in corrosion scales. These corrosion scales became more stable than the ones that formed in the AR treated with Cl_2 alone. O_3/Cl_2 disinfection inhibited corrosion and iron release efficiently by changing the content of corrosion-related bacteria. Moreover, ozone disinfection inactivated or damaged the opportunistic pathogens due to its strong oxidizing properties. The damaged bacteria resulting from initial ozone treatment were inactivated by the subsequent chlorine disinfection. Compared with the AR treated with Cl_2 alone, the opportunistic pathogens *M. avium* and *L. pneumophila* were not detectable in effluents of the AR treated with O_3/Cl_2, and decreased to (4.60 ± 0.14) and (3.09 ± 0.12) $\log_{10}$ (gene copies/g corrosion scales) in biofilms, respectively. The amoeba counts were also lower in the AR treated with O_3/Cl_2. Therefore, O_3/Cl_2 disinfection can effectively control opportunistic pathogens in effluents and biofilms of an AR used as a model for a drinking water distribution system.

Introduction

The opportunistic pathogens in drinking water distribution systems (DWDSs) present an emerging health risk to humans, especially immunocompromised populations (Thomas and Ashbolt, 2011). Recently, opportunistic pathogens including *Legionella pneumophila*, *Mycobacterium avium*, *Pseudomonas aeruginosa*, and the free-living amoeba, such as *Acanthamoeba* spp. and *Naegleria fowleri*, have been found in DWDSs and tap water (Campese et al., 2011; Wang et al., 2013; Morgan et al., 2016). Tap water is a direct route for human exposure to opportunistic pathogens, typically by inhalation of aerosols or skin contact (Wang et al., 2012a). Amoebae can slough off or migrate into the bulk water from biofilms and make their way

* *Corresponding author*. E-mail: huchun@rcees.ac.cn (Chun Hu).

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into contact with humans through the DWDSs (Miller et al., 2015; Morgan et al., 2016).

Bacterial growth in DWDSs includes regrowth in bulk water and formation of biofilms (Langmark et al., 2007; Liu et al., 2014). Biofilms in DWDSs provide microenvironments for opportunistic pathogen growth (Berry et al., 2006). Moreover, biofilms are always formed in the corrosion scales of metal pipes (Wang et al., 2014). Gomez-Smith et al. (2015) have found many opportunistic pathogens, such as *M. avium*, in the biofilms of corrosion scales. The opportunistic pathogens in biofilms can release into the bulk water during metal release from corrosion scales, resulting in higher numbers of opportunistic pathogens in the bulk water of DWDSs with iron pipes compared to those with PVC pipes (Wang et al., 2012b). Yang et al. (2012) have indicated that thick and densely distributed corrosion scales with higher Fe_3O_4 content are more stable, and thin corrosion scales with higher α-FeOOH and $FeCO_3$ content more easily release iron into the distributed water. More iron release will result in discoloration of tap water, leading to customer complaints (Li et al., 2010). Therefore, both the iron release and opportunistic pathogens growth will affect the bulk water quality of DWDSs.

In order to control the bacterial growth in DWDSs, the final disinfection step typically involves the addition of chlorine or chloramines, and a constant disinfectant residual concentration is also required (Hwang et al., 2012; Mi et al., 2015; Moradi et al., 2017). However, opportunistic pathogens possess several adaptive features, including resistance to disinfection and tendency to form biofilms, to aid their survival (Wang et al., 2013), and many opportunistic pathogens are found in tap water (Wang et al., 2012a; Delafont et al., 2014; Thomas et al., 2014). Therefore, more powerful disinfection technologies should be applied to control opportunistic pathogens in DWDSs. Advanced disinfection methods, including ozone, hydrogen peroxide, ultraviolet (UV) and electrochemical treatment, are promising technologies for removing microorganisms (Li et al., 2011; Sun et al., 2017). Among these advanced disinfection technologies, ozone has gained attention for inactivating microorganisms in drinking water treatment, due to its strong oxidizing properties (Gunten, 2003; Alexander et al., 2016). However, ozone also needs to be employed together with chlorine to maintain a disinfectant residual in DWDSs. Ozone followed by chlorine disinfection has been used to inactivate *Cryptosporidium parvum oocysts* and *Bacillus subtilis* spores in drinking water (Corona-Vasquez et al., 2002; Cho et al., 2003). Currently, there is little known about the effect of sequential ozone and chlorine disinfection on the opportunistic pathogens in DWDSs. Moreover, the addition of ozone to disinfection will affect the biofilms bacterial community in DWDSs, which can affect the corrosion process and iron release in DWDSs (Wang et al., 2012c). Reports about the effects of O_3/Cl_2 disinfection on the corrosion and iron release in DWDSs are also scarce.

Therefore, the objective of this study is to investigate the effects of O_3/Cl_2 disinfection on corrosion and opportunistic pathogens growth in DWDSs, with Cl_2 disinfection alone acting as a reference. The mechanism for control of iron release and opportunistic pathogens growth in DWDSs by O_3/Cl_2 disinfection is also discussed.

1. Materials and methods

1.1. Materials and model distribution systems

Two annular reactors (ARs) (Model 1320LJ, BioSurface Technologies Co., USA) were used to simulate DWDSs, as described in other studies (Murphy et al., 2008; Wang et al., 2012c). The schematic of the experimental set-up was shown in Appendix A Fig. S1. In the ARs, there were two concentric glass cylinders and a rotating inner drum that supported 20 cast iron coupons. The cast iron coupons with an elemental composition (wt%) of C 3.25%, O 1.63%, Si 2.23%, P 0.08%, S 0.10%, Fe 90.48%, Cu 0.76%, Mn 0.72%, and Zn 0.75% were used. Each coupon had an exposed surface area of 17.5 cm^2 for biofilms growth. The two ARs were arranged in parallel and operated at a rotational speed of 50 r/min, according to the conditions employed to simulate DWDSs using ARs in a previous study (Murphy et al., 2008). The hydraulic retention time (HRT) of the reactors was 6 hr, which translated to a total flow rate of 2.8 mL/min into the ARs. The HRT was also consistent with the previous study using ARs (Murphy et al., 2008).

In one AR, the test water used as influent was treated with O_3 for a contact time of 12 min in a 6-L reactor. Approximately 46 mg of gaseous O_3/L oxygen-ozone was bubbled into the reactor through a porous plate in the reactor bottom at a flow rate of 200 mL/min. The residual ozone dosage in the water was about 0.55 mg/mg dissolved organic carbon (DOC). After 2 hr, the residual ozone dosage decayed to zero. Then, the AR was exposed to chlorine as the second disinfectant. Chlorine was dosed from a stock solution of NaClO. After chlorination for 4 hr, the water was pumped into the AR. The second AR was operated with chlorine as the only disinfectant. Before day 60, the initial chlorine concentration in both ARs was 2.3 mg/L, and it was increased to 2.6 mg/L from day 60 to day 240 in order to increase the chlorine residual in the effluents of both ARs. The chlorine residual and total iron concentration in effluents of both ARs were analyzed in triplicate. The results are shown in Fig. 1.

1.2. The tested water and water quality

The tested raw water was collected from a drinking water treatment plant in north of China, which was treated with coagulation using polyaluminum chloride, sedimentation, sand filtration, and biologically-activated carbon filtration (prior to entering the chlorine contact tanks). Water quality parameters (Appendix A Table S1) were measured according to standard methods (EPA of China, 2002). pH was measured using a Mettler Toledo pH Meter (FE20K, China). The total iron concentration was analyzed by an Inductively Coupled Plasma Optical Emission Spectrometer (SHIMADZU, ICPE-9820, Japan). The initial chlorine concentration and chlorine residual were measured using a HANNA HI93711 spectrophotometer (Italy) according to the DPD (*N*, *N*-diethyl-ρ-phenylenediamine) colorimetric method. DOC was analyzed *via* a total organic carbon analyzer (TOC-V_{CPH}, SHIMADZU, Japan). Differences in water quality parameters between the two ARs were analyzed using analysis of variance (ANOVA) with a significance threshold of α = 0.05.

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