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Impacts of bacteria and corrosion on removal of natural organic matter and disinfection byproducts in different drinking water distribution systems

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ABSTRACT

Impacts of bacteria and corrosion on removal of natural organic matter (NOM) and disinfection byproducts (DBPs) were studied in simulated drinking water distribution systems (DWDSs) using two annular reactors (ARs) with different disinfection. The results verified that the removal of NOM and DBPs was related very well to the corrosion and bacterial regrowth in both ARs. The initial stages (before 50 days) at which the rate of corrosion is higher in both ARs is considered as stage I. At stage I, UV controlled bacterial regrowth and more chlorine reacted with iron to promote corrosion rate, inducing the higher removal of NOM and DBPs in AR treated with UV/Cl_2 than that treated with Cl_2 alone. At stage II (from 50 days to 250 days), comparing with Cl_2 disinfection alone, UV/Cl_2 decreased the number of the total bacteria in effluents and the dehalogenation related bacteria in biofilms effectively. Moreover, UV/Cl_2 induced the bacterial genus *Dechloromonas* dominant in the biofilms, resulting in the lower corrosion rate. The lower bacterial regrowth and corrosion rate caused the less removal of NOM and DBPs. These results are very helpful for the control of NOM and DBPs in DWDSs with different disinfection.

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1. Introduction

Chlorine has been used in the disinfection of drinking water for more than 100 years (Abdullah et al., 2009). However, during chlorine disinfection process, chlorine can react with natural organic matters (NOM) to form disinfection byproducts (DBPs), including trihalomethanes (THMs) and haloacetic acids (HAAs) (Lyon et al., 2014).

After disinfection, the remaining bacteria in drinking water may result in biofilms formation on the surface of pipes when the water goes through the drinking water distribution systems (DWDSs) (Usher et al., 2014). Therefore, a constant disinfectant residual concentration is needed to control the bacterial regrowth and biofilms formation in DWDSs (Hwang et al., 2012). Moreover, because the UV/Cl_2 disinfection can effectively inactivate the microorganisms and UV can decrease the initial Cl_2 concentration which would minimize DBPs formation, UV/Cl_2 disinfection is always used in the water disinfection (Shah et al., 2011).

UV irradiation might change NOM structure and split large NOM molecules into organic acids with lower molecular weight (Choi and Choi, 2010). The changes of NOM structure resulting from the UV application might affect subsequent DBPs formation when the sequential Cl_2 disinfection was used. Liu et al. (2006, 2012) have found that UV irradiation changed NOM structure and increased chloroform, dichloroacetic acid and trichloroacetic acid formation from subsequent chlorine disinfection. However, Liu et al. (2002) and Dotson et al. (2010) have reported little effect on THMs and HAAs formation when UV was applied at 40–186 mJ/cm^2 doses.

Moreover, iron pipes are still being used in DWDSs in many countries now, and the iron pipes are usually subject to corrosion (Sarin et al., 2001; Zarasvand and Rai, 2014). The main iron corrosion products were goethite (α-FeOOH), magnetite (Fe_3O_4), lepidocrocite (β-FeOOH) and green rust (Chun et al., 2005). Many studies have reported the adsorption of NOM by Fe^0 and its corrosion products (Tsang et al., 2009; Rao et al., 2009). Some studies have also demonstrated that a variety of chlorinated and brominated DBPs are susceptible to reduction by many iron oxides such as Fe^{II}/goethite, Fe^{II}/magnetite and carbonate green rust (Chun

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et al., 2007; Lee et al., 2008). In addition, many halogenated DBPs can be reduced by zero-valent iron (Fe^0) (Hozalski et al., 2001; Lee et al., 2007). Moreover, pipe corrosion can also protect microorganisms in biofilms from disinfection, and some bacteria including *Afipia* and *Methylobacterium* in biofilms can reduce some DBPs (Zhang et al., 2009). Therefore, Fe^0, corrosion products and the biofilms in the cast iron pipes may play great roles on the changes of NOM and DBPs in DWDSs.

Although some studies have evaluated the spatial and temporal variability of DBPs in small and real DWDSs (Shanks et al., 2013; Guilherme and Rodriguez, 2015), there is little known about the reason for removal of NOM and DBPs in DWDSs with different disinfection. Therefore, impacts of bacteria and corrosion on the removal of NOM and DBPs in DWDSs with Cl_2 disinfection alone and UV/Cl_2 disinfection were studied in this paper. The relationship among NOM and DBPs removal, corrosion process and bacterial regrowth in DWDSs was also discussed.

2. Materials and methods

2.1. The simulated drinking water distribution systems

The simulated DWDSs were set up using two annular reactors (ARs) (Model 1320LJ, BioSurface Technologies Co., USA). Twenty cast iron coupons were housed in the rotating inner drum of the AR. The exposed surface area of each cast iron coupon was 17.5 cm^2, and the main composition of the coupon was Fe (90.48%, wt%). Both ARs were run at a rotational speed of 50 rpm, and the hydraulic retention time for both ARs was 6 h, which was determined by the flow rate of 2.5 mL/min.

Two ARs were run in parallel for 250 days. The water as influents in one AR was treated with chlorine disinfection alone, and the other one was treated with UV/Cl_2 disinfection. The fluence of the UV lamp was determined by ultraviolet irradiation meter (Handy, Beijing, China), and a fluence of 40 mJ/cm^2 was used in this study. The solution of sodium hypochlorite (NaClO) was used as the chlorine disinfectant. Because free chlorine could react with ammonia nitrogen in the water to form chloramines, total chlorine concentration was measured using HANNA HI93711 spectrophotometer (Italy). In order to control different total chlorine concentration in the effluents of both ARs, different concentration of chlorine disinfectant was prepared and added into the disinfection tank, and then the total chlorine concentration in the influents and effluents of both ARs was measured. Before 50 days, the corrosion in both ARs was very fast because new cast iron coupons were used. This stage was stage I, and the initial total chlorine concentration in the influents of both ARs was about 0.85 mg/L. After 50 days, the relatively stable corrosion scales were formed on the surface of cast iron coupons in both ARs, and ARs were run into stage II. At stage II, the initial total chlorine concentration in the influents of both ARs were different, however, both of the total chlorine concentration in effluents of two ARs was controlled at 0.08 mg/L. This stage continued for 200 days.

2.2. NOM and DBPs analysis

The tested water was collected from a drinking water treatment plant in Beijing city of China. The water was treated by coagulation-settlement, sand filtration, and ozonization-biological activated carbon. The tested water had a pH 7.16–7.77, turbidity 0.08–0.25 NTU, alkalinity 104–150 mg $CaCO_3$/L, DOC 1.78–2.19 mg/L, Cl^- 19.8–32.7 mg/L, NO_3^- 6.21–10.4 mg/L and SO_4^{2-} 50.4–103 mg/L. The water was disinfected and pumped into both ARs in laboratory. The effluents of both ARs were taken weekly, and NOM and DBPs were analyzed.

A total organic carbon analyzer (TOC-V CPH, Shimadzu, Japan) was used to detect the dissolved organic carbon (DOC) concentration. High performance size exclusion chromatography (HPSEC) was used to measure the molecular size distribution of NOM. Weight-averaged molecular weight (*Mw*) was calculated according to the methods of Her et al. (2002). Fourier transform infrared (FTIR) spectroscopy (Thermo Nicolet NEXUS 670) was used to analyze the structure of NOM, and it was set to scan from 4000 to 400 cm^{-1}.

DBPs including THMs and HAAs were detected by a gas chromatograph equipped with an electron capture detector (GC-ECD, 6890N; Agilent). THMs were measured using the following temperature program: hold at 35 °C for 4 min and ramp to 65 °C at 2 °C/min. HAAs were measured using the following temperature program: hold at 35 °C for 4 min, ramp to 260 °C at 10 °C/min and hold for 4 min.

Differences in water quality parameters between the two ARs were analyzed using analysis of variance (ANOVA) with a significance threshold of $\alpha = 0.05$.

2.3. Corrosion process and corrosion products characterization

The corrosion rate of the cast iron coupons in different ARs was characterized by weight loss method according to our previous works (Wang et al., 2015). The scraped coupons were freeze-dried under vacuum conditions, and weighted to determine the weight loss.

Moreover, the corrosion behavior of the cast iron coupons were undertaken on coupons of the size 1 × 1 cm embedded in a three-electrode electrochemical cell. Working electrode potentials were referred to a saturated calomel electrode (SCE). The Tafel curves were recorded by scanning the potential −1000 mV and 0 mV versus the open circuit potential (OCP) at a sweep rate of 2 mV/s. The Electrochemical Workstation CHI 660 D (CH Instrument, China) was used to analyze the corrosion current densities (i_{corr}).

The X-ray powder diffractometer (XRD, X'Pert PRO MPD; PANalytical, The Netherlands) was used to determine the chemical composition of the corrosion products on the surface of cast iron coupons in different ARs, using Cu Kα radiation (λ = 1.5406 Å) at a scanning range of 2θ = 10–70°.

Moreover, the total iron concentration and turbidity in effluents of both ARs were measured according to standard methods (EPA of China, 2002). The differences of the water quality parameters between the two ARs at different stages were also analyzed using analysis of variance (ANOVA) with a significance threshold of $\alpha = 0.05$.

2.4. Quantitative real time PCR (qPCR) and 454 pyrosequencing

A FastDNA spin kit for soil (Qbiogene, Solon, OH) was used to extract the DNA of all samples. DNA quantity and quality were determined by a NanoDrop (ND-1000, NanoDrop, USA). The primer pairs 1369F (5′-CGGTGAATACGTTCYCGG-3′) and 1492R (5′-ACGGCTACCTTGTTACGACT-3′) with the probe 1389F (5′- CTTGTACACACCGCCCGTC-3') were used to quantify the 16S rRNA of total bacteria (Suzuki et al., 2000). qPCR of all samples was performed in triplicate with the Applied Biosystems 7300 system, and the annealing temperature was 60 °C. The R^2 and amplification efficiency values for quantification were 0.992 and 96.5%, respectively.

For 454 pyrosequencing, DNA was amplified by PCR with forward primer 341F (5′-CCTACGGGAGGCAGCAG-3′) and reverse primer 1073R (5′-ACGAGCTGACGACARCCATG-3'). The PCR products were purified with AxyPrep DNA Gel Extraction Kit (Axygen, USA). Roche massively parallel 454 GS-FLX Titanium sequencer (Roche 454 Life Sciences, Branford, CT, USA) was used for the

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