Chemosphere 85 (2011) 1146–1153

Contents lists available at SciVerse ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Chlorine residuals and haloacetic acid reduction in rapid sand filtration

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ARTICLE INFO

Article history:
Received 10 May 2011
Received in revised form 15 August 2011
Accepted 18 August 2011
Available online 4 October 2011

Keywords:
Rapid sand filtration
Biodegradation
Chlorine residual
Haloacetic acid
Backwash

ABSTRACT

It is quite rare to find biodegradation in rapid sand filtration for drinking water treatment. This might be due to frequent backwashes and low substrate levels. High chlorine concentrations may inhibit biofilm development, especially for plants with pre-chlorination. However, in tropical or subtropical regions, bioactivity on the sand surface may be quite significant due to high biofilm development—a result of year-round high temperature. The objective of this study is to explore the correlation between biodegradation and chlorine concentration in rapid sand filters, especially for the water treatment plants that practise pre-chlorination. In this study, haloacetic acid (HAA) biodegradation was found in conventional rapid sand filters practising pre-chlorination. Laboratory column studies and field investigations were conducted to explore the association between the biodegradation of HAAs and chlorine concentrations. The results showed that chlorine residual was an important factor that alters bioactivity development. A model based on filter influent and effluent chlorine was developed for determining threshold chlorine for biodegradation. From the model, a temperature independent chlorine concentration threshold ($Cl_{threshold}$) for biodegradation was estimated at 0.46–0.5 mg L^{-1}. The results imply that conventional filters with adequate control could be conducive to bioactivity, resulting in lower HAA concentrations. Optimizing biodegradable disinfection by-product removal in conventional rapid sand filter could be achieved with minor variation and a lower-than-$Cl_{threshold}$ influent chlorine concentration. Bacteria isolation was also carried out, successfully identifying several HAA degraders. These degraders are very commonly seen in drinking water systems and can be speculated as the main contributor of HAA loss.

1. Introduction

It is common to use pre-oxidation for algae and microbial control in areas with higher average temperature (subtropical or tropical weather) for surface water treatment. In places where water resources are limited to river and reservoirs, the major problems are usually the high organic contents and algae. In such cases, pre-chlorination practise is an essential step to prevent algal growth in the treatment train.

Most water treatment plants (WTPs) in Taiwan practise conventional treatment processes which include pre-chlorination, coagulation, sedimentation, rapid sand filtration and post-chlorination. Chlorine gas and sodium hypochlorite are the most common disinfectants used for both pre- and post-chlorination. Therefore, chlorinated disinfection by-products (C-DBPs) such as trihalomethanes (THMs), haloacetic acids (HAAs), haloketones and haloacetaldehydes are commonly found in pre-chlorinated water.

A significant amount of C-DBPs is formed during pre-chlorination process and cannot be removed effectively by conventional treatment processes. These DBPs may increase the DBP levels in distribution systems compared to plants without pre-chlorination. As indicated in previous studies, for THMs and HAAs combined (all chlorinated and brominated species), it accounts for 80% and 84% (by weight) of total detectable DBPs in drinking water systems for Korea and USA (Utah), respectively (Neiminski et al., 1993; Shin et al., 1999). National-wide investigations in the UK revealed that 27.6–50.9 and 35–96 μg L^{-1} of mean total THMs and HAA, and an average total HAA concentration of 21.3 μg L^{-1} were observed in the drinking water (Malliarou et al., 2005; Zhang et al., 2010). In Taiwan, the total THMs and HAA_5 (monochloroacetic acid, MCAA; dichloroacetic acid, DCAA; trichloroacetic acid, TCAA; dibromoacetic acid, DBAA; bromochloroacetic acid, BCAA) concentrations ranged from 0–119.0 and 0.2–46.7 μg L^{-1} in the distribution systems (Chang et al., 2010).

The biodegradability of THMs and HAAs have been well-documented. An aerobic biodegradation of THMs is not thermodynamically favorable due to its high oxidization states (Vogel et al., 1994). On the other hand, HAA is shown to be biodegradable. According to some studies on HAA degradation in WTPs and distribution systems, biodegradation was the major removal

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doi:10.1016/j.chemosphere.2011.08.037

process. HAA may undergo biodegradation within locations where high bioactivity existed, such as distribution systems, storage tanks (Singer et al., 1993; Chen and Weisel, 1998; Landmeyer et al., 2000), granular activated carbon filters (Xie and Shou, 2002; Kim and Kang, 2008) and sand filtrations (Rodriguez et al., 2007).

Biological activity is commonly found in slow sand filters. DBP formation potentials reduction and HAA degradation were observed in slow sand filtration process (Collins et al., 1992; Rodriguez et al., 2007). In contrast, the removal of HAA was not commonly observed in rapid sand filtration with only few exceptions which reported the observation of DCAA degradation in warm seasons (Rodriguez et al., 2007). Other researches indicated that HAA concentrations remained in similar levels before and after rapid sand filtration year around (Rodriguez et al., 2007; Kim and Kang, 2008). Slow sand filtration has longer contact time and higher biofilm formation due to typically no backwash in slow sand filters; on the contrary, rapid sand filtration has shorter contact time (min) and frequent backwashes which inhibit the biofilm development.

Several factors are associated with the biological activity in drinking water treatment systems. Studies have shown that both low chlorine residuals and high heterotrophic bacteria counts (HPCs) were associated with HAA biodegradation (Speight and Singer, 2005; Tung and Xie, 2009). Although low chlorine concentration is a necessary condition, it cannot ensure a HAA decrease (Speight and Singer, 2005). Neither can HPC value act alone as an index for HAA loss (Tung and Xie, 2009). Information regarding factors of biodegradation in conventional rapid sand filters has been rather limited. Therefore, it is the objectives of this study to explore these factors, particularly on chlorine concentrations and its impacts, which affect bioactivity in rapid sand filters.

In this study, both lab-scale column experiments and field investigations have been conducted to explore the biodegradation in rapid sand filters, using HAA as an example. A simulative model based on measurements of chlorine concentration and biodegradation kinetics has been developed to predict biodegradation performance. In addition, several bacteria contributing to HAA biodegradation have been successfully isolated. These results could provide a better understanding of those factors affecting bioactivity in rapid sand filtration. They can also be applied to help control the biodegradable DBPs in finished water.

2. Materials and methods

2.1. WTPs and sampling methods

Eighteen WTPs (Plant A–Plant R) in Taiwan have been investigated. In Supplementary material (SM), Table SM-1 shows the general water parameters of the target plants. Plant A to Plant L practise conventional treatment processes with both pre- and post-chlorination. For Plant M to Plant R, chlorine is applied only at pre-chlorination step and the dosage is high enough to maintain the chlorine residual regulation in the finished water (free chlorine as 0.2–1.0 mg L^{-1}).

Samples were collected from the influent (effluent of sedimentation) and the effluent of rapid sand filters. To avoid interference, samples were taken at least 8 h after filter backwash. General water parameter including pH, temperature, free and total chlorine residuals were measured on site at the time of sampling. Free and total chlorine residuals were measured by N,N-diethyl-p-phenylenediamine colorimetric method (515 nm) (Hach Company, Loveland, CO, USA). Water samples for HAA analysis were stored in 40 mL glass vials with 65 mg ammonium chloride to quench the free chlorine. Samples were sealed after acidified by sulfuric acid and were stored at 4 °C until laboratory analysis was conducted (<14 d).

2.2. Sample analysis

HAA_5 concentrations were analyzed by the USEPA method 552.3 (USEPA, 2006), with the minimum detection limits (MDL) in μg L^{-1} of 0.8 for MCAA; 1.1 for DCAA; 0.7 for TCAA and BCAA; 0.6 for DBAA. The dissolved organic carbon (DOC) concentrations (filtrated by 0.5 μm glass fiber filter) were analyzed, using wet-oxidation method with a total organic carbon analyzer (OI-Analytical, Model 1010TOC, College Station, TX, US).

2.3. Column studies

Column studies were prepared to explore the chlorine residual effects on HAA biodegradation and biodegradation kinetics. These experiments were conducted by using the same column setup with different experimental conditions. Before the experiment, a batch of sand (0.3–1 mm in diameter) freshly collected from plant A was packed into glass columns (30 cm in height and 1.75 cm id) and equilibrated with synthetic influent, sterilized tap water with 0.4 mg L^{-1} DOC and spiked with 50 μg L^{-1} each HAA for 48 h. The bioactivity of sands in each column was assumed to be the same initially (0 h).

2.3.1. Column study with constant influent chlorine concentrations

To explore the chlorine residual effects on HAA biodegradation, 12 columns were used. The control column was sterilized to eliminate bioactivity by autoclaving at 121 °C for 20 min. The column influent was prepared by using sterilized tap water (DOC = 0.4 - mg L^{-1}) spiked with three HAAs (MCAA, DCAA, and TCAA 50 μg L^{-1} each). Different chlorine concentration was applied for each column, and the chlorine concentrations throughout the study have been monitored periodically. The empty bed contact time (EBCT) for each column was around 10–13 min (0.37–0.45 m^3 m^{-2} h^{-1}). Samples were taken periodically up 36 h for each column from both column influents and effluents.

2.3.2. Column study with gradient influent chlorine concentrations

Column studies were applied to explore the effects of chlorine variation on microbial activity. Two columns were setup for this experiment, the influents were sterilized tap water spiked with three HAAs (50 μg L^{-1} each) as in Section 2.3.1. One of them was sterilized as the control. The EBCT was 10 min. The influent was chlorine free in the beginning of the experiment. It was gradually increased for every 8–10 h (samples were taken at the 6 and 8 h) until the total chlorine residuals reached 1.13 mg L^{-1}. When influent chlorine residual reached its maximum concentration at 1.13 mg L^{-1}, the effluent total chlorine residual was 0.91 mg L^{-1}. At the end of the experiment, the influent was switched back to chlorine free condition.

2.4. Microbial isolation for HAA degrading bacteria

Silica sand collected from Plant A was used for bacterial cultivation and isolation. The biomass on the silica sand was retrieved by ultrasound bath method described elsewhere (Buesing and Gessner, 2002) with slight modification. In short, 20 mL of phosphate buffered saline (PBS; Bioman, Taiwan) buffer was mixed with 30 g fresh sand in a 200 mL-presterilized-beaker. Sonication (130 W) was applied for detaching biomass. The supernatant was taken for further microbial cultivation and isolation. 50 μL bacterial suspension with different dilutions were spread directly on Reasoner's 2A (R2A) (van der Linde et al., 1999) medium and were incubated at 30 °C for up to 10 d. Viable colonies were further purified at least 3 times by streak plate method.

Bacterial isolates were obtained by differentiating the viable colonies, using ribotyping analysis. Polymerase chain reaction