Chemosphere 136 (2015) 190–197

Contents lists available at ScienceDirect

Chemosphere

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

Disinfection byproduct formation during biofiltration cycle: Implications for drinking water production

R. Delatolla [a,*], C. Séguin [a], S. Springthorpe [b], E. Gorman [c], A. Campbell [c], I. Douglas [c]

[a] Department of Civil Engineering, University of Ottawa, Ottawa, Ontario K1N 6N5, Canada
[b] Center for Research on Environmental Microbiology, University of Ottawa, Ottawa, Ontario K1H 8M5, Canada
[c] City of Ottawa, Ottawa, Ontario K1P 1J1, Canada

HIGHLIGHTS

- TOC and DOC showed slight decrease in concentration after full-scale biofiltration.
- Full-scale biofiltration showed a reduction in TTHM and HAA5 formation potentials.
- Shorter filtration cycle times promote the largest reduction in $TTHM_{fp}$ and $HAA5_{fp}$.
- Higher quantities of biofilm and attached cells correspond to higher DBP_{fp}.
- Biofilm growth in biofilters lowers effectiveness of biofiltration on DBP removal.

ARTICLE INFO

Article history:
Received 25 July 2014
Received in revised form 15 March 2015
Accepted 2 May 2015
Available online 22 May 2015

Keywords:
Disinfection byproducts
Biofiltration
Filtration cycle
Biofilm
Total organic carbon

ABSTRACT

The goal of this study was to investigate the potential of biofiltration to reduce the formation potential of disinfection byproducts (DBPs). Particularly, the work investigates the effect of the duration of the filter cycle on the formation potential of total trihalomethanes (TTHM) and five species of haloacetic acids (HAA5), dissolved oxygen (DO), organic carbon, nitrogen and total phosphorous concentrations along with biofilm coverage of the filter media and biomass viability of the attached cells. The study was conducted on a full-scale biologically active filter, with anthracite and sand media, at the Britannia water treatment plant (WTP), located in Ottawa, Ontario, Canada. The formation potential of both TTHMs and HAA5s decreased due to biofiltration. However the lowest formation potentials for both groups of DBPs and or their precursors were observed immediately following a backwash event. Hence, the highest percent removal of DBPs was observed during the early stages of the biofiltration cycle, which suggests that a higher frequency of backwashing will reduce the formation of DBPs. Variable pressure scanning electron microscopy (VPSEM) analysis shows that biofilm coverage of anthracite and sand media increases as the filtration cycle progressed, while biomass viability analysis demonstrates that the percentage of cells attached to the anthracite and sand media also increases as the filtration cycle progresses. These results suggest that the development and growth of biofilm on the filters increases the DPB formation potential.

1. Introduction

A dilemma exists in current water treatment practice between microbial control through disinfection and the formation of harmful disinfection byproducts (DBP) (Rook et al., 1982; Arora et al., 2001; Hua and Reckhow, 2007; Badawy et al., 2012; Richardson and Postigo, 2012). In North America, chlorination has been the primary method of disinfection used for drinking water. In recent years, both the US and Canada have applied more stringent regulations with respect to the concentration of total trihalomethanes (TTHM) and five species of haloacetic acids (HAA5) (Richardson and Postigo, 2012; Health Canada, 2012), due to their direct correlation with higher incidences of cancer as well as adverse reproductive effects (Nieuwenhuijsen et al., 2000; Porter et al., 2005). The maximum allowable concentration (MAC) for TTHMs and HAA5 in the US drinking water is 0.080 mg/L and 0.060 mg/L, respectively (USEPA, 2006). Canada has slightly higher MACs of 0.100 mg/L and 0.080 mg/L for TTHMs and HAA5s, respectively

* Corresponding author at: Department of Civil Engineering, University of Ottawa, 161 Louis Pasteur, Ottawa, Ontario K1N 6N5, Canada.
E-mail address: robert.delatolla@uottawa.ca (R. Delatolla).

http://dx.doi.org/10.1016/j.chemosphere.2015.05.001
0045-6535/

(Health Canada, 2012). The European Union has an established MAC for TTHMs of 0.100 mg/L and has yet to set the limit on HAA5 (Council Directive 98/83/EC).

DBP presence, variety and quantity is a complex function of many interacting factors, including the disinfectant used, the source, composition and concentration of the natural organic matter (NOM) along with the microbes present and their state of activity (Krasner, 2009). The multiple chemical and biological sources and pathways for DBP formation are all potentially confounded, and also overlap with the mechanisms for DBP elimination. As such the relative contribution of any particular component or mechanism to DBP formation or destruction is often difficult to quantify and hence the contribution to DBP formation is commonly presented as the disinfection byproduct formation potential (DBP_{fp}).

NOM is a known precursor for DBP formation in drinking water sources and is found to exist in concentrations that range between 2 and 10 mg/L. NOM in source waters consists mainly of microorganisms, complex organic matter including humic and fulvic acids, as well as naturally occurring degradation products, including amino acids, fatty acids, phenols, sterols, sugars, hydrocarbons, urea, porphyrins and polymers (Gopal et al., 2007). Dissolved organic matter (DOM) is considered to constitute the majority of NOM; where a significant number of the smaller sized bacteria present in natural waters (<0.45 μm) would be included in DOM measurements (Hobbie et al., 1977).

Thus, microorganisms in source water and potable water might ultimately contribute to the formation of DBP during disinfectant contact (Wang et al., 2012, 2013). However, bacteria and fungi can also degrade components of DOM and DBP precursors (Hur et al., 2013), in addition to degrading DBP once formed (McRae et al., 2004). To take advantage of these metabolic activities, biofiltration is becoming increasingly adopted or trialed in distributed potable water to not only enhance the production of biologically stable waters but to also promote the degradation of DOM and DBP precursors (Carlson and Amy, 1998; Lou et al., 2009; Liao et al., 2013). Although biologically active filtration has been shown to remove disinfectant by-product formation potential during the treatment process (Graham, 1999; Hozalski et al., 1999; Simpson, 2008), current research has demonstrated variable rates of DBP precursor removal along with increases in DBP precursor formation due to biological filtration (Toor and Mohseni, 2007; Bond et al., 2009). This Increase in DBP formation during biofiltration has been recently correlated to backwashing events and biofilm growth in a granulated activated carbon media filter (Liu et al., 2010).

The overall objective of this work was to study the effect of elapsed biofiltration time between backwashing events on TTHM and HAA5 formation potential in a full-scale water treatment plant (WTP) that operates a passive biofiltration unit composed of anthracite and sand media. Particularly, the study investigates the effects of biofiltration duration throughout a filter cycle on (i) the formation potential of TTHMs and HAA5s; (ii) dissolved oxygen (DO), organic carbon, nitrogen and total phosphorous concentrations; and (iii) biofilm coverage of the filter media and biomass viability of the cells embedded in the attached biofilm.

2. Materials and methods

2.1. Water treatment plant and source water description

The Britannia WTP services the City of Ottawa, Ontario, Canada with a design treatment rate of 360,000 m^3/d. The Britannia treatment train consists of the multiple treatment units, as shown in Supporting Information (Fig. S1). Source water is drawn solely from the Ottawa River. The treatment train includes flocculation (with aluminium sulfate (alum), pH adjustment and activated silica), sedimentation, biofiltration via dual-media (anthracite/sand) filters and disinfection (chlorine, pH correction and ammonia addition); chloramine is used as a secondary disinfectant and fluoride is added.

The average empty bed contact time (EBCT) for the filters during the study was approximately 20 min. The two filters used for this study have a surface area of 135 m^2 each and are comprised of 0.56 m of anthracite overlaying 0.30 m of sand. The velocity rise rate of the filters during the study period was approximately 2.5 m/h. The backwash events for the filters occurred every 72 h. The slow filtration rates and the correspondingly longer filtration cycles of the Britannia WTP are representative of water conservation efforts of the population served by the drinking water plant. Non-chlorinated, filtered water was used as backwash water. Each backwash event included 2 min of fluidization at a low reverse flow rate of 16.9 m/h followed by 3 min of fluidization at a high reverse flow rate of 44.6 m/h and 7 min of non-fluidized wash at a reverse flow rate of 13.8 m/h.

2.2. Experimental plan and analytical methods

Influent and effluent samples were collected from two filters, over the course of two filtration cycles, between June and July at the Britannia WTP. Filter influent samples were collected after 0, 24, 48 and 69 h of filtration. Effluent samples were collected one EBCT after the collection of the influent to minimize effects of influent variance. The samples were analyzed for various constituents including DO, total organic carbon (TOC), dissolved organic carbon (DOC), ammonia (NH_4^+/NH_3), nitrite (NO_2^{2-}), nitrate (NO_3^-), and total phosphorous (TP). Samples were also analyzed for TTHM formation potential ($TTHM_{fp}$) and HAA5 formation potential ($HAA5_{fp}$). In addition, 10 filter media samples of anthracite and sand were collected from filter 1 before and after the backwash cycles. These samples were analyzed for biofilm coverage of the biofilter media and cell viability analysis of the attached bacteria.

DO was measured using an Orion RDO probe and meter (Thermo Fisher Scientific, Walthan (MA), USA). NH_4^+/NH_3, NO_2^-, NO_3^-, TP, TOC, DOC, $TTHM_{fp}$ and $HAA5_{fp}$ samples were collected and stored in the refrigerator at 4 °C and analyzed within 24 h of sampling. NH_4^+/NH_3, NO_2^- and NO_3^- were analyzed according to *Standard Methods for the Examination of Water and Wastewater* (APHA, 1989), methods 4500-NH_3 C, 4500-NO_2^- B and 4500-NO_3^- B, respectively. TP was analyzed using the Ontario Ministry of the Environment (MOE) method RTNP-E3367 (February 12, 2007) acid digestion in combination with an auto-analyzer (Skalar, Breda, Netherlands) (MOE, 2010). TOC, DOC, $TTHM_{fp}$ and $HAA5_{fp}$ samples were collected in preserved, headspace-free, cleaned vessels. TOC and DOC measurements were performed according to *Standard Methods for the Examination of Water and Wastewater*, method 5310 B (APHA, 1989), using the Apollo 9000 TOC/TN Analyzer (Teledyne Tekmar, Mason (OH), USA). DOC samples were first filtered using a 0.45 μm filter and then measured according to the same method as TOC.

DBP_{fp} ($TTHM_{fp}$ and $HAA5_{fp}$) were determined using *Standard Methods for the Examination of Water and Wastewater* (APHA, 1989), method 6251B with a contact time of 1 h; which is representative of the contact time used at the Britannia WTP. The contact time of one hour used in this study is shorter than conventionally used; however it was selected in this study to maintain a direct correlation between the protocol and the operation of the plant. The samples were filtered with a 0.45 μm filter prior to dosing the sample to 1.5 mg/L of chlorine and storing the sample in the dark for 1 h prior to DPB analysis. THM concentrations were measured according to U.S. EPA Method 524.4 (EPA, 2013), with this method showing a relative standard deviation of