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Pilot investigation of two-stage biofiltration for removal of natural organic matter in drinking water treatment

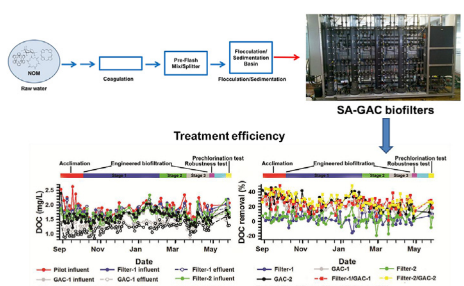
Jie Fu [a, c], Wan-Ning Lee [a], Clark Coleman [b], Melissa Meyer [a], Jason Carter [b], Kirk Nowack [b], Ching-Hua Huang [a, *]

[a] School of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, GA 30332, United States
[b] ARCADIS U.S., Inc, 2410 Paces Ferry Rd., Suite 400, Atlanta, GA 30339, United States
[c] Department of Environmental Science & Engineering, Fudan University, Shanghai 200433, China

HIGHLIGHTS

- A pilot study employing SA-GAC biofilters was conducted to remove NOM in water.
- GAC contactors played the dominant role in NOM removal.
- Middle-upper part was the critical zone in GAC contactors for the removal of NOM.
- Nutrients supplementation slowed down the DOC breakthrough.
- The most explanatory water parameter in GAC contactors was UV_{254}.

GRAPHICAL ABSTRACT



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ABSTRACT

A pilot study employing two parallel trains of two-stage biofiltration, i.e., a sand/anthracite (SA) biofilter followed by a biologically-active granular activated carbon (GAC) contactor, was conducted to test the efficiency, feasibility and stability of biofiltration for removing natural organic matter (NOM) after coagulation in a drinking water treatment plant. Results showed the biofiltration process could effectively remove turbidity (<0.1 NTU in all effluents) and NOM (>24% of dissolved organic carbon (DOC), >57% of UV_{254}, and >44% of $SUVA_{254}$), where the SA biofilters showed a strong capacity for turbidity removal, while the GAC contactors played the dominant role in NOM removal. The vertical profile of water quality in the GAC contactors indicated the middle-upper portion was the critical zone for the removal of NOM, where relatively higher adsorption and enhanced biological removal were afforded. Fluorescence excitation-emission matrix (EEM) analysis of NOM showed that the GAC contactors effectively decreased the content of humic-like component, while protein-like component was refractory for the biofiltration process. Nutrients (NH_4-N and PO_4-P) supplementation applied upstream of one of the two-stage biofiltration trains (called engineered biofiltration) stimulated the growth of microorganisms, and showed a modest effect on promoting the biological removal of small non-aromatic compositions in NOM. Redundancy analysis (RDA) indicated influent UV_{254} was the most explanatory water quality parameter for GAC contactors' treatment performance, and a high load of UV_{254} would result in significantly reduced removals of UV_{254} and $SUVA_{254}$.

* Corresponding author.
E-mail address: ching-hua.huang@ce.gatech.edu (C.-H. Huang).

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1. Introduction

Natural organic matter (NOM) has a wide variety of chemical compositions and molecular sizes, and is ubiquitous in all surface, ground, and soil waters (Matilainen and Sillanpää, 2010). NOM in water is generally derived from living or decayed vegetation and microbial decomposition processes, having both aquagenic and pedogenic sources (Goel et al., 1995). The water-derived NOM (such as algal and cyanobacterial biomass) is low in phenolic and aromatic constituents, whereas soil-derived NOM (from terrestrial vegetation) tends to have a higher lignin content and aromatic fraction (Rashid, 1985). In terms of hydrophobicity, the components in NOM can be divided into hydrophobic and hydrophilic fractions. The hydrophobic fraction is generally humic substances, comprising of humic acids (HA), fulvic acids (FA) and humin, which makes up approximately 50% of the total organic carbon (TOC) in water (Thurman, 1985). Compared with hydrophobic NOM of high content of aromatic carbon, phenolic structures and conjugated double bonds, hydrophilic NOM contains more aliphatic carbon and nitrogenous compounds, such as carbohydrates, sugars and amino acids (Matilainen and Sillanpää, 2010).

NOM may cause many problems in water treatment processes and distribution networks. It leads to undesirable color, taste and odor, and reacts with common disinfectants to produce a variety of toxic disinfection by-products (DBPs) including trihalomethanes (THMs) and haloacetic acids (HAAs) (Sadiq and Rodriguez, 2004). Residual NOM can promote bacterial regrowth and pipe corrosion in the drinking water distribution system (Meylan et al., 2007). NOM can also reduce the overall efficiency of a water treatment plant through increased chemical dosages, interference with the removal of other contaminants, and filter fouling (De Ridder et al., 2011; Shen and Schäfer, 2015). Due to the air and surface water temperature rising, increase in rainfall intensity, atmospheric carbon dioxide (CO_2) increase and decline in acid deposition, a considerable increase in the content of NOM (especially for the hydrophobic fraction) in surface waters has been observed to occur during the past 20 years (Korth et al., 2004; Worrall and Burt, 2009).

NOM is a key parameter with respect to design and operation of water treatment processes, and the most common and economically feasible processes to remove NOM are coagulation and flocculation followed by sedimentation/flotation and granular media filtration (Matilainen and Sillanpää, 2010). Coagulation removes most of the NOM and especially its hydrophobic fraction, which is generally more aromatic and larger in molecular size than the hydrophilic fraction (Sharp et al., 2006). Unfortunately, the residual NOM after coagulation generally retains significant DBP formation potential (DBPFP), which is largely due to the hydrophilic fraction of lower size and charge. In some cases, compliance with existing DBP regulations (e.g., Stage 2 DBP Rule of US EPA) necessitates additional treatment to remove this residual NOM. This may involve nanofiltration (NF)/reverse osmosis (RO) (Shen and Schäfer, 2015), activated carbon adsorption (Letterman, 1999), and advanced oxidation processes (AOPs) (Matilainen and Sillanpää, 2010).

Granular activated carbon (GAC) is one option for removing NOM as well as taste and odor compounds, and is recommended by the US EPA for the control of DBP precursors (Kim and Kang, 2008). The removal of NOM by GAC is through reversible and irreversible physical adsorption caused by non-specific mechanisms, such as van der Waals forces, dipole interactions and hydrophobic interactions (Gauden et al., 2006). There are two options for locating a GAC treatment unit in water treatment plants, i.e., (1) post-filtration adsorption, where the GAC unit is located after the conventional filtration process; and (2) filtration-adsorption, in which some or all of the filter media in a granular media filter is replaced with GAC. Compared with filter-adsorbers, the post-filtration application provides the most flexibility for handling GAC and for designing specific adsorption conditions, and thus often allows for lower operational costs. Many bench-, pilot- and full-scale studies have indicated GAC is a promising method to effectively remove NOM (Kim and Kang, 2008; Sorlini et al., 2014; Yang et al., 2010). However, a major constraint in operating GAC contactors is the cost of routinely replacing the GAC media due to the loss of adsorption capacity that occurs as the GAC becomes saturated. It is possible to substantially reduce the GAC replacement costs by implementing a biofiltration process, i.e., biological activated carbon (BAC) process, where microbial activity on activated carbon possibly extends GAC adsorption capacity via *in situ* regeneration of adsorption sites on the external surface or in inner pores through the biodegradation of previously adsorbed organic matter (Kim et al., 1997). The BAC process combines both biosorption/sorption and biodegradation functions, providing many benefits for the water treatment (Gibert et al., 2013).

Currently, biofiltration in drinking water treatment is largely operated as a passive process, and the design and operational parameters are generally limited to media configuration, backwash strategy and loading rate (Lauderdale et al., 2012). In recent years, an alternative approach has been proposed to move the practice of biofiltration from a passive process to a purposefully operated biological system, i.e., engineered biofiltration. The goal of engineered biofiltration is to target multiple water quality objectives while maintaining or improving hydraulic performance, which can be achieved by providing specific conditions that improve the biological performance such as nutrient enhancement. In a biofilter system, the ratio of bioavailable organic carbon (C) to ammonium nitrogen (NH_4-N) to orthophosphate phosphorus (PO_4-P), i.e., C:N:P, has been shown to be important in microbial growth (Lauderdale et al., 2012). An optimal molar ratio of C:N:P = 100:10:1 is generally used and recommended for biodegradation in an aerobic process (Leys et al., 2005). However, NH_4-N and PO_4-P nutrient concentrations limitations have been usually shown to minimize microbial activity levels (LeChevallier et al., 1991; Nishijima et al., 1997). Thus, the supplementation of NH_4-N and PO_4-P in the feed water is expected to promote the microbial growth and biofilm formation in the biofilter, subsequently enhancing the removal of NOM. To date, only a few studies have investigated the engineered biofiltration (Azzeh et al., 2015; Lauderdale et al., 2012; McKie et al., 2015; Rahman et al., 2016), and the information on such processes remains rather limited.

This pilot study employed a two-stage BAC biofiltration to remove the residual NOM after the coagulation process. The research objectives were to (1) test the efficiency, feasibility and stability of biofiltration as a treatment step for further NOM removal; (2) evaluate the impact of nutrient supplementation on the performance of biofiltration; and (3) identify the critical factors that affect the performance of a two-stage biofiltration process.

2. Materials and methods

2.1. Pilot-scale test

The pilot study was conducted in a water treatment plant near Atlanta, Georgia, United States. The plant has a treatment capacity of 0.27 million cubic meters per day and its process train includes pre-oxidation, coagulation, flocculation, sedimentation, sand/anthracite filtration, and post-filter GAC (Figure S1, Supporting Information). The feed water for the pilot skid was pumped from the effluent channel of the full-scale sedimentation basins (i.e., the skid was supplied with clarified water). A schematic of the pilot skid is shown in Fig. 1. The pilot skid included four columns