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Evaluation of backwash strategies on biologically active carbon filters by using chloroacetic acids as indicator chemicals

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ABSTRACT

Disinfectant-enhanced backwash is frequently required to control the over-growth of biomass in biologically active carbon (BAC) filters for drinking water treatment. This study was conducted to investigate the impact of different backwashing strategies on the biodegradation and adsorption performance of BAC filters and attached biomass concentration in the filters. The biodegradation performance was evaluated using the three chloroacetic acids (CAAs) as indicator chemicals. Results showed that both free chlorineand chloramines-enhanced backwashes could significantly increase the removal efficiency of attached biomass, but they also impaired the CAA degradation in BAC filters. The deterioration of CAA degradation could not be correlated with the removed attached biomass. Use of CAAs was a feasible approach to evaluate the biodegradation performance of BAC filters either during operation or after backwash. Chloramines-enhanced backwash is suggested to be employed to sustain the operation of BAC filters when excessive biomass growth takes place, due to its higher efficiency in removing attached biomass and lower adverse impact on BAC adsorption properties compared with free chlorine-enhanced backwash. However, the more pronounced adverse impact on organic matter degradation and the inconvenience of using chloraminated water must be considered.

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1. Introduction

Filtration with biologically active carbon (BAC), usually following sand filtration as a separate unit operation or used as a top layer of a dual or triple media filter, has been widely practiced all around the world to improve the finished drinking water qualities. BAC filtration is the later stage of granular activated carbon (GAC) filtration when biodegradation predominates over adsorption in removing contaminants [1–3]. BAC filtration is usually very effective to remove ammonia, odor- and taste-causing substances, trace organic compounds and the biodegradable fraction of natural organic matter (BOM) primarily attributed to the biofilm grown on the BAC grains [4–7]. In practical applications, it is however necessary to limit the BAC biomass content, which, when in excess, can

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http://dx.doi.org/10.1016/j.procbio.2016.03.016 1359-5113/© 2016 Elsevier Ltd. All rights reserved. lead to a substantial increase of the water head loss and decrease of mass transfer efficiency. More catastrophically, it can raise up the concern about the carryover of too many microorganisms into the filtrate. Microorganisms could be continuously detached with the flowing of water over a biofilm [5]. Depending on the state of the filters, the filtrate biomass concentration could be at $\sim 10^4$ cfu/mL or higher [8]. A higher biomass concentration in the filtrate necessitates a higher disinfectant dosage (leading to a higher formation of disinfection by-products (DBPs)) and results in a higher water turbidity. In addition, some microfauna (such as protozoa and metazoa) could also proliferate in BAC filters in some humid tropical and subtropical regions, which further increases the health risk of BAC filtrate [5]. Therefore, BAC filters need to be regularly backwashed to prevent the above problems. In actuality, a number of previous studies have emphasized the importance of backwash to sustain the operation of BAC filters [9–14].

The BAC filtrate was conventionally used to backwash the filters (termed as disinfectant-free backwash in this study).

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Previous studies however showed that disinfectant-free backwash was sometimes not sufficiently effective to regulate the BAC biomass content [10,13,14], especially when the filter influent contains a high concentration of BOM and the water temperature is favorable for microorganism proliferation. This backwashing approach is also highly water-intensive. A pre-stage of air scouring was later applied to increase the backwashing efficiency and to decrease the backwashing water demand [9,10,12]. Collapsepulsing backwash is a second substitute for the disinfectant-free backwash [9,15]. Alternatively, disinfectants including free chlorine and chloramines can be added into the backwashing water to enhance the biomass control efficiency (termed as disinfectantenhanced backwash). However, one probable problem of adding disinfectants is the attack on the GAC grains which may reduce not only the adsorption capacity but also mechanical strength [13]. In this regard, chloramines may be a better choice due to their weaker reactivity. A potentially more serious problem of adding disinfectants is the deterioration of the biodegradation performance due to the excessive decrease of biomass concentration and/or activity. Previous studies have showed that an overdose of free chlorine could lead to an immediate loss of attached biomass on the top of a BAC filter and a concomitant deterioration of the assimilable organic carbon (AOC) degradation, which was hardly recovered in a short period of time [16]. It was even worse for the degradation of the relatively poorly biodegradable organic matter [13].

To optimize the backwashing conditions for BAC filters, it is therefore very important to evaluate the biodegradation performance before and after the backwash. Both aggregate parameters and individual chemical compounds were utilized as indicators in a number of researches. To assess the filter performance, the aggregate parameters including total or dissolved organic carbon (TOC/DOC), biodegradable DOC (BDOC) and AOC were often used [10,16]. However, these aggregate parameters were found to lack sufficient sensitivity. For example, the removal ratio for TOC by a typical BAC filter was within 13-23%, which was not expected to be significantly changed after each backwash [9]. The measurement of BDOC and AOC was very time-consuming and may also suffer from a lot of systemic errors. Some individual chemical compounds which constitute and represent the BOM were later used for the evaluation. These compounds were mostly ozonation by-products and might include oxalate, acetate, formate, formaldehyde and glyoxal [9,14,16,17]. Some of the advantages of using individual compounds are that both readily and poorly biodegradable organic matter could be used (and as such the backwashing effect could be evaluated in more details) and that the evaluation is accurate and reproducible because the compounds can be analytically measured with high accuracies. Nevertheless, most ozonation by-products were readily biodegradable, and the information regarding backwashing effect was consequently limited.

In this study, three chloroacetic acids (CAAs) were used as indicator chemicals to evaluate the effects of the disinfectant-free backwash, and free chlorine- and chloramines-enhanced backwash on the biodegradation performance of a BAC filter. CAAs are used primarily because they are relatively easy to measure with several standard methods available [18]. In addition, previous studies revealed that both mono- and di-chloroacetic acids (MCAA and DCAA) are readily biodegradable while trichloroacetic acid (TCAA) is less biodegradable [19-21]. Moreover, CAAs can be a major BOM component in the influent water to BAC filters when pre- and intermediate chlorination are applied to control algae growth and/or to enhance coagulation and filtration efficiency in drinking water treatment plants [22]. BAC filters were found to be capable of reducing CAAs levels in the finished water [22,23]. The effect of backwash on the biomass concentrations on the BAC grains and in the filtrate was also tested. The aim of this study is to find an appropriate backwashing approach (and the corresponding conditions) by which

the removal of excessive attached biomass is maximized while the impact on the filter performance, in terms of BOM removal and GAC adsorption capacity, is minimized.

2. Materials and methods

2.1. BAC filters, operation and backwash

Two identical lab-scale acrylic columns with an inner diameter of 4.0 cm and a length of 2.0 m were constructed as duplicate BAC filters. A perforated plate (with holes of 1.0 mm in diameter) was sitting at the bottom of the column, on which a bed of cylindrical BAC grains (with a nominal size of $1.0 \text{ mm} \times 3.0 \text{ mm}$ according to the supplier) was packed to a height of 100 cm. (Noted is that wall effects could be negligible in the filter columns in that the columnto-grain diameter ratio was larger than a minimum value of 10 [24,25]). There were a number of sampling ports (A–E) located at the depths of the BAC bed of 10, 30, 50, 80,100 cm, corresponding to an empty bed contact times (EBCTs) of 2, 6, 10, 16, 20 min, respectively (at a flow rate of 60 mL/min). Two ports (F and G) which were at the top (at 5 cm depth) and bottom (at 95 cm depth) of the filter bed were used for sampling of BAC grains (Fig. 1). The BAC grains were collected from a full-scale BAC filter at a local drinking water treatment plant (Beijing, China). The plant adopted a treatment process consisting of pre-chlorination, coagulation, clarification, anthracite filtration, BAC filtration and disinfection. The BAC in the filter had been in operation for more than three years, during which it was regularly backwashed about once every week by using the disinfectant-free BAC filtrate.

The influent (feed water) to the BAC filters was a mixed solution of the three CAAs (i.e. MCAA, DCAA and TCAA) (Sinopharm, China) freshly prepared in a local groundwater at \sim 150 µg/L for each. The groundwater did not contain any DBPs initially and was sufficiently clean with a TOC concentration of about 0.5 mg/L. The influent had a pH value of 7.5 and an ionic strength of 780–830 µS/cm. The water temperature was controlled within 18–22 °C by using a heating rod.

The BAC filters were pre-operated for two months (as the startup period) to reach a quasi-steady removal state. During the startup period, the filters were backwashed by using the filtrate every one or two weeks. Water samples were taken from the sampling ports A and E, corresponding to an EBCT of 2 and 20 min, respectively. It was observed that, for an EBCT of 20 min, the removal ratios for MCAA and DCAA could reach ~100% in less than 5 days while that for TCAA could reach \sim 100% in 15 days (Fig. S1a). When the EBCT was 2 min, it took 20 days for the removal ratios for MCAA and DCAA to reach ${\sim}100\%$, while it took about 35 days for TCAA (Fig. S1b). It was believed that the quasi-steady state was reached after 35 d operation, after which the CAAs removal rates were not significantly changed. The fast startup of the filters was owing to the use of BAC grains collected from the full-scale plant with prechlorination. At the steady state, all three CAAs were predominantly removed by biodegradation rather than adsorption in the BAC filters, because decrease of the removal ratios was only observed at the very beginning of the startup period that took only for 1–2 days.

When the steady state was reached, a number of backwashing schemes, i.e. disinfectant-free backwash and free chlorineand chloramines-enhanced backwashes with different disinfectant dosages, were tested individually. Free chlorine was dosed by adding an appropriate volume of concentrated sodium hypochlorite (reagent grade, Sigma-Aldrich, USA) into the BAC filtrate. Chloramines were dosed by adding an appropriate volume of concentrated monochloramine solution, which was prepared freshly by slowly adding sodium hypochlorite solution into an ammonium chloride solution (molar ratio of free chlorine to ammonium was 1.2) [26]. Within each backwashing cycle, the same operational

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