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Short communication

Comparisons between biological filtration and coagulation processes for the removal of dissolved organic nitrogen and disinfection by-products precursors

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ABSTRACT

Effects of biological aerated filter (BAF), sand filter (SF), biological activated carbon (BAC), alum coagulation, and *Moringa oleifera* coagulation on the removal of DOC and DON from reservoir water were investigated. The DOC removal efficiency was in the following sequence: BAC (56%), BAF (51%), SF (45%), alum (27%), and *M. oleifera* (22%). The trend for DON removal efficiency was similar to that of DOC. The better performance by BAC was attributed to the effective microbial breakdown of organic matter and the adsorption of some molecules onto activated carbon. The organic content with a molecular weight (MW) of less than 10 kDa accounted for more than 45% of the total DOC, whereas a MW of less than 1 kDa accounted for 50% of the total DON. The biofilters removed the entire MW range of organic contents, unlike the coagulants which only removed organics with a MW of more than 10 kDa. Consistent with greater DOC and DON removal, BAC led to greater removal of DBP precursors as shown by DBP formation potentials.

1. Introduction

The elevated level of dissolved organic carbon (DOC), dissolved organic nitrogen (DON), and particulate matter present in natural water sources, particularly river, lake, and reservoir waters can cause harmful effects to human and aquatic living organisms due to their chronic toxicity, persistence, and bioaccumulation (Willett et al., 1998; Zhou et al., 2006). Dissolved organic matter (DOM) consists of a wide range of different compounds, such as humic substances, hydrophilic acids, carbohydrates, amino acids, and carboxylic acids (Thurman, 1985). Some components of the DOM can react with disinfectants and thus form disinfection by-products (DBPs), such as trihalomethanes (THMs) and haloacetic acids (HAAs). These carbonaceous DBPs are potentially carcinogenic (Nikolaou et al., 2004), and therefore, the removal of their precursors is required to eliminate potential health risks and threats.

Studies showed that the presence of dissolved organic nitrogen (DON) in drinking water has several adverse impacts as it produces nitrogenous DBPs (NDBPs), particularly N-nitrosodimethylamine (NDMA) (Lee et al., 2007; Krasner et al., 2009). NDBPs are considered to have higher carcinogenicity than the carbonaceous DBPs (Plewa et al., 2004; Lee et al., 2007). DON comprises a relatively small portion of the DOM and includes amino acids, amides, amino sugars, peptides, and heterocyclic-N compounds (Fuhrman, 1990). It is an essential nutrient for microbial survival and growth (Pehlivanoglu and Sedlak, 2004). Although the removal of DOC has been widely investigated, less work has been done on DON and the nitrogenous DBPs.

Coagulation using iron and alum salts is commonly employed in water treatment facilities for the removal of DOM as well as particulate matter. However, the use of coagulants causes a secondary issue, i.e., it produces chemical sludge. Alternative natural coagulants such as *Moringa oleifera* seeds may be more cost-effective (Omm-e-hany et al., 2013; Mohamed et al., 2014). Mohamed et al. (2014) found that *M. oleifera seeds* reduced the turbidity, COD, and phosphorus of wastewater by 90%, 60% and 75%, respectively.

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The use of this natural coagulant for organics removal is limited. However, filtration processes such as biological activated carbon (BAC) and sand filtration (SF) have been widely employed for drinking water treatment. They have the merits of low capital cost and ease of maintenance. Biological aerated filters (BAF) have been reported to be effective for wastewater treatment (Biplob et al., 2011) due to the large surface area for the growth of biofilm. However, the BAF process has seldom been investigated for organic matter removal from drinking water. Studies have been conducted on the characterisation of DOC (Thurman, 1985; Frimmel et al., 2002) and DON (Pehlivanoglu-Mantas and Sedlak, 2008) in drinking water. However, little comparison of the removal of the various DOC and DON fractions by different treatments is known.

The objective of this study was to compare the performance of a range of water treatment processes: coagulation, BAC, SF, and BAF, in terms of the removal of the different DOC and DON fractions and the resultant impact on carbonaceous DBP formation potentials (THMFP) and HAAFP) and nitrogenous DBPs potential (NDBPFP). The natural coagulant (*M. oleifera*) was compared with alum, the conventional coagulant. Membranes with different molecular weight cut-offs were used to separate the DOC and DON fractions in reservoir water and thus to understand the effect of each treatment on the removal of the DOM size fractions.

2. Materials and methods

2.1. Feed water

The feed water was collected from a reservoir. Samples were stored at 4 °C in the laboratory and then equilibrated to room temperature (22 ± 2 °C) prior to all experimental tests.

2.2. Treatment processes

The BAF was constructed in a PVC chamber with an internal diameter of 8 cm and an effective media height of 60 cm. Plastic media (Kaldness) with length, diameter, density, and an internal surface area of 3 mm, 5 mm, 0.42–0.46 g/cm^3, and 305 m^2/m^3, respectively, were used for the BAF. Prior to packing the BAF reactor, the plastic media was inoculated with 3 L activated sludge, provided with additional nutrient sources (N, P and C) to promote the rapid growth of biofilm on the media. It was then gently washed with tap water to remove excess biofilm and transferred to a BAF reactor, and feed was commenced. The DOC removal was stable after 40 days of BAF operation, indicating that the biofilm was well established on the surface of the media.

Glass column reactors for SF and BAC were constructed with an internal diameter of 4.5 cm and an effective bed height of 50 cm. The surface area, total pore volume, and micropore volume of the activated carbon used were 800 m^2/g, 0.865 cm^3/g, and 0.354 cm^3/g, respectively. After seeding with activated sludge, the DOC levels of the BAC and SF effluents became constant after 35 and 60 days of operation, respectively.

All the filters were backwashed for 20 min every week to avoid physical clogging. They were operated in a downflow mode with an empty bed contact time of 30 min.

Alum (Al_2 $(SO_4)_3 \cdot 18H_2O$) and *M. oleifera* seeds were used as coagulants. The seeds of *M. oleifera* were extracted from the dry pods and it was then ground into a fine powder by using a blender, and then sieving through 0.8 mm mesh. Afterwards, Milli-Q water was added to the fine powder to make 1% suspension. The suspension was stirred using a magnetic stirrer for 30 min to extract the active components of seed (coagulant proteins) in water and this was then filtered through 125 mm filter paper. The solutions were shaken prior to use.

Both alum and *M. oleifera* coagulations were performed using a jar test apparatus (Phipps and Bird, PB-900). The samples were mixed for 5 min at 300 rpm and then mixed for the next 20 min at 40 rpm. After 1-hour settling by gravity the supernatant was tested for water quality. Preliminary tests with the *M. oleifera* seeds coagulant (dosage range 50–100 mg/L), showed the optimum condition for DOC removal to be 70 mg/L. The optimum dose for alum was determined to be 7 mg/L. These experiments were conducted without pH adjustments as the pH of the feed water was low (6.58).

2.3. Analytical methods

DOC concentration was determined using a total organic carbon analyser (TOC-5000A, Shimadzu). Potassium hydrogen phthalate (1, 5, 10 and 25 mg C L^{-1}) was used for calibration of TOC analyser. The UV absorbance at 254 nm (UV_{254}) was determined using a spectrophotometer (UV-16000, Shimadzu) and Milli-Q water was used as a reference. A Hach spectrophotometer (model DR/4000) was used for determining the colour at 455 nm. Before these analyses, all samples were filtered through 0.45 μm filter.

Turbidity, dissolved oxygen (DO), and pH of the water samples were measured using a turbidity meter (Eutech, TN100IR), a DO meter (IC-MW-600), and a pH meter (Mettler Toledo), respectively. The pH meter was calibrated with standard solutions of pH 4, 7 and 10. Formazin standards (20, 100 and 800 NTU) were used to calibrate the turbidimeter.

The carbohydrate and protein concentrations of the water samples were measured using the phenol-sulfuric (DuBois et al., 1956) and the bicinchoninic acid (BCA) method, respectively. D-glucose was used as the standard for carbohydrate, and QPBCA QuantiPro™ BCA Assay Kit (Sigma Aldrich) and bovine serum albumin (Sigma Aldrich) was used as the standard for protein. 1 mL sample was used for both protein and carbohydrate analysis. The detection range of this method for protein content was 0.5–30 mg/L.

The apparent molecular weight distributions of the samples were determined using ultrafiltration. A series of ultrafiltration (UF) membranes with molecular weight cut-off (MWCO) of 1000 Da (YM1), 3000 Da (YM3), 10,000 Da (YM10), and 30,000 Da (YM30) were used and then the DOC level of each fraction collected was determined. All samples (2 L) were pre-filtered (0.45 μm).

The THM and HAA formation potentials were determined using a gas chromatograph (GC-8A, Shimadzu, Japan) (Agilent 6890N) (APHA, 1998). THMs and HAAs were quantified using a fused silica capillary column and a DB5 capillary column, respectively. The standard deviation of the mean concentrations of seven replicate analyses was calculated to determine the method detection limit for each of the THM and HAA species. The detailed procedure can be found elsewhere (Tubić et al., 2011). The ammonia concentration was measured using automated phenate method 4500-NH_3 (APHA, 1998). Nitrate and nitrite were determined using an ion chromatograph (HIC-6A, Shimadzu, Japan). Total dissolved nitrogen (TDN) was analysed by a coupled TOC-VCSH and nitrogen analyser (TNM-1, Shimadzu). The DON values (DON = TDN − nitrate − nitrite − ammonia) were then determined according to the method described by Lee and Westerhoff (2005). The concentration of NDMA precursors in water samples was determined using a gas chromatograph equipped with a mass spectrometer detector (QP5050A, Shimadzu, Japan). The detailed procedure can be found elsewhere (Mitch et al., 2003).

3. Results and discussion

3.1. Effect of treatment processes on water quality

The characteristics of the water samples after different treatments are shown in Table 1. The BAC process achieved the greatest