*Desalination and Water Treatment*
www.deswater.com
doi: 10.1080/19443994.2014.929976

55 (2015) 2069–2078
August

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# Profile of dissolved organic nitrogen (DON) in full-scale ozone and biological activated carbon filter

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Received 9 August 2013; Accepted 24 May 2014

ABSTRACT

The profile of dissolved organic nitrogen (DON) was investigated in a full-scale two-stage integrated process of ozonation and biological activated carbon (BAC) filtration. The variation of DON along the media depth of BAC filters was studied comprehensively. The results showed that 48.3% of DON was removed in the two-step processes. Ozone can mineralize organic-N into inorganic-N or nitrogen gas, removing approximately 31% of DON. To determine the variation of DON in the BAC filtration process, DON and other related parameters at different media depths of a BAC filter were studied. The results showed that the concentration of DON decreased from 1.6 to 1.2 mg/L in media depth of 0–10 cm and increased gradually from 1.2 to 1.4 mg/L at deeper depths of the media (10–200 cm). Similarly, the biomass concentration and microbial activity first increased rapidly and then decreased gradually along the media depth of the BAC filter. The proportion of the small molecular weight (<6 kDa) DON in influent, 10 cm media depth and effluent of the BAC filter increased from 91 to 93% and then decreased from 93 to 87%. In contrast, the variation of the large molecular weight (>20 kDa) DON had the opposite trend. The middle molecular weight fraction (6–20 kDa) of DON was almost unchanged. Soluble microbial products (SMPs) released by bacterial metabolisms might be the main source of DON variation in BAC filters. These SMPs contained aromatic protein-like fractions, which were confirmed by EEM analysis.

*Keywords:* Dissolved organic nitrogen (DON); Soluble microbial products (SMPs); Ozonation; Biological activated carbon filter; EEM

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## 1. Introduction

In order to eliminate pathogenic microbes and ensure drinking water safety, chlorine or chloramine is often used in drinking water treatment [1]. However, dissolved organic matter (DOM) in drinking water can react with chlorine or chloramine to produce undesirable disinfection by-products (DBPs) [2]. These include trihalomethanes and haloacetic acids which are most frequently detected as well as other hazardous halogenated DBPs which are considered carcinogens or suspected carcinogens [3–5]. Furthermore, nitrogenous fractions of DOM were discovered to react with chlorine or chloramine to produce nitrogenous disinfection by-products (N-DBPs) [6–9]. N-DBPs were found to have much higher toxicity and mammalian cell genotoxicity than DBPs without nitrogen [6,10]. Nitrogenous fractions account for a relatively small portion of DOM and are designated as dissolved organic nitrogen (DON). The removal of DON in drinking water and wastewater treatment processes is potentially critical due to the formation of N-DBPs.

DON should be removed during the drinking water treatment process to prevent the production of N-DBPs. Treatment processes that involve ozonation integrated with biological activated carbon (BAC) filtration have been increasingly used for drinking water treatment to reduce the concentration of DOM. It was found that organic matter was effectively removed by the ozone and BAC filter processes [11–13]. Refractory DOM was transformed to biodegradable forms by ozonation and then was biodegraded by BAC filtration. It was reported that the ozone and BAC filter processes decreased the precursors of regulated DBPs and N-DBPs [14,15]. However, profiles of DON during the two-stage ozone and BAC filter processes in full-scale drinking water treatment facilities have rarely been reported. Moreover, it remains unclear how the concentration and molecular weight of DON changes along the media depth of BAC filters within these types of systems.

Generally, DON in surface water is from soluble microbial products (SMPs) of bacterial metabolisms, the extracellular organic material of algae, forest litter, and agriculture fertilizers [16]. When biological treatment processes were widely used to remove organic matter and $NH_4^+$-N, SMPs were produced and were regarded as the main source of DON [16]. It has been reported that SMPs can be released in biofilters as microbes utilize substrates and that SMPs concentration can increase gradually along the media depth of the biofilter [17]. BAC filters, which are frequently used in biological treatment processes, might exhibit similar patterns. Therefore, the variation of DON in BAC filters used for drinking water treatment deserves our attention.

In our work, the DON profile in a typical two-stage ozone and BAC filtration system is investigated. The profile of DON at different media depths of one BAC filter is formulated and was studied. Biomass, microbial activity, and the molecular weight distribution of DON are investigated. Furthermore, three-dimensional excitation–emission matrix (EEM) fluorescence spectroscopy is applied to determine the mechanism of the DON variation in the BAC filter.

## 2. Materials and methods

### 2.1. Drinking water treatment processes

This study was carried out at the Pinghu drinking water treatment plant ($5.0 \times 10^4$ $m^3/d$) located in Zhe Jiang Province, China. A study published in 2012 reported that the concentrations of DON and dissolved organic carbon (DOC) were approximately 2.00 and 8.22 mg/L, respectively, in the source water [18]. Fig. 1 shows the flow chart of the water treatment plant. The treatment processes include bio-pretreatment, coagulation–sedimentation, sand filtration, the first ozonation, the first BAC filtration, the second ozonation, the second BAC filtration, and disinfection. The BAC filter was 5.5 m in height and contained 2.0 m of granular activated carbon (GAC). The filtration velocity was 10.0 m/h and the empty bed contact time was 12 min. The dosages of ozone in the first and second ozonation processes were approximately 3.5 and 1.5 mg/L, respectively.

### 2.2. Sampling

Samples (Sample 1–5) were taken at different points in the treatment train (Fig. 1). To investigate the variation of DON in the BAC filter, samples at different media depths (influent, 10, 20, 50, 75, 100 cm, and effluent) from the first BAC filter were also taken. Before analysis, all water samples were filtered using 0.45-μm pore-sized membranes. GAC in the first BAC filter at different depths (0, 10, 20, 50, 75, and 100 cm) was sampled for the determination of biomass and microbial activity.

### 2.3. Analytical methods

DOC was determined using the Shimadzu TOC-VCHS analyzer. UV absorbance at 254 nm ($UV_{254}$) was determined using a spectrophotometer (752 N/UV-2101PC).

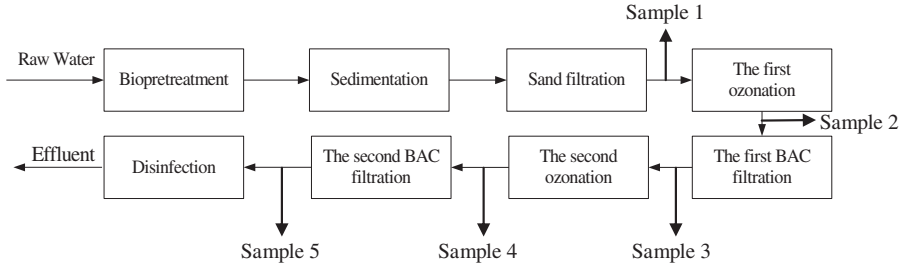


Fig. 1. Flow chart of drinking water treatment plant.

The special UV absorbance (SUVA) was calculated as $UV_{254}$ divided by mg/L DOC concentration ($UV_{254}$/DOC). $NH_4^+$-N was measured using the salicylate–hypochlorite method. $NO_2^-$-N was measured using the N-(1-naphthyl)-ethylenediamine photometric method. $NO_3^-$-N was determined using a UV spectrophotometry method. Total dissolved nitrogen (TDN), sum of $NH_4^+$-N, $NO_2^-$-N, $NO_3^-$-N, and DON were measured using an alkaline potassium persulfate digestion-UV spectrophotometric method. All the determinations were done according to the Chinese National Standard Methods [19]. DON was quantified as the difference between TDN and total dissolved inorganic nitrogen (TDIN, sum of $NH_4^+$-N, $NO_3^-$-N, and $NO_2^-$-N). Biomass was measured using the phospholipid method [20]. Microbial activity was determined using the Specific Oxygen Uptake Rate (SOUR) method [21].

### 2.4. Molecular weight fractionation

The DON was fractionated by molecular sieves. Two types of regenerated cellulose membranes (Millipore Corp) were used: (1) a membrane with a 20,000 nominal molecular weight limit (NMWL) and (2) a membrane with a 6,000 NMWL. Two hundred milliliters of the water samples were filtered through the first and second membrane in series. Fifty milliliters of raw water and each filtrate was retained for further analysis. The percentage of DON in each size range was calculated as follows:

$$\%<6\ \text{kDa} = \frac{C_{6K\ \text{permeate}}}{C_{\text{raw}}} \times 100\% \quad (1)$$

$$\%<6-20\,\text{kDa} = \frac{C_{20K\ \text{permeate}} - C_{6K\ \text{permeate}}}{C_{\text{raw}}} \times 100\% \quad (2)$$

$$\%>20\,\text{kDa} = \frac{C_{\text{raw}} - C_{20K\ \text{permeate}}}{C_{\text{raw}}} \times 100\% \quad (3)$$

where *Ci* is the measured parameter of fraction *i*. Thus, we calculated the percentage difference between the original total mass and the sum of the masses for each mass fraction.

### 2.5. EEM fluorescence spectroscopy

Three-dimensional EEM fluorescence spectroscopy (F-4600 FL Spectrophotometer, Hitachi, Japan) was used to characterize DOM in water samples. The excitation (Ex) wavelength was set from 200 to 500 nm at 5 nm sampling intervals, corresponding to emission (Em) wavelengths from 280 to 500 nm at the same sampling intervals. The Ex and Em slits were set at 5 nm and the scanning speed was set at 1,200 nm/min. The spectrum of double distilled water was recorded as the blank. The software Origin 7.5 (OriginLab Inc, USA) was employed to process the data. The EEM spectra were plotted as the elliptical shape of contours. The *X*-axis represented the Em spectra from 280 to 500 nm, and the *Y*-axis represented Ex from 200 to 500 nm. As a third dimension, a contour line was shown for each EEM spectra to express the fluorescence intensity at an interval of 5 nm.

## 3. Results

### 3.1. DON and related species at different sampling points in the treatment train

As shown in Fig. 2(A), DON concentrations decreased from 2.39 to 1.42 mg/L after the first-stage ozonation and BAC filtration processes. Approximately 29.3 and 16.0% of DON were removed by the first-stage ozonation and BAC filtration processes, respectively. After the second-stage ozonation and BAC filtration processes, the concentrations of DON decreased from 1.42 to 1.22 mg/L. Approximately 4.9 and 9.6% of DON were removed by the second-stage ozonation and BAC filtration processes, respectively.

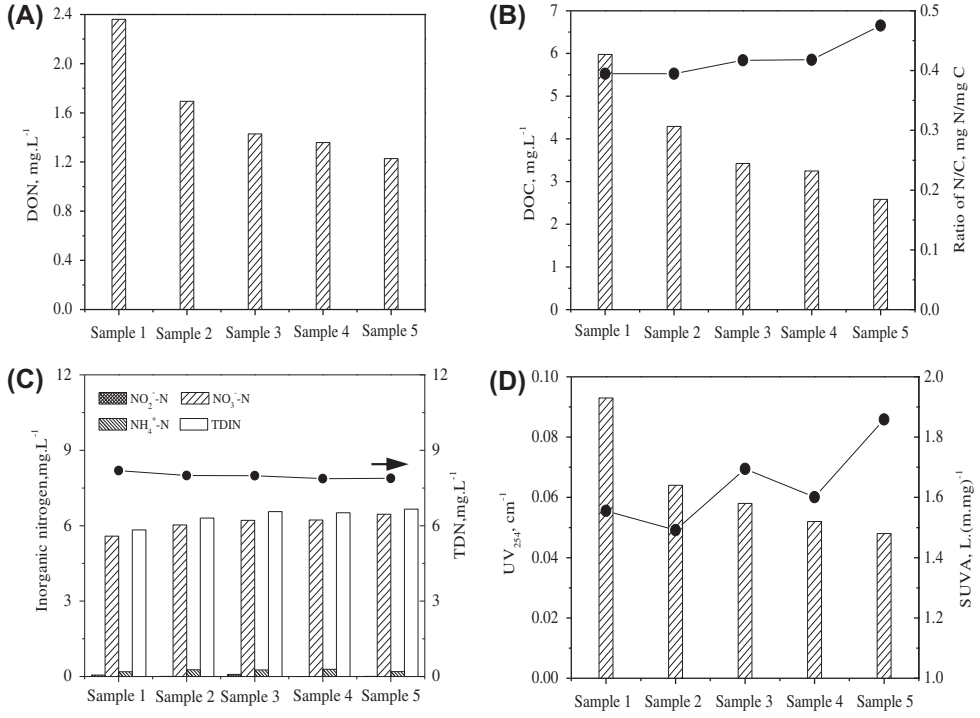


Fig. 2. Variation of DON, DOC, the N/C ratios, TDN, DIN, $UV_{254}$, and SUVA concentrations at different points along the treatment train. (A) DON concentration; (B) DOC concentration and Ratio of N/C; (C) DIN and TDN concentration; (D) $UV_{254}$ and SUVA. The data presented in the figures are averages of three observations and their relative standard deviations ($n = 3$) are below 8%.

The total removal of DON in the two-step ozonation and BAC filtration process was approximately 48.3%.

Fig. 2(B) shows the variations of DOC and the ratio of N/C in the two-stage ozonation-biological treatment. DOC decreased gradually from 5.78 to 2.58 mg/L, and the removal of DOC was 55.4%. The ratio of N/C increased from 0.39 to 0.46 mg N/mg C. It should be noted that only BAC filtration increased the ratio of N/C.

$NH_4^+$-N, $NO_3^-$-N, $NO_2^-$-N, TDIN, and TDN during the two-step process were determined and are shown in Fig. 2(C). Since the concentrations of $NH_4^+$-N and $NO_2^-$-N were relatively low, their variations were not distinct. The concentration of $NO_3^-$-N increased gradually from 5.58 to 6.66 mg/L. The concentration of TDIN increased slightly from 5.83 to 6.30 mg/L in the first-ozonation process and was almost unchanged in the three following processes. However, the concentration of TDN decreased slightly from 8.19 to 7.99 mg/L in the first-ozonation process and remained unchanged subsequently.

Fig. 2(D) shows the trends of $UV_{254}$ and SUVA in the treatment process. The $UV_{254}$ concentration decreased gradually from 0.093 to 0.048 $cm^{-1}$ and the process removed 48.4% of the initial amount. The ozonation process increased the concentration of SUVA, while the BAC filtration decreased it.

### 3.2. DON and related parameters in the first BAC filter

As mentioned above, a decrease in DON concentration was observed during the BAC filtration process. However, this result did not indicate the trend of DON in the BAC filter. Hence, profiles of DON, DOC, and other related parameters along the media depth in the first BAC filter were evaluated.

As shown in Fig. 3(A), the concentration of DON in the influent of first BAC filter was approximately 1.62 mg/L. The DON concentration initially decreased with an increase in the media depth and reached a minimum of 1.28 mg/L at the depth of 10 cm. Subsequently, the concentration of DON increased gradually to 1.43 mg/L in the effluent of the filter.

Fig. 3(B) shows that the DOC concentration decreased continuously in the biofilter from 4.33 to 3.50 mg/L. In addition, the rate of decrease was much faster at the media depth of 0–20 cm where DOC concentration was reduced from 4.43 to 3.71 mg/L. The ratio of N/C was decreased at the media depth of

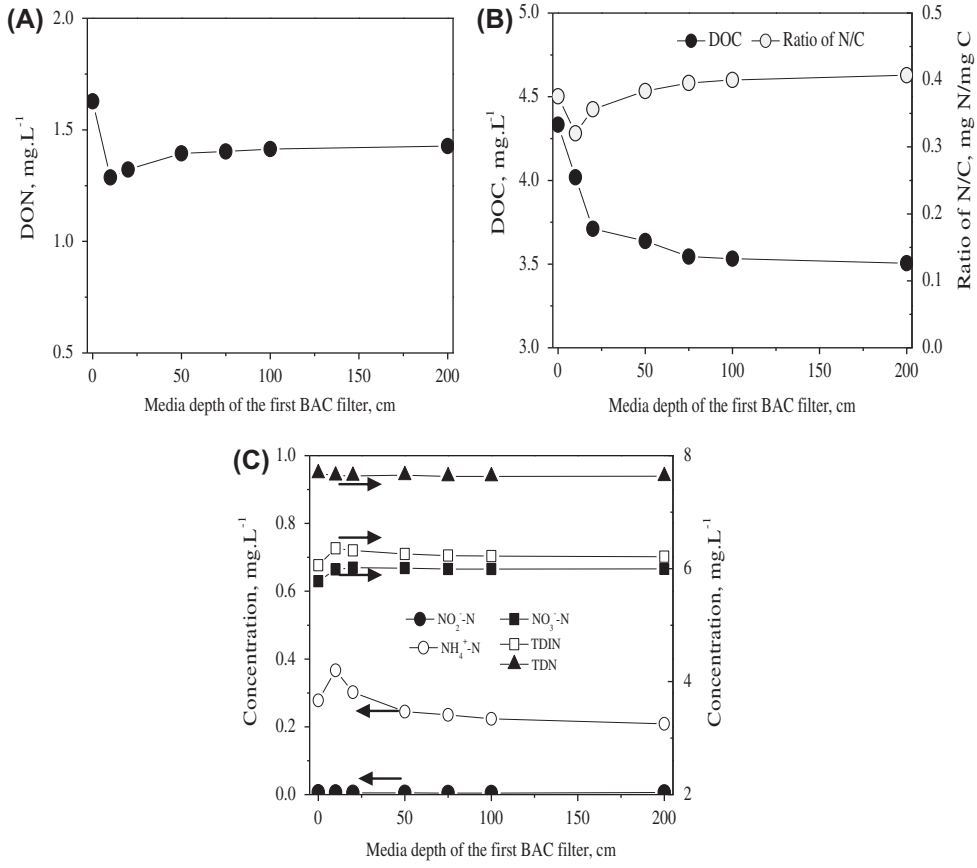


Fig. 3. Profiles of DON concentration and related parameters along the media depth in the first BAC filter. (A) DON concentrations; (B) DOC concentrations and Ratio of N/C; (C) DIN, TDIN, and TDN concentrations. The data presented in the figures are averages of three observations and the relative standard deviations of the data ($n = 3$) are below 10%.

0–10 cm and then increased gradually at media depth greater than 10 cm.

The concentrations of $NH_4^+$-N, $NO_3^-$-N, $NO_2^-$-N, TDIN, and TDN along the media depth are shown in Fig. 3(C). The variation trends of $NH_4^+$-N, $NO_3^-$-N, and TDIN concentrations were similar. Along the media depth range of 0–10 cm, the concentrations of $NH_4^+$-N, $NO_3^-$-N, and TDIN increased from 0.27, 5.77, and 6.06 to 0.36, 5.98, and 6.35 mg/L, respectively. Finally, these concentrations decreased gradually to 0.20, 5.98, and 6.21 mg/L. $NO_2^-$-N and TDN concentrations were almost unchanged along the media depth in the $O_3$-BAC filter.

### 3.3. Biomass and microbial activity in the first BAC filter

The variations in the biomass concentration and microbial activity along the media depth were also measured (Fig. 4). Biomass increased from 73.7 to 127.5 nmole lipid P/cm$^3$ along the media depth of

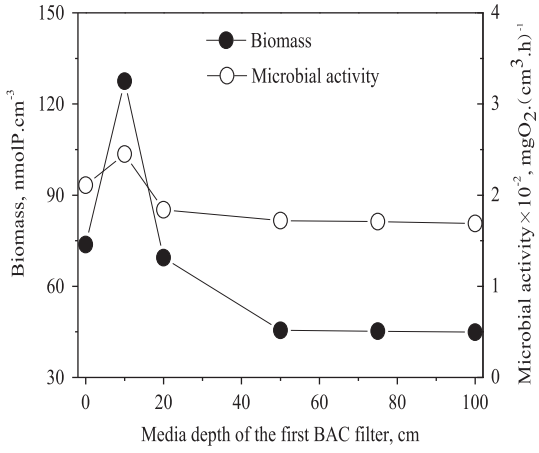


Fig. 4. Biomass and microbial activity in the first BAC filter. The data presented in the figures are averages of three observations and the relative standard deviations of the data ($n = 3$) are below 10%.

0–10 cm. Biomass then decreased continuously from 127.5 to 44.8 nmole lipid P/$cm^3$ along the media depth of 10–100 cm. Microbial activity followed a similar trend. Microbial activity first increased from $2.1 \times 10^{-2}$ to $2.5 \times 10^{-2}$ mg $O_2$/($cm^3$ of media × h) along the media depth of 0–10 cm. Then, it gradually decreased from $2.5 \times 10^{-2}$ to $1.6 \times 10^{-2}$ mg $O_2$/($cm^3$ of media × h) along the media depth of 10–100 cm.

### 3.4. Molecular weight fractionation of DON in the first BAC filter

Molecular weight fractionation of DON in the influent, water at the media depth of 10 cm, and the effluent of the first $O_3$-BAC filter is shown in Fig. 5. The molecular weight distribution of the DON in the influent showed that the small (<6 kDa) and middle (6–20 kDa) molecular weights accounted for 91 and 7% of the total and that the large DON (>20 kDa) only accounted for about 2%. At the media depth of 10 cm, the fraction of <6 kDa DON increased from 91 to 93%, while the fraction of >20 kDa decreased from 2 to 0.8%. The variation of the middle molecular weight fraction (6–20 kDa) was not so distinct. There was a slow decrease in the middle fraction from 7.0 to 6.4%. The molecular weight in the effluent exhibited a decrease from 93 to 87% in the fraction of <6 kDa, an increase from 0.8 to 5.9% in the fraction of >20 kDa, and an increase from 6.3 to 7.1% in the fraction of 6–20 kDa.

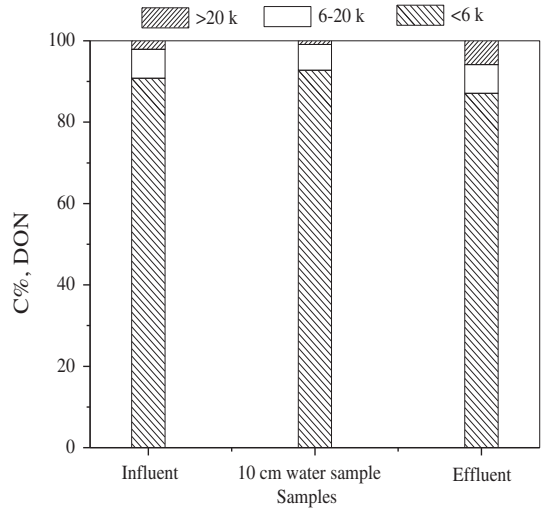


Fig. 5. Profiles of molecular weight fractionation of DON in influent, 10 cm water sample, and effluent of the $O_3$-BAC filter 1. The presented data in the figure are averages of three observations and the relative standard deviations of the data ($n = 3$) are below 9%.

### 3.5. Characterization of DON

The variation of DON in the BAC filter suggests that nitrogen enriched compounds might be removed or regenerated. To seek evidence of DON formation, dissolved organic substances in water samples were characterized by EEM. The EEM fluorescence spectra of the different samples, including influent of the BAC filter, effluent of the BAC filter, and water at the media depth of 10 cm are shown in Fig. 6. We identified two main peaks (Peak A and Peak B) in the EEM spectra. It is worth noting that there were many peaks with relatively low intensity which could not be clearly identified.

Peak A was located at the Ex/Em wavelengths of 230/340 nm and Peak B was located at the Ex/Em wavelengths of 275/310 nm. It has been reported that these two main peaks correspond to aromatic protein-like substances and tryptophan protein-like substances, respectively [22,23]. The fluorescence parameters of the spectra including peak locations, fluorescence intensity, and peak intensity ratios are summarized in Table 1. In the BAC filtration process, the intensity of peaks A and B decreased at first and then increased. Similarly, the intensity ratios of Peak A/Peak B reduced from 1.41 to 1.35 and then increased from 1.35 to 1.39. This suggests that the molecular structure of organic compounds in the water changed during the course of BAC filtration.

## 4. Discussion

Ozonation plays an important role in the removal of DOM in two-stage ozonation and BAC filtration processes, probably because organic matter containing organic-N and organic-C is mineralized by ozonation. The high value of SUVA represented hydrophobic and large molecular weight NOM, such as ionization of carboxyl groups and humic substances. The ozone can make the organic molecules smaller, more oxidized, and more biodegradable by separating the unsaturated bonds of organic matter [14]. Hence, the concentrations of DON, DOC, $UV_{254}$, and SUVA decreased, which are consistent with a previous report [15]. From Fig. 2(B), it can be seen that the first- and second-stage ozonation processes removed 28 and 5% of DOC, respectively. Some removed substances contained nitrogenous functional groups, and thus can be classified as DON. DON in this process was co-removed, and the removal of DON (about 28 and 5%) was close to that of DOC. Thus, the ratio of N/C was almost unchanged. During ozonation, it is possible that DON was transferred to DIN or nitrogen gas by the

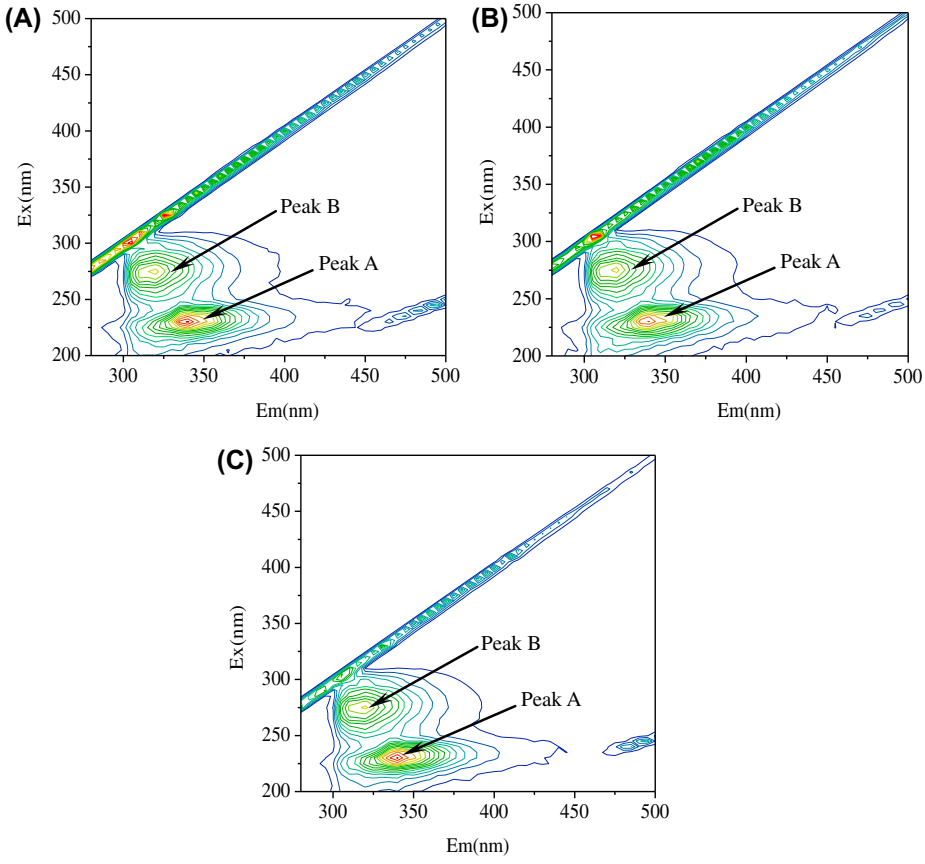


Fig. 6. EEM spectra of the different samples in the first $O_3$-BAC filter. (A) Influent; (B) 10 cm water sample; (C) Effluent.

Table 1
Fluorescence spectral parameters of DOM samples

| Sample | Peak A | | Peak B | | Peak A/Peak B |
|---|---|---|---|---|---|
| | Ex/Em | Intensity | Ex/Em | Intensity | |
| Influent | 230/340 | 2,636 | 275/310 | 1,870 | 1.41 |
| 10 cm water sample | 230/340 | 2,482 | 275/310 | 1,836 | 1.35 |
| Effluent | 230/340 | 2,570 | 275/310 | 1,843 | 1.39 |

oxidation, and therefore, TDN concentration decreased and TDIN increased.

Generally, ozone-BAC filtration is designed to further remove DOC via biodegradation and adsorption [24–27]. The concentration of DOC was reduced along the media depth of the filter in this study (Fig. 3(B)). However, the DON concentration decreased first and then increased gradually during the BAC filtration treatment (Fig. 3(A)). A variety of organic compounds (SMPs) can be formed and released into the extracellular solution during the process of BAC filtration. SMPs are nitrogen enriched compounds which contain organic matter such as polysaccharides, proteins, nucleic acids, amino acids, and steroids [28]. SMPs present higher ratios of N/C and biodegradability [29]. Therefore, the release of SMPs due to biological activity might be the cause of the variation of DON in the BAC filtration process.

The performance of BAC filters in removing bioavailable materials depends on the microbial biomass and activity on the media of the filter. At the media depth of 0–10 cm, the microbial biomass and activity increased gradually (Fig. 4), corresponding to a fast removal of organic compounds containing DON. The rate of DON utilization tended to be higher than that of SMPs generation, which resulted in the reduction of

the DON concentration. Furthermore, it has been reported that growth related utilization associated products (UAPs) and non-growth related biomass associated products (BAPs) constitute SMPs [17]. The formation of UAPs is directly associated with the rate of substrate utilization, while the formation of BAPs is associated with the endogenous respiration of the cell [17]. Therefore, UAPs could predominate over BAPs at the media depth of 0–10 cm. It has also been reported that UAPs present significantly better biodegradability than BAPs [17]. This characteristic of UAPs might lead to more degradation of SMPs and contribute to a reduction of DON at this level of the media.

At the media depth of 10–200 cm, biomass concentration and microbial activity decreased gradually. The levels of biodegradable organic matter and other nutrients decreased with media depth. Hence, the behavior of the microbes transformed gradually from substrate utilization to endogenous respiration, leading to the formation of more residual dead cells. More BAPs were produced, leading to poor overall SMP biodegradability. The rate of SMPs generation was much higher than that of substrate utilization. SMPs accumulated gradually along the media depth of the BAC filter, resulting in an increase of the DON concentration at the media depth of 10–200 cm.

It has been reported that SMPs have a broad spectrum of molecular weight [30]. UAPs were mainly micro-molecules resulting from the substrate utilization, while BAPs from cellular decay were larger in molecular weight [30]. It can be seen from Fig. 5 that small molecular weight (<6 kDa) DON in 10 cm media depth was higher than that of the influent. Biomass concentration and microbial activity reached their maximum at the 10 cm media depth. This might be because at the 10 cm media depth microbes consumed many more organic compounds and thus caused higher production of UAP with lower molecular weight. Subsequently, the large molecular weight (>20 kDa) DON in the effluent was increased. With increasing media depth, the behavior of the microbes was gradually transformed from substrate utilization to endogenous respiration. The BAPs gradually increased and accumulated, resulting in a significant increase of the organic matter with large molecular weight. By this mechanism, the large molecular weight of DON in effluent was distinctly increased compared with the molecular weight distribution of DON in influent and in samples from the 10 cm media depth.

In the media depth of 0–10 cm, the DON was oxidized to $NH_4^+$-N by the ammonification process, and nitrifying bacteria oxidized $NH_4^+$-N to $NO_3^-$-N via the process of nitrification, i.e. DON→$NH_4^+$-N→$NO_2^-$-N→$NO_3^-$-N. Other pathways, such as microbial assimilation may be involved in the transformation of $NH_4^+$-N [31]. A portion of the $NH_4^+$-N might be assimilated to form cell components, then becoming SMPs and finally, being released into the water as DON. Therefore, the rate of DON oxidized was higher than the $NH_4^+$-N assimilation rate, and the concentration of $NH_4^+$-N, $NO_3^-$-N, and TDIN was increased in the 0–10 cm media depth (Fig. 3(C)). Along the media depth of the BAC filter, the rate of $NH_4^+$-N assimilation was higher than the DON oxidation rate, leading to a decrease in $NH_4^+$-N, $NO_3^-$-N, and TDIN.

As shown in Fig. 6 and Table 1, the EEM parameters from influent to effluent obviously suggest that there is a good relationship between the DON concentration and the intensity of Peak A. This indicates that the variation of DON in the BAC filtration process depended heavily on the amount of aromatic protein-like substances. Previous studies confirmed that aromatic protein-like substances had a direct relationship with microbial activity. These protein-like substances might be transported into the system (allochthonous) or be created by microbial activity within the system (autochthonous) [32,33]. Therefore, aromatic protein-like substances generated during biofiltration should be SMPs. Our results indicated that SMPs account for the variation of DON during BAC filtration.

## 5. Conclusions

The profile of DON in a two-stage ozonation and BAC filtration drinking water treatment process was investigated in this study. Particular focus was paid to the variation of DON in the first BAC filter. Approximately 48.3% of DON was removed in this two-step ozonation and BAC filtration process. At 0–10 cm media depth of the BAC filter, DON was rapidly utilized which resulted in a reduction of the DON concentration. However, the accumulation of SMPs along the media depth caused the DON concentration to increase gradually. The trends of biomass and microbial activity were almost the same in the BAC filter. The biomass concentration and microbial activity first increased rapidly and then decreased gradually along the media depth. The small (<6 kDa) and the large (>20 kDa) molecular weight distribution of DON in influent, 10 cm media depth and effluent showed obviously variations that reflected the DON concentration in different media depths of the BAC filter. EEM results confirmed that the nitrogen enriched aromatic protein-like fractions of SMPs contributed to the variation of the DON concentration in the BAC filter. From the point of view of water quality management, the BAC filtration drinking water treatment process should be reconsidered to reach the minimum

concentration of DON in effluent because of its potential contribution to N-DPBs formation.

## Acknowledgments

The authors are grateful for the financial support for this work provided by the National Natural Science Foundation of China (NSFC) (No. 51208448), the Key Program of He'nan Educational Committee (No. 14A610004), the Key laboratory of the Three Gorges Reservoir Region's Eco-Environment, Chongqing University, the Xinyang Normal University Ph.D. Research Fund (No. 91231), the Key Laboratory of Geospatial Technology for Middle and Lower Yellow River Regions (Henan University), the Ministry of Education (GTYR2011006) and the Xinyang Normal University Youth Teachers Scheme (No. 120511).

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