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Molecular weight distributions of effluent organic matters and the biodegradability assessment of a typical advanced drinking water treatment plant in South China

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ABSTRACT

Biodegradability and molecular weight (MW) distributions of effluent organic matters were studied. Sample units of advanced drinking water treatment processes, namely, pre-ozonation, conventional water treatment (CWT), and main ozonation/biological activated carbon (BAC), were obtained from the drinking water treatment plant in South China. Biodegradable dissolved organic carbon (BDOC) and assimilable organic carbon (AOC), which is a component of BDOC, were measured as indicators of organic matter biodegradability and biological stability in drinking water. Results showed that the high-MW organic matter in raw water was oxidized into low-MW organic matter (MW < 3 kDalton) through preozonation process, thereby enhancing the BDOC content in raw water. BDOC components were removed by the subsequent CWT process. Components of the effluent organic matters obtained from the CWT process were completely decomposed through the main ozonation process, and the remaining organic matters were simultaneously oxidized into low-MW organic matters, thus increasing the BDOC content and improving the biodegradability. Finally, the low-MW BDOC was efficiently removed through BAC process. The AOC content in user water was over 100 μg L^{-1}, indicating that the factors contributing to biological instability remained. Additional attention should be given to BAC protection in China.

Keywords: Advanced water treatment; Biodegradability; Molecular weight; Biodegradable dissolved organic carbon; Assimilable organic carbon

1. Introduction

The safety of drinking water in relation to public health is an issue that is receiving increasing attention with an improvement of living standards and the implementation of the Drinking Water Quality Standard (GB5749-2006) in China. In particular, human activities contribute to an increasing amount of contaminants entering the water supply, resulting in further water deterioration [1,2]. However, the current

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conventional water treatment (CWT) process used in China cannot ensure the safety of drinking water. Therefore, advanced water treatment processes should be implemented to obtain excellent drinking water [2–4].

The ozone/biological activated carbon (BAC) advanced drinking water treatment process was developed in Europe during the 1970s [5]. Activated carbon water purification technology was introduced in China during the 1960s and the ozone/BAC process was investigated during the 1980s. At present, the ozone/BAC process is an effective advanced water treatment technology used to remove organic pollutants. Ozone/BAC process is also a hot topic in advanced drinking water treatment research [2,5–8]. Ozone/activated carbon technology has been extensively researched and applied in water purification processes in China [9–12]. The results showed that using this advanced technology ensures the drinking water safety in China.

Advanced water treatment processes remove the organic pollutants from water. The biological treatment process in advanced water treatment guarantees the complete removal of the biodegradable and low-/medium-molecular weight (MW) components of organic matters. Removal efficiency is limited if the biodegradable and MW organic matter components are low. Dissolved organic carbon (DOC) can be divided into nonbiodegradable dissolved organic carbon and biodegradable dissolved organic carbon (BDOC) according to microorganism utilization efficiency, and the BDOC component that can be used by bacteria to generate a cell body is known as assimilable organic carbon (AOC). AOC and BDOC are closely related to the growth of heterotrophic bacteria in the drinking water distribution system, which is the focus of the biological stability analysis of drinking water. AOC is an organic matter component that can be easily assimilated by bacterial organisms; BDOC is the substance and energy source used by bacteria and other microorganisms in the water for metabolic processes, such as assimilation and alienated consumption. A lower AOC or BDOC content in the water corresponds to a more difficult microorganism growth and reproduction [13]. AOC or BDOC content is related to the growth potential of heterotrophic bacteria in the drinking water pipe networks; therefore, either AOC or BDOC can be used as indicators in evaluating the biological stability in drinking water [12,13]. Van der Kooij [14] showed that the number of heterotrophic bacteria in a non-chlorinated system does not increase when AOC levels are lower than 10 $\mu g\,L^{-1}$. LeChevallier et al. [15–17] suggested that the regrowth in a system maintaining residual disinfectant may be limited when AOC levels are lower than 50–100 $\mu g\,L^{-1}$.

Therefore, the biodegradability, the MW distributions of effluent organic matters, and the AOC levels and their relationships in drinking water pipe networks in advanced water treatment processes should be investigated to evaluate the drinking water biological stability [18–20]. These critical investigations should be conducted in light of the lack of related research in China on the applicability of the ozone/BAC process [18,21].

This study investigates and evaluates the biodegradability and MW distributions of the effluent organic matters obtained from a typical advanced water treatment plant in South China. Preozonation, CWT, and ozonation/BAC process units were investigated in this study. The biological stability (AOC content) of drinking water was also evaluated to assess the feasibility of the processes.

2. Materials and methods

2.1. Sampling locations

The drains of the advanced water treatment processes of the drinking water treatment plant examined in our study and the sampling locations (a–h) are shown in Fig. 1.

Batches of 10 L water samples were obtained on the same day. Raw water samples were collected from the micro-polluted lake, which is the water source of the water treatment plant. Water user samples were obtained from a tap connected to the pipe network. Other water samples were collected from special

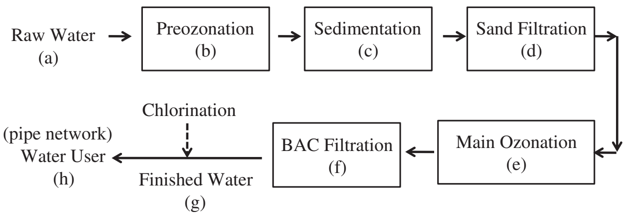

Fig. 1. A schematic diagram of the advanced treatment processes drains and sampling locations. (a) Raw water from the micro-polluted lake; (b) Preozonation concentration: 0.5 mg L^{-1}, contact time: 4 min; (c) Coagulation with AlO3, 18 g m^3, coagulation time: 28 min, Sedimentation tank: water flow rate at 10–25 mm s^{-1}; (d) Sand Filtration: quartz sand with particle size 0.9–1.35 mm, flow rate at 8 m h^1; (e) Main Ozonation concentration: 1.5 mg L^{-1}, contact time: 10 min; (f) BAC Filtration: filtration rate 11–12 m h^1, contact time 10–11 min, BAC particle size 0.9–1.35 mm, filtration period 2–5 d; (g) Chlorination concentration: 2 mg L^{-1}.

sampling points at each process unit. All the water samples were placed in an icebox immediately after collection and analyzed within 24 h.

2.2. MW fractioning

The measurement method developed by Wang et al. [21] was used in this study. The MW distribution was obtained by means of parallel filtration method using regenerated cellulose acetate ultrafiltration (UF) membranes with different molecular cut-offs. The DOC contents were measured by using a total organic carbon analyzer (Jena multi N/C 2100, Germany) to determine the various organic matter content in the filtrates.

The membranes were first immersed in ultrapure water and then placed in a refrigerator (4℃) for preservation. Prior to use, the membranes were leached with ultrapure water until the UV_{254} values of the effluents (measured using Spectrophotometer-725, Shanghai Optical Instrument, China) were equal to that of the ultrapure water. Parallel method was conducted as follows. First, the water samples were filtered using 0.45 μm mixed cellulose ester microfiltration membranes (Millipore, USA). The water samples were then filtered using the Millipore PL series YM100, YM10, YM5, YM3, YM1, and YC500 UF membrane models with corresponding molecular cut-offs measuring 100, 10, 5, 3, 1, and 0.5 kDa, respectively (1 kDa viz. 1,000 Dalton). Finally, the DOC content in the UF filtrates with various MW distribution ranges was measured using subtraction method.

2.3. BDOC determination

The measurement procedure developed by Liu et al. [22] and Tihomirova et al. [23] was used in this study. The principle behind dynamic cycle method for determining BDOC is to cycle the water samples through biologically active vector particles. Therefore, the biodegradable organic matter was fully decomposed until the DOC value in the reactor effluent reached a constant. The DOC in water samples was measured at certain times of this procedure. BDOC value is typically determined by the difference between the initial and final DOC concentrations.

The ceramisites were first inoculated with bacteria to form biofilms. Each water sample was filtered using 0.45 μm microfiltration membrane to obtain 2 L filtrate. The filtrate flowed through the biological media, which had a diameter of 200 m and a height of 1.5 m, at a speed ranging from 3 to 5 mL min^{-1}. Samples (sample volume: 10 mL) were collected from the effluents every 60 min. Stable DOC was obtained approximately 27 h later. BDOC was determined by the difference in DOC content value between the initial DOC and the stable DOC. A detection limit of 0.1 mg L^{-1} was used in the BDOC bioassay.

2.4. AOC determination [13]

Water samples measuring 600 mL were collected in clean, carbon-free Erlenmeyer flasks, pasteurized for 30 min at 60℃ in a water bath, and then rapidly cooled to 15℃ in an ice bath to eliminate influential factors. Phosphate salt (CAS No: 7664-38-2, 99.9%) was added to the samples to obtain a final concentration of 400 μg L^{-1} $K_2HPO_4 \cdot 3H_2O$. Sodium thiosulfate (CAS No: 7772-98-7, 99.8%) measuring 50 mg L^{-1} was added to the 600 mL flask to neutralize chlorine in the water samples. All of the chemicals were of analytical reagents (Chemical Co, Beijing).

The *Pseudomonas fluorescens* strain P17 created in the ozone oxidation process consumed most of the BDOC in the water samples, with the exception of oxalic acid (CAS No.: 144-62-7, 99.6%). Therefore, the *Spirillum* strain NOX, which contains oxalic acid as a growth substrate, was also selected for AOC content determination. First, P17 was introduced into the samples. The amount of bacteria at the bio-stable period was recorded. The samples were then pasteurized and inoculated with NOX. The amount of P17 and NOX bacteria was converted to AOC–acetic carbon content and AOC–oxalic carbon content, respectively.

3. Results and discussion

3.1. MW distributions

Figs. 2 and 3 exhibit various MW distributions of the effluents obtained from preozonation, conventional treatment, and ozonation/BAC process units, as well as various MW distribution percentages of the effluents obtained from each process unit.

The MW distributions of organic matters obtained from various types of raw waters in the drinking water treatment process were markedly different. However, the water treatment process units displayed similar characteristics in the removal of organic matters with different MWs [19,21]. The results showed that high-MW organic matters in raw water were oxidized into organic matters with low MW (< 3 kDa) after undergoing preozonation process. The amount of organic matters with MW < 3 kDa increased by 12.87% (Figs. 2 and 3). High-MW organic matters were effectively removed after coagulation

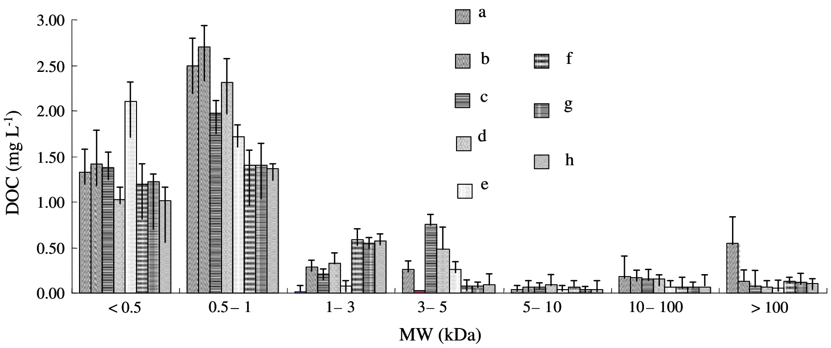

Fig. 2. DOC content and MW distributions in the effluent of each process unit.

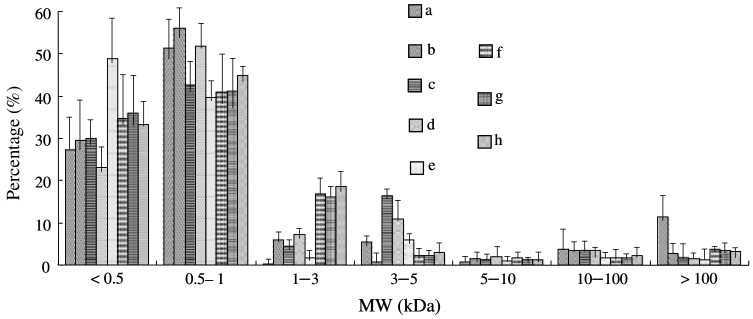

Fig. 3. Percentage of MW distributions in the effluent of each process unit.

and sedimentation because strongly hydrophobic and high-MW organic matters easily form into large flocs, which are adsorbed into the solid–liquid interface [24]. Therefore, the high-MW organic matters were effectively removed through sedimentation. The low-MW (0.5–1 kDa) organic matters were easily removed because they were prone to adsorption by the high-MW organic matters as a result of preozonation, consequently decreasing the ζ-potential of the organic particles. Thus, the colloids in the water are stabilized. Additional components of the high/medium-MW organic matters were removed after filtration; however, the efficiency in the removal of organic matters with MW < 3 kDa was low, particularly when MW ranged from 0.5 to 3 kDa. The low-MW organic matters displayed a high hydrophilic performance; therefore, forming large flocs for interception or adsorption was difficult. The low-MW organic matters were likely to be eliminated from surface of the filter media because they were affected by either flow shear stress or competitive adsorption. Accordingly, a negative removal rate trend was determined, and the amount of organic matters with MW < 3 kDa slightly decreased. The removal rate of DOC in the effluents of the preozonation, coagulation–sedimentation, and filtration process units was only 8.44%.

The dissolved organic matters with MW < 3 kDa were a major component of raw water. These dissolved matters were inefficiently removed by the preozonation, coagulation–sedimentation, and filtration processes (Fig. 2). This inefficient removal was the main reason why the dissolved organic matter removal efficiency of conventional processes was low. The MWs of the organic matters were notably reduced to the low MW ranges, and the efficiencies in the removal of DOC with MW ranging from 1 to 3 kDa and MW ranging from 3 to 100 kDa were significantly higher than that of DOC with MWs > 100 kDa. The total DOC removal rate after main ozonation was low (3.15%), implying that the DOC with high-MW organic matters has been removed in the previous processes.

High efficiency in the removal of the dissolved organic matters with MW values in the remaining distribution ranges was achieved after activated carbon adsorption, with the exception of the efficiency in the removal of the dissolved organic matters with MWs > 100 kDa. Microscopy revealed a large number of bacteria, native micelles, and metazoans on the granular-activated carbon surface. The microorganisms effectively degrade low-MW organic matters because main ozonation significantly decomposes organic matters. Consequently, organic matter biodegradability significantly improves, and adequate dissolved oxygen is provided for the microbial activities in activated carbon. The increase in the amount of DOC with MWs > 100 kDa after activated carbon adsorption can be attributed to the dissolution of microbial metabolite residues in the water [25].

The amount of DOC with MWs > 100 kDa slightly decreased after finished water chlorination. The chlorine oxidation effect decomposed the high-MW organic matters into medium-/low-MW organic matters. The amount of DOC with MWs < 1 kDa in the user water was also decreased. This result is attributed to the consumption of low-MW organic matters by the growth of microorganisms in the pipeline network. A similar phenomenon was observed with BDOC.

3.2. Biodegradability assessment

Prominent changes occurred in the BDOC and AOC of each unit under the normal operational conditions of the water treatment plan (Fig. 1). The changes in the biodegradability of the organic matters in the effluents (from a to h) obtained from the advanced water treatment processes are shown in Fig. 4 and Tables 1 and 2.

BDOC content in the effluent obtained from preozonation process significantly increased because of the strong ozone oxidation that decomposed the high-MW, hydrophobic organic matters into the low-MW, hydrophilic organic matters (Fig. 4). Approximately 30% of the BDOC was removed by the complex precipitation in the sedimentation tank. An additional 20% of the remaining BDOC was continually removed through sand filtration and microorganism degradation. Components of the non-biodegradable organic matters in the effluent were oxidized into biodegradable organic matters through main ozonation after sand filtration, and the effluent organic matters were oxidized further into low-MW organic matters. This oxidation resulted in a further increase in BDOC content in the effluent obtained from the main ozonation process. The water conditions after the ozonation process were ideal for microorganism growth and reproduction in the subsequent BAC process. Approximately 30 to 40% of the BDOC were removed after this process.

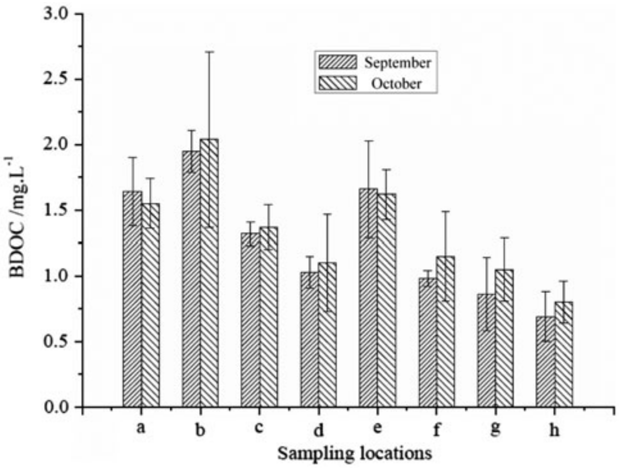

Fig. 4. BDOC content in the effluent of each process unit.

Table 1
AOC content (mean ± SD) (measured on 31 May) ($n = 3$) (operational conditions shown in Fig. 1)

Samples	AOC$_{P17}$ (μg L^{-1})	AOC$_{NOX}$ (μg L^{-1})	AOC (μg L^{-1})
Raw water (a)	65.00 ± 0.94	36.46 ± 0.61	101.45 ± 1.48
Sand filtration water (d)	61.56 ± 0.61	35.39 ± .046	96.96 ± 1.05
Finished water (g)	82.84 ± 1.82	44.27 ± 0.67	127.11 ± 1.15
User water (h)	73.92 ± 1.05	40.01 ± 0.34	113.93 ± 1.03

Table 2
AOC (mean ± SD) content (measured on 8 October) ($n = 3$)

Samples	AOC$_{P17}$ (μg L^{-1})	AOC$_{NOX}$ (μg L^{-1})	AOC (μg L^{-1})
Raw water (a)	75.90 ± 1.48	20.60 ± 0.30	96.50 ± 2.64
Sand filtration water (d)	72.40 ± 0.86	16.60 ± 0.24	89.00 ± 1.12
Finished water (g)	45.60 ± 0.49	46.40 ± 0.31	92.00 ± 0.93
User water (h)	61.80 ± 0.19	27.80 ± 0.17	89.60 ± 0.86

AOC content was measured on 31 May and 8 October (Tables 1 and 2). The results showed that the AOC$_{P17}$ and AOC$_{NOX}$ contents in the effluent obtained from the sand filtration process were both low.

Jiang and Xu [26] determined that the flocculation/sedimentation processes can remove –255.7 to 79.3% of AOC, whereas the filtration process can remove –353.1 to 67.5% of AOC. The two processes displayed low AOC removal efficiencies, although the AOC removal rates were all positive.

Finished water chlorination causes an increase in AOC content; however, the main ozonation process oxidizes high-MW organic matters into low-MW organic matters and increases the amount of the AOC component removed by BAC (Tables 1 and 2).

The AOC measured on 31 May was not efficiently removed after the BAC process. The disinfection of the finished water with chlorine even enhanced the increase in AOC content. This phenomenon is mainly caused by chlorine, which is a strong oxidant used to kill bacteria. Accordingly, chlorine reacts easily with the organic matters in drinking water, resulting in the disinfection of by-products. Low-MW organic matters such as carboxylic acids, amino acids, aldehydes, and ketones constitute additional AOC [27]. The AOC content of 127.11 and 113.93 $\mu g\,L^{-1}$ in the finished water and user water, respectively, suggested that the drinking water was biologically instable as per LeChevallier et al. [15–17], who determined that regrowth may be limited by AOC levels lower than 50 to 100 $\mu g\,L^{-1}$ in a system maintaining residual disinfectants.

The AOC content (8 October) of 92 $\mu g\,L^{-1}$ in finished water indicated that the drinking water was biologically stable. However, the microbial degradation of organic matters declined with the temperature decline in October. BDOC content in finished water reached 1.05 $mg\,L^{-1}$. The ratio of AOC and BDOC contents was only 8.8%, thus showing that the AOC removal efficiency of BAC was rather low. This result is consistent with the conclusions of Shu et al. [28]. The removal rate of AOC by the entire preozonation + CWT + Ozonation/BAC process was only –25.3 to 4.66%. AOC content in the finished water released by the plant was relatively low, whereas BDOC content was relatively high when the water temperature dropped. Thus, the factors contributing to the biological instability of drinking water should be given particular attention.

At present, the recommended AOC level limit is 200 $\mu g\,L^{-1}$ or less in consideration of the social development and economic condition of China [17]. The advanced drinking water processes examined in our study satisfy the current basic requirements of Chinese residents.

4. Conclusions

Preozonation process oxidized relatively high-MW organic matters in raw water into hydrophilic organic matters with MWs < 3 kDa. Main ozonation process lowered the MW of effluent organic matters into the low MW ranges. DOC matters with various MW distribution ranges were efficiently removed through the BAC process, with the exception of DOC matters with MWs > 100 kDa. Preozonation process significantly increases BDOC content, whereas CWT process removes part of the BDOC.

The AOC_{P17} and AOC_{NOX} contents in the effluent obtained from sand filtration process were both less than 100 $\mu g\,L^{-1}$, implying that the presence of AOC is guaranteed when the advanced water treatment processes are implemented. The advanced drinking water processes examined in our study satisfy the current basic requirements of Chinese residents. However, the factors contributing to biological instability remain; thus, additional attention must be given to BAC protection.

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