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Removal of viruses and disinfection by-products at two drinking water treatment plants in southern China

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ABSTRACT

The removal of viruses and disinfection by-products (DBPs) at two water plants (A and B) in a city of southern China was investigated. Coliphages MS2 and Phix174 were used as indicators of viruses. The results clearly indicated that the removal efficiency of MS2 and Phix174 was over 99% by conventional coagulation/sedimentation with polyaluminium sulfate chloride (PACS) at Plants A and B. MS2 and Phix174 were not detected in the effluent of these two plants. However, the removal efficiency of UV_{254} and TOC by coagulation/sedimentation was only 22.05% and 64.57% at Plant A, and 17.64% and 11.89% at Plant B, respectively. The species and amount of DBPs and their formation potential had no significant decrease after coagulation/sedimentation. In biological filtration, UV_{254} and TOC could be removed 6.62% and 24.50%, respectively. However, more soluble microbial products (SMP) and extracellular polymeric substances (EPS), the precursors of DBPs, were produced. Granular activated carbon (GAC) was efficient in the removal of UV_{254} and TOC, with an efficiency of 16.18% and 14.85%, respectively. Sand filtration, the final process, performed poorly in removing viruses and DBPs precursors. Both the processes of Plants A and B were not efficient in the removal of DBPs and their precursors.

Keywords: Drinking water; Viruses; Disinfection by-products (DBPs); Removal efficiency; Water plants; Southern China

1. Introduction

The microbiological safety of drinking water is key for human health and widely investigated in the world [1–4]. It is important to consider enteric viruses in water quality studies; not only because of their incidence as causal agents for diarrhea disease [5], but also due to their characteristics which allow them to survive in the environment for long periods of time, and tolerate changing environmental conditions [6,7]. The MS2 and Phix174 coliphages have been typically proposed as indicators of enteric viruses for drinking water treatment [8,9].

Disinfection is essential to inactivate microbial pathogens in the water supply and remove certain physical–chemical contaminants. However, potentially harmful disinfection by-products (DBPs) are produced during the disinfection process [10]. Epidemiological studies have shown most of the DBPs present in drinking water are either carcinogenic or potential carcinogenic [11,12]. In order to prevent the production

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of DBPs, one strategy is to decrease their formation potential through the removal of precursors before disinfection.

The reduction of risks from pathogenic microorganism and DBPs exposure is very important for protecting human health. But previous research has mostly focused on pathogenic microorganisms or DBPs individually [13–17]. They did not comprehensively research the removal of both pathogenic microorganisms and DBPs using different treatment technologies in the drinking water treatment process, especially in at the drinking water plant.

The present study comprehensively investigated how two water treatment plants, which had different water sources and treatment technology, performed in the removal of viruses and the DBPs formation potential at several successive stages. The goal was to identify the key steps in the treatment process which could remove viruses and DBPs efficiently. The results provide a deeper understanding of the performances of individual treatment processes, and provide a reference to help ensure the safety of drinking water.

2. Materials and methods

2.1. Water treatment plants and water sources

The water treatment plants investigated for this study are referred to as Plant A and Plant B, with capacities of 1,600,000 $m^3\ d^{-1}$and 40,000 $m^3\ d^{-1}$, respectively. Both plants, which have different water sources, are located in southern China. Plant A is surrounded by many factories which discharged an increasing amount of wastewater directly into the local river. Therefore, the river water quality is deteriorating shown by the increase in organics, the rise in ammonia concentration, and the decrease of dissolved oxygen. The water source of Plant B is an inland river with several cargo handling wharves located in the upstream. In addition, chemical fertilizers are widely used in agriculture in that area. Therefore, the quality of water source of Plant B is mainly influenced by agriculture pollution and inland waterway transportation.

The purification processes of Plants A and B are shown in Fig. 1. That of Plant A is a conventional coagulation/sedimentation – sand filtration – chloramine disinfection process. About 20 mg/L of coagulant, (PACS) and 12% sodium hypochlorite (NaClO) are added together in the pipeline. The V-filter tank is filled to a 1200 mm height with an even-granular quartz sand medium of 0.8–1.2 mm diameter. Free chlorine and ammonia are added to the connecting pipe between the filtration tank and clean water reservoir. The dosage of NaClO in chloramine disinfection is adjusted to maintain a residual chlorine level of 1.5–2.0 mg/L in the effluent, which would guarantee the residual chlorine level to be over the 0.05 mg/L level required by the national water standard at the end of the large distribution system.

As shown in Fig. 1, the water purification process of Plant B is a biological filtration–coagulation/sedimentation–GAC–chloramine disinfection–sand filtration process. The biological filter is filled to a 2000 mm height with biostyrene material of 5 mm diameter. The dosage of coagulant PACS ranges from 30–120 mg/L, which is adjusted by the change of water quality and temperature. The GAC tank contains dual media of sand and GAC. The upper GAC medium, with a depth of 1500 mm, has a particle size of 0.8–1.5 mm. This is followed by a lower sand medium which has a particle size of 2.0–16 mm and a depth of 500 mm. NaClO and ammonia are added to the disinfection tank with 120 min of contact time. The dosage of NaClO and ammonia are adjusted to maintain a residual chlorine level of 2.0 mg/L in the effluent. The sand filtration tank is filled with quartz sand with particle size and depths from top to bottom are as follows: 0.6–1.2 mm/900 mm, 1.0–2.0 mm/100 mm, 4.0–8.0 mm/100 mm, 8.0–16.0 mm/100 mm and 16.0–32.0 mm/300 mm. The water samples were taken from the effluent of every process section in Plants A and B.

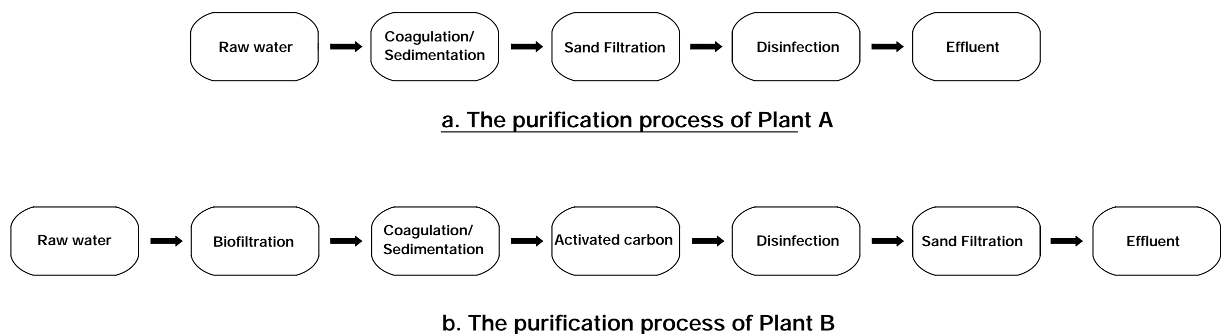

Fig. 1. A flowchart of water treatment processes at Plant A and Pant B.

2.2. Enumeration of virus

As referred to above, coliphages are similar in size, shape, components with human enteric viruses and show similar resistance to environment and water treatment processes. Therefore, the MS2 and Phix174 coliphages are used as indicators of viruses.

The MS2 and Phix174 coliphages are concentrated by a new electropositive filter [18]. NanoCeram (Argonide, Sanford, FL) and Zeta Plus Virosorb 1MDS (Cuno, Meriden, CT) electropositive cartridge filters were used in this study. The enumeration method of MS2 and Phix174 coliphages was performed according to BSI methodology as follows [19]: The concentrated sample is mixed with a small volume of semi-solid nutrient medium; a culture of host strain is added and plated on a solid nutrient medium; Finally incubation and reading of plates for visible plaques is performed. The results are expressed as the number concentration of plaque-forming particle (C_{pfp}) per unit of volume.

2.3. Analysis of DBPs formation potential

2.3.1. Chlorination experiments

The chlorination experiments were conducted in 50 mL glass bottles. The bottles were pre-soaked in a potassium dichromate–sulfuric acid solution for 30 min, rinsed with tap water, then triple-rinsed with Milli-Q water. Each water sample was dosed with a sodium hypochlorite solution (Clorox Bleach, 52.8 g/L) at Cl_2/TOC (total organic carbon) = 4. All samples were stored without headspace for 1 day at 25 °C in the dark and after which each 15 mL samples of the chlorinated water were used to determine the species of trihalomethanes (THMs) and haloacetic acids (HAAs) [20,21]. The free chlorine was standardized by the *N*,*N*-diethyl-*p*- phenylenediamine titrimetric method [22]. TOC was determined using the TOC analyzer (TOC-VCPN, Shimadzu, Japan).

2.3.2. DBPs extraction and analysis

The species of THMs and HAAs were investigated at first, and the results were the basis of following quantitative analysis. Qualitative analysis of THMs and HAAs were processed according to the standard method [22,23]. GC/MS analyses with electron ionization (70 eV) and GC column (VF-WAXms 30 m long, 0.25 mm internal diameter, 0.25 μm coating) were performed on a Finnigan Voyager gas chromatograph/mass spectrometer using the NIST and NBS library database. The oven temperature was programmed as follows: an initial temperature of 50 °C was held for 2 min, and then increased at a rate of 20 °C per min to 250 °C for 10 min. The temperature of the injector was set at 230 °C. Helium was used as the carrier gas at a rate of 10 mL/min and with a split ratio of 15:1.

The standard substances of THMs and HAAs were purchased according to the results of qualitative analysis. Quantitative analysis of THMs and HAAs were processed according to the EPA standard method [24]. GC-ECD (HP6890) with HP-5 capillary column (30m × 0.25 mm × 0.25 μm) was used for the determination of THMs. The oven temperature was programmed as follows: an initial temperature of 40 °C was held for 2 min, and then increased at a rate of 10 °C per min to 280 °C for 10 min. The temperature of the injector was set at 230 °C. Nitrogen was used as the carrier gas at a rate of 10 mL/min and with a split ratio of 10:1. GC-ECD (HP6890) with HP-5 capillary column was used for the determination of HAAs too. The oven temperature was programmed as follows: an initial temperature of 40 °C was held for 5 min, and then increased at a rate of 10 °C per min to 230 °C for 5 min. The temperature of the injector was set at 220 °C. Nitrogen was used as the carrier gas at a rate of 10 mL/min and with a split ratio of 10:1.

3 Results and discussion

3.1. Removal of viruses

The removal performance of viruses, using MS2 and Phix174 coliphages as indicators, by processes of Plant A is listed in Table 1. The titer of MS2 and Phix174 in the raw water reached 2.77×10^4 PFU/100 L and 8.71×10^3 PFU/100 L, respectively, which could be harmful to human health because of the higher concentration. After the coagulation/sedimentation process, levels of MS2 and Phix174 were decreased 3log (99.91%) and 2log (99.86%), respectively. In this procedure, MS2 and Phix174 were removed significantly by sedimentation

Table 1
The removal of two coliphages by the processes at Plant A

Plant A	MS2 titer(PFU/100 L)	Removal efficiency (%)	Phix174 titer (PFU/100 L)	Removal efficiency (%)
Raw water	2.77×10^4	–	8.71×10^3	–
Sedimentation	2.45×10^1	99.91	1.23×10^1	99.86
Sand filtration	0	100.00	0	100.00
Effluent	0	100.00	0	100.00

of coagulant PACS and inactivation of the disinfectant NaClO which was added together with coagulant. However, the removal efficiency of the two coliphages did not reached the drinking water standard of the USEPA, which demands the inactivation or removal efficiency of coliphages in drinking water treatment system must achieve 4log (99.99%) [25].

A previous study showed that optimized granular media filtration preceded by coagulation/flocculation could effectively remove viral indicators such as the MS2 coliphage in surface water filtration [26]. In the present study, MS2 and Phix174 were not detected after sand filtration and in the effluent, and the total removal efficiency of them both reached 100%. However, in fact, the actual removal amount of MS2 and Phix174 by sand filtration was very poor at only 2.45×10^1 and 1.23×10^1 PFU/100 L, respectively. The result of the present study coincides with Nasser et al. [14], who employed a coagulation and high rate filtration process to treat surface water samples containing various pathogenic viruses. They observed this process was efficient for the removal of pathogenic viruses from surface water as 93% MS2 and 80% poliovirus was eliminated. But the removal of MS2 coliphage by high rate filtration without the addition of alum was very poor, and the addition of alum enhanced the removal of viruses by high rate filtration.

In contrast to the treatment processes of Plant A, the process of biological filtration and GAC was employed in Plant B. The removal efficiency of MS2 and Phix174 by the processes of Plant B is listed in Table 2. The titer of MS2 and Phix174 in raw water reached 5.00×10^3 and 5.95×10^4 PFU/100 L, respectively. The biological filter was applied to treat the ammonia and organic loading in raw water. After biological pretreatment, the number of MS2 and Phix174 increased due to the mass multiplication of their host strain. Because various bacteria were liable to exist and propagate in the biological filter where dissolved oxygen was abundant and organic compounds present, MS2, Phix174 and other bacteria reproduced.

In the coagulation/sedimentation unit the removal amount of MS2 and Phix174 was 1.75×10^4 and 7.34×10^4 PFU/100 L, the removal efficiency which was relative to biological filter effluent was 99.95% and 99.94%, respectively. However, owing to increase of MS2 and Phix174 in the biological filter unit, the total removal efficiency only reached to 2log (98.29%) and 2log (99.38%), respectively. For deeply treating ammonia nitrogen and organic compounds, GAC was applied during the Plant B process. The results listed in Table 2 show that the titer of MS2 and Phix174 changed little during the process of GAC. The removal efficiency was only increased to 98.57% and 99.82%, and corresponding removal amount was 1.43×10^1 and 2.64×10^2 PFU/100 L, respectively. Table 2 also shows that after disinfection and in the effluent of sand filtration, or in the finished water, MS2 and Phix174 were not detected, the removal efficiency of them both reached 100%.

Due to the two plants having different water sources, the titer of MS2 was higher than Phix174 in raw water of Plant A, but the result of Plant B was opposite (Tables 1 and 2). It is obvious that the processes of Plants A and B could remove both MS2 and Phix174 efficiently with different titer in raw water, and the coagulation/sedimentation unit was the key step. But the function of coagulation/sedimentation in the removal of MS2 and Phix174 was not clear for Plant A which employed pre-chlorination and coagulation together. The size of MS2 and Phix174 are too small, only 32 nm and 26 nm, respectively [9], to act as the cohesive core of coagulation process. But in Plant B, it is clear that MS2 and Phix174 were removed mostly by the coagulation/sedimentation unit. It can be seen in Table 3 that turbidity was also removed mostly in the coagulation/sedimentation unit with a removal efficiency of 55.61%. Therefore, it can be inferred that the coliphages may adsorb on suspended particles in raw water and be removed with turbidity in coagulation/sedimentation unit. As for the function of the sand filter in the removal of MS2 and Phix174, the removal amount of two coliphages by the sand filtration unit of Plants A and B was zero.

Table 2
The removal of two coliphages by the processes at Plant B

Plant B	MS2 titer (PFU/100 L)	Removal efficiency (%)	Phix174 titer (PFU/100 L)	Removal efficiency (%)
Raw water	5.00×10^3	–	5.95×10^4	–
Biological filtration	1.76×10^4	–	7.38×10^4	–
Sedimentation	8.57×10^1	98.29	3.71×10^2	99.38
GAC	7.14×10^1	98.57	1.07×10^2	99.82
Disinfection	0	100.00	0	100.00
Sand filtration	0	100.00	0	100.00
Effluent	0	100.00	0	100.00

Table 3
The removal of turbidity, UV_{254} and TOC by the processes at Plant B

Plant B	Turbidity (NTU)	Removal efficiency (%)	UV_{254} (cm^{-1})	Removal efficiency (%)	TOC (mg/L)	Removal efficiency (%)
Raw water	31.1	–	0.136	–	8.08	–
Biological filtration	17.8	42.77	0.127	6.62	6.10	24.50
Sedimentation	0.505	98.38	0.103	24.26	5.11	36.39
GAC	0.201	99.35	0.081	40.44	3.94	51.24
Disinfection	0.242	99.22	0.086	36.76	2.38	70.54
Sand filtration	0.202	99.25	0.085	37.50	2.30	71.53
Effluent	0.238	99.23	0.091	33.09	2.36	70.79

3.2. Removal of DBPs and their formation potential

3.2.1. Removal of DBPs and their formation potential at Plant A

The species of THMs and HAAs in the original water samples of Plant A were different at every process (Table 4). It was shown that $CHCl_3$ and $CHBr_3$ were detected in the processes but other THMs were not found. It can be inferred that $CHCl_3$ existed widely in the water source of Plant A because only $CHCl_3$ was detected in raw water. The $CHCl_2COOH$ (DCAA), which is one type of HAAs, was detected in the processes of Plant A yet no HAAs were detected in the raw water.

The chlorination experiment was conducted to investigate the species of THMs and HAAs formation potential, as shown in Table 4. Four species of THMs formation potential ($CHCl_3$, $CHCl_2Br$, $CHClBr_2$ and $CHBr_3$) and four species of HAAs formation potential (CCl_3COOH (TCAA), DCAA, CHClBrCOOH (BCAA) and $CHBr_2COOH$ (DBAA)) existed in the source water. However, after pre-chlorination at Plant A which was conducted with coagulation, only two species of THMs ($CHCl_3$ and $CHBr_3$) and one species of HAAs (DCAA) were detected in the original water sample of the coagulation/sedimentation process. One reason for this was that the concentration of NaClO was slightly lower at Plant A than in the laboratory. Another possible reason was that the concentration of the other species of THMs ($CHCl_2Br$ and $CHClBr_2$) and three species of HAAs (TCAA, BCAA and DBAA) formation potential in the raw water were possible lower.

After coagulation/sedimentation, sand filtration and in the final effluent, the species of THMs and HAAs formation potential was the same as in raw water. This result indicated that the conventional coagulation/sedimentation and sand filtration did not efficiently remove the species of THMs and HAAs formation potential. The previous research has shown that natural organic compounds (NOM), such as humic acid and fulvic acid, had been identified as primary precursors for chlorinated DBPs [27,28]. These organic compounds components were difficult to remove through the conventional coagulation/sedimentation process [29]. TOC concentration of water was generally a good indicator to determine the amount of THMs and other DBP precursors present [30]. The absorption of a solution at 254 nm (UV_{254}) is also commonly used as a surrogate parameter of DBPs precursor [31,32]. The removal of TOC and UV_{254} by processes of Plant A is listed in Table 5. According to Table 5, only 4.25 mg/L of TOC was removed in the end, and the removal efficiency was 65.18%. The removal amount of TOC by coagulation/sedimentation and sand filtration was 4.21 mg/L and 0.33 mg/L and the corresponding removal efficiency was 64.57% and 4.56%, respectively. The removal of UV_{254} by coagulation/sedimentation and sand filtration was 0.028 and 0 cm^{-1}, which corresponded to removal efficiencies of 22.05% and 0%, respectively. These results show that coagulation/sedimentation was not efficient in the removal of DBPs precursors which were reflected by TOC and UV_{254}. Sand filtration had no effect on the removal of DBPs precursors. It is reasonable to conclude from these data that the conventional coagulation/sedimentation and sand filtration could not remove THMs and HAAs precursors effectively, and did not decrease the species of THMs and HAAs formation potential.

According to the results of qualitative analysis, four HAAs ($CH_2ClCOOH$ (MCAA), $CH_2BrCOOH$ (MBAA), DCAA and TCAA) were measured in present study. As presented in Fig. 2, only one species of THMs ($CHCl_3$) and two species of HAAs (DCAA, TCAA) existed in raw water. The concentration of $CHCl_3$ was higher than others significantly which was up to 7.58 μg/L. Due to pre-chlorination employed together with coagulation, after coagulation/sedimentation process, $CHCl_3$ and TCAA decreased to 4.13 μg/L and 0 μg/L, respectively, $CHCl_2Br$, $CHClBr_2$, $CHBr_3$, DCAA and MBAA increased to 2.39 μg/L, 2.17 μg/L, 1.33 μg/L, 4.97 μg/L and 1.49 μg/L,

Table 4
The species of DBPs in the original and chlorinated drinking water samples of Plant A

Plant A		$CHCl_3$	$CHCl_2Br$	$CHClBr_2$	$CHBr_3$	CCl_3COOH	$CHCl_2COOH$	CHClBrCOOH	$CHBr_2COOH$
Raw water	Original sample	D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.	D.
Coagulation/ sedimentation	Original sample	D.	N.D.	N.D.	D.	N.D.	D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.	D.
Sand filtration	Original sample	D.	N.D.	N.D.	D.	N.D.	D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.	D.
Effluent	Original sample	D.	N.D.	N.D.	D.	N.D.	D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.	D.

D is a written abbreviation for 'detectable'.
N.D. is a written abbreviation for 'not detectable'.

Table 5
The removal of turbidity, UV_{254} and TOC by the processes at Plant A

Plant A	Turbidity (NTU)	Removal efficiency (%)	UV_{254} (cm^{-1})	Removal efficiency (%)	TOC (mg/L)	Removal efficiency (%)
Raw water	13.5	–	0.127	–	6.52	–
Sedimentation	0.193	98.57	0.099	22.05	2.31	64.57
Sand filtration	0.232	98.28	0.099	22.05	1.98	69.13
Effluent	0.254	98.12	0.108	14.96	2.27	65.18

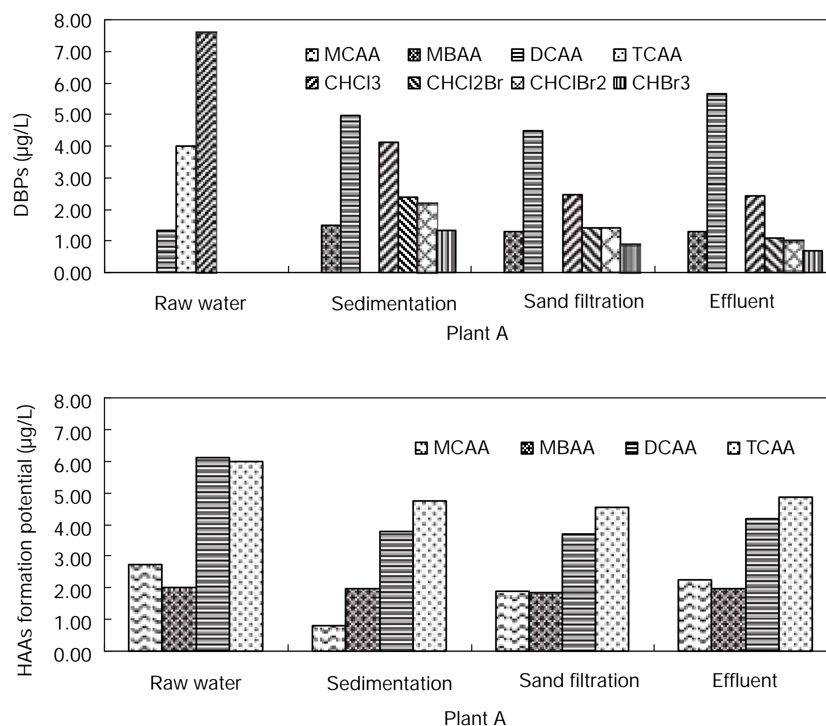

Fig. 2. The variation of DBPs and HAAs formation potential at several stages of Plant A.

respectively. After sand filtration and in the final effluent, all four THMs and two HAAs changed slightly. The effluent had meted the standards for drinking water quality of China [33]. As shown in Fig. 2, after the raw water was treated by the processes of Plant A, the formation potential of MCAA, MBAA, DCAA and TCAA only decreased from 2.72 µg/L to 2.26 µg/L, 2.00 µg/L to 1.95 µg/L, 6.09 µg/L to 4.18 µg/L and 5.97 µg/L to 4.88 µg/L. It is obvious that the conventional coagulation/sedimentation and sand filtration processes are not efficient in the removal of DBPs and their formation potential.

3.2.2. Removal of DBPs and their formation potential at Plant B

The species of THMs and HAAs in the original and chlorinated drinking water samples of Plant B is listed in Table 6. This data shows that $CHCl_3$ was the only species of DBPs in the raw water of Plant B, and no HAAs were detected. Four species of THMs ($CHCl_3$, $CHCl_2Br$, $CHClBr_2$ and $CHBr_3$) and three species of HAAs (DCAA, BCAA and DBAA) formation potential existed in the water source. Table 6 demonstrates that four species of THMs ($CHCl_3$, $CHCl_2Br$, $CHClBr_2$ and $CHBr_3$) and three species of HAAs (DCAA, BCAA and DBAA) were detected in all chlorinated samples of Plant B. The processes of Plant B did not efficiently decrease the species of THMs and HAAs formation potential. The removal efficiency of TOC and UV_{254} by the processes of Plant B, which are the most important two surrogate parameters for DBPs formation potential, is listed in Table 3. TOC decreased from 8.08 mg/L to 2.36 mg/L (removed 70.97%) by the integrated water treatment system of Plant B. In particular it was removed efficiently by the biological filtration unit, which specifically removed 1.98 mg/L. UV_{254} decreased from 0.136 cm^{-1} to 0.091 cm^{-1} (removal of 33.09%) by the processes of Plant B. The removal efficiency of UV_{254} by the biological filtration unit was 6.62%. Furthermore, as shown in Table 6, the species of DBPs formation potential had no change between raw water and the biological filter effluent.

Fig. 3 shows the infrared spectrogram of raw water and biological filter effluent of Plant B. This comparison shows that the organic functional groups, with the wave numbers of 3000–2800 cm^{-1} in the biological filter effluent spectrogram, had increased. Two peaks of organic groups were observed at 2922 cm^{-1} and 2853 cm^{-1} in the raw water. After biological filtration, the 2922 cm^{-1} peak moved to a higher wave number, the 2853 cm^{-1} peak had no change, and a new peak at 2956 cm^{-1} had emerged. This result is presumably due to the soluble microbial products (SMP) and extracellular polymeric substances (EPS) which are related to bacteria metabolism and cell lysis produced in biological filter. The different DBPs

Table 6
The species of DBPs in the original and chlorinated drinking water samples of Plant B

Plant B		$CHCl_3$	$CHCl_2Br$	$CHClBr_2$	$CHBr_3$	$CHCl_2COOH$	$CHClBrCOOH$	$CHBr_2COOH$
Raw water	Original sample	D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.
Biological filtration	Original sample	D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.
Coagulation/ sedimentation	Original sample	D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.
GAC	Original sample	D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.
Disinfection	Original sample	D.	D.	D.	D.	D.	D.	D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.
Sand filtration	Original sample	D.	D.	D.	D.	D.	D.	D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.
Effluent	Original sample	D.	D.	D.	D.	D.	D.	D.
	Chlorinated sample	D.	D.	D.	D.	D.	D.	D.

D. is a written abbreviation for 'detectable'.
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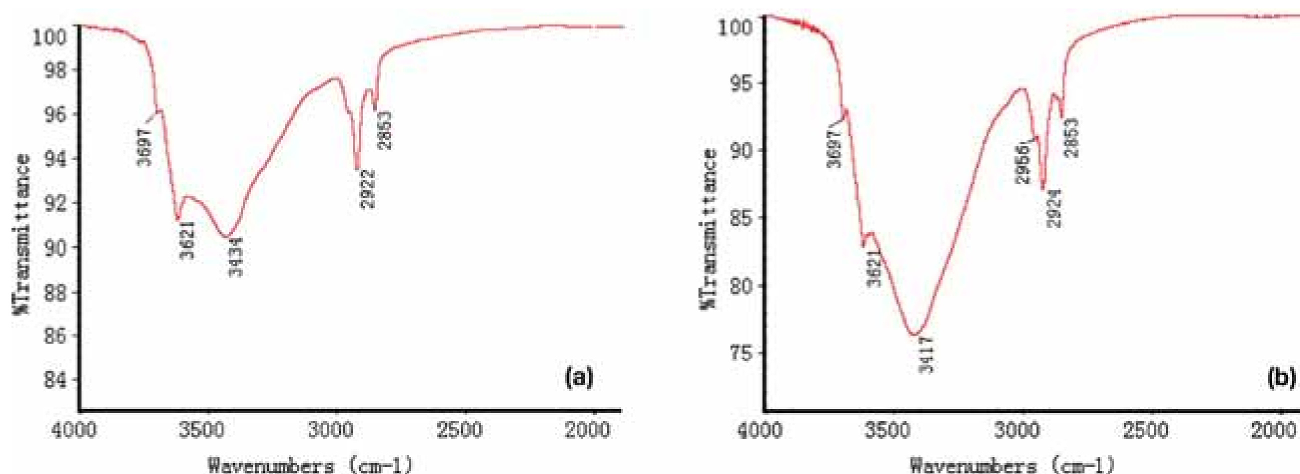

Fig. 3. The infrared spectrogram of raw water (a) and the biological filter effluent (b) of Plant B.

species came from different components of organic precursors. Although UV_{254} and TOC were decreased by bacteria, the decomposition products of organic precursors in raw water, SMP and EPS, could also react with disinfectant to form DBPs. Therefore, the species of DBPs formation potential had no change after biological filtration.

As presented, the removal amount of TOC was 0.99 mg/L and 1.17 mg/L in the coagulation/sedimentation and GAC unit, and the corresponding removal efficiency was 11.89% and 14.85%, respectively. The decrease of organic compounds was achieved through the highly efficient adsorption of GAC. The previous study showed similar results, where 20–30% of the dissolved organic carbon (DOC) was removed by the adsorption of GAC [34]. According to Table 3, UV_{254} was removed mostly in the coagulation/sedimentation and GAC units, and the corresponding removal efficiency of UV_{254} was 17.64% and 16.18%, respectively by these units. However, Table 6 demonstrated that the species of THMs and HAAs formation potential had no obvious difference among the biological filtration, sedimentation and GAC effluent. Coagulation/sedimentation and GAC almost did not remove the species of DBPs formation potential. Seven species of DBPs and their formation potential were detected in the chlorinated and original water samples of the disinfection unit at Plant B. However, after sand filtration, both the species of DBPs and their formation potential had no change. The corresponding removal efficiency of UV_{254} and TOC by sand filtration was only 0.74% and 0.99%. Sand filtration also was not efficient in the removal of the species of DBPs formation potential.

As shown in Fig. 4, DBPs were removed efficiently by biological treatment. Although TCAA increased from 6.11 μg/L to 6.23 μg/L, DCAA only decreased from 2.07 μg/L to 2.04 μg/L. However, $CHCl_3$ was removed effectively which decreased from 2.61 μg/L to 1.15 μg/L. After disinfection process, $CHCl_3$, $CHCl_2Br$, $CHClBr_2$, MBAA, DCAA and TCAA increased from 0.87 μg/L to 1.10 μg/L, 0 μg/L to 0.70 μg/L, 0 μg/L to 0.46 μg/L, 0 μg/L to 1.74 μg/L, 1.86 μg/L to 5.46 μg/L and 5.60 μg/L to 9.83 μg/L, respectively. Toxicological studies of individual DBPs suggested that brominated DBPs might be more carcinogenic than chlorinated analogues [35]. Therefore, the brominated DBPs which increased significantly are particularly important to consider. The effluent had met the standards for drinking water quality of China [33]. Fig. 4 also shows the variation of HAAs formation potential at several stages of Plant B. After the raw water was treated by the processes of Plant B, the total formation potential of HAAs was up to 24.11μg/L.

A comparison of Plants A and B demonstrates that both the processes of Plants A and B could not decrease the species and amount of DBPs efficiently. And also the species of DBPs formation potential showed no difference throughout the entire processes of Plants A and B. Therefore the technologies of both Plants A and B were not efficient in the removal of the species of DBPs formation potential. However, UV_{254} and TOC were removed efficiently in the processes of Plant B. We then conclude that the combined technologies of Plant B might more efficient in the removal of DBPs precursors than the conventional technology of Plant A. But from Figs. 2 and 4, we found the HAAs formation potential only decreased from 16.78 μg/L to 13.28 μg/L at Plant A and increased from 12.41 μg/L to 24.11 μg/L at Plant B. It was concluded that the processes of Plants A and B are not efficient in the removal of HAAs formation potential.

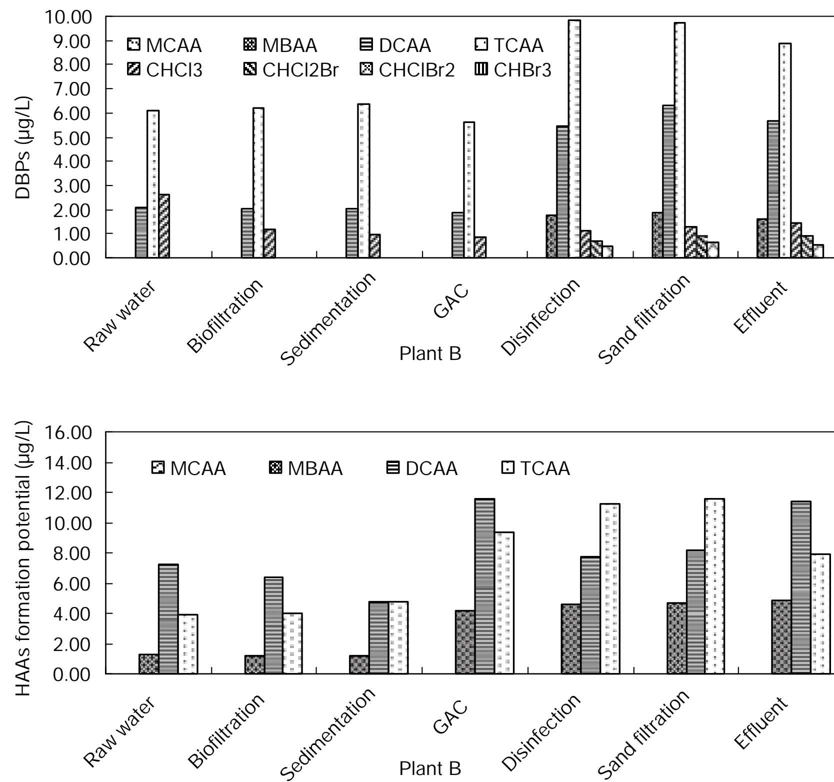

Fig. 4. The variation of DBPs and HAAs formation potential at several stages of Plant B.

4. Conclusions

$CHCl_3$ existed widely in the water sources of Plants A and B. The concentration of $CHCl_3$ was 7.58 μg/L and 2.61 μg/L in the raw water of Plants A and B, respectively. Coagulation/sedimentation could not efficiently remove DBPs precursors, which was reflected by UV_{254} and TOC. However, MS2 and Phix174 coliphages were removed efficiently. The removal of UV_{254} and TOC by coagulation/sedimentation was 22.05% and 64.57% at Plant A, and 17.64% and 11.89% at Plant B, respectively. The removal of MS2 and Phix174 coliphages by coagulation/sedimentation reached 99.91% and 99.86% at Plant A, and 99.95% and 99.94% at Plant B, respectively.

GAC was efficient in the removal of UV_{254} and TOC, with an efficiency of 16.18% and 14.85%, respectively. However, GAC was not efficient in the removal of DBPs and their formation potential. The role of sand filtration in removal of DBPs and coliphages can be neglected. The removal of UV_{254} and TOC by sand filtration was 0% and 4.56% at Plant A, and 0.74% and 0.99% at Plant B, respectively. The species and amount of DBPs and their formation potential showed no obvious change after sand filtration. The removal of MS2 and Phix174 coliphages by sand filtration was 0.09% and 0.14% at Plant A, and 0% and 0% at Plant B, respectively.

Biological filtration produced the new DBPs precursors SMP and EPS potentially which should be emphasized. UV_{254} and TOC were removed by 6.62% and 24.50% through biological filtration, respectively. But after biological filtration a new organic functional group shown at wave numbers of 2956 cm^{-1} emerged and may be included with SMP and EPS, and react with disinfectant to form DBPs.

Both the processes of Plants A and B were not efficient in the removal of DBPs and HAAs formation potential. The total amount of DBPs decreased from 12.89 μg/L to 12.19 μg/L at Plant A and increased from 10.79 μg/L to 19.11 μg/L at Plant B, respectively, and the total amount of HAAs formation potential decreased from 16.78 μg/L to 13.28 μg/L at Plant A and increased from 12.41 μg/L to 24.11 μg/L at Plant B.

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