



“Gheorghe Asachi” Technical University of Iasi, Romania



## BEHAVIOUR OF OZONATION BY-PRODUCTS DURING ADVANCED DRINKING WATER TREATMENT WITH PEARL RIVER WATER

Yue Wu<sup>1,2,3\*</sup>, Chun-De Wu<sup>1,2</sup>, Zhi-Lin Zhang<sup>1,2</sup>, Fauzia Naluswata<sup>1</sup>,  
Bo-Jie Yuan<sup>1</sup>, Jia-Li Liang<sup>1</sup>

<sup>1</sup>College of Environmental Science and Engineering, South China University of Technology, Guangzhou 510006, China

<sup>2</sup>The Key Laboratory of Pollution Control and Ecosystem Restoration in Industry Clusters of Ministry of Education, Guangzhou 510006, China

<sup>3</sup>Shantou Polytechnic, Shantou 515041, China

### Abstract

Simulation experiments were conducted to investigate the behaviour of ozonation by-products and the removal of organic matter in the treatment of the Pearl River raw water. The treatment processes include pre-ozonation, conventional treatment processes (coagulation/sedimentation and sand filtration), post-ozonation and granular activated carbon (GAC) filtration. Treatment efficiency of each unit process was evaluated by using several parameters such as permanganate index ( $\text{COD}_{\text{Mn}}$ ), ultraviolet absorbance at 254 nm ( $\text{UV}_{254}$ ), bromate ( $\text{BrO}_3^-$ ) and formaldehyde. The overall conversion rates of  $\text{BrO}_3^-$  in the six water samples were 0.43~5.54 %. Treated water flowed through the pre-ozonation unit process in which  $\text{COD}_{\text{Mn}}$  and  $\text{UV}_{254}$  were greatly removed. The conventional treatment processes had poor ability to remove  $\text{BrO}_3^-$ , but were effective in the removal of formaldehyde. In the post-ozonation unit process, the concentrations of  $\text{BrO}_3^-$  and formaldehyde reached the highest value. GAC filtration enhanced the removal efficiency of  $\text{BrO}_3^-$  compared with the conventional treatment processes. Water samples from the Xijiang and Beijiang River of the Pearl River basin contained higher concentrations of  $\text{BrO}_3^-$  and lower values of  $\text{COD}_{\text{Mn}}$ ,  $\text{UV}_{254}$  and formaldehyde in the final effluents than those from the Dongjiang River.

*Key words:* bromate, formaldehyde, granular activated carbon, ozone, Pearl River raw water

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### 1. Introduction

The Pearl River Basin is the second largest river area in China and an important water source in South China. It mainly consists of the Dongjiang River, Xijiang River, and Beijiang River with extremely abundant tributaries. In China, most drinking water treatment plants have adopted the typical conventional water treatment processes including coagulation/sedimentation, sand filtration, and disinfection (Xia et al., 2004). In recent years, the surface waters in the Pearl River Delta have been polluted by increasing discharges of industrial wastewater, domestic wastewater, and agricultural

activities, leading to micropollutant loads as well as conventional pollutants (Gan et al., 2013). Organics in drinking water can cause odor, taste, color, and bacterial regrowth, which cannot be effectively removed by the conventional treatment processes. Meanwhile, bromide concentrations would increase in raw waters once the seawater intruded, which might increase the total disinfection by-products (DBPs) yield during chlorine disinfection (Yang et al., 2010). Nowadays, new strict regulations for water quality and water sources have been imposed in the world (Fabian et al., 2016; Pérez et al., 2016). Therefore, it is essential to upgrade the conventional drinking water treatment processes, and it is urgent to add advanced

\* Author to whom all correspondence should be addressed: e-mail: 5.yue@163.com

water treatment processes into the existing drinking water facilities at present.

The combination of ozone and granular activated carbon (GAC) (hereafter referred as  $O_3/GAC$ ), which is available to remove taste, odor, ammonia nitrogen, organic matters, and the precursors of disinfection by-products (DBPs) such as trihalomethanes (THMs) and haloacetic acids (HAAs), is widely used for the advanced drinking water treatment (Badawy et al., 2012; Gümüő and Akbal, 2017; Gurzäu et al., 2013; Kim et al., 1997; Kim and Hyun, 2012; Kurniawan et al., 2006). Based on the previous studies, ozone was a powerful oxidant and disinfectant in drinking water treatment (Badawy et al., 2012). Pre-ozonation prior to coagulation was to decrease color, turbidity, taste, odor, and total THMs formation potential, and would also result in considerable COD reduction and coagulation enhancement (Hua and Reckhow, 2013; Kim and Hyun, 2012; Selcuk et al., 2005). Several reports have demonstrated that coagulation was a typical and basic unit process in water purification for removing suspended solids (SS) and natural organic matter (NOM) (Xiao et al., 2010, 2013). In post-ozonation, ozone was added after sand filtration process and prior to GAC process, which would result in enhancing the absorption capacity of GAC by converting larger molecules into smaller ones (Amy et al., 1991; Chiang et al., 2002). However, there would be by-products, such as bromate ( $BrO_3^-$ ) and aldehydes, formed during ozonation in bromide containing water (Hua and Reckhow, 2013; Kim and Hyun, 2012; Wang et al., 2017). It has been confirmed that bromate is regarded as the 2B carcinogen by the International Agency for Research on Cancer (Delker et al., 2006). It was the

only ozonation disinfection by-product (DBP) which was regulated in drinking water. As such, a maximum limit of  $10 \mu g \cdot L^{-1}$  has been set for this substance in drinking water (Xu et al., 2012). Formaldehyde, which is widely known as a toxic chemical for human, can enter drinking water supplies by the oxidation of NOM during ozonation (Hill et al., 2009; Lewis and Chestner, 1981). The health-based guideline value of  $0.9 mg \cdot L^{-1}$  for formaldehyde in drinking water has been derived by World Health Organization (WHO, 1996). So far, there are few reports about the variation of ozonation by-products formed during the advanced drinking water treatment of the Pearl River raw water.

The main objectives of this paper are to investigate the behaviours of bromate and formaldehyde in raw waters along the overall process including pre-ozonation, conventional treatment processes (coagulation/sedimentation and sand filtration), post-ozonation, and GAC filtration by simulated experiments, and to explore the feasibility of the processes used to purify raw waters.

## 2. Experimental

### 2.1. Water samples

Six water samples were collected from the source water in Guangdong Province of the Pearl River Basin during the period of May 2011 to June 2011. A location map of the sampling sites was depicted as Fig. 1. Six water samples were taken from the Dongjiang River, Xijiang River, and Beijiang River, respectively. The water samples were collected during the rainy season. Typical characteristics of the Pearl River raw water was given in Table 1.

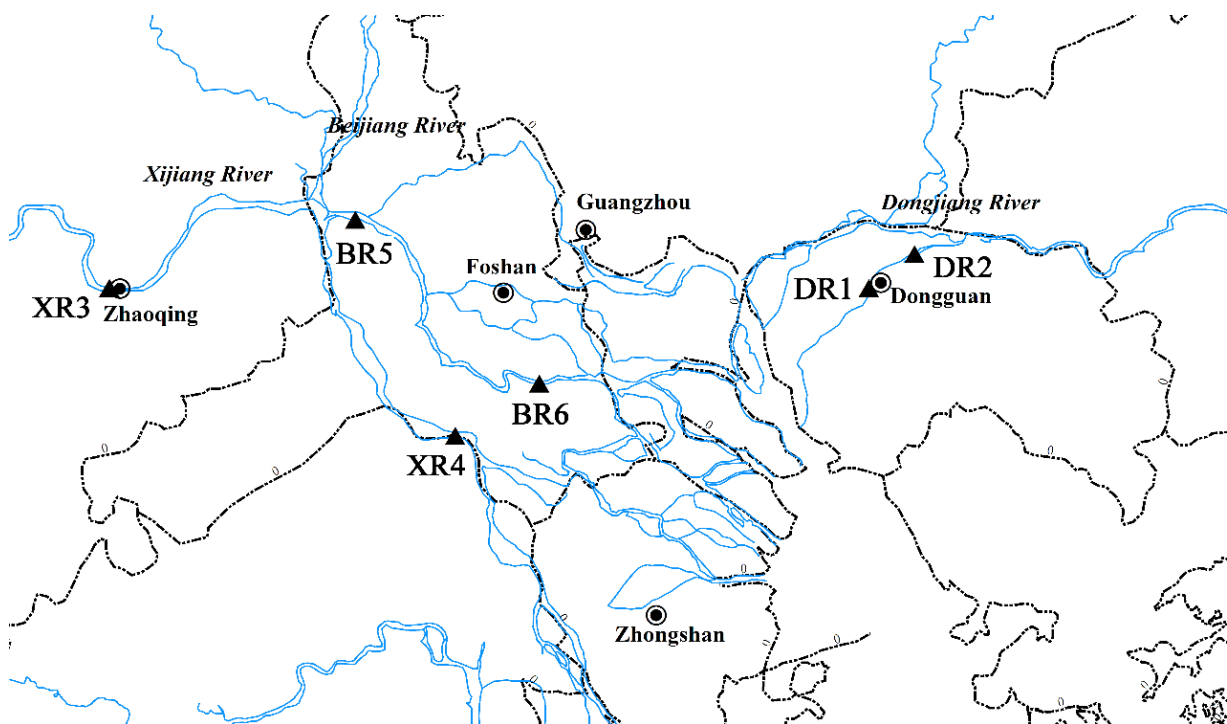


Fig. 1. Map showing the sampling area in Guangdong Province. “▲” are the sampling sites of raw waters within the Pearl River. “○” are cities. “—” are the Pearl River (DR: Dongjiang River, XR: Xijiang River, BR: Beijiang River)

## 2.2. Experimental methods

The samples of the raw water were treated by an advanced drinking water treatment process composed of pre-ozonation, conventional treatment processes (coagulation/sedimentation and sand filtration), post-ozonation, and GAC filtration. The overall treatment process was illustrated in Fig. 2.

Before the analysis, all the water samples were filtered with 0.45  $\mu\text{m}$  membrane films to remove suspended particles. Due to the low content of  $\text{Br}^-$  (only 3.56–6.09  $\mu\text{g}\cdot\text{L}^{-1}$ ) and the precursors of formaldehyde during the rainy season, six water samples were supplemented by adding 400  $\mu\text{g}\cdot\text{L}^{-1}$   $\text{Br}^-$  and 2  $\text{mg}\cdot\text{L}^{-1}$  acrylic acid, respectively.

Pre- and post-ozonation were employed using an ozonation system, which consisted of an ozone generator (Ozone Generator, HF-3), a reactor and an ozone destructor. Ozone was produced from air by the ozone generator and both pre- and post-ozone dosage were 2  $\text{mg}\cdot\text{L}^{-1}$ . The reactor had a total volume of 2 L with an inner diameter of 8 cm and a height of 40 cm, and was equipped with a sampling port. The outlet gas from the reactor was passed into the ozone destructor (2 % KI) in order to decompose the excess ozone to molecular oxygen before being discharged into the atmosphere.

Firstly,  $\text{O}_3$  from the ozone generator flowed into the glass reactor containing 1.5 L of water sampler, with 0.02  $\text{m}^3\cdot\text{h}^{-1}$  large contaminants flux and 1 min 55 sec aeration time, and then stopped when the concentration of  $\text{O}_3$  reached to a determined value. The ozone gas was continuously bubbled into the water through a sand core aerator at the bottom of the reactor. At the same time, the magnetic stirrer was switched on and started to record. Samples were taken ten minutes later. The concentration of  $\text{O}_3$  was evaluated and the concentrations of  $\text{BrO}_3^-$  and  $\text{Br}^-$  were also tested after the residual ozone quenched by 1.5  $\text{g}\cdot\text{L}^{-1}$  sodium nitrite solution.

Jar tests were performed on each water sample (4  $\times$  1.0 L) for coagulation process at room temperature (about 25  $^\circ\text{C}$ ), using a six-paddle stirrer (ZR4-6, CHN). 30  $\text{mg}\cdot\text{L}^{-1}$  Polyaluminium Chloride (PAC) as a coagulant was added in the rapid mixing basin. The contents of the jars were rapidly mixed at a paddle speed of 200 rpm for 2 min, followed by slow mixing at 50 rpm for 10 min and settling for 30 min.

The design of the sand filtration and GAC

filters allowed for water sampling over the filter bed height, as well as influent and effluent water samples. The sand filtration was packed to 0.8 m with sand media and had an effective size of 1.0 mm.

The filtration velocity was about 8  $\text{m}\cdot\text{h}^{-1}$ . GAC process was carried out with a granular activated carbon column, which was made of organic glass and had a height of 1 m and an inner diameter of 5 cm. The GAC column was filled with 0.8 m of GAC and had the effective sizes ranging from 1.0 to 2.5 mm. The loading density, filtration velocity, and residence time were 500  $\text{g}\cdot\text{L}^{-1}$ , 8  $\text{m}\cdot\text{h}^{-1}$ , and 10 min, respectively. Sand and GAC filters were equipped with backwashing devices, which enhanced the operational stability.

## 2.3. Analytical methods

Ozone was monitored in aqueous solution by its UV absorbance at 258 nm ( $\epsilon = 2950 \text{ L}\cdot\text{mol}^{-1}\cdot\text{cm}^{-1}$ ). The concentrations of  $\text{BrO}_3^-$  and  $\text{Br}^-$  were obtained using an ion chromatograph (Model ICS-1000, Dionex, CA), which was equipped with IonPac AS23 analytical column (250 mm  $\times$  4 mm)/IonPac AG23 guard column (50 mm  $\times$  4 mm) and AMMS-III anion suppressor (4 mm). The leachate consisted of a mixture of 0.8 mM sodium hydrogen carbonate ( $\text{NaHCO}_3$ ) and 4.5 mM sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) delivered at the flow rate of 1  $\text{mL}\cdot\text{min}^{-1}$ . An autosampler was assembled with a 200  $\mu\text{L}$  injection loop and the lowest detection limit for  $\text{BrO}_3^-$  and  $\text{Br}^-$  were less than 2.00  $\mu\text{g}\cdot\text{L}^{-1}$  (EPA, 2000).

Formaldehyde was analyzed by gas chromatography (HP 5890 Series II Plus, USA) with an electron capture detector (GC/ECD) according to the USEPA Method 556 (EPA, 1998). The lowest detection limit of formaldehyde was less than 0.11  $\mu\text{g}\cdot\text{L}^{-1}$ .

$\text{COD}_{\text{Mn}}$  was determined according to the national standard method of the People's Republic of China in 1989 (GB11892, 1989). The lowest detection limit of this method was 0.5  $\text{mg/L}$ . UV-254 absorbance was measured using a UNIC 2100 spectrophotometer (USA) at a wavelength of 254 nm with a 1 cm quartz cell. Turbidity was measured in a HACH 2100P TURBIDIMETER (USA). pH values were measured at 25  $^\circ\text{C}$  using a pHs-3C meter (CHN).  $\text{NH}_3\text{-N}$  was analyzed according to the USEPA standard methods (EPA, 2000).

**Table 1.** Typical characteristics of raw waters from the Pearl River

| Sample number | Water temperature ( $^\circ\text{C}$ ) | pH   | Turbidity (NTU) | $\text{COD}_{\text{Mn}}$ ( $\text{mg}\cdot\text{L}^{-1}$ ) | $\text{NH}_3\text{-N}$ ( $\text{mg}\cdot\text{L}^{-1}$ ) | $\text{UV}_{254}$ ( $\text{cm}^{-1}$ ) | $\text{Br}^-$ ( $\mu\text{g}\cdot\text{L}^{-1}$ ) |
|---------------|--|------|-----------------|--|--|--|---|
| DR1           | 25.52                                  | 6.88 | 43.7            | 2.76   | 1.83   | 0.042                                  | 3.92  |
| DR2           | 25.56                                  | 6.63 | 43.6            | 2.63   | 1.89   | 0.039                                  | 4.15  |
| XR3           | 25.35                                  | 7.21 | 20.8            | 1.67   | 0.97   | 0.026                                  | 3.56  |
| XR4           | 26.21                                  | 7.23 | 9.2             | 1.59   | 0.87   | 0.028                                  | 6.09  |
| BR5           | 26.68                                  | 7.35 | 31.2            | 1.84   | 0.48   | 0.026                                  | 4.77  |
| BR6           | 28.26                                  | 7.24 | 13.2            | 1.76   | 0.54   | 0.033                                  | 5.38  |

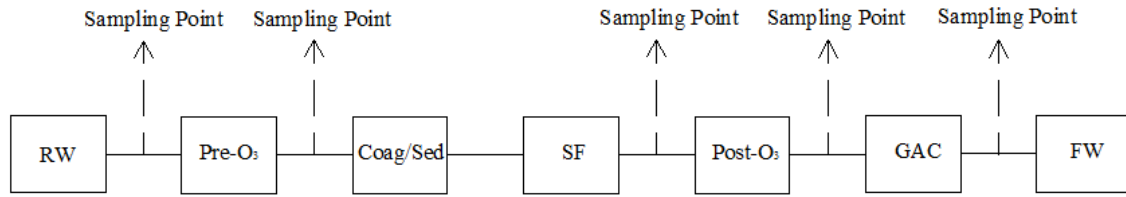


Fig. 2. The treatment processes in the experiment (RW: Raw Water, O<sub>3</sub>: Ozonation, Coag: Coagulation, Sed: Sedimentation, SF: Sand Filtration, GAC: Granular Activated Carbon, FW: Finished Water)

### 3. Results and discussions

#### 3.1. Effect of unit process on variations of Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup>

Bromate is an inorganic ozonation by-product of considerable health risk. Fig. 3 showed the variation of Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> concentrations in the overall process of raw and treated water samples. Fig. 3(a) showed that the variation trends of Br<sup>-</sup> in the six water samples throughout the overall process were opposite to BrO<sub>3</sub><sup>-</sup>. As shown in Fig. 3(b), though there were no BrO<sub>3</sub><sup>-</sup> existed in all the raw water samples, the amount of BrO<sub>3</sub><sup>-</sup> in the finished water from the overall process in the six water samples were in the order of BR5 > XR4 > BR6 > XR3 > DR1 > DR2, which corresponded to the overall conversion rates of 5.54, 4.95, 3.61, 1.35, 0.48, and 0.43 %, respectively.

The range of the conversion rates were 0.43~5.54 % and their average value was 2.73 %. Siddiqui et al. (1994) observed that up to 30 μg·L<sup>-1</sup> of BrO<sub>3</sub><sup>-</sup> might be formed by ozonation treatment, with an average of initial bromide concentration 100 μg·L<sup>-1</sup> in many drinking water supplies. It can also be found from Fig. 3(b) that the percentages of bromate formation of those samples were 0.37~7.21 % in pre-ozonation process and 0.76~13.71 % in post-ozonation process using ozone dosage 2 mg·L<sup>-1</sup> for each ozonation process. The conversion rates of BrO<sub>3</sub><sup>-</sup> in post-ozonation were higher than those in pre-ozonation. Pre-ozonation in this drinking water

treatment process resulted in a slight increase in BrO<sub>3</sub><sup>-</sup> concentration. The raw water samples BR5 and BR6 from the Beijing River, which had lower contents of organic matters and ammonia nitrogen than other samples, achieved higher concentrations of BrO<sub>3</sub><sup>-</sup> after pre-ozonation than those from the Dongjiang River. For the Sample XR4, the low concentrations of organic matters, ammonia nitrogen, and turbidity might be responsible to the high concentration of formed BrO<sub>3</sub><sup>-</sup>. Molnar et al. (2013) reported that the ozone-

NOM reaction was faster than the ozone-bromide reaction, resulting in the inhibition of BrO<sub>3</sub><sup>-</sup> formation by NOM during ozonation of bromide containing water. On the other hand, ozone could more rapidly oxidize ammonia in the presence of Br<sup>-</sup> which acted as a catalyst than that in its absence (Haag et al., 1984). In the post-ozonated water, the amount of BrO<sub>3</sub><sup>-</sup> in those water samples were in the order of BR6 > BR5 > XR4 > XR3 > DR2 > DR1. It is observed that the concentrations of BrO<sub>3</sub><sup>-</sup> were the highest. The formation concentration of BrO<sub>3</sub><sup>-</sup> in the Sample BR6 achieved the highest 58.24 μg·L<sup>-1</sup> among all the samples. This result was similar to the study taken by Kim and Hyun (2012). The BrO<sub>3</sub><sup>-</sup> concentrations of the six water samples were reduced to 4.6~69.5 % by the conventional treatment processes. GAC filtration enhanced the BrO<sub>3</sub><sup>-</sup> removal efficiency in comparison with the conventional treatment processes by adsorbing BrO<sub>3</sub><sup>-</sup> onto the pore surface of the activated carbons.

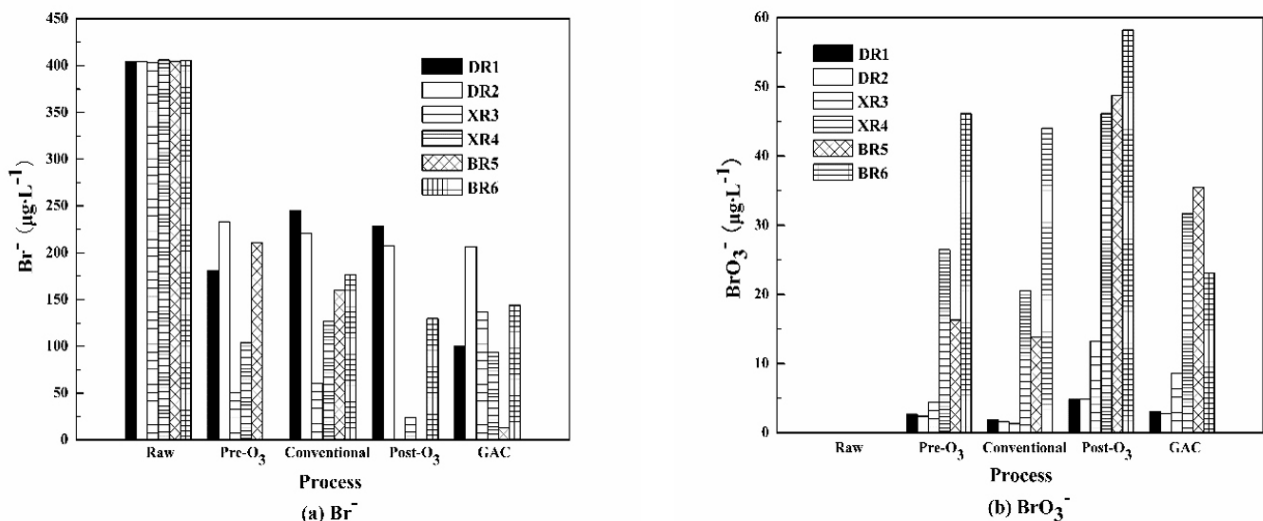


Fig. 3. The concentrations of Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> after each unit

In this study, the concentrations of  $\text{BrO}_3^-$  in those water samples during GAC filtration process were decreased from 4.80, 4.84, 13.27, 46.14, 48.75, and 58.24  $\mu\text{g}\cdot\text{L}^{-1}$  before inletting GAC reactor to 3.07, 2.78, 8.66, 31.66, 35.48, and 23.13  $\mu\text{g}\cdot\text{L}^{-1}$ , which corresponded to the removal percentages of 36.04, 42.56, 34.74, 31.38, 27.22, and 60.29 %, respectively. The results revealed that GAC could play an important role in the removal of  $\text{BrO}_3^-$  during the overall process. However, Results obtained by Asami et al. (1999) indicated that the removal rate of  $\text{BrO}_3^-$  apparently decreased during transition from new GAC to biological activated carbon (BAC) after 3 months.

### 3.2. Effect of unit process on variations of formaldehyde

Formaldehyde can be a useful indicator for the characteristics of organic by-products produced by ozonation. Fig. 4 showed the performance of each unit process for formaldehyde control in the six water samples. As a result, the variation trends of formaldehyde in those samples throughout the process were consistent with each other.

The results also showed that formaldehyde was not detected in each raw water and the concentrations of formaldehyde in the final effluents of those samples were 0.004, 0.003, 0.000, 0.002, 0.000, and 0.002  $\text{mg}\cdot\text{L}^{-1}$ , respectively. Post-ozonation caused formaldehyde formation except for the Sample XR3, which reached a peak value, followed by pre-ozonation unit. Melin and Odegaard (2000) have shown that reactions between ozone and humus could form low molecular weight compounds like aldehydes which were polar and present at low concentrations. Both the conventional treatment and GAC process were effective in the removal of formaldehyde in drinking water. According to Table 1 and Figs. 4,5 it was found that the formaldehyde concentration in finished water of the Dongjiang River, which had higher content of organic matters and ammonia nitrogen than those of the Xijiang River and Beijiang River, was the highest.

### 3.3. Effect of unit process on reductions of $\text{UV}_{254}$

$\text{UV}_{254}$ , which utilizes light at the UV 254nm wavelength to detect organic matters in water, is a water quality test parameter. This is due to the fact that the pollutants with conjugated double bonds and aromatic hydrocarbons strongly absorb light at the wavelength of 254 nm. The variation trends of  $\text{UV}_{254}$  values in raw and treated water were presented in Fig. 6. It can be seen that the removal pattern of  $\text{UV}_{254}$  along the overall process was similar to the removal trend of  $\text{COD}_{\text{Mn}}$ . Furthermore, the removal efficiency of  $\text{UV}_{254}$  was higher than that of  $\text{COD}_{\text{Mn}}$ . It was thought that unsaturated compounds, nitrogen compounds, and aromatic organics were easily

oxidized by ozone in comparison with the saturated aliphatic compounds.

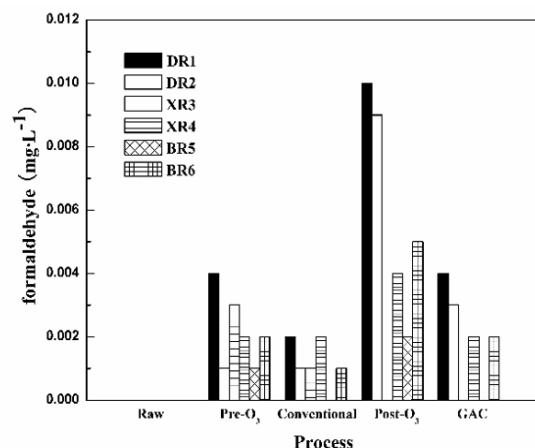


Fig. 4. The concentration of formaldehyde after each unit

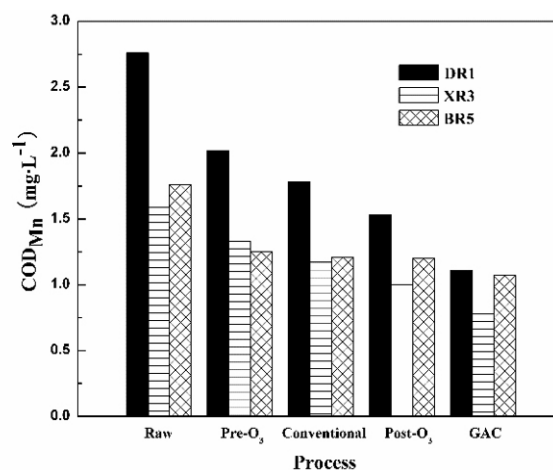


Fig. 5. The concentrations of  $\text{COD}_{\text{Mn}}$  in different units

The variation trends of  $\text{UV}_{254}$  values of six water samples throughout the process were consistent with each other. The  $\text{UV}_{254}$  values of those waters were 0.042, 0.039, 0.028, 0.026, 0.033, and 0.026  $\text{cm}^{-1}$ , respectively, while those of the finished water decreased to 0.011, 0.010, 0.001, 0.002, 0.001, and 0.002  $\text{cm}^{-1}$ , respectively.

As for the overall process, the total removal efficiencies of  $\text{UV}_{254}$  were in the order of BR5 (97.0 %) > XR3 (96.4 %) > XR4 (92.3 %) = BR6 (92.3 %) > DR2 (74.4 %) > DR1 (73.8%), of which pre-ozonation accounted for 54.8, 66.7, 75.0, 61.5, 72.7, and 65.4 %, respectively, making a great contribution to the total removal of  $\text{UV}_{254}$ .

The significant decrease of  $\text{UV}_{254}$  could be attributed to the degradation of the double bonds such as  $-\text{C}=\text{O}$  by ozonation. The addition of  $\text{O}_3$ -GAC treatment to the conventional treatment processes indeed improved the reduction of organic matters. From the results shown in Table 1 and Fig. 6, the  $\text{UV}_{254}$  values in finished water of the Dongjiang River

with higher contents of ammonia nitrogen and organic matters, was higher than those of others.

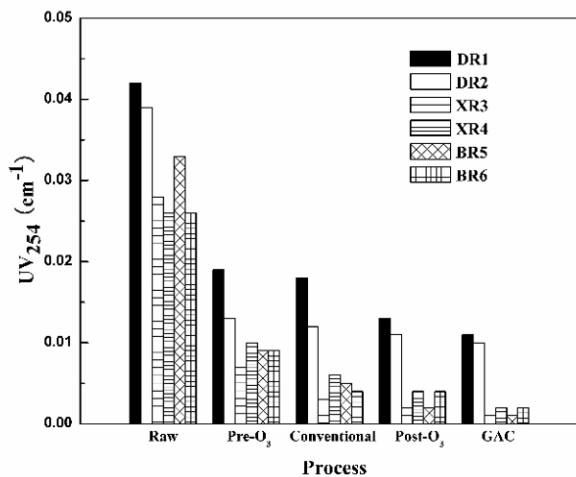


Fig. 6. The absorbance of UV<sub>254</sub> after each unit

#### 4. Conclusions

The following major conclusions were drawn from this study.

1. BrO<sub>3</sub><sup>-</sup> and formaldehyde concentrations were found to reach a peak value in post-ozonation, followed by pre-ozonation. GAC filtration enhanced the removal efficiencies of BrO<sub>3</sub><sup>-</sup> and formaldehyde in comparison with the conventional treatment processes.

2. COD<sub>Mn</sub> and UV<sub>254</sub> were greatly removed by pre-ozonation. The addition of O<sub>3</sub>/GAC treatment to the conventional treatment process indeed improved the reduction of organic matters. The reduction percentage of UV<sub>254</sub> was higher than that of COD<sub>Mn</sub>.

3. The raw waters from the Xijiang River and the Beijiang River, which had lower contents of organic matters and ammonia nitrogen, achieved higher concentrations of BrO<sub>3</sub><sup>-</sup> and lower values of COD<sub>Mn</sub>, UV<sub>254</sub> and formaldehyde after GAC process than those from the Dongjiang River.

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#### References

- Amy G.L., Tan L., Davis M.K., (1991), The effects of ozonation and activated carbon adsorption on trihalomethane speciation, *Water Research*, **25**, 191-202.
- Asami M., Aizawa T., Morioka T., Nishijima W., Tabata A., Magara Y., (1999), Bromate removal during transition from new granular activated carbon (GAC) to biological activated carbon (BAC), *Water Research*, **33**, 2797-2804.
- Badawy M.I., Gad-Allah T.A., Ali M.E.M., Yoon Y., (2012), Minimization of the formation of disinfection by-products, *Chemosphere*, **89**, 235-240.

- Chiang P.-C., Chang E.-E., Liang C.-H., (2002), NOM characteristics and treatabilities of ozonation processes, *Chemosphere*, **46**, 929-936.
- Delker D., Hatch G., Allen J., Crissman B., George M., Geter D., Kilburn S., Moore T., Nelson G., Roop B., Slade R., Swank A., Ward W., DeAngelo A., (2006), Molecular biomarkers of oxidative stress associated with bromate carcinogenicity, *Toxicology*, **221**, 158-165.
- EPA, (1998), Determination of carbonyl compounds in drinking water by pentafluorobenzylhydroxylamine derivatization and capillary gas chromatography with electron capture detection, Office of Research and Development, USEPA Method 556, Revision 1.0, U.S. Environmental Protection Agency, Cincinnati, Ohio 45268, On line at: <http://www.epa.gov/safewater/methods/pdfs/methods/met556.pdf>.
- EPA, (2000), Determination of inorganic anions by ion chromatography, USEPA Method 9056A, Revision 1.0, U.S. Environmental Protection Agency, Washington, D.C., On line at: [http://media.wix.com/ugd//6cb017\\_23c7c3a5ed8f22129f2362cf84dfa39b.pdf](http://media.wix.com/ugd//6cb017_23c7c3a5ed8f22129f2362cf84dfa39b.pdf).
- Fabian F., Fiore S., Genon G., Panepinto D., Nedeff V., Panainte M., (2016), Preliminary evaluation of the environmental impact of water treatment process, *Environmental Engineering and Management Journal*, **15**, 1867-1872.
- Gan W.-H., Guo W.-H., Mo J.-M., He Y.-S., Liu Y.-J., Liu W., Liang Y.-M., Yang X., (2013), The occurrence of disinfection by-products in municipal drinking water in China's Pearl River Delta and a multipathway cancer risk assessment, *Science of the Total Environment*, **447**, 108-115.
- GB1892, (1989), Water quality-Determination of permanganate index, the State Environmental Protection Administration of China.
- Gümüş D., Akbal F., (2017), A comparative study of ozonation, iron coated zeolite catalyzed ozonation and granular activated carbon catalyzed ozonation of humic acid, *Chemosphere*, **174**, 218-231.
- Gurzău A.E., Roman C.D., Dumitraşcu I., Gurzău E.S., (2013), Trihalomethanes variation in urine under different conditions of human exposure, *Environmental Engineering and Management Journal*, **12**, 257-263.
- Haag W.R., Hoigné J., Bader H., (1984), Improved ammonia oxidation by ozone in the presence of bromide ion during water treatment, *Water Research*, **18**, 1125-1128.
- Hill A.A., Lipert R.J., Fritz J.S., Porter M.D., (2009), A rapid, simple method for determining formaldehyde in drinking water using colorimetric-solid phase extraction, *Talanta*, **77**, 1405-1408.
- Hua G.-H., Reckhow D.A., (2013), Effect of pre-ozonation on the formation and speciation of DBPs, *Water Research*, **47**, 4322-4330.
- Kim Y., Lee Y., Gee C.S., Choi E., (1997), Treatment of taste and odor causing substances in drinking water, *Water Science and Technology*, **35**, 29-36.
- Kim Y.-J., Hyun K.-S., (2012), Characteristics of disinfection by-products reduction in the processes of drinking water treatment system using Nakdong river water, *Desalination and Water Treatment*, **43**, 159-166.
- Kurniawan T.A., Lo W.-H., Chan G.Y.S., (2006), Degradation of recalcitrant compounds from stabilized landfill leachate using a combination of ozone-GAC adsorption treatment, *Journal of Hazardous Materials*, **137**, 443-455.

- Lewis B.B., Chestner S.B., (1981), Formaldehyde in dentistry: a review of mutagenic and carcinogenic potential, *Journal of the American Dental Association*, **103**, 429-434.
- Melin E.S., Odegaard H., (2000), The effect of biofilter loading rate on the removal of organic ozonation by-products, *Water Research*, **34**, 4464-4476.
- Molnar J., Agbaba J., Dalmacija B., Tubić A., Krčmar D., Maletić S., Tomašević D., (2013), The effects of matrices and ozone dose on changes in the characteristics of natural organic matter, *Chemical Engineering Journal*, **222**, 435-443.
- Pérez A., Torres P., Escobar J.C., (2016), Hazard identification in watersheds based on water safety plan approach: Case study of Cali-Colombia, *Environmental Engineering and Management Journal*, **15**, 861-872.
- Selcuk H., Vitosoglu Y., Ozaydin S., Bekbolet M., (2005), Optimization of ozone and coagulation processes for bromate control in Istanbul drinking waters, *Desalination*, **176**, 211-217.
- Siddiqui M., Amy G., Ozekin K., Westerhoff P., (1994), Empirically and theoretically-based models for predicting brominated ozonated by-products, *Ozone: Science & Engineering*, **16**, 157-178.
- Wang F.-F., van Halem D., Ding L., Bai Y., Lekkerkerker-Teunissen K., van der Hoek J.P., (2017), Effective removal of bromate in nitrate-reducing anoxic zones during managed aquifer recharge for drinking water treatment: Laboratory-scale simulations, *Water Research*, **130**, 88-97.
- WHO, (1996), Guidelines for drinking-water quality-second edition-Volume 2-Health criteria and other supporting information, ISBN 92 4 154480 5, World Health Organization, Geneva, On line at: [http://www.who.int/water\\_sanitation\\_health/dwq/2edv012p1.pdf](http://www.who.int/water_sanitation_health/dwq/2edv012p1.pdf).
- Xia S.-J., Nan J., Liu R.-P., Li G.-B., (2004), Study of drinking water treatment by ultrafiltration of surface water and its application to China, *Desalination*, **170**, 41-47.
- Xiao F., Simcik M.F., Gulliver J.S., (2013), Mechanisms for removal of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) from drinking water by conventional and enhanced coagulation, *Water Research*, **47**, 49-56.
- Xiao F., Zhang X.-R., Zhai H.-Y., Yang M.-T., Lo I.M.C., (2010), Effects of enhanced coagulation on polar halogenated disinfection byproducts in drinking water, *Separation and Purification Technology*, **76**, 26-32.
- Xu C.-H., Shi J.-J., Zhou W.-Z., Gao B.-Y., Yue Q.-Y., Wang X.-H., (2012) Bromate removal from aqueous solutions by nano crystalline akaganeite ( $\beta$ -FeOOH)-coated quartz sand (CACQS), *Chemical Engineering Journal*, **187**, 63-68.
- Yang J.-S., Yuan D.-X., Weng T.-P., (2010), Pilot study of drinking water treatment with GAC, O<sub>3</sub>/BAC and membrane processes in Kinmen Island, Taiwan, *Desalination*, **263**, 1-3.