

## Removal of bromide and bromate from drinking water using granular activated carbon

Yong-qing Zhang, Qing-ping Wu, Ju-mei Zhang and Xiu-hua Yang

### ABSTRACT

Granular activated carbon (GAC) was used to remove bromide ( $\text{Br}^-$ ) and bromate ( $\text{BrO}_3^-$ ) from drinking water in both bench- and pilot-scale experiments. The present study aims to minimize  $\text{BrO}_3^-$  formation and eliminate  $\text{BrO}_3^-$  generated during the ozonation of drinking water, particularly in packaged drinking water. Results show that the  $\text{Br}^-$  and  $\text{BrO}_3^-$  levels in GAC-treated water decreased in both bench- and pilot-scale experiments. In the bench-scale experiments, when the empty bed contact time (EBCT) was 5 min, the highest reduction rates of  $\text{Br}^-$  in the mineral and ultrapure water were found to be 74.9% and 91.2%, respectively, and those of  $\text{BrO}_3^-$  were 94.4% and 98.8%, respectively. The GAC capacity for  $\text{Br}^-$  and  $\text{BrO}_3^-$  removal increased with the increase in EBCT. Reduction efficiency was better in ultrapure water than in mineral water. In the pilot-scale experiments, the minimum reduction rates of  $\text{Br}^-$  and  $\text{BrO}_3^-$  were 38.5% and 73.2%, respectively.

**Key words** | bromate, bromide, granular activated carbon, ozone, reduction

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### INTRODUCTION

Ozone, used as an alternative to chlorine in water treatment, has the advantages of significant effectiveness without generation of the disinfection by-products (DBPs) trichloromethane, haloacetic acids and other regulated DBPs. However, a DBP of bromate ( $\text{BrO}_3^-$ ), which is a suspected carcinogen, is generated during the ozonation of bromide ( $\text{Br}^-$ )-containing water. Thus, a maximum contaminant level of 10  $\mu\text{g/L}$  for  $\text{BrO}_3^-$  was established by the United States Environmental Protection Agency and by the drinking water standards of China (GB 5749–2006 and GB 8537–2008). A review of  $\text{BrO}_3^-$  occurrence also indicated that there have been issues of excessive  $\text{BrO}_3^-$  in drinking water in many countries (Xie & Shang 2006a).

To solve the excessive  $\text{BrO}_3^-$  problem in drinking water, a number of possible solutions for minimizing  $\text{BrO}_3^-$  formation, such as adding ammonia (Von Gunten 2003), reducing the pH (Von Gunten 2003), chlorine–ammonia process (Buffle *et al.* 2004; Wert *et al.* 2007) and optimizing the ozonation conditions (Bouland *et al.* 2004; Van der Helm *et al.* 2005), have been tested. A granular activated

carbon (GAC) treatment method also has been used for the removal of  $\text{BrO}_3^-$  generated during the ozonation of  $\text{Br}^-$ -containing water (Mills *et al.* 1996; Siddiqui *et al.* 1996; Bao *et al.* 1999; Huang *et al.* 2004a, b; Huang & Cheng 2008). These strategies are always applied for  $\text{BrO}_3^-$  control in domestic drinking water.

The Chinese standards of drinking natural mineral water (GB 8537–2008) and hygienic specifications of factory for drinking natural mineral water (GB 16330–1996) stipulated that ozone is the only disinfectant except ultra-violet irradiation to be used in the drinking natural mineral water treatment process. A survey showed that 64% of the 108 source water samples of natural mineral water contained  $\text{Br}^-$  levels above 50  $\mu\text{g/L}$  (Meng 2007). Von Gunten (2003) proposed that in waters with  $\text{Br}^-$  levels above 50  $\mu\text{g/L}$ ,  $\text{BrO}_3^-$  formation may exceed the maximum contaminant level of the drinking water standard under certain treatment conditions. Thus, the high  $\text{Br}^-$  concentration in the source water and the ozone application made  $\text{BrO}_3^-$  a serious issue in the mineral water industry in China.

The present study was performed to elucidate the capacity of GAC to remove  $\text{Br}^-$  and  $\text{BrO}_3^-$  from waters in bench-scale experiments. The effects of empty bed contact time (EBCT) and source waters on the levels and reduction rates of  $\text{Br}^-$  and  $\text{BrO}_3^-$  were evaluated. The efficiency of GAC in removing  $\text{Br}^-$  and  $\text{BrO}_3^-$  from water was also observed in the pilot-scale experiments. The results should provide some new references for controlling the occurrence of  $\text{BrO}_3^-$  during the ozonation of water with  $\text{Br}^-$ .

## METHODS

### Source water

The mineral water used in the study was five gallons of packaged drinking water, containing  $7.07 \mu\text{g/L}$  of  $\text{Br}^-$  and  $6.76 \mu\text{g/L}$  of  $\text{BrO}_3^-$ , and was purchased from the market (Guangzhou, China). The ultrapure water, which had no detectable  $\text{Br}^-$  and  $\text{BrO}_3^-$ , was produced using deionization and nanopure purification coupled with a Milli-Q purification system (Milli-Q system, Millipore) and had a resistivity of  $18.2 \text{ M}\Omega \text{ cm}$ . Domestic drinking water, containing  $18\text{--}20 \mu\text{g/L}$   $\text{Br}^-$  and no detectable  $\text{BrO}_3^-$ , was used in the pilot-scale experiments.

### Bench-scale experiments

The bench-scale experiments were performed using a  $4.5 \text{ cm} \times 80 \text{ cm}$  glass column packed with  $400 \text{ g}$  GAC. The GAC used in the experiment was the same as the GAC3 reported by Zhang *et al.* (2011) with  $648.42 \text{ mg/g}$  iodine adsorption value. The length of the GAC bed was  $38 \text{ cm}$ . Both the mineral and ultrapure waters were used as source waters. The source waters were spiked with various levels of  $\text{Br}^-$  or  $\text{BrO}_3^-$  to produce test solutions, which were carefully pumped into and out of the GAC column using YZ1515x peristaltic pumps (Baoding Longer Precision Pump Co., Ltd, Hebei, China). The EBCT was controlled by the water flow rate using the peristaltic pumps. The influent and effluent samples were periodically collected for  $\text{Br}^-$  and  $\text{BrO}_3^-$  detection and were taken in parallel every time.

### Pilot-scale experiments

The pilot-scale experiments were carried out using domestic drinking water as the source water and were performed in a set of  $2 \text{ T/h}$  equipment for mineral water treatment. The equipment was as follows: three storage tanks for the source water, three sets of ultra-violet irradiation apparatus, a tank of sand for filtration, two tanks of GAC, a set of instruments for fine filtration comprising sequential  $10$ ,  $5$ ,  $1$  and  $0.22 \mu\text{m}$  filters, a set of ozone generation instruments, two sets of ozone contact instruments, apparatus for the addition of  $\text{CO}_2$  and a storage tank for the treated water.

The pilot-scale experiments for determining GAC capacity for  $\text{Br}^-$  reduction were initiated after the addition of various concentrations of  $\text{KBr}^-$  into the source water until the mixture was fully stirred, then the solutions were treated by using the sand filter, GAC, fine filter and ozonation. The samples were periodically collected and examined before and after GAC treatment. Two samples were taken in parallel every time. The GAC used in the experiment was the same as the GAC11 reported by Zhang *et al.* (2011) with  $953.27 \text{ mg/g}$  iodine adsorption value.

The pilot-scale experiments for detecting GAC capacity for  $\text{BrO}_3^-$  reduction was initiated after the addition of  $60 \mu\text{g/L}$  of  $\text{KBrO}_3^-$  into the water source until the mixture was fully stirred, then the solution was treated as in the  $\text{Br}^-$  reduction experiments. The  $\text{BrO}_3^-$  concentration in the influent sample of the GAC system was controlled using the ozone dose during ozonation. The samples were periodically collected and examined before and after GAC treatment. Two samples were taken in parallel every time. The GAC used in the experiment was the same as the GAC2 reported by Zhang *et al.* (2011) with  $832.96 \text{ mg/g}$  iodine adsorption value.

### Analytical method

$\text{Br}^-$  and  $\text{BrO}_3^-$  levels were measured using a Dionex ion chromatograph (ICS1500) equipped with AS19 analytical/AG19 guard columns, an ASRS300 anion suppressor and an AS40 automatic sampler. A value of  $0.50 \mu\text{g/L}$  was used to facilitate statistical analysis if the detected result was lower than the detection limit of  $1 \mu\text{g/L}$ . The reduction

rates of  $\text{Br}^-$  or  $\text{BrO}_3^-$  were calculated as:  $([\text{influent Br}^- \text{ or BrO}_3^-] - [\text{effluent Br}^- \text{ or BrO}_3^-]) / ([\text{influent Br}^- \text{ or BrO}_3^-]) \times 100\%$ .

## RESULTS AND DISCUSSION

### Bench-scale experiments

#### Removal of $\text{Br}^-$ from the mineral water

In mineral water with an initial  $\text{Br}^-$  content of 50.00 and 105.59  $\mu\text{g/L}$ , the  $\text{Br}^-$  contents of the treated water were respectively 36.84 and 66.14  $\mu\text{g/L}$  after 2 min, and 24.09 and 43.14  $\mu\text{g/L}$  after 5 min. When the initial  $\text{Br}^-$  was 192.57, 264.47 and 596.13  $\mu\text{g/L}$ , the  $\text{Br}^-$  contents of the treated water were 59.56, 114.59 and 149.39  $\mu\text{g/L}$  after 5 min. The  $\text{Br}^-$  reduction rates were between 26.3 and 41.9% after 2 min and between 51.8 and 74.9% after 5 min (Figure 1).

Other studies suggested that when the  $\text{Br}^-$  levels in water are greater than 50  $\mu\text{g/L}$ , the  $\text{BrO}_3^-$  produced under certain treatment conditions may exceed the drinking water standard of 10  $\mu\text{g/L}$  (Amy et al. 2000; Von Gunten & Pinkernell 2000). Thus, this method cannot remove the excessive  $\text{BrO}_3^-$  when the initial level of  $\text{Br}^-$  in the source water is greater than 190  $\mu\text{g/L}$ . Other methods should be

considered to control  $\text{BrO}_3^-$  produced during the ozonation of drinking water with  $\text{Br}^-$ .

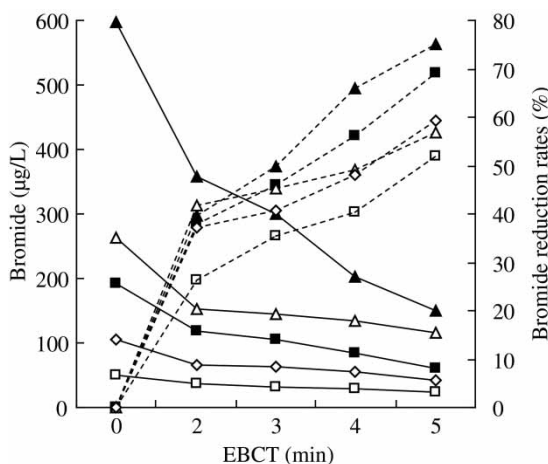
#### Removal of $\text{Br}^-$ from the ultrapure water

In ultrapure water with an initial  $\text{Br}^-$  content of 30.62 and 91.27  $\mu\text{g/L}$ , the contents of the treated water were respectively 11.71 and 24.28  $\mu\text{g/L}$  after 2 min, and 11.56 and 16.35  $\mu\text{g/L}$  after 5 min. When the initial  $\text{Br}^-$  was 192.60, 395.98 and 599.39  $\mu\text{g/L}$ , the  $\text{Br}^-$  contents of the treated water was respectively 18.23, 34.95 and 72.19  $\mu\text{g/L}$  after 5 min. The  $\text{Br}^-$  reduction rates were between 61.74 and 86.58% after 2 min and between 62.26 and 91.17% after 5 min (Figure 2).

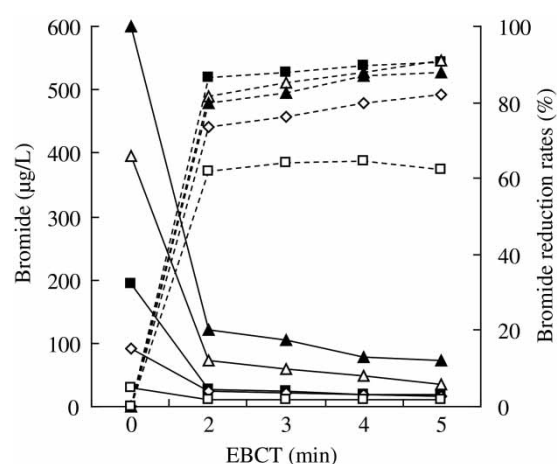
The GAC capacity for  $\text{Br}^-$  reduction was better in the ultrapure water than in the mineral water, which may be attributable to the more complex properties of the latter compared with the former. Meanwhile, GAC efficiency for  $\text{Br}^-$  removal increased when EBCT was increased for both mineral and ultrapure waters. This may provide greater chances for GAC to absorb  $\text{Br}^-$ .

#### Removal of $\text{BrO}_3^-$ from the mineral water

In the mineral water with an initial  $\text{BrO}_3^-$  content of 18.12, 31.48, 57.40, 81.10 and 110.01  $\mu\text{g/L}$ , the  $\text{BrO}_3^-$  levels in the treated water were respectively 4.36, 7.81, 14.29, 20.39 and 10.47  $\mu\text{g/L}$  after 2 min, and 0.71, 3.33, 3.51, 12.69 and



**Figure 1** | Effect of GAC on  $\text{Br}^-$  in mineral water. Unbroken lines:  $\text{Br}^-$  ( $\mu\text{g/L}$ ); dotted lines:  $\text{Br}^-$  reduction rates (%). Initial  $\text{Br}^-$  in water: ( $\square$ ) 50.00  $\mu\text{g/L}$ ; ( $\diamond$ ) 105.59  $\mu\text{g/L}$ ; ( $\blacksquare$ ) 192.57  $\mu\text{g/L}$ ; ( $\triangle$ ) 264.47  $\mu\text{g/L}$ ; and ( $\blacktriangle$ ) 596.13  $\mu\text{g/L}$ .



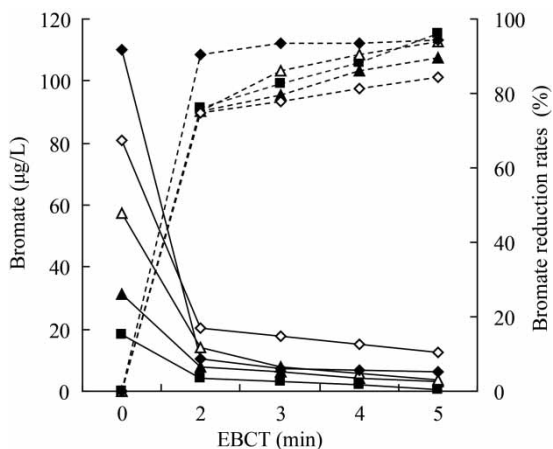
**Figure 2** | Effect of GAC on  $\text{Br}^-$  in ultrapure water. Unbroken lines:  $\text{Br}^-$  ( $\mu\text{g/L}$ ); dotted lines:  $\text{Br}^-$  reduction rates (%). Initial  $\text{Br}^-$  in water: ( $\square$ ) 30.62  $\mu\text{g/L}$ ; ( $\diamond$ ) 91.27  $\mu\text{g/L}$ ; ( $\blacksquare$ ) 192.60  $\mu\text{g/L}$ ; ( $\triangle$ ) 395.98  $\mu\text{g/L}$ ; and ( $\blacktriangle$ ) 599.39  $\mu\text{g/L}$ .

6.13  $\mu\text{g/L}$  after 5 min. The removal rates were greater than 75% when EBCT was longer than 2 min, and the maximum reduction rate was 94.4% (Figure 3). With increasing EBCT, the  $\text{BrO}_3^-$  levels of the treated water decreased, and the removal rates increased. These results were consistent with the report of Huang *et al.* (2004a, b) stating that  $\text{BrO}_3^-$  removal improves when EBCT is increased.

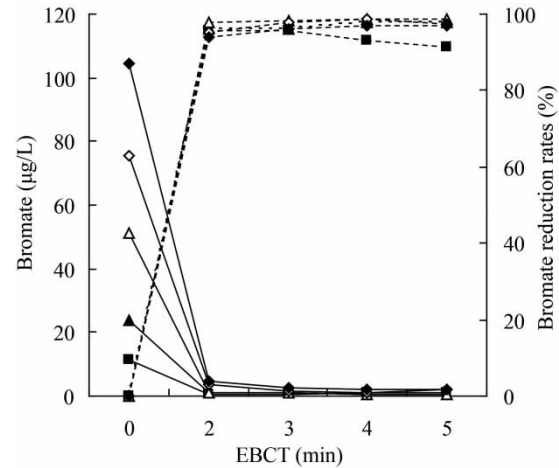
### Removal of $\text{BrO}_3^-$ from the ultrapure water

In the ultrapure water, the initial  $\text{BrO}_3^-$  contents were respectively 11.23, 24.05, 51.05, 75.63 and 104.65  $\mu\text{g/L}$ . When EBCT was 2 min, the  $\text{BrO}_3^-$  levels in the treated water were efficiently removed, and the desired criterion of less than 10  $\mu\text{g/L}$  was achieved. The reduction rates were between 87.8 and 98.8% after 5 min (Figure 4). When EBCT was increased, the  $\text{BrO}_3^-$  levels of the treated water decreased, and the removal rates increased.

These results suggest that the GAC capacity for  $\text{BrO}_3^-$  and  $\text{Br}^-$  removal was better in ultrapure water than in mineral water. This result may be attributed to the competitive adsorption between  $\text{Br}^-/\text{BrO}_3^-$  and natural organic matter or other anions, such as  $\text{Cl}^-$ ,  $\text{Br}^-$  and  $\text{SO}_4^{2-}$ . Competitive adsorption has been reported to reduce the capability of GAC for absorbing  $\text{BrO}_3^-$  (Mills *et al.* 1996; Bao *et al.* 1999; Huang *et al.* 2004a, b; An *et al.* 2008).



**Figure 3** | Effect of GAC on  $\text{BrO}_3^-$  in mineral waters. Unbroken lines:  $\text{BrO}_3^-$  ( $\mu\text{g/L}$ ); dotted lines:  $\text{BrO}_3^-$  reduction rates (%). Initial  $\text{BrO}_3^-$  in water: (■) 18.12  $\mu\text{g/L}$ ; (▲) 31.48  $\mu\text{g/L}$ ; (△) 57.40  $\mu\text{g/L}$ ; (◇) 81.10  $\mu\text{g/L}$ ; and (◆) 110.01  $\mu\text{g/L}$ .



**Figure 4** | Effect of GAC on  $\text{BrO}_3^-$  in ultrapure water. Unbroken lines:  $\text{BrO}_3^-$  ( $\mu\text{g/L}$ ); dotted lines:  $\text{BrO}_3^-$  reduction rates (%). Initial  $\text{BrO}_3^-$  in water: (■) 11.23  $\mu\text{g/L}$ ; (▲) 24.05  $\mu\text{g/L}$ ; (△) 51.05  $\mu\text{g/L}$ ; (◇) 75.63  $\mu\text{g/L}$ ; and (◆) 104.65  $\mu\text{g/L}$ .

### Pilot-scale experiments

#### Removal of $\text{Br}^-$

Domestic drinking water with different initial  $\text{BrO}_3^-$  content was used as the source water. The results show that the  $\text{Br}^-$  concentrations in the treated water decreased (Table 1). Therefore, GAC may be used for the removal of  $\text{Br}^-$  from source waters.

#### Removal of $\text{BrO}_3^-$

The source water was used to make a solution containing 60  $\mu\text{g/L}$  of  $\text{BrO}_3^-$ . Table 2 shows that when the ozone concentration is 0.73 mg/L, the concentration of produced  $\text{BrO}_3^-$  increased steeply and exceeded 10  $\mu\text{g/L}$  standard. Nevertheless, the excessive amount of  $\text{BrO}_3^-$  decreased to less than 10  $\mu\text{g/L}$  after GAC treatment. Therefore, GAC may be used to eliminate the  $\text{BrO}_3^-$  generated during the ozonation of drinking water.

**Table 1** | Effects of GAC on  $\text{Br}^-$  in water

Influent content ( $\mu\text{g/L}$ )	Effluent content ( $\mu\text{g/L}$ )	Reduction rates (%)
65.87	37.84	42.56
80.51	47.37	41.17
97.10	59.67	38.54

**Table 2** | Effect of GAC on BrO<sub>3</sub><sup>-</sup> in water

Ozone doses (mg/L)	Influent content (µg/L)	Effluent content (µg/L)	Reduction rates (%)
0.11	0.50	0.50	0.00
0.30	2.77	0.50	81.95
0.43	6.27	1.43	77.25
0.55	5.84	1.57	73.15
0.73	18.80	1.41	92.48

For both Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> the reduction rates were lower in the pilot-scale than in the bench-scale experiments. The variations in the results may be attributable to the different source waters and GAC used in the experiments.

In the present study, GAC appeared to be capable of reducing both Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> concentrations in drinking water, which may be attributable to the similar inorganic characteristics of Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup>. Nevertheless, the removal mechanism of BrO<sub>3</sub><sup>-</sup> using GAC should be different from that of Br<sup>-</sup>. The process of BrO<sub>3</sub><sup>-</sup> removal by GAC was reported to initially be an adsorption, followed by a two-step reduction reaction (Siddiqui *et al.* 1996). A limited amount of data is available on investigations of the GAC mechanism for Br<sup>-</sup> elimination. Therefore, studying the GAC mechanisms for Br<sup>-</sup> removal is necessary.

Both the physical and chemical effects simultaneously act on the adsorption–reduction process during the GAC treatment for BrO<sub>3</sub><sup>-</sup> reduction (Huang & Cheng 2008). Source water quality and EBCT also affect GAC capacity for BrO<sub>3</sub><sup>-</sup> removal (Bao *et al.* 1999). Therefore, ascertaining the optimum EBCT and investigating how both GAC and water properties affect the removal of Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> are important.

In addition, other strategies for the reduction of the BrO<sub>3</sub><sup>-</sup> generated during the ozonation of the water with Br<sup>-</sup>, such as ultra-violet irradiation (Meunier *et al.* 2006), ion exchange membrane bioreactor (Matos *et al.* 2005) and BrO<sub>3</sub><sup>-</sup> reduction using zerovalent iron (Fan *et al.* 2006; Xie & Shang 2006b) and TiO<sub>2</sub> (Noguchi *et al.* 2002), are also available. However, a number of problems, such as high cost and the quality of the treated water, limit the use of these methods, especially for packaged drinking water.

Based on the current experiments, GAC was effective in removing Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> contents from drinking water. GAC technology may be used to reduce BrO<sub>3</sub><sup>-</sup> formation and to

eliminate BrO<sub>3</sub><sup>-</sup> generation during the ozonation of water with Br<sup>-</sup>. However, further investigations on the actual conditions and influence factors of using GAC technology should be conducted because of the limited data available.

## CONCLUSIONS

In the present study, Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> levels in the solutions of water sources were efficiently eliminated using GAC. In the bench-scale experiments, the removal contents and rates of Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> increased with increasing EBCT, and the GAC capacity to reduce Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> contents was better in ultrapure water than in mineral water. Although the effects of GAC on Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> were weaker in the pilot-scale experiments than in the bench-scale experiments, GAC still showed the capability of eliminating Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> contents in the pilot-scale experiments. The minimum reduction rates of Br<sup>-</sup> and BrO<sub>3</sub><sup>-</sup> were 38.5% and 73.2%, respectively. Therefore, GAC can be employed in water treatment facilities to minimize BrO<sub>3</sub><sup>-</sup> formation by decreasing Br<sup>-</sup> contents and to eliminate the BrO<sub>3</sub><sup>-</sup> generated in ozonated water.

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