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Addition of hydrogen peroxide for the simultaneous control of bromate and odor during advanced drinking water treatment using ozone

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ABSTRACT

Complete removal of the characteristic septic/swampy odor from Huangpu River source water could only be achieved under an ozone dose as high as 4.0 mg/L in an ozone-biological activated carbon (O_3-BAC) process, which would lead to the production of high concentrations of carcinogenic bromate due to the high bromide content. This study investigated the possibility of simultaneous control of bromate and the septic/swampy odor by adding H_2O_2 prior to the O_3-BAC process for the treatment of Huangpu River water. H_2O_2 addition could reduce the bromate concentration effectively at an H_2O_2/O_3 (g/g) ratio of 0.5 or higher. At the same time, the septic/swampy odor removal was enhanced by the addition of H_2O_2, although optimization of the H_2O_2/O_3 ratio was required for each ozone dose. At an ozone dose of 2.0 mg/L, the odor was removed completely at an H_2O_2/O_3 ratio of 0.5. The results indicated that H_2O_2 application at a suitable dose could enhance the removal of the septic/swampy odor while suppressing the formation of bromate during ozonation of Huangpu River source water.

Introduction

Ozonation integrated with biological activated carbon (O_3-BAC) has been widely used for drinking water treatment because of its effectiveness in removing disinfection byproduct precursors (Chu et al., 2012; Ratasuk et al., 2008), odorants such as 2-methylisoborneol (2-MIB), geosmin and β-ionone (Peter and von Gunten, 2007), and many other odor causing compounds. The discovery of the formation of potentially carcinogenic bromate (BrO_3^-) during ozonation of bromide-containing source water (von Gunten and Hoigné, 1994), however, has greatly compromised the merits of ozone. Many efforts have thus been devoted to the control of bromate formation during ozonation (von Gunten and Oliveras, 1998).

Addition of ammonia or hydrogen peroxide (H_2O_2) has been proposed as a practical approach in controlling the formation of bromate (von Gunten and Hoigné, 1994; Mizuno et al., 2011). Ammonia could react with $HOBr/OBr^-$, an important ozonation intermediate for bromate production (von Gunten and Oliveras, 1998), to form bromamines (NH_2Br, $NHBr_2$ and NBr_3) (Hofmann and Andrews, 2001). Although quite effective in suppressing bromate formation, this approach may compromise the oxidation power of ozone due to the consumption of ozone by bromamines, resulting in the release of Br^- and NO_3^- (Hoigné and Bader, 1978; Haag et al., 1984). On the other hand, H_2O_2 addition at certain H_2O_2/O_3 ratios has been proved to be effective in reducing bromate formation by decomposing molecular ozone and consuming $HOBr/OBr^-$ (Mizuno et al., 2011; Croue et al., 1996). The combination of O_3 and H_2O_2, at the same time, has been known as an advanced oxidation process, which was widely used for micropollutant removal (von Gunten and

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Oliveras, 1998). Therefore, compared with the ammonia addition approach, the addition of H_2O_2 may have the potential merit of enhancing the oxidation of pollutants in source water through the generation of hydroxyl radicals.

Odor control has long been an important issue for drinking water treatment. Ozonation has been proved to be effective for some odor problems (Peter and von Gunten, 2007). In a previous study, however, ozonation could only reduce the earthy-musty odor intensity from 10 to 6 according to the flavor profile analysis method (Bruchet et al., 2004). H_2O_2 addition was found to be able to enhance the reduction of odor intensity by ozonation (Glaze et al., 1990; Bruchet et al., 2004). Thus there is the possibility to control the formation of bromate and enhance the removal of odorants at the same time by adding H_2O_2 in an ozonation process.

The Huangpu River, which is known for its septic/swampy odor associated with seasonal earthy/musty odor (Yu et al., 2009), is an important drinking water source for Shanghai. The removal of odor has long been an important issue for the treatment of Huangpu River source water. However, with a high bromide concentration (204.9–394. 6 μg/L) (Huang et al., 2010), bromate control must be considered if ozone is adopted for odor removal. In this study, the possibility of simultaneous control of bromate and odor by adding H_2O_2 prior to the O_3-BAC process was evaluated in a pilot study, and a suitable H_2O_2/O_3 (g/g) ratio was proposed. The results of this study will be helpful for better application of ozone for drinking water treatment.

1 Materials and methods

1.1 Setup for the pilot study

Figure 1 shows the schematic diagram of the O_3-BAC system (120 L/hr) used for this study. Huangpu River water was treated in a conventional treatment process including coagulation, sedimentation and sand filtration, and then fed into the O_3-BAC system. Ozone gas was supplied into the ozonation column (height, 1500 mm; inner diameter, 130 mm; effective volume, 18.1 L) from the gas diffuser located at the bottom. The hydraulic retention time (HRT) was 9.1 min for ozonation. H_2O_2 was added into water before ozonation using a pump. Ozonated water flowed into the BAC column (carbon height, 1000 mm; inner diameter, 130 mm) in a down-flow mode with an empty bed contact time of 12.0 min. BAC of approximately 2.5 years age was taken from the BAC tank of a full scale waterworks using surface water as raw water.

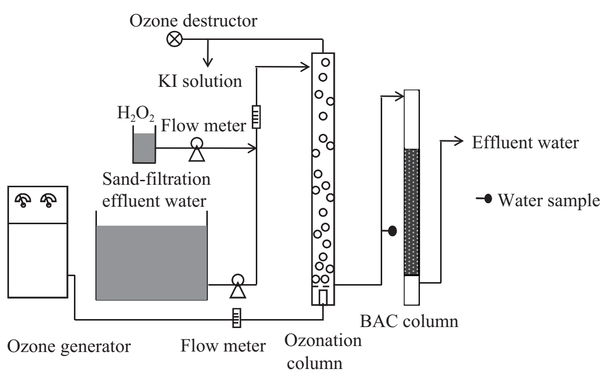

Fig. 1 Schematic diagram of the O_3-BAC system.

The experiment was performed from September to November in 2010 after a two-month pre-operation. The characteristics of raw water and sand filtered water are shown in **Table 1**. All the samples were stored at 4°C and analyzed in one week.

1.2 Analytical methods

All reagents applied in the experiment were of analytical grade, and all stock solutions were prepared with Milli-Q water (Millipore).

The indigo method was employed to measure the aqueous ozone concentrations (Bader and Hoigné, 1981), and gaseous phase ozone was quantified by the iodometric method (APHA, 1995). Ozone consumption was defined as the difference between the influent and effluent gaseous ozone. Hydrogen peroxide was determined by the peroxidase-DPD method (Bader et al., 1988). The DOC was determined using a Shimadzu TOC analyzer (TOC-Vcph). Bromate and bromide concentrations were analyzed by ion chromatography (Dionex 3000) using an AC9-SC analytical column with a detection limit of 2.0 μg/L and 10.0 μg/L, respectively.

The taste & odor intensity of water samples was determined using the flavor profile analysis method, which was described in standard methods (APHA, 1995). In this test, four panelists were trained for the panel. They assigned an intensity rating to each water sample using a

Table 1 Characteristics of Huangpu River raw water and sand filtered water

Parameter	Raw water	Sand filtered water
Temperature (°C)	13–18	14.2–18.5
Turbidity (NTU)	16.4–39.6	0.26–0.96
Ammonia (mg/L)	0.51–0.84	0.17–0.23
DOC (mg/L)	4.98–5.21	3.44–3.71
THMFP (μg/L)	207.7–286.2	103.7–236.1
HAAFP (μg/L)	256.2–291.9	178.31–202.0
2-MIB (ng/L)	6.13–17.08	6.86–20.66
Geosmin (ng/L)	2.68–3.79	1.95–4.06
Bromide (μg/L)	215.2–417.3	215.2–380.5
Septic/swampy odor intensity	8.0–10.0	6.0–9.0

* Septic/swampy odor intensity was analyzed by the flavor profile analysis method. THMFP: trihalomethane formation potential; HAAFP: haloacetic acid formation potential.