

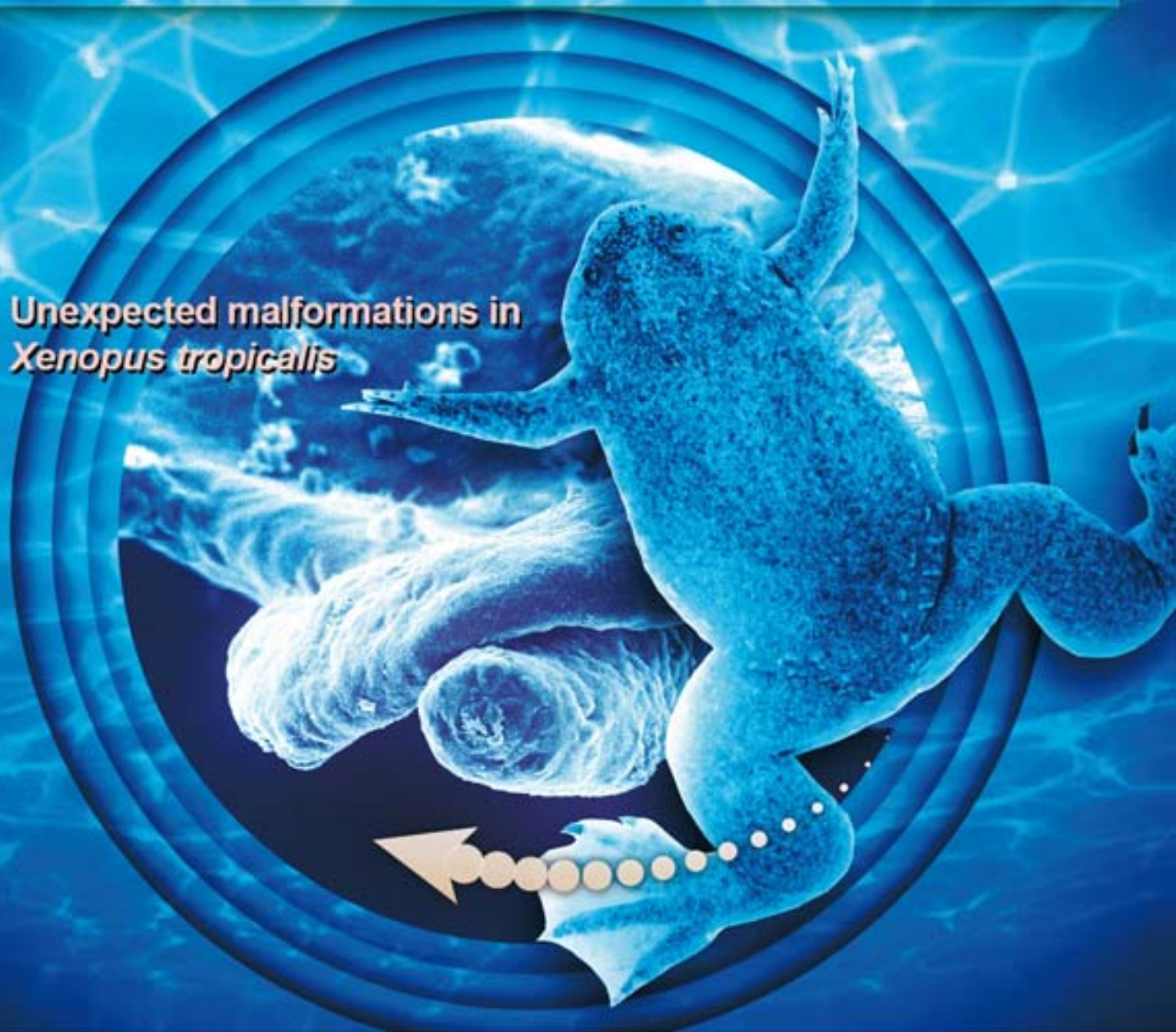
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Unexpected malformations in
Xenopus tropicalis



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CONTENTS

Aquatic environment

Metal composition of layered double hydroxides (LDHs) regulating ClO_4^- adsorption to calcined LDHs via the memory effect and hydrogen bonding Yajie Lin, Qile Fang, Baoliang Chen	493
Limitation of spatial distribution of ammonia-oxidizing microorganisms in the Haihe River, China, by heavy metals Chao Wang, Baoqing Shan, Hong Zhang, Yu Zhao	502
Temperature sensitivity of organic compound destruction in SCWO process Yaqin Tan, Zheming Shen, Weimin Guo, Chuang Ouyang, Jinping Jia, Weili Jiang, Haiyun Zhou	512
Influence of moderate pre-oxidation treatment on the physical, chemical and phosphate adsorption properties of iron-containing activated carbon Zhengfang Wang, Mo Shi, Jihua Li, Zheng Zheng	519
Reduction of DOM fractions and their trihalomethane formation potential in surface river water by in-line coagulation with ceramic membrane filtration Pharkphum Rakruam, Suraphong Wattanachira	529
N_2O emission from nitrogen removal via nitrite in oxic-anoxic granular sludge sequencing batch reactor Hong Liang, Jiaoling Yang, Dawen Gao	537
Influence of stabilizers on the antimicrobial properties of silver nanoparticles introduced into natural water Aleksandra Burkowska-But, Grzegorz Sionkowski, Maciej Walczak	542
Addition of hydrogen peroxide for the simultaneous control of bromate and odor during advanced drinking water treatment using ozone Yongjing Wang, Jianwei Yu, Dong Zhang, Min Yang	550
Nitric oxide removal by wastewater bacteria in a biotrickling filter Hejingying Niu, Dennis Y C Leung, Chifang Wong, Tong Zhang, Mayngor Chan, Fred C C Leung	555
Elucidating the removal mechanism of <i>N,N</i> -dimethyldithiocarbamate in an anaerobic-anoxic-oxic activated sludge system Yongmei Li, Xianzhong Cao, Lin Wang	566
Influencing factors of disinfection byproducts formation during chloramination of Cyclops metabolite solutions Xingbin Sun, Lei Sun, Ying Lu, Jing Zhang, Kejing Wang	575

Atmospheric environment

Sources of nitrous and nitric oxides in paddy soils: Nitrification and denitrification Ting Lan, Yong Han, Marco Roelcke, Rolf Nieder, Zucong Cai	581
Upper Yellow River air concentrations of organochlorine pesticides estimated from tree bark, and their relationship with socioeconomic indices Chang He, Jun Jin, Bailin Xiang, Ying Wang, Zhaohui Ma	593
Mechanism and kinetic properties of NO_3^- -initiated atmospheric degradation of DDT Cai Liu, Shanqing Li, Rui Gao, Juan Dang, Wenxing Wang, Qingzhu Zhang	601
Sorption and phase distribution of ethanol and butanol blended gasoline vapours in the vadose zone after release Ejike Ugwoha, John M. Andresen	608

Terrestrial environment

Effects of temperature change and tree species composition on N_2O and NO emissions in acidic forest soils of subtropical China Yi Cheng, Jing Wang, Shenqiang Wang, Zucong Cai, Lei Wang	617
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Environmental biology

Influence of sunlight on the proliferation of cyanobacterial blooms and its potential applications in Lake Taihu, China Qichao Zhou, Wei Chen, Kun Shan, Lingling Zheng, Lirong Song	626
Bioavailability and tissue distribution of Dechloranes in wild frogs (<i>Rana limnocharis</i>) from an e-waste recycling area in Southeast China Long Li, Wenyue Wang, Quanxia Lv, Yujie Ben, Xinghong Li	636

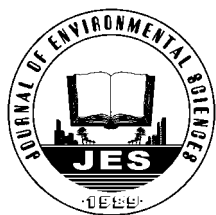
Environmental health and toxicology

Unexpected phenotypes of malformations induced in <i>Xenopus tropicalis</i> embryos by combined exposure to triphenyltin and 9- <i>cis</i> -retinoic acid Jingmin Zhu, Lin Yu, Lijiao Wu, Lingling Hu, Huahong Shi	643
Expression of sulfur uptake assimilation-related genes in response to cadmium, bensulfuron-methyl and their co-contamination in rice roots Jian Zhou, Zegang Wang, Zhiwei Huang, Chao Lu, Zhuo Han, Jianfeng Zhang, Huimin Jiang, Cailin Ge, Juncheng Yang	650

Environmental catalysis and materials

Reaction mechanism and metal ion transformation in photocatalytic ozonation of phenol and oxalic acid with Ag ⁺ /TiO ₂ Yingying Chen, Yongbing Xie, Jun Yang, Hongbin Cao, Yi Zhang	662
Effect of TiO ₂ calcination temperature on the photocatalytic oxidation of gaseous NH ₃ Hongmin Wu, Jinzhu Ma, Changbin Zhang, Hong He	673
Effects of synthesis methods on the performance of Pt + Rh/Ce _{0.6} Zr _{0.4} O ₂ three-way catalysts Zongcheng Zhan, Liyun Song, Xiaojun Liu, Jiao Jiao, Jinzhou Li, Hong He	683
Catalytic combustion of soot over ceria-zinc mixed oxides catalysts supported onto cordierite Leandro Fontanetti Nascimento, Renata Figueredo Martins, Rodrigo Ferreira Silva, Osvaldo Antonio Serra	694
Effects of metal and acidic sites on the reaction by-products of butyl acetate oxidation over palladium-based catalysts Lin Yue, Chi He, Zhengping Hao, Shunbing Wang, Hailin Wang	702
Mechanism of enhanced removal of quinonic intermediates during electrochemical oxidation of Orange II under ultraviolet irradiation Fazhan Li, Guoting Li, Xiwang Zhang	708

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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 (O₃-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 H₂O₂ prior to the O₃-BAC process for the treatment of Huangpu River water. H₂O₂ addition could reduce the bromate concentration effectively at an H₂O₂/O₃ (g/g) ratio of 0.5 or higher. At the same time, the septic/swampy odor removal was enhanced by the addition of H₂O₂, although optimization of the H₂O₂/O₃ ratio was required for each ozone dose. At an ozone dose of 2.0 mg/L, the odor was removed completely at an H₂O₂/O₃ ratio of 0.5. The results indicated that H₂O₂ 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 (O₃-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 (BrO₃⁻) 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 (H₂O₂)

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 (NH₂Br, NHBr₂ and NBr₃) (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 NO₃⁻ (Hoigné and Bader, 1978; Haag et al., 1984). On the other hand, H₂O₂ addition at certain H₂O₂/O₃ 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 O₃ and H₂O₂, 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 $\mu\text{g/L}$) (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

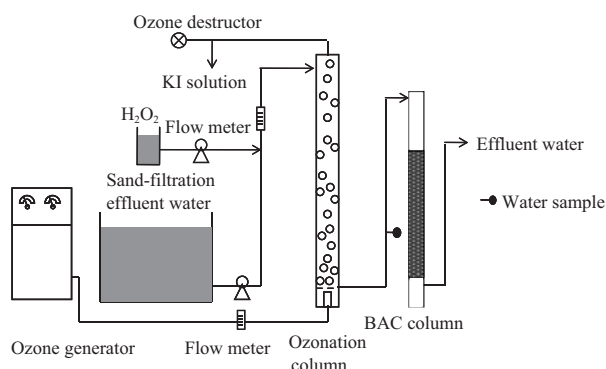


Fig. 1 Schematic diagram of the O_3 -BAC system.

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.

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 $\mu\text{g/L}$ and 10.0 $\mu\text{g/L}$, 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 ($^{\circ}\text{C}$)	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 ($\mu\text{g/L}$)	207.7–286.2	103.7–236.1
HAAFP ($\mu\text{g/L}$)	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 ($\mu\text{g/L}$)	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.

7-point category scale: 0 = not detectable odor, 2 = very weak, 4 = weak, 6 = weak-to-moderate, 8 = moderate, 10 = moderate-to-strong, 12 = strong. The descriptor and intensity for each sample was then evaluated and recorded. 2-MIB and geosmin concentrations of water samples were analyzed based on the processes of a previous study (Yu et al., 2009).

2 Results and discussion

2.1 Odor removal and bromate formation during O₃-BAC treatment

Offensive odor of the septic/swampy type was found to be the characteristic odor in Huangpu River water (Yu et al., 2009), with an intensity of 8.0–10.0 during the study, which was decreased slightly to 6.0–9.0 after conventional treatment (Table 1). As shown in Fig. 2, the septic/swampy odor was removed from an intensity of 6.8 to 2.6 with an ozone dose of 3.8–4.9 mg/L, showing that this odor was very difficult to remove. The subsequent BAC treatment could remove the odor effectively from an intensity of 6.8 to 2.4 even without ozonation. It is not clear that what were the major compounds responsible for the septic/swampy odor, although some sulfur-containing compounds such as dimethyl trisulfide were suspected to contribute to this odor (Yu et al., 2009). It has been found that dimethyl trisulfide could be removed effectively by a biologically active sand filter (Lundgren et al., 1988). At the same time, adsorption may also be important for the odor removal (Hrudey et al., 1995). However, even with the combination of O₃ and BAC, complete odor removal was only achieved at an ozone dose of 4 mg/L.

2-MIB was detected with a concentration of 6.86–20.66 ng/L in sand-filtered water. As shown in Fig. S1, the 2-MIB

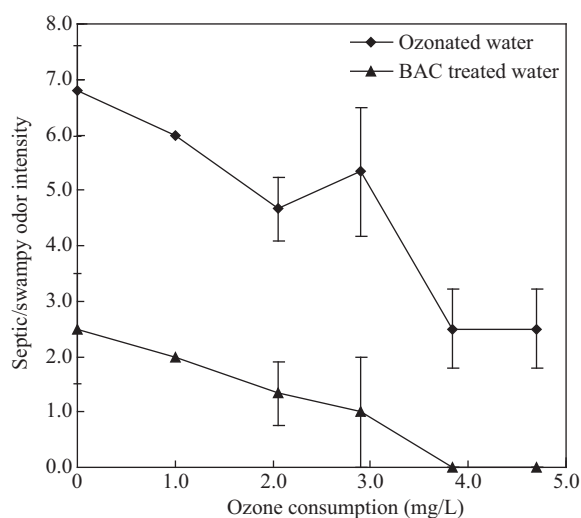


Fig. 2 Change of septic/swampy odor intensity as a function of ozone dose.

concentration in both ozonated and BAC-treated water decreased with the increase of ozone dose, and was below detection limit in the BAC-treated water at an ozone dose of 2.0 mg/L.

Figure 3 shows the formation of bromate as a function of ozone dose. With bromide concentration of 215.2–380.5 µg/L in the sand-filtered water, the bromate concentration increased quickly with increasing ozone dose. The bromate concentration corresponding to a frequently applied ozone dose of 3.0 mg/L was 17.2 µg/L, much higher than the standard level of 10 µg/L. Even at an ozone dose of 2.0 mg/L, the bromate concentration (11.4 µg/L) could exceed the standard level, and it could not be removed during the BAC treatment processes. However, an ozone dose of 2.0 or 3.0 mg/L could not remove the septic/swampy odor effectively by ozonation alone, as exhibited in Fig. 2. Weak septic/swampy odor could be detected even after the BAC treatment. Thus further efforts are required to remove the septic/swampy odor while controlling the bromate formation simultaneously.

2.2 Effects of H₂O₂ addition on odor removal and bromate control

Figure 4 displays the bromate concentration as a function of H₂O₂/O₃ ratio at two ozone doses (2.0 and 3.0 mg/L). With an H₂O₂/O₃ (g/g) ratio of 0.5, the bromate concentration decreased significantly from 11.5 µg/L and 17.2 µg/L to 1.9 µg/L and 1.8 µg/L, respectively, at the two ozone doses, indicating that H₂O₂ addition was very effective in suppressing bromate formation (Daniel et al., 1995). This H₂O₂/O₃ ratio was lower than that suggested by Mizuno et al. (2011), which may result from the relatively high residual H₂O₂ in the reaction system. After ozonation, more than 30% of H₂O₂ remained in the treated water (inset Fig. 4) and H₂O₂ could react with HOBr⁻/OBr⁻, which is an important intermediate in forming bromate, with

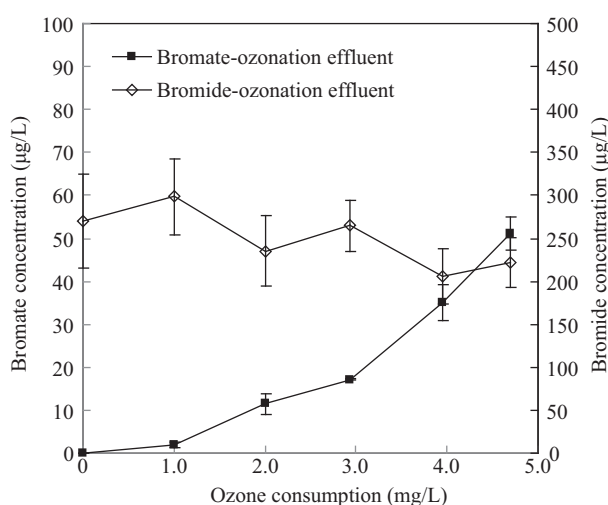


Fig. 3 Bromate formation as a function of ozone dose.

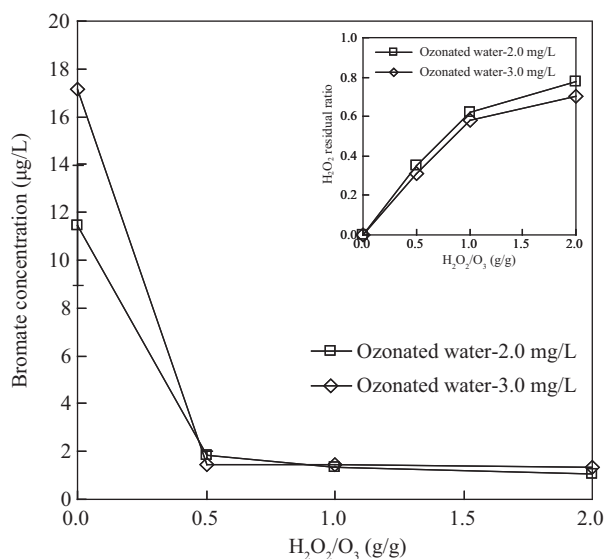


Fig. 4 Effect of H₂O₂ addition on bromate control. The inset is the H₂O₂ residual ratio H₂O₂ concentration/initial H₂O₂ concentration at different H₂O₂/O₃ (g/g) ratios.

reaction rate constant of 7.6×10^8 mol/(L·sec) (von Gunten and Oliveras, 1997), inducing the lower concentration of bromate. The optimal H₂O₂/O₃ ratio for bromate control may be different for different source water because many factors such as bromide, ammonia, background natural organic materials, and temperature could affect the ozone decomposition and hydroxyl radical concentration in water (von Gunten and Oliveras, 1998; von Gunten and Hoigné, 1994).

Figure 5 displays the decrease of the septic/swampy odor intensity as a function of H₂O₂/O₃ ratio. It was clearly demonstrated that the addition of H₂O₂ could enhance the removal of the septic/swampy odor. However, there existed an optimum H₂O₂/O₃ ratio for each ozone dose. At an

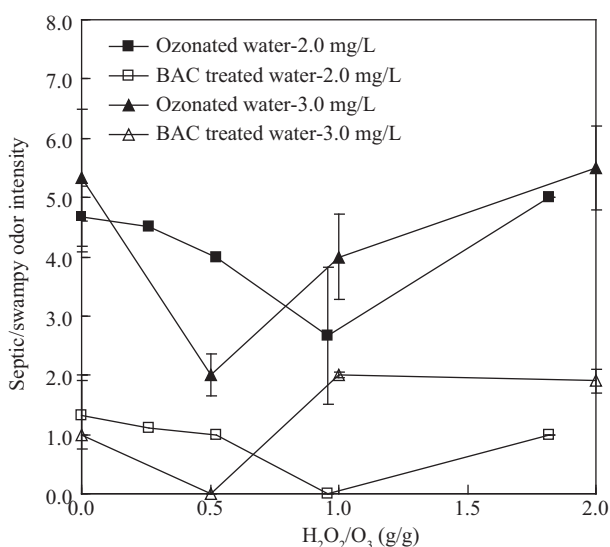


Fig. 5 Effect of H₂O₂ addition on septic/swampy odor removal.

ozone dose of 2.0 and 3.0 mg/L, the best odor removal efficacy was obtained at an H₂O₂/O₃ ratio of 0.5 and 1.0, respectively. The enhancement in odor removal decreased at higher H₂O₂/O₃ ratio, which might be the consequence of reactions between excessive amounts of H₂O₂ (inset **Fig. 4**) and OH radicals (von Gunten and Oliveras, 1998; Kleiser and Frimmel, 2000).

Figure S2 displays the 2-MIB residual ratio (2-MIB/2-MIB₀) in the H₂O₂-O₃ treated water at the ozone doses of 2.0 and 3.0 mg/L. The residual ratio decreased as the H₂O₂ doses increased from 0 to 1.0, indicating that the removal of 2-MIB by ozonation was enhanced as H₂O₂ was applied. However, MIB could not be detected in the BAC treated water at the different H₂O₂ doses.

As discussed above, the addition of H₂O₂ could not only suppress the formation of bromate, but also enhance the removal of the septic/swampy odor. However, a suitable H₂O₂ dose should be determined to achieve the best performance.

3 Conclusions

This study investigated the possibility of simultaneous control of bromate and odor by adding H₂O₂ into an ozonation process for the treatment of Huangpu River water. The following conclusions were obtained: (1) Septic/swampy odor as the major odor problem for Huangpu River water could be completely removed by the combination of O₃ and BAC at an ozone dose as high as 4 mg/L, which would produce high concentrations of bromate. (2) By adding H₂O₂ with an H₂O₂/O₃ ratio of 0.5, it was possible to remove the septic/swampy odor and suppress the formation of bromate by the combination of O₃ and BAC at an ozone dose of 2.0 mg/L. The suitable H₂O₂ dose should be determined according to raw water quality and the ozone dose.

Acknowledgment

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Supporting materials

Supplementary data associated with this article can be found in the online version.

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Supporting materials

This supplementary information provides the effectiveness of O₃-BAC treatment and H₂O₂ addition on the odors removal.

Parameters of the conventional treatment processes

The pilot-scale conventional drinking water treatment processes consisted of coagulation, sedimentation and sand filtration. Coagulation was performed in a four-chamber mechanical flocculation system under the following conditions: 6.0 mg/L of polyaluminium chloride (Al₂O₃ content, 11%); fast mixing (200 r/min) for 1min; slow mixing for 15 min (150 r/min, 100 r/min and 50 r/min for 5 min respectively). The sedimentation time was 48 min. Sand filtration was conducted in a stainless-steel column (inner diameter, 300 mm; sand depth, 900 mm; sand diameter, 0.95-1.35 mm) at a filtration velocity of 8.0 m/hr.

Removal of 2-MIB by O₃-BAC treatment

Although 2-MIB was detected with concentrations of 6.86--20.66 ng/L in the sand-filtered water, earthy/musty odor was very weak to be detected because of the high septic/swampy intensity. **Figure S1** shows the results of 2-MIB removal by O₃-BAC process. Ozonation alone could remove 2-MIB effectively for the oxidation of OH radicals produced by ozone decomposition (Westerhoff et al., 2006), with the destruction percent of about 60% at the 1.0 mg/L ozone. The oxidation efficacy of 2-MIB increased with greater ozone doses, which was consistent with other research (Westerhoff et al., 2006). At the ozone dose of 2.0 mg/L, no 2-MIB could be detected in the BAC treated water.

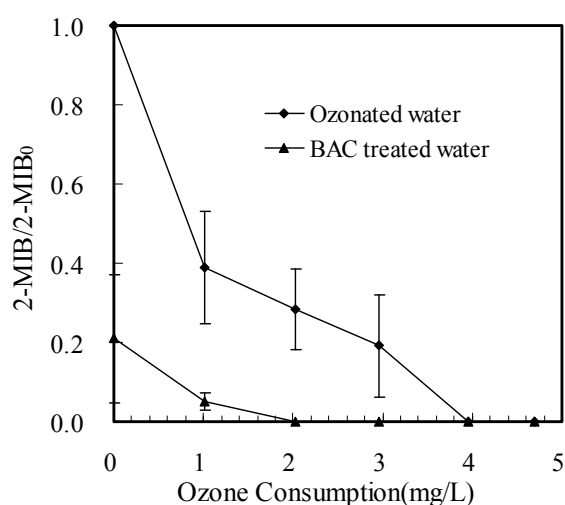


Fig. S1 2-MIB degradation in the O₃-BAC treated water as a function of ozone doses.

Effect of H₂O₂ addition on the 2-MIB removal

Figure S2 displays the 2-MIB remaining ratio (2-MIB/2-MIB₀) in the ozonated water at the ozone doses of 2.0 and 3.0 mg/L. The remaining ratio decreased as the H₂O₂ doses increased from 0 to 1.0, and then increased. However, MIB could not be detected in the BAC treated water at different H₂O₂ doses.

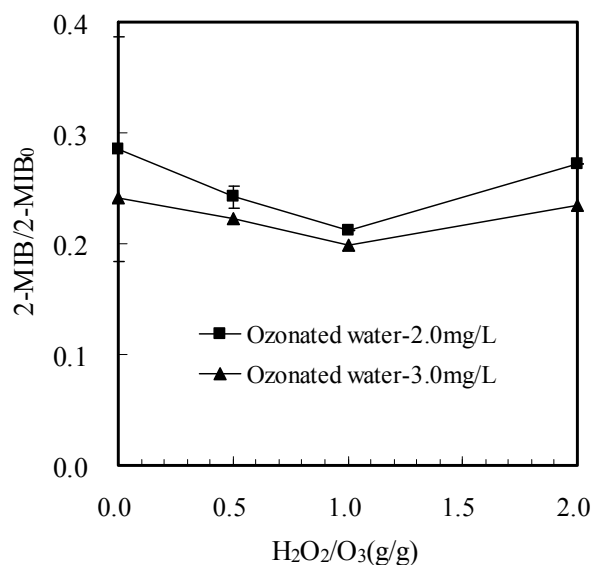


Fig. S2 2-MIB degradation in the ozonated water as a function of H₂O₂/O₃ (g/g) ratio.

Bromate concentration in ozonated water and BAC treated water

During the period of experiment, different ozone doses (0-4.6mg/L) and H₂O₂/O₃ (g/g) ratios were applied for the sand filtrated water treatment. Thus bromate concentration in ozonated water and BAC treated water varied as different doses of oxidant used. **Figure S3** displays the bromate concentration in both waters during the period. It shows that BAC treatment could not remove bromate effectively.

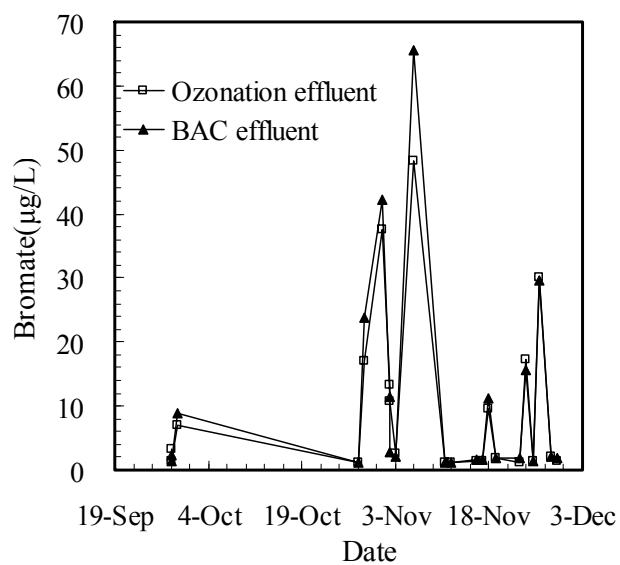


Fig. S3 Bromate concentration in the ozonated water and BAC treated water.

References

Westerhoff, P., Nalinakumari, B., Pei P, 2006. Kinetics of MIB and geosmin oxidation during ozonation. *Ozone: Science & Engineering* 28(5): 277-286.

Supporting materials

This supplementary information provides the effectiveness of O₃-BAC treatment and H₂O₂ addition on the odors removal.

Parameters of the conventional treatment processes

The pilot-scale conventional drinking water treatment processes consisted of coagulation, sedimentation and sand filtration. Coagulation was performed in a four-chamber mechanical flocculation system under the following conditions: 6.0 mg/L of polyaluminium chloride (Al₂O₃ content, 11%); fast mixing (200 r/min) for 1min; slow mixing for 15 min (150 r/min, 100 r/min and 50 r/min for 5 min respectively). The sedimentation time was 48 min. Sand filtration was conducted in a stainless-steel column (inner diameter, 300 mm; sand depth, 900 mm; sand diameter, 0.95-1.35 mm) at a filtration velocity of 8.0 m/hr.

Removal of 2-MIB by O₃-BAC treatment

Although 2-MIB was detected with concentrations of 6.86--20.66 ng/L in the sand-filtered water, earthy/musty odor was very weak to be detected because of the high septic/swampy intensity. **Figure S1** shows the results of 2-MIB removal by O₃-BAC process. Ozonation alone could remove 2-MIB effectively for the oxidation of OH radicals produced by ozone decomposition (Westerhoff et al., 2006), with the destruction percent of about 60% at the 1.0 mg/L ozone. The oxidation efficacy of 2-MIB increased with greater ozone doses, which was consistent with other research (Westerhoff et al., 2006). At the ozone dose of 2.0 mg/L, no 2-MIB could be detected in the BAC treated water.

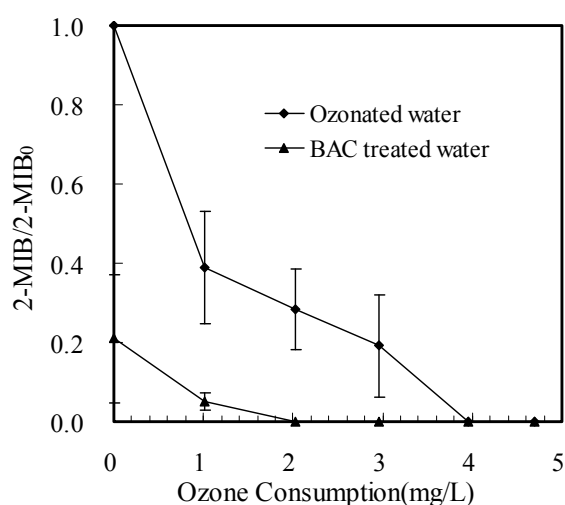


Fig. S1 2-MIB degradation in the O₃-BAC treated water as a function of ozone doses.

Effect of H₂O₂ addition on the 2-MIB removal

Figure S2 displays the 2-MIB remaining ratio (2-MIB/2-MIB₀) in the ozonated water at the ozone doses of 2.0 and 3.0 mg/L. The remaining ratio decreased as the H₂O₂ doses increased from 0 to 1.0, and then increased. However, MIB could not be detected in the BAC treated water at different H₂O₂ doses.

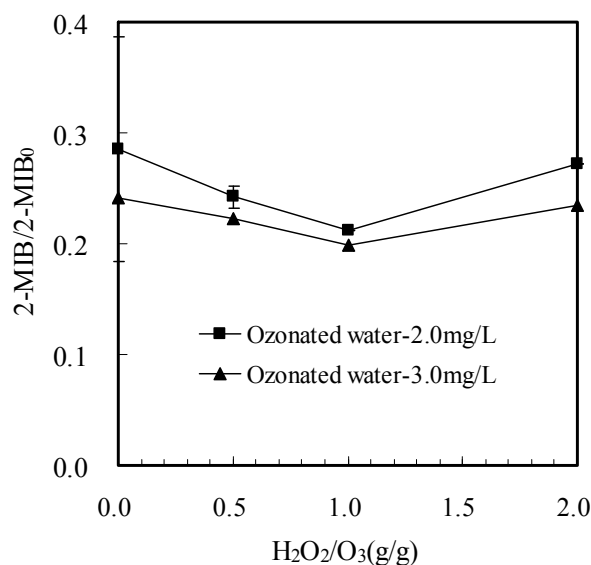


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Bromate concentration in ozonated water and BAC treated water

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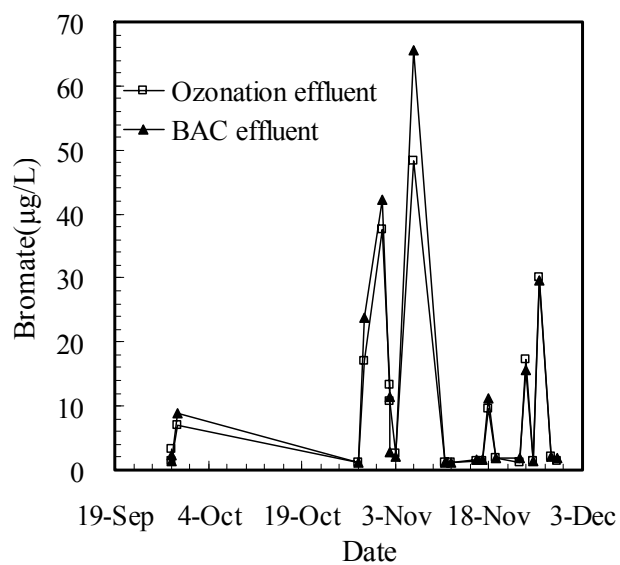


Fig. S3 Bromate concentration in the ozonated water and BAC treated water.

References

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