



Effects of microbubble ozonation on the formation of disinfection by-products in bromide-containing water from Tai Lake

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ABSTRACT

Because it promotes efficient gas solubility, microbubble aeration has been widely used in ozonation of water systems. In conventional ozone treatment, disinfection by-products (DBPs) are a significant concern during subsequent chlorination, especially when raw water contains bromide. However, the effect of microbubbles ozonation on the formation of DBPs is not well characterized. In this study, we investigated the formation of DBPs during chlorination of bromide-containing water from Tai Lake after microbubble ozonation, and compared this to conventional bubbling. The results suggest that more bromate and bromo-organic DBP precursors were produced after microbubble ozonation, which creates a challenge to the practical implementation of microbubble ozonation in water treatment. Since bromate is the main DBP of concern in the presence of bromide during ozonation, we also explored the effect of natural organic matter and hydroxyl radicals on bromate formation. Finally, we found that the addition of ammonia reduced bromate formation during microbubble ozonation to levels even lower than with conventional bubble ozonation.

1. Introduction

Ozonation of water has been widely used in water treatment because it is both highly effective and environmentally friendly. However, more widespread application of ozone is limited by its short retention time and low mass transfer coefficient. Microbubbles, with a diameter less than 50 μm , which are much smaller than conventional bubble (diameters larger than 1 mm), represent a possible solution to these problems according to mass transfer theory. The large specific surface area resulting from their small size is one of the most important characteristics of microbubbles, as this feature results in significant improvement of gas-liquid mass transfer. Under the influence of surface tension, microbubbles will constantly shrink as they rise in the water column, with the increase of internal pressure caused by self-pressurization. As a result, the gas mass transfer efficiency is increased, which further increases the surface tension, thus leading to further contraction and final annihilation of the bubbles [1]. Many researchers have reported an acceleration of hydroxyl radicals' generation during the collapse of air and ozone microbubbles [2–7] as detected by the electron-spin resonance method [5,8,9]. Hydroxyl radicals can react with a wide range of dissolved refractory compounds, while ozone is a highly selective oxidant [10,11]. Therefore, microbubbles offer a promising enhancement of current water treatment methods.

Although ozonation is effective in water treatment, one serious

drawback is the formation of disinfection by-products (DBPs) during subsequent chlorination [12], especially brominated DBPs. Bromide is generally present in raw water in China; for example, the bromide level in the Tai River can be as high as 200 $\mu\text{g/L}$ year-round. Bromide can react with ozone leading to the undesired formation of bromate and intermediate products which then could form multiple bromo-organic by-products during subsequent chlorination. Brominated DBPs are dozens to hundreds of times more toxic than conventional chlorinated DBPs [13], thus increasing the threat to human health. And bromate, owing to its ubiquity and high carcinogenicity, has become the DBP of most concern during ozonation of bromide-containing raw water. The maximum level of bromate in drinking water has been set at 10 $\mu\text{g/L}$ both in the United States and the European Union [14]. Therefore, DBP, and especially brominated DBP, formation is a considerable concern when microbubbles are used in ozonation.

In this study, we applied a typical microbubble generation system to examine the role of microbubbles ozonation in the formation of DBPs during subsequent chlorination in bromide-containing water from Tai Lake. Since bromate is the main DBP of concern in the presence of bromide, we also aimed to investigate and limit the extent of bromate formation during microbubble ozonation.

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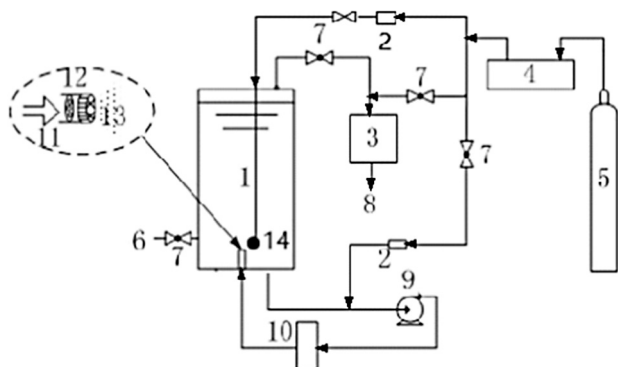


Fig. 1. Schematic of the experimental setup. 1. Reaction tank (40 L); 2. Flow meter; 3. Gaseous ozone monitor; 4. Ozone generator; 5. Oxygen cylinder; 6. Wastewater discharge port; 7. Valve; 8. Ozone emission port; 9. Centrifugal pump; 10. Dissolving tank (1 L); 11. Gas-containing water; 12. Microbubble-generating nozzle; 13. Microbubble-containing water; 14. Porous stones.

2. Experimental methods

2.1. Water samples

Water samples were collected from a drinking water treatment plant where the source water came from Tai Lake. Grab samples were collected after coagulation-sedimentation and gravity filtration treatment. All experiments were performed using freshly sampled water unless noted otherwise. Water quality features were constant throughout the period of the experiments. The DOC was 3.04 ± 0.1 mgC/L, UV254 was 0.045 ± 0.005 cm⁻¹, pH was 7.12 ± 0.05 , the concentration of Br⁻ was 178 ± 10 µg/L and the temperature remained 13 ± 1 °C.

2.2. Experimental method

All ozonation experiments were carried out using a semi-batch system. A schematic of the experimental apparatus is shown in Fig. 1. The microbubble aeration system contains a centrifugal pump, a dissolution tank and a microbubble-generating nozzle (MF5, Shanghai Xingheng Technology, Inc.), which generates microbubbles by applying a high pressure (0.4 MPa), as described previously [15]. Bubble diameters were determined using a Malvern Mastersizer, and ranged from 10 µm to 50 µm. Water was circulated by the pump at a flow rate of 12 L/min for microbubbles. In conventional bubble experiments, ozone gas was directly blown into the reaction tank through four stone balls (Φ 20 mm), which is the most widely used method in water treatment plants.

Ozone was generated from oxygen by a corona-discharge ozone generator (COM-AD-01, Anseros). The ozone gas concentration was monitored by a UV-absorption ozone meter (Model-600, HARE) and kept at 14.0 ± 0.1 mg/L during ozonation experiments. The outlet ozone gas after oxidation reactions was also analyzed before feeding into a destructor. The ozone gas flow rate was 0.3 L/min during all ozonation experiments. Before dosing with ozone, the solution was aerated by air for 30 min to check the effect of air microbubbles. To test the formation of halogen organic DBPs, samples were subjected to 30 min of chlorination (1.5 mg/L) after ozonation.

2.3. Analytical methods

Water quality features tested included dissolved organic carbon (DOC), pH, temperature, UV254, trihalomethanes (THMs) including Chloroform, bromodichloromethane, dibromochloromethane and three methyl bromide, haloacetonitriles (HANs) including chloroacetic acid, bromoacetic acid and dibromoacetic acid, haloacetic acid (HAA) including chloroacetonitrile, bromide chloride acetonitrile and dibromo

acetonitrile, bromide and bromate. DOC was measured by a TOC analyzer (TOC-L, Shimadzu). UV254 was detected by a UV spectrophotometer (DR-6000, HACH). THMs, HANs and HAA were quantified via liquid-liquid extraction with methyl tert-butyl ether (MTBE) as the extracting solvent, after which the samples were analyzed using a gas chromatograph (GC; 890B, Agilent) with an electron capture detector. The analysis of HAA incorporated anhydrous sodium sulfate as the drying agent and MTBE was used as the extracting solvent, with 1, 2-dibromopropane as the internal standard. A 20 ml sample was extracted with 4 ml extracting solvent. One micro liter of the extract was then injected into a GC with a DB-5 silica capillary column (30 m × 0.25 mm × 0.25 µm). For THMs and HANs, the GC temperature program started at 35 °C for 10 min, increased to 80 °C at 10 °C/min, then further increased to 150 °C at 20 °C/min for 1 min. For HAA, the GC temperature program started at 35 °C for 10 min, increased to 40 °C at 2 °C/min for 1 min, increased to 75 °C at 5 °C/min for 15 min, then increased to 100 °C at 40 °C/min for 15 min, and further increased to 135 °C at 40 °C/min. Bromate was determined by real-time sampling. Br⁻ and BrO₃⁻ concentrations were both analyzed via ion chromatography after chemical ion suppression (ICS-1000, Dionex). A Dionex AS19 column (internal diameter, 4 mm; length, 250 mm) and a Dionex AG19 guard column (internal diameter, 4 mm; length, 50 mm) were used. The mobile phase was 20 mM NaOH solution and the injection volume was 150 µL. Dissolved ozone was quantified using the indigo method [16].

3. Results and discussion

3.1. Formation of bromate and bromo-organic DBP precursors using different ozone-aeration methods

Ozonation of sand-filtered water from Tai Lake using either microbubbles or conventional bubbles was carried out (Fig. 2). Ozone dosage was calculated using the equation:

$$\int O_3 dt = \int_0^t (C_{in} - C_{out}) \times Q dt / V$$

where C_{in} is the influent ozone gas concentration at time t (mg/L), C_{out} is the outlet ozone gas concentration at time t (mg/L), Q is the gas flow rate (L/min), and V is the solution volume (L). As shown in Fig. 2a, bromate formation increased with increasing ozone dosage, reaching a plateau at about 20 µg/L, when ozone dosage exceeded 0.10 mmol/L under both aeration conditions. However, more bromate was generated with microbubble ozonation than with conventional bubbles at the early stages of the experiment. In Fig. 2b, formation rates of bromate, r_A , at different Br⁻ concentrations were obtained by a tangent line method. From the linear relationship of r_A and Br⁻ concentration, the homologous reaction rate constant k can be obtained. The bromate formation rate under microbubble aeration conditions, reaching 0.0962 min⁻¹, was clearly higher than with conventional bubbling (0.0247 min⁻¹). In theory, for a given ozone dosage rate and bromide concentration, the same amount of bromate should be formed regardless of process. Clearly, however, this is not what we observe and other factors must account for the difference. One hypothesis might be that free radicals are generated during the collapse of microbubbles; another might be that microbubbles increase the ozone concentration in the reaction region because the pressure inside a microbubble is higher than that outside [17–19].

Halogen DBP formation with chlorination after both ozone aeration methods is shown in Fig. 2c. Microbubble aeration results in the formation of a far higher level of halogen DBPs precursors than conventional bubbling, possibly because microbubbles provide a greater number of reactive intermediates such as ·OH [15] and thus a higher degree of oxidation. With ozone microbubbles, halogen DBP formation after chlorination increased rapidly and achieved a maximum at an ozone dosage of about 0.03 mmol/L, then decreased slightly as ozone

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