



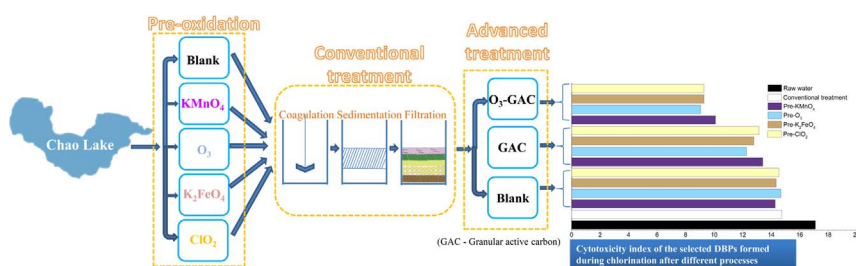
Comparison of drinking water treatment processes combinations for the minimization of subsequent disinfection by-products formation during chlorination and chloramination

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GRAPHICAL ABSTRACT



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ABSTRACT

Micro-ozone, permanganate, ferrate, and chlorine dioxide were applied as pre-oxidants to meet conventional requirements for water quality and algal control. The formation of typical carbonaceous disinfection by-products (C-DBPs) and emerging nitrogenous disinfection by-products (N-DBPs) during both chlorination and chloramination were investigated with combinations of different processes: (i) coagulation-sedimentation-filtration (CSF), (ii) pre-KMnO₄/O₃/K₂FeO₄/ClO₂ matched with CSF, and (iii) pre-KMnO₄/O₃/K₂FeO₄/ClO₂ coupled with CSF and followed by granular activated carbon (GAC)/O₃-GAC advanced treatment. All conventional water quality indexes met the requirements of the national standards for drinking water quality of China (GB5749-2006) when the system was under stable operation; only NH₄⁺-N, which only met the requirement with either pre-oxidation or post-ozonation, failed to do so. Micro-ozone pre-treatment coupled with CSF and O₃-GAC showed the best performance in removing chloroform (CF) and dichloroacetonitrile (DCAN) precursors. Pre-oxidation had a negative impact on chloropicrin (TCNM) and chloral hydrate formation, but a positive effect on DCAN. The trade-off analysis based on the toxicity of the selected DBPs indicated that pre-oxidation (KMnO₄, O₃, K₂FeO₄, and ClO₂) had a positive influence on the control of overall cytotoxicity, and O₃ pre-oxidation coupled with CSF followed by O₃-GAC performed the best. However, for the control of overall genotoxicity, the ClO₂ pre-treatment showed best results, whether or not GAC or O₃-GAC was involved. These results are important for optimizing the design of the treatment processes in drinking water treatment plants using eutrophic lake water where precursors for regulated C-DBP and unregulated N-DBP formation are prevalent.

Abbreviations: C-DBPs, carbonaceous disinfection by-products; CF, chloroform; HALs, haloaldehydes; CSF, coagulation-sedimentation-filtration; DBPs, disinfection by-products; DCACAm, dichloroacetamide; DOC, dissolved organic carbon; DON, dissolved organic nitrogen; DWTPs, drinking water treatment plants; FPs, formation potentials; HAAs, haloacetic acids; HAcAms, haloacetamides; N-DBPs, nitrogen-containing disinfection by-products; TCACAm, trichloroacetamide; THMs, trihalomethanes

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1. Introduction

Disinfection of drinking water has been an indispensable treatment process since the early 20th century as it has reduced the incidence of waterborne diseases including cholera, typhoid fever, and amoebic dysentery [1,2]. However, there are numerous organic materials (i.e. humus, amino acids, proteins, environmental pollutants) in natural water that can react with free chlorine to yield halogenated organics [3,4]. Ever since disinfection by-products (DBPs) were first related to chronic health concerns, rather than acute effects, in the early 1970s [5], approximately hundreds of DBPs have been reported in the literature [4,6–8]. More attention has been focused on nitrogen-containing DBPs (N-DBPs), including haloacetonitriles (HANs), halonitromethanes (HNMs), and haloacetamides (HAcAms), because they are actually more cytotoxic and genotoxic in mammalian cell assays than the regulated carbonaceous DBPs (C-DBPs), including trihalomethanes (THMs), haloacetic acids (HAAs), and chloral hydrate (CH) [8–10]. When raw water contains bromine or/and iodine, brominated and iodinated analogues with much higher cytotoxicity and genotoxicity than chlorinated DBPs may also occur [3].

Removing precursors of DBPs using pre-treatment or advanced post-treatment technologies before disinfection is a relatively effective way to control the formation of DBPs [11,12]. Dissolved organic nitrogen (DON) is a major precursor of N-DBPs. Compared to dissolved organic carbon (DOC), the main precursors of C-DBPs [13–15], DON is greatly impacted by algal blooms and fertilizer runoff [12,16]. A median DON concentration of 0.37 mg/L of N was reported in surface water and 0.24 mg/L of N in shallow groundwater [14]. The DOC concentration is usually much higher than DON, and in earlier studies DOC/DON averaged 18 mg DOC per mg DON [14,17].

Previous studies indicated that Al and Fe salts used as coagulants do not efficiently coagulate organic nitrogen compounds or proteins [17–19]. Algal-derived DOC enriched in nitrogen, in particular, is reportedly difficult to be removed during coagulation [20]. Many studies have shown that the use of ozone, permanganate, ferrate or chlorine dioxide as pre-oxidants can enhance the removal of algae [21,22]. Apart from its application as a disinfectant, ozone has been used as a pre-oxidant, or in advanced treatment, due to its high oxidation potential [23,24]. It can oxidize a wide range of nuisance or potential toxicants, including substances with natural colour and compounds associated with unpleasant tastes or odours [23,25–28]. Ozone, integrated with granular activated carbon (GAC) filtration, is commonly used to degrade toxic micro-pollutants, remove precursors of THMs, and increase bio-degradability in GAC filtration [26,29–31]. Taking the cost and practicability into consideration, low doses of ozone (micro-ozone) are also used as pre-oxidants. Pre-oxidation with potassium permanganate, despite not being a cure-all technology, has long been considered an alternative to pre-chlorination, not merely for lowering THM concentrations but also for eliminating compounds that produce tastes and odours [32,33]. Ferrate is a strong oxidant; its reduction potential can reach as high as +2.2 V in an acidic environment, which makes it even more powerful than ozone and potassium permanganate [34]. Studies have also shown that pre-treatment with potassium ferrate could enhance coagulation efficiency and increase algae removal [35–37]. Chlorine dioxide is becoming a more popular agent in drinking water treatment. As reported before, chlorine dioxide used as pre-oxidant can reduce the formation of THMs and HAAs [38,39].

Taking into account the environment impacts, low consumption, and low operation costs, low doses of these oxidants were recommended for pre-oxidation processes in many drinking water treatment plants (DWTPs) of China, besides the quality of drinking water must meet the requirements of national standards for drinking water quality of China (GB5749-2006). However, little is known on the formation of typical C-DBPs and N-DBPs during chlorination and chloramination after these pre-treatments and conventional treatments, used alone or followed by O₃-GAC. Therefore, in the present study, the

formation of typical C-DBPs and emerging N-DBPs during both chlorination and chloramination was investigated with three different process combinations: (i) coagulation-sedimentation-filtration (CSF), (ii) pre-KMnO₄/O₃/K₂FeO₄/ClO₂ matched with CSF, and (iii) pre-KMnO₄/O₃/K₂FeO₄/ClO₂ coupled with CSF and followed by GAC/O₃-GAC advanced treatment. Conventional water quality indexes (turbidity, permanganate index (COD_{Mn}), UV₂₅₄ and TOC), the formation potentials (FPs) of typical DBPs (THMs, CH, HANs, HNMs, and HAcAms) and the reduction of DBP FPs were all examined.

2. Materials and methods

2.1. Materials

The pilot experiment was carried out in October 2016. All the raw water was sampled through the intake of a drinking water plant located in Hefei Province (China) which draws water from Lake Chao. This lake has long been considered one of the five largest freshwater lakes in China (with maximum capacity 4.81 billion cubic meters and an average water depth of 3 m), and is a major source of the potable water used in Hefei Province. However, the lake has been subjected to eutrophication and frequent algal blooms for years, due to the enrichment of the nitrogen and phosphorus in the Chao Lake from the residents' raw sewage, agricultural non-point source pollution and waste water from the factory [40–42]. In order to deal with the water quality problems caused by algae, the DWTPs using eutrophic lake water usually add pre-treatment and/or advanced treatment (e.g., O₃-GAC) to ensure the safety of drinking water quality.

This study measured THMs, CH, HANs, HNMs, and HAcAms. DCACAm (98.5%) and TCACAm (98.5%) standards were purchased from Alfa Aesar (Karlsruhe, Germany). Free chlorine stock solution was prepared using sodium hypochlorite solution (active chlorine > 5%, Sinopharm Chemical Reagent Co., Ltd., China). All other chemical reagents were of analytical grade at least and were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China) unless noted. Ultrapure water was obtained from a Millipore Milli-Q Gradient water purification system (18 MΩ·cm, Billerica, MA, USA).

2.2. Pilot-plant process flows

The pilot plant is located in a typical DWTP near Lake Chao. The process flows included pre-treatment, coagulation, sedimentation, sand filtration, post-ozonation, and GAC filtration. Pre-treatment included four different oxidants: KMnO₄, O₃, K₂FeO₄, and ClO₂ pre-oxidation. The design flow velocity of the pilot plant used in this study was 1 m³/h.

2.2.1. Pre-treatment

The pre-treatment mainly included ozone, potassium permanganate, ferrate, and chlorine dioxide. Ozone is generated by a domestic ozone generator (GuoLin, Qingdao, China). A pre-ozone reaction column (0.16 m [Φ] × 2.75 m [H]) was used to ensure sufficient contact and dissolution of ozone in the water. The hydraulic retention time (HRT) was 10 min and co-flow ambient of ozone and water was selected. Except for ozone, each oxidant (potassium permanganate, ferrate, or chlorine dioxide) was auto-added from a barrel under control of a metering pump and controlled at about 1 mg/L in the water.

2.2.2. Coagulation-sedimentation-filtration (CSF)

With poly aluminium ferric sulphate dosed at 10 mg/L, the coagulation process was achieved in a three-stage series flocculation chamber (1.25 m [L] × 0.4 m [W] × 0.6 m [H]). The average velocity gradients in the three chambers, which had speed-regulating stirring devices, were 50, 20, and 8 s⁻¹, respectively. A sedimentation tank (1.25 m [L] × 0.73 m [W] × 1.65 m [H]) with up-flow inclined-tube settler was in front of the filter. The filtration column (0.4 m [Φ] × 1.7 m [H]) was

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