



The enhanced removal of carbonaceous and nitrogenous disinfection by-product precursors using integrated permanganate oxidation and powdered activated carbon adsorption pretreatment



Wenhai Chu^{a,b,*}, Dechang Yao^a, Naiyun Gao^{a,*}, Tom Bond^c, Michael R. Templeton^c

^a State Key Laboratory of Pollution Control and Resources Reuse, College of Environmental Science and Engineering, Tongji University, Shanghai 200092, China

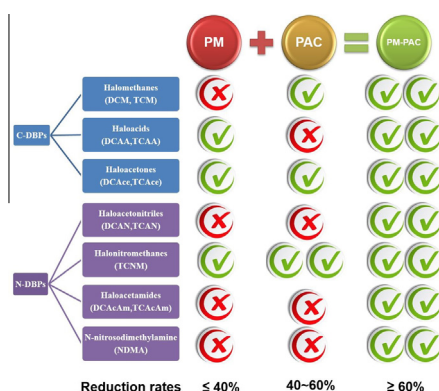
^b Research & Service Center for Environmental Industry, Yancheng 224051, China

^c Department of Civil and Environmental Engineering, Imperial College London, London SW7 2AZ, UK

HIGHLIGHTS

- PM oxidation and PAC adsorption both improved the removal of DOC and DON compounds.
- PM oxidation has better performance on reducing HAA formation than PM adsorption.
- PAC adsorption has better performance on reducing N-DBP and THM formation than PM oxidation.
- PM improved the performance of PAC on removal of C-DBP and N-DBP precursors.
- PM–PAC process is a promising pretreatment technology in integrated control of DBPs.

GRAPHICAL ABSTRACT



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ABSTRACT

Pilot-scale tests were performed to reduce the formation of a range of carbonaceous and nitrogenous disinfection by-products (C-, N-DBPs), by removing or transforming their precursors, with an integrated permanganate oxidation and powdered activated carbon adsorption (PM–PAC) treatment process before conventional water treatment processes (coagulation–sedimentation–filtration, abbreviated as CPs). Compared with the CPs, PM–PAC significantly enhanced the removal of DOC, DON, $\text{NH}_3\text{-N}$, and algae from 52.9%, 31.6%, 71.3%, and 83.6% to 69.5%, 61.3%, 92.5%, and 97.5%, respectively. PM pre-oxidation alone and PAC pre-adsorption alone did not substantially reduce the formation of dichloroacetonitrile, trichloroacetonitrile, N-nitrosodimethylamine and dichloroacetamide. However, the PM–PAC integrated process significantly reduced the formation of both C-DBPs and N-DBPs by 60–90% for six C-DBPs and 64–93% for six N-DBPs, because PM oxidation chemically altered the molecular structures of nitrogenous

Abbreviations: C-DBPs, carbonaceous disinfection by-products; CF, chloroform; DBPs, disinfection by-products; CPs, conventional water treatment processes; DCAcAm, dichloroacetamide; DCAA, dichloroacetic acids; DCAN, dichloroacetonitrile; DCAc, dichloroacetone; DCM, dichloromethane; DIN, dissolved inorganic nitrogen; DOC, dissolved organic carbon; DOM, dissolved organic matter; DON, dissolved organic nitrogen; DWTPs, drinking water treatment plants; GC/MS, gas chromatograph/mass spectrometry; HAAs, haloacetic acids; HACAm, haloacetamides; HANs, Haloacetonitriles; HNMs, halonitromethanes; HPI, hydrophilic; HPO, hydrophobic; N-DBPs, nitrogenous disinfection by-products; NDMA, N-nitrosodimethylamine; PAC, powdered activated carbon; PM, permanganate; TCAcAm, trichloroacetamide; TCAA, trichloroacetic acid; TCAc, trichloroacetone; TCAN, trichloroacetonitrile; TCNM, trichloronitromethane; THMs, trihalomethanes; UFC, uniform formation conditions..

* Corresponding authors at: State Key Laboratory of Pollution Control and Resources Reuse, College of Environmental Science and Engineering, Tongji University, Shanghai 200092, China (W. Chu).

E-mail addresses: 1world1water@tongji.edu.cn, feedwater@126.com (W. Chu), gaonaiyun@126.com (N. Gao).

Permanganate
Powdered activated carbon
Integrated process

organic compounds and increased the adsorption capacity of the DBP precursors, thus highlighting a synergistic effect of PM and PAC. PM–PAC integrated process is a promising drinking water technology for the reduction of a broad spectrum of C-DBPs and N-DBPs.

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1. Introduction

The presence of nitrogenous disinfection by-products (N-DBPs) in drinking water has gained attention because these compounds are significantly more genotoxic and cytotoxic than the currently regulated carbonaceous DBPs (C-DBPs), such as trihalomethanes (THMs) and haloacetic acids (HAAs) (Muellner et al., 2007; Plewa et al., 2008a,b; Richardson et al., 2007; Richardson and Ternes, 2014). Moreover, water resource shortages and growing water demands have forced utilities to exploit source waters influenced by wastewater effluents and/or algal blooms, both of which are known to be key sources of dissolved organic nitrogen (DON) and in turn N-DBP precursors. Among N-DBP families of current interest are halonitriles (e.g., dichloroacetonitrile [DCAN]), halonitroalkanes (e.g., trichloronitromethane [TCNM]), haloamides (e.g., dichloroacetamide [DCAcAm]), and N-nitrosamines (e.g., N-nitrosodimethylamine [NDMA]) (Fig. SM1) (Shah and Mitch, 2012).

Numerous DBPs can be controlled in drinking water systems by removal of dissolved organic matter (DOM)—the main precursors of DBPs—through physicochemical treatment processes, such as enhanced coagulation, upstream of the disinfection process. However, in polluted source waters featuring high levels of nitrogenous compounds, including dissolved inorganic nitrogen (DIN) and DON, conventional water treatment processes (abbreviated as CPs, e.g., coagulation–sedimentation–filtration) (Chu et al., 2011) are typically ineffective in removing these precursors. Additional processes are therefore often applied to improve the removal of algae, taste and odor compounds, $\text{NH}_4^+\text{-N}$ and other pollutants, including chemical processes (e.g., permanganate [PM] oxidation) and physical processes (e.g., powdered activated carbon [PAC] adsorption).

In recent years, PM had been used widely as a pre-treatment process to strengthen the subsequent CPs in drinking water treatment plants (DWTPs). PAC is also usually used as an emergency measure to remove algae, taste and odor compounds and other pollutants in raw water with high concentration (Westerhoff et al., 2005; Zhang et al., 2010). Previous studies have reported that PM oxidation or PAC adsorption can reduce and/or increase the formation of several regulated C-DBPs and unregulated N-DBPs by removing and/or chemically transforming precursors prior to disinfection (Chen and Valentine, 2008; Chu et al., 2011; Kristiana et al., 2011; Jiang et al., 2014). However, very little information is available concerning the control of a range of N-DBPs in N-rich waters pre-treated by coupling PM oxidation and PAC adsorption (PM–PAC). Therefore, the objective of this study was to examine the removal efficiencies of the precursors for six representative unregulated N-DBPs (DCAN, trichloroacetonitrile [TCAN], TCNM, DCAcAm, trichloroacetamide [TCAcAm], and NDMA) and six C-DBPs (dichloromethane [DCM], chloroform [CF], dichloroacetic acid [DCAA] and trichloroacetic acid [TCAA], 1,1-dichloroacetone [DCAce], and 1,1,1-trichloroacetone [TCAce]) with PM and/or PAC pre-treatment processes prior to a conventional surface water treatment train (coagulation–sedimentation–filtration). The formation of brominated DBPs was not the focus of the study due to the relatively low bromide levels in the filtered waters throughout the experimental period.

2. Materials and methods

2.1. Materials

Standards for two HANs (DCAN and TCAN), one HNM (TCNM), one N-nitrosamine (NDMA), two halomethanes (DCM and CF), two HAAs (DCAA and TCAA), and two halogenated acetones (DCAce and TCAce) were purchased from Sigma–Aldrich (Oakville, ON, Canada). Standards for two HACams (DCAcAm and TCAcAm) were obtained from Alfa Aesar (Karlsruhe, Germany). A sodium hypochlorite solution (active chlorine >5%, Sinopharm Chemical Reagent Co., Ltd., China) was used to prepare free chlorine stock solutions. Ultrapure water was produced with a Millipore Milli-Q Gradient water purification system (Billerica, MA, USA). All other materials were at least analytical grade and obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China) unless otherwise noted.

2.2. Pilot-plant process flow

The pilot plant was located in a conventional drinking water treatment plant (DWTP) situated near Taihu Lake. The three pilot plant pre-treatment process flows (PAC adsorption, PM oxidation, PM–PAC integrated process) were operated with downstream conventional water treatment processes (coagulation–sedimentation–filtration–chlorination) at a design flow of $1\text{ m}^3/\text{h}$, as shown in Fig. 1. The raw water used for the pilot-scale treatment processes was obtained from an intake in the east of Taihu Lake, which is the third largest freshwater lake in China and the potable water source of many cities and towns in Eastern China. In recent years, the accumulation of nutrient-rich sewage and agricultural runoff in this shallow lake has caused severe eutrophication and nuisance algae blooms (Yang et al., 2008). Therefore, the pre-treatment process was added before conventional treatment processes in most of DWTPs in these cities, to improve the final treated drinking water quality.

The pre-treatment process was composed of PM oxidation alone, PAC adsorption alone, and the PM–PAC integrated process. For PM oxidation, 1.0 mg/L KMnO_4 was added to the influent of the raw water tank. For PAC adsorption, 20 mg/L PAC was dosed to the effluent of the raw water tank from a hydrated PAC tank by a metering pump, and was transported to the coagulation tank (three-stage flocculation plant) continuously. For PM–PAC

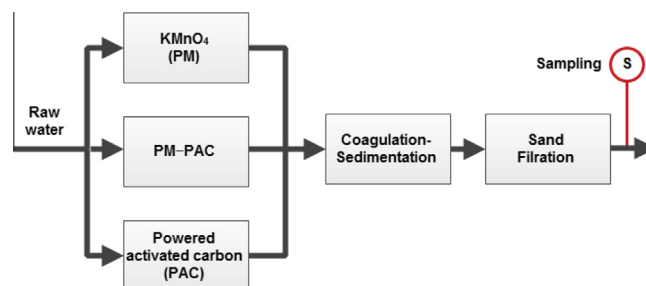


Fig. 1. Pilot plant process flow diagrams.

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