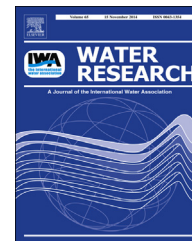




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The control of emerging haloacetamide DBP precursors with UV/persulfate treatment

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ABSTRACT

Haloacetamides (HACAm), an emerging class of nitrogen-containing disinfection byproducts (N-DBPs) of health concern in drinking water, have been reported to occur in treated drinking waters at low $\mu\text{g/L}$ levels typically. The objective of this study was to examine the potential of an ultraviolet light/persulfate (UV/PS) oxidation technology to reduce the precursors of HACAm and also minimize the formation of other N-DBPs upon subsequent chlorination. Low-pressure UV photolysis alone and PS pre-oxidation alone did not significantly affect HACAm formation, however UV/PS pre-oxidation achieved a statistically significant reduction in HACAm formation and also reduced bromine incorporation into the HACAm. UV/PS also showed a good performance in removing the precursors of haloacetonitriles and halonitromethanes prior to chlorination. Therefore, UV/PS has the potential to minimize the formation of a range of N-DBPs in organic nitrogen-rich waters where N-DBP precursors are prevalent. However, these benefits should be weighed against the potential drawbacks of increased bromate and sulfate formation, particularly in high-bromide waters.

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1. Introduction

Haloacetamides (HACAm), an emerging class of halogenated nitrogen-containing disinfection byproducts (N-DBPs), have been found to be highly cytotoxic and genotoxic in mammalian cell assays, i.e. $142\times$ more cytotoxic and $12\times$ more genotoxic than regulated haloacetic acids (HAAs) (Plewa et al., 2007; Richardson et al., 2007). Their elevated toxicity was also observed in a recent study based on metabonomics

(Zhang et al., 2013). Most of the chlorinated HACAm (Cl-HACAm) are less cytotoxic and genotoxic than their brominated and iodinated analogues (Plewa et al., 2008), which is similar to toxicity relationships for other DBPs (Yang and Zhang, 2013; Richardson and Ternes, 2014).

Five HACAm, including chloro- (Cl-HACAm), dichloro- (DCl-HACAm), trichloro- (TCl-HACAm), bromo- (Br-HACAm), and dibromoacetamide (DBr-HACAm), were quantified as part of a U.S. nationwide DBP occurrence study, and the highest total concentration of these HACAm in the finished drinking waters

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was 7400 ng/L (median = 1400 ng/L) (Krasner et al., 2006). Afterwards, other brominated HACams (Br-HACams), including bromochloro- (BCAcAm), bromodichloro- (BDCAcAm), and dibromochloroacetamide (DBCACAm) (Richardson et al., 2008; Pressman et al., 2010), and an iodinated HACAm (I-HACAm) bromoiodoacetamide (BIAcAm) (Plewa et al., 2008) were identified in drinking water by broad-screen analyses using gas chromatography/mass spectrometry (GC/MS). The occurrence of tribromoacetamide (TBACAm) has also been reported in swimming pool water (Richardson et al., 2010). Recently, a method for the trace determination of 13 HACams, including three new I-HACams (iodo- [IACAm], chloroiodo- [CIACAm], diiodoacetamide [DIACAm]), in seven drinking water treatment plants (DWTPs) in China was reported (Chu et al., 2012a). Using that method, a number of Br- and I-HACams, including BcAcAm, BdAcAm, DbcAcAm, TbaAcAm, and CiaAcAm, were detected in chlor(am)inated drinking water for the first time (Chu et al., 2012a).

Generally, HACAm precursors are characterized by low molecular weight (MW), low hydrophobicity, and poor removal efficiencies by a conventional DWTP using coagulation-sedimentation-filtration (Chu et al., 2010, 2011). An effective strategy to control the formation of DBPs is to remove their precursors before chlorination disinfection (Bond et al., 2011; Gan et al., 2013; Krasner et al., 2013). UV is a widely used technology for disinfection and photochemical oxidation of water and wastewater (Li et al., 2012; Poepping et al., 2014). Several studies reported the impact of direct UV photolysis on N-DBP formation upon subsequent chlorination, and found that the formation of N-DBPs depended on the lamp type (Liu et al., 2006; Bond et al., 2011). Medium pressure UV irradiation alone increases the formation of some DBPs during subsequent chlorination and can simultaneously reduce the formation of other DBPs (Shah et al., 2011; Reckhow et al., 2010). However, low pressure UV irradiation at the commonly applied UV disinfection dose range (e.g. 40–100 mJ/cm²) has little impact on N-DBP formation (Sharpless and Linden, 2003; Krasner et al., 2013). Xie et al. (2013) investigated the impact of ozone and permanganate on the subsequent formation of some chlorinated DBPs (including Cl-HACams) during chlorination, and found ozonation and permanganate oxidation had less impact on the formation of Cl-HACams compared to other N-DBPs. However, there has been very little research to examine the effect of PS oxidation on HACAm formation.

Recently, an innovative oxidation technology based on the generation of sulfate radical anions (SO₄⁻) by ultraviolet light-activated persulfate (UV/PS) has been studied (Antoniou et al., 2010; Gao et al., 2012; Grčić et al., 2012). Because of its moderate cost and relatively high stability and solubility, PS is an ideal source of SO₄⁻ through activation by UV. Sulfate radicals have a high reduction potential of 2.6 V, which is only slightly lower than that of hydroxyl radicals (2.9 V), but they are more stable in water than hydroxyl radicals (Antoniou et al., 2010; Tan et al., 2013). Moreover, sulfate radicals exhibit a nonselective oxidation pattern and can quickly decompose a wide range of organic pollutants in water (Gao et al., 2012). However, when this technology is used to reduce target organic contaminants, natural organic matter will be simultaneously altered, which is likely to result in the changes in the

speciation and quantities of DBPs formed upon subsequent chlorination. Previous study has found that hydroxyl radicals, formed from ultraviolet light-hydrogen peroxide, could increase THM formation at a UV dose of 1000 mJ/cm² (Dotson et al., 2010). However, there has been little study to examine the impact of UV/PS on DBP formation, and especially, no information is available concerning the formation of Cl-, Br- and I-HACams and other N-DBPs in drinking waters treated by UV/PS and subsequent chlorination.

The objective of this study was therefore to examine the potential of ultraviolet light/persulfate (UV/PS) oxidation technology to reduce the precursors of HACams, by determining HACAm formation upon subsequent chlorination in three filtered waters with different dissolved organic matter (DOM) characteristics, dissolved organic nitrogen (DON), bromide, and iodide levels. In addition, the impact of UV/PS on the formation of other halogenated N-DBPs, including dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), dichloronitromethane (DCNM), and trichloronitromethane (TCNM), upon subsequent chlorination was also examined.

2. Materials and methods

2.1. Materials

CACAm (98.5%), DCACAm (98.5%), and TCACAm (99%) standards were obtained from Alfa Aesar (Karlsruhe, Germany). BACAm and IACAm standards were purchased from Sigma-Aldrich (St. Louis, Missouri, USA). DBACAm, BCACAm, BDCACAm, DBCACAm, TBACAm, DIACAm, CIACAm, and BIAcAm standards were purchased from Orchid Cellmark (New Westminster, BC, Canada). N-DBPs (DCAN, TCAN, DCAN, and TCNM) and the model compounds (free tyrosine, boc-tyrosine, and tyrosine tert-butyl ester) were purchased from Sigma-Aldrich (Oakville, ON, Canada). 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). Sodium persulfate (Na₂S₂O₈ ≥99.5%) and other materials were at least analytical grade and obtained from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China) unless otherwise noted.

2.2. Sampling

Water samples were collected from three Chinese DWTPs. The water samples were collected after coagulation, sedimentation, and filtration. The plants did not use any pre-oxidant before filtration, so the collected samples had never been exposed to any oxidant. Also, analyses of the waters did not show any detectable HACAm background levels. 'MH' DWTP located in southern China treated water from the Yangtze River. 'SY' DWTP located in northern China treated water from the Yellow River. The raw waters of MH and SY had a high and low specific UV absorbance at 254 nm (SUVA₂₅₄), respectively. In addition, 'ZQ' DWTP, which treats Taihu Lake water, characterized by high DON, was also selected for the study. These DWTPs were selected because of differences in their DOM characteristics (i.e. SUVA₂₅₄, DON, as

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