Water Research 107 (2016) 141-150

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

Copper increases reductive dehalogenation of haloacetamides by zero-valent iron in drinking water: Reduction efficiency and integrated toxicity risk

Wenhai Chu ^{a, *}, Xin Li ^a, Tom Bond ^b, Naiyun Gao ^a, Xu Bin ^a, Qiongfang Wang ^a, Shunke Ding ^a

 ^a State Key Laboratory of Pollution Control and Resources Reuse, Institute of Disinfection By-product Control in Water Treatment, College of Environmental Science and Engineering, Tongji University, Shanghai, 200092, China
^b Department of Civil and Environmental Engineering, Imperial College London, London, SW7 2AZ, UK

ARTICLE INFO

Article history: Received 21 June 2016 Received in revised form 16 October 2016 Accepted 19 October 2016 Available online 19 October 2016

Keywords: Disinfection byproducts Haloacetamides Dechlorination Zero-valent iron/copper Integrated toxicity risk Drinking water

ABSTRACT

The haloacetamides (HAcAms), an emerging class of nitrogen-containing disinfection byproducts (N-DBPs), are highly cytotoxic and genotoxic, and typically occur in treated drinking waters at low μ g/L concentrations. Since many drinking distribution and storage systems contain unlined cast iron and copper pipes, reactions of HAcAms with zero-valent iron (ZVI) and metallic copper (Cu) may play a role in determining their fate. Moreover, ZVI and/or Cu are potentially effective HAcAm treatment technologies in drinking water supply and storage systems. This study reports that ZVI alone reduces trichloroacetamide (TCAcAm) to sequentially form dichloroacetamide (DCAcAm) and then monochloroacetamide (MCAcAm), whereas Cu alone does not impact HAcAm concentrations. The addition of Cu to ZVI significantly improved the removal of HAcAms, relative to ZVI alone. TCAcAm and their reduction products (DCAcAm and MCAcAm) were all decreased to below detection limits at a molar ratio of ZVI/Cu of 1:1 after 24 h reaction (ZVI/TCAcAm = 0.18 M/5.30 μ M). TCAcAm reduction increased with the decreasing pH from 8.0 to 5.0, but values from an integrated toxic risk assessment were minimised at pH 7.0, due to limited removal MCAcAm under weak acid conditions (pH = 5.0 and 6.0). Higher temperatures (40 °C) promoted the reductive dehalogenation of HAcAms. Bromine was preferentially removed over chlorine, thus brominated HAcAms were more easily reduced than chlorinated HAcAms by ZVI/Cu. Although tribromoacetamide was more easily reduced than TCAcAm during ZVI/Cu reduction, treatment of tribromoacetamide resulted in a higher integrated toxicity risk than TCAcAm, due to the formation of monobromoacetamide (MBAcAm).

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1. Introduction

Nitrogenous disinfection by-products (N-DBPs) have received increasing attention because they are more toxic than the regulated trihalomethanes (THMs) and haloacetic acids (HAAs) (Plewa et al., 2007, 2008; Yang and Zhang, 2013; Liu and Zhang, 2014; Chu et al., 2015a). As a result of rapid population growth and rising water demand, drinking water source waters are facing threats of insufficiently treated wastewater discharges and algal blooms (Shah and Mitch, 2012; Chu et al., 2015b). These polluted water sources are characterized by higher levels of dissolved organic nitrogen (Dotson and Westerhoff, 2009; Liu et al., 2013),, which is linked to their frequent occurrence in chlor(am)inated drinking water (Krasner et al., 2006, 2013; Richardson et al., 2008, Richardson and Ternes, 2014; Goslan et al., 2009; Bond et al., 2011, 2015; Chu et al., 2012; Hou et al., 2012). Among these N-DBPs, haloacetamides (HAcAms) were found to be highly cytotoxic and genotoxic in mammalian cell assays, i.e. $142 \times$ more cytotoxic and $12 \times$ more genotoxic than HAAs (Plewa et al., 2007; Richardson et al., 2007). They are the most cytotoxic of all DBP classes measured to-date, and are the second-most genotoxic DBP class, just behind the halonitriles (Plewa and Wanger, 2015; Richardson and Postigo, 2015). Among HAcAms, dichloroacetamide (DCAcAm) is typically







^{*} Corresponding author. College of Environmental Science and Engineering, Tongji University, Room 308 Mingjing Building, 1239 Siping Road, Yangpu District, Shanghai, 200092, China.

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

found at the highest concentration, with trichloroacetamide (TCAcAm) and monochloroacetamide (MCAcAm) reported infrequently in low-bromide sources (Krasner et al., 2006; Richardson et al., 2007; Chu et al., 2012). The elevated toxicity of TCAcAm and DCAcAm were also observed in the recent studies based on metabonomics (Zhang et al., 2013; Yu et al., 2015), and DCAcAm presented significantly higher cytotoxicity and genotoxicity than halomethanes (Yang et al., 2014). While chlorinated HAcAms tend to occur at higher concentrations than their brominated analogues in drinking water (Krasner et al., 2006; Bond et al., 2011; Chu et al., 2012), the latter are actually more cytotoxic and genotoxic (Plewa et al., 2008), Therefore, it is important to control both chlorinated and brominated HAcAms in drinking water.

Generally, DBP control strategies can be divided into three categories: (1) removal of DBP precursors prior to disinfection, (2) modification of disinfection practices to minimize DBP formation, and (3) removal of DBPs after formation. It has been reported that various pre-treatments in drinking water treatment plants (DWTPs) can achieve good removal of HAcAm precursors (Chu et al., 2011, 2014, 2015a; Xie et al., 2013), as well as for other N-DBPs (Bond et al., 2011; Shah et al., 2012). Efforts have also been made to study HAcAm formation mechanisms with the purpose of acquiring the knowledge to reduce their formation during chlorination (Shah and Mitch, 2012; Huang et al., 2012; Wang et al., 2014; Chu et al., 2015b). However, DBP formation can also occur in water distribution systems, due to reactions involving residual chlorine (Rossman et al., 2001). There remains an information gap on the removal of N-DBPs after their formation.

The polar nature of HAcAms (Fig. SM1), including MCAcAm, DCAcAm and TCAcAm, suggests that reductive dehalogenation is a potential route for their removal. Furthermore, the end products of dehalogenation (i.e. non-chlorinated acetamide) are of low-toxicity to humans and the environment (Plewa et al., 2008). This motivated us to consider the potential of reductive dehalogenation technologies to reduce the levels of HAcAms in drinking water.

Since Gillham and O'Hannesin discovered that metallic iron fillings could be utilized in groundwater remediation (Gillham and O'Hannesin, 1994), the use of zero-valent iron (ZVI, Fe⁰) for in-situ remediation of groundwater contaminated by chlorinated organic compounds has received considerable attention (Dries et al., 2004; He et al., 2010; Kohn et al., 2005; Liu and Lowry, 2006; Zhang et al., 2011). Moreover, to enhance dehalogenation and prolong the activity of ZVI, a second transition metal (e.g. Pd, Ni, Ru and Pt) with high hydrogenation ability can be incorporated in a bimetal system, such as applied for the remediation of carbon tetrachloride (Feng and Lim, 2007; Wang et al., 2009), chloroform (Feng and Lim, 2007; Wang et al., 2009), dichloromethane (Feng and Lim, 2007; Wang et al., 2009), tetrachloroethylene (Lien and Zhang, 2001; Zhang et al., 1998), trichloroethylene (Lien and Zhang, 2001; Tee et al., 2009; Zhang et al., 1998), dichloroethene (Lien and Zhang, 2001; Zhang et al., 1998), 1,1-dichloroethane (Cwiertny et al., 2006), 1,1,1-trichloroethane (Cwiertny et al., 2006; Lien and Zhang, 2001), and 1,1,2,2-tetrachloroethane (Lien and Zhang, 2005).

Finished drinking water is often distributed from DWTPs to consumers by unlined cast iron pipes, and sometimes copper pipes (Ridgway et al., 1981; McNeill and Edwards, 2001; Lin et al., 2001; Niquette et al., 2000). During distribution, drinking water is also often stored in storage ponds made from iron materials, with copper materials also sometimes used to inhibit the growth of microorganisms (Kooij et al., 2005; Teng et al., 2008; Critchley et al., 2001; Lehtola et al., 2004). It is important to proactively examine the potential of ZVI, Cu, and their combination (ZVI/Cu) for the reductive dehalogenation of HAcAms and possible application in drinking water distribution and storage systems. Therefore, the objective of this study was to examine the potential of three

reductive dehalogenation technologies – ZVI alone, Cu alone, and combined ZVI/Cu – for removing HAcAms in drinking water. This is the first study to report findings about N-DBP removal by reductive dehalogenation methods. Both chlorinated and brominated HAcAms were considered. The changes of HAcAms concentrations and integrated toxic risk caused by HAcAms were also examined.

2. Materials and methods

2.1. Materials

MCAcAm (98.5%), DCAcAm (98.5%) and TCAcAm (99%) standards were obtained from Alfa Aesar (Karlsruhe, Germany), and five bromine-containing HAcAms (bromochloroacetamide [BCAcAm], dibromoacetamide [DBAcAm], bromodichloroacetamide [BDCAcAm], dibromochloroacetamide [DBCAcAm] and tribromoacetamide [TBAcAm]) were purchased from Orchid Cellmark (New Westminster, BC, Canada). Monobromoacetamide (MBAcAm) and three HAAs (monochloroacetic acid [MCAA], dichloroacetic acid [DCAA] and trichloroacetic acid [TCAA]) were supplied by Sigma-Aldrich (St Louis, Missouri, USA). Sodium acetate anhydrous, 2-(N-morpholino)ethanesulfonic acid (MES, 99%) and tris(hydroxymethy)aminomethane (TRIS, 99.9%) were purchased from Aladdin Industrial Inc (Shanghai, China). The ZVI (299.99%) and copper (≥99.99%) powders were also supplied by Sigma-Aldrich (St Louis, Missouri, USA). Ultrapure water was produced with a Millipore Milli-Q Gradient water purification system (Billerica, MA, USA). All the other chemical reagents were at least of analytical grade, and obtained from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China) unless otherwise noted.

2.2. Experimental procedure

Batch reduction tests were performed by adding selected doses of ZVI and/or Cu powder (doses range: ZVI = 0.2–10.0 g/L [0.004–0.179 M]; ZVI/Cu molar ratio = 1:1, except as noted) to 250-mL sealed glass bottles containing 50 mL one of the following trihalogenated HAcAms: TCACAM, BDCAcAM, DBCAcAM or TBAcAM (5.30 μ M, except as noted). Before each experiment the reaction bottles were purged with nitrogen to remove air. Each run lasted for 24 h and the bottles were placed on a water bath shaker at 150 r/m to make the solid and liquid well-mixed and keep the temperature constant at 25 \pm 0.2 °C unless otherwise noted. Sodium acetate (0.1 M), MES (0.03 M) and Tris (0.05 M) were used as a pH 7.0 \pm 0.2 buffer (Keenan and Sedlak, 2008) unless otherwise noted.

2.3. Analytical methods

At selected time intervals, 10 mL of reaction solution was filtered through 0.22 µm syringe filters and 4 g anhydrous sodium sulfate was added into the filtered sample. Then HAcAms were immediately extracted into 2 mL methyl tert-butyl ether (MTBE), and were analyzed using gas chromatography (GC) coupled with an electron capture detector (ECD) (QP 2010 plus, Shimadzu, Japan). Injections of 1.0 µL of the extract were introduced via a splitless injector onto a GC column (RTX-5, 30-m \times 0.25-mm ID, 0.25- μ m film thickness). The column oven was initially held at 40 °C for 10min, ramping to 200 °C at 20 °C/min and holding for 2 min. The injection port was controlled at 180 °C and the ECD detector temperature was 320 °C. Three HAAs (MCAA, DCAA and TCAA) were also analyzed by the GC/ ECD (QP 2010 plus, Shimadzu, Japan) according to US EPA method 552.2 (USEPA, 1995; Chu et al., 2014). The detection limits for TCAcAm, DCAcAm, MCAcAm, TCAA, DCAA, and MCAA were 0.01, 0.01, 0.05, 0.05, 0.05, and 0.07 μ M, respectively. Brominated HAcAms, were analyzed by combining high pressure liquid Download English Version:

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