



Removal of *N*-nitrosodimethylamine precursors with powdered activated carbon adsorption



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ABSTRACT

The main objective of this study was to examine the roles of powdered activated carbon (PAC) characteristics (i.e., surface chemistry, pore size distribution, and surface area) in the removal of *N*-nitrosodimethylamine (NDMA) formation potential (FP) in surface and wastewater-impacted waters. Also, the effects of natural attenuation of NDMA precursors in surface waters, NDMA FP concentration, and carbon dose on the removal of NDMA FP by PAC were evaluated. Finally, the removal of NDMA FP by PAC at two full-scale DWTPs was monitored. Wastewater-impacted and surface water samples were collected to conduct adsorption experiments using different PACs and activated carbon fibers (ACFs) with a wide range of physicochemical characteristics. The removal efficiency of NDMA FP by PAC was significantly higher in wastewater-impacted than surface waters. Adsorbable NDMA precursors showed a size distribution in the waters tested; the adsorbable fraction included precursors accessing the pore size regions of 10–20 Å and <10 Å. Basic carbons showed higher removal of NDMA FP than acidic carbons on a surface area basis. The overall removal of NDMA FP by PAC on a mass basis depended on the surface area, pore size distribution and pH_{PZC}. Thus, PACs with hybrid characteristics (micro and mesoporous), higher surface areas, and basic surface chemistry are more likely to be effective for NDMA precursor control by PAC adsorption. The application of PAC in DWTPs for taste and odor control resulted in an additional 20% removal of NDMA FP for the PAC doses of 7–10 mg/L. The natural attenuation of NDMA precursors through a combination of processes (biodegradation, photolysis and adsorption) decreased their adsorbability and removal by PAC adsorption.

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1. Introduction

Since 1990s, drinking water facilities in the United States (U.S.) have increased their use of chloramination to reduce the formation of regulated disinfection by-products (DBPs) trihalomethanes (THMs) and haloacetic acids (HAAs). One survey indicated that U.S. drinking water utilities increased their use of chloramination by 37% between 2007 and 2010, and approximately 68 million people in the U.S. were estimated to be consuming chloraminated water (Li, 2011). However, one unintended consequence of chloramination is the possibility of nitrosamine formation (Najm and Trussel, 2001; Choi and Valentine, 2002; Mitch et al., 2003; Mitch and Sedlak, 2004; Sedlak et al., 2005). *N*-nitrosodimethylamine (NDMA) has been the most frequently detected nitrosamine in

distribution systems in the U.S. (Russell et al., 2012), and is classified as probable human carcinogen with a 10⁻⁶ lifetime cancer risk associated with a drinking water concentration of 0.7 ng/L (U.S. EPA, 1993a). Although there are currently no U.S. Federal drinking water regulations for nitrosamines, the frequent detection of NDMA in distribution systems has prompted the Department of Health Service in California and the Massachusetts Department of Environmental Protection to implement an action level of 10 ng/L (MassDEP, 2004; OEHHA, 2006), and the Ontario Ministry of the Environment and Climate Change to establish a maximum allowable concentration of 9 ng/L for NDMA (MOE, 2003) in their drinking water supplies. Furthermore, the U.S. Environmental Protection Agency (EPA) has recently identified nitrosamines as one of three potential groups of contaminants highlighted for possible regulatory action (Roberson, 2011).

One approach for NDMA control is to remove its precursors from water before disinfecting with chloramines; therefore, different strategies have been investigated. Coagulation with ferric

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chloride and alum has shown low degree (<20%) of NDMA precursor removal (Krasner et al., 2008; Sacher et al., 2008; Mitch et al., 2009; Knight et al., 2012), while some of the cationic polymers and ion exchange resins have been reported to release NDMA precursors (Najm and Trussel, 2001; Kohut and Andrews, 2003; Wilczak et al., 2003; Gan et al., 2013). NDMA precursors and model precursor compounds were effectively removed (98–99%) with reverse osmosis (RO) treatment from different background waters (Mitch and Sedlak, 2004; Snyder et al., 2006; Miyashita et al., 2009; Krauss et al., 2010; Farré et al., 2011), while rejections higher than 98.5% of nitrosamine precursors were reported with nanofiltration (Miyashita et al., 2009). Powdered activated carbon (PAC) is widely used in drinking water treatment plants (DWTPs) mainly to remove taste and odor causing-compounds, as well as synthetic organic chemicals (SOCs). To date, only a small number of studies have examined the removal of NDMA precursors by PAC adsorption during water treatment. The reported NDMA formation potential (FP) removals were (i) >73% from an effluent organic matter (EfOM) with 50 mg/L of PAC (F400, Calgon Carbon Corp., Pittsburgh, PA) and 7 d of contact time (Krasner et al., 2008); (ii) 50% in river waters with 5 mg/L of PAC (F300, Calgon Carbon Corp., Pittsburgh, PA) and 7 d of contact time, $\geq 90\%$ with 20 mg/L of PAC (Sacher et al., 2008); and (iii) 40% in a secondary wastewater effluent with 3 mg/L of PAC (WPH, Calgon Carbon Corp., Pittsburgh, PA) and 4 h of contact time (Hanigan et al., 2012). A dose of 75 mg/L of WPH had similar removals (approximately 90%) in secondary wastewater effluents with different NDMA FP (544 and 1470 ng/L) (Hanigan et al., 2012). Recently, Hanigan et al. (2015) found similar removals (17–34%) of watershed-derived NDMA precursors by 3 mg/L of a bituminous-based PAC (WPH, Calgon Carbon Corp., Pittsburgh, PA) from blends of a treated secondary wastewater effluent and Colorado River water. Additionally, they reported that all PAC doses investigated (3–20 mg/L) with different contact times (15–120 min) decreased NDMA FP, but contact times longer than 15 min did not provide additional removal of watershed-derived precursors.

Although some studies have investigated the removal of NDMA precursors by PAC, the effects of the PAC characteristics on the removal of NDMA precursors have not been systematically examined. Therefore, the main objective of this study was to examine the roles of PAC surface chemistry, pore size distribution, and surface area in the removal of NDMA FP from surface and wastewater-impacted waters. Furthermore, the effects of natural attenuation of NDMA precursors in natural waters, NDMA FP concentration, and carbon dose on the removal of NDMA FP by PAC were evaluated. Finally, the removal of NDMA FP by PAC was monitored at two full-scale DWTPs.

2. Materials and methods

2.1. Water samples

Water samples were obtained from six different sources, and classified as surface waters (SW) or wastewater impacted (IW). Three surface waters (SW-1, SW-2, and SW-3) are used by DWTPs, and four IW samples were collected from: (i) 8 km downstream of WWTP-A (tertiary treatment and UV disinfection) in two different months, October (IW1-Oct) and January (IW1-Jan); (ii) the treated effluent of WWTP-B (conventional secondary activated sludge); and (iii) the treated effluent of WWTP-C (primary clarifier, trickling filters, rotating biological contactors, and sand filters). The IW2 and IW3 samples were prepared in the laboratory by making 1.5-fold dilution of the treated effluents of WWTP-B and WWTP-C, respectively, with the upstream water of each one of the WWTPs

discharges. Upon arrival to the laboratory, all samples were filtered with 0.2 μm pore size Whatman[®] Polycap 150 TC filters, and kept at 4 °C until experiments were conducted (usually a maximum of 2–3 days).

2.2. Adsorbents

Two acidic PACs with high surface areas (A1 and A2), and two basic PACs with lower surface areas (B1 and B2) were selected for the adsorption experiments at bench scale. In the full-scale plants, DWTP-A applied A2 and B3 (a basic PAC with low surface area), and DWTP-B applied A1 only (Table 1). A1, A2 and B3 PACs were more mesoporous, whereas the B1 and B2 carbons had a higher micropore volume. Four of the PACs (A1, A2, B2 and B3) are currently used for taste and odor control at different DWTPs, and B1 was available in our laboratory from previous research. The PACs with different physical and chemical properties were used to assess the role of carbon characteristics on the NDMA FP removal. All the carbons were sieved through a U.S. Mesh Sieve size No. 400 (37 μm diameter opening) prior to use. Additionally, two activated carbon fibers (ACFs) previously characterized in our research group (Karanfil et al., 2006) were also used to examine the pore size regions that NDMA precursors can access. ACF10 consists mostly of primary micropores, with 91% of pores less than 10 Å. ACF20H has 47% of its pore volume in pores less than 10 Å, and 50% volume in the 10–20 Å pore size range. Both fibers have a basic pH_{pzc} (pH of the point of zero charge) but ACF20H has twice the surface area of ACF10 (Table 1).

2.3. Laboratory adsorption experiments

A 30 mg/L dose of each PAC was added to amber borosilicate bottles and filled with 1 L of water sample adjusted to pH (6.0 ± 0.2) using 1 M HCl and/or 1 M NaOH. This pH was selected for this study because PAC is added to water prior to coagulation/flocculation at the full-scale water treatment plants monitored in this study, where coagulation pH has ranged between 5.3 and 6.4. The bottles were placed on a tumbler for 4 h, a contact time to mimic a PAC application. PAC was then separated from the water using 0.45 μm pre-washed membrane filters (Supor[®] 450, Pall). NDMA FP tests were carried out with the filtered samples. The PAC dose of 30 mg/L, a high value for practical applications, was intentionally selected for our laboratory experiments to magnify the removal of NDMA precursors and better evaluate the trends. NDMA FP tests were used to determine the NDMA precursor levels in the samples in the presence of excess amount of monochloramine. NDMA FP was conducted in 500-mL amber borosilicate bottles. The pH of the samples was maintained at 7.8 in the presence of 20 mM phosphate buffer solution. All of the samples were spiked with 100 mg/L of Cl_2 from a monochloramine stock solution and stored for five days. To prepare preformed monochloramine stock solution, the pH of sodium hypochlorite (NaOCl) and ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) was adjusted to 9.0 by addition of HCl and/or NaOH. Then, the NaOCl solution was added dropwise into the $(\text{NH}_4)_2\text{SO}_4$ solution to achieve a 4:1 (Cl_2 :N) weight ratio (Hong et al., 2007).

All samples were prepared and analyzed in duplicates; raw water samples in triplicates, then each extract was analyzed on GC/MS/MS with two injections. The analytical variability across duplicates had relative deviation of less than 15%.

2.4. Full-scale water treatment plant monitoring

Two DWTPs were monitored for 18 months for the removal of NDMA FP during 2012–2014 period. In both DWTPs, PAC was occasionally used for taste and odor control. Out of 18 months, PAC

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