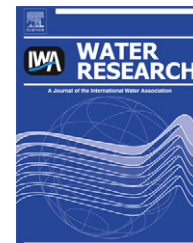


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The control of N-DBP and G-DBP precursors with MIEX[®]

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ABSTRACT

The objective of this study was to examine the potential of Magnetic Ion Exchange (MIEX[®]) technology for removing nitrogenous disinfection byproduct (N-DBP) precursors while minimizing carbonaceous DBP (C-DBP) precursors in (i) surface waters, and (ii) effluent impacted waters. Samples were collected from several drinking water source waters and wastewater treatment plant effluents. The effluent impacted source waters were simulated in the laboratory by mixing treated wastewater effluents with the same source water. Formation potential (FP) tests were conducted for regulated trihalomethanes (THMs), haloacetic acids (HAAs), and selected N-DBPs (nitrosamines and halonitromethanes (HNMs)) before and after the MIEX[®] treatment. The MIEX[®] process substantially lowered UV absorbance, total organic carbon, and THM and HAA FPs in all examined water samples, ranging from 39 to 87% reduction. A relatively small portion (9–33%) of HNM precursors was removed by the MIEX[®] treatment. On the other hand, an increase in N-nitrosodimethylamine (NDMA) FP was observed after the MIEX[®] process but only for the effluent impacted waters. Soluble metals, inorganic nitrogen, and bromide in effluent impacted waters did not correlate with the increase in NDMA FP. There was no effect of MIEX[®] treatment on the removal of other nitrosamine species precursors. Simulations of typical water treatment and distribution systems scenarios showed that NDMA concentrations remained below 10 ng/L, when chlorine alone or 40 min chlorine contact time prior to ammonia addition were employed for post-disinfection. However, when chlorine and ammonia were added simultaneously, NDMA concentration reached 36 ng/L for the water tested in the study.

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1. Introduction

Disinfection byproducts (DBPs) are formed as a result of reactions between organic matter in natural waters and oxidants used during drinking water treatment. To meet the stringent Stage 2 Disinfectants and Disinfection Byproduct (D/DBP) rule, some water utilities in the United States (US) have been switching from chlorine to alternative disinfectants to

lower the concentrations of regulated carbonaceous DBPs (C-DBPs: trihalomethanes (THMs) and haloacetic acids (HAAs)) in distribution systems. Although alternative disinfectants such as chloramine and ozone significantly reduce the formation of regulated THMs and HAAs, formation of some nitrogenous DBPs (N-DBPs) has been reported at higher levels than found with disinfection using chlorine. Chloramination increases the concentration of nitrosamines (Mitch et al., 2003; Chen

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and Valentine, 2006; Nawrocki and Andrzejewski, 2011). Increased concentrations of halonitromethanes (HNMs) have been observed during ozonation-chlorination as compared to chlorination alone (Hoigne and Bader, 1988; Krasner et al., 2006; Hu et al., 2010).

Research has shown that some of the unregulated N-DBPs exhibit orders of magnitude higher cyto- and geno-toxicity than the regulated C-DBPs (Wilbourn et al., 1999; Richardson et al., 2007). Nitrosamines are a group of compounds classified as probable human carcinogens in water with concentrations as low as 0.2 ng/L associated with a 10^{-6} lifetime cancer risk (USEPA, 1993). Although there are currently no federal regulations for nitrosamines in drinking water in the U.S., the Environmental Protection Agency (USEPA) has recently identified nitrosamines as one of three potential groups of contaminants slated for possible regulatory action in the near future (Roberson, 2011). USEPA included *N*-nitrosodimethylamine (NDMA), *N*-nitrosopyrrolidine (NPNR), *N*-nitrosodiethylamine (NDEA), *N*-nitrosomethylethylamine (NMEA), *N*-nitrosodi-*n*-propylamine (NDPA), and *N*-nitrosodi-*n*-butylamine (NDBA) in the Unregulated Contaminant Monitoring Rule 2 (UCMR 2) (USEPA, 2006), and NDMA, NDEA, NDPA, NPNR, and *N*-nitrosodiphenylamine (NDPhA) in the Contaminant Candidate List 3 (CCL 3) (USEPA, 2009). These trends suggest that US water utilities may need to comply with both C-DBP and N-DBP regulations simultaneously in the near future. Since the use of one or a combination of disinfectants/oxidants will not eliminate the formation of DBPs, applying treatment technologies maximizing the removal of DBP precursors from water before oxidant addition has been explored for DBP control.

The magnetic ion exchange (MIEX[®]) process has been increasingly used in recent years for DBP precursor control. It is applied as a pretreatment before oxidant addition to remove dissolved organic matter (or dissolved organic carbon, DOC), thus reducing the formation of DBPs. The smaller particle size, higher density, magnetic properties and simple regeneration features of MIEX[®] provide higher exchange efficiency and rapid clarification compared to traditional resins (Singer and Bilyk, 2002; Fearing et al., 2004; Boyer and Singer, 2005; Mergen et al., 2008). The MIEX[®] process has been demonstrated to be effective for THM and HAA control in the treatment of a wide range of waters (Singer and Bilyk, 2002; Drikas et al., 2003; Fearing et al., 2004; Boyer and Singer, 2005). Removal of UV₂₅₄ absorbance, DOC, THM and HAA precursors in a pilot study of four drinking waters with SUVA₂₅₄ ranging from 1.2 to 7.7 L/mg*m have been 55–88%, 35–67%, 38–77%, and 44–74%, respectively (Singer and Schneider, 2007). Research showed improved DOC removal and DBP reduction with inclusion of the MIEX[®] process prior to coagulation, and the advantage of the MIEX[®] process was more obvious in treating low SUVA waters (Drikas et al., 2003; Boyer and Singer, 2005). Preferential removal of DBP precursors over bulk DOC by MIEX[®] has also been reported (Singer and Bilyk, 2002; Drikas et al., 2003).

While the effectiveness of MIEX[®] for the removal of C-DBP precursors has been well established in many surface waters, understanding of its effectiveness for removing the precursors of the emerging N-DBPs is quite limited. Although MIEX[®] was capable of removing some dissolved organic nitrogen (DON) components (Boyer et al., 2008), understanding of its

effectiveness for removing the precursors of different classes of N-DBPs is limited. This is mainly due to the weak correlations between DON and different classes of N-DBPs (Chen and Westerhoff, 2010).

The main objective of this study was to examine the removal of NDMA and HNM precursors by MIEX[®] while removing THM and HAA precursors from waters. The MIEX[®] process was evaluated in (i) drinking waters (DW), and (ii) effluent impacted waters (EFIW). These sources represent two different types of N-DBP precursors. While natural organic matter (NOM) and sometimes algal organic matter are the major components of surface waters, wastewater effluents consist of soluble microbial products and the residual NOM remaining in water after drinking water treatment. Due to growing water demand and drought conditions, some surface water sources are increasingly impacted by the upstream wastewater treatment plant discharges. Although several studies have been conducted for the MIEX[®] process on surface waters (Mergen et al., 2008; Singer et al., 2009), its performance on effluent impacted waters has not been well documented.

2. Materials and methods

2.1. Water samples

In this study, water samples were obtained from six different sources in South Carolina (USA): three source water samples were collected from the inlets of drinking water treatment plants (DWTPs) located in Charleston, Lyman, and Myrtle Beach, and three treated effluents (i.e., prior to disinfection) were obtained from municipal wastewater treatment plants (WWTPs) located in Anderson, Greenville, and Pendleton. WWTP A process train consists of a primary clarifier, trickling filters, rotating biological contactors, and sand filters. WWTP B is a conventional secondary activated sludge plant. WWTP C has a biological nutrient removal activated sludge process with nitrification and denitrification units, and deep sand filters. All samples were immediately filtered through 0.2 μm membrane filters upon arrival at the laboratory, and they were characterized for DOC, UV₂₅₄ absorbance, dissolved nitrogen (DN), pH, ammonia, nitrate, nitrite, bromide, and sulfate. Samples were stored at 4 °C in the dark until use. The EFIW samples were prepared in the laboratory by mixing treated effluents with the same drinking water (i.e., DW 1 in Table 1) at the ratio of 1:1.

2.2. MIEX[®] treatment

MIEX[®] resin was obtained from Orica Watercare. Before an experiment, the resin was placed in a fluidized bed column and rinsed with 600 bed volumes of distilled and deionized water (DDI) for cleaning. For selected water samples, kinetic experiments were initially conducted to determine the optimal treatment conditions. A range of resin doses (5–20 mL/L) were tested based on the DOC concentration of the raw water. One liter of raw water was mixed with MIEX[®] at 100 rpm in a Phipps & Bird jar, and samples were withdrawn from each jar at 5, 10, 20, 30 and 60 min, and filtered through pre-rinsed 0.45 μm membrane filter for UV₂₅₄ and DOC analysis. Given that the majority of removal occurred in the first

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