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Formation of disinfection by-products after pre-oxidation with chlorine dioxide or ferrate





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ABSTRACT

The effect of pre-oxidation with chlorine dioxide (ClO₂) or ferrate (Fe(VI)) on the formation of disinfection by-products (DBPs) during chlorination or chloramination was tested with natural waters from 12 sources (9 surface waters, 1 groundwater, and 2 wastewater effluents). DBPs investigated included trihalomethanes (THM), chloral hydrate (CH), haloketones (HK), haloacetonitriles (HAN) and trichloronitromethane (TCNM), chlorite and chlorate. Chlorite and chlorate were found in the ClO₂-treated waters. Application of 1 mg/L ClO₂ ahead of chlorination reduced the formation potential for THM by up to 45% and the formation of HK, HAN and TCNM in most of the samples. The CH formation results were mixed. The formation of CH and HK was enhanced with low doses of Fe(VI) (1 mg/L as Fe), but was greatly reduced at higher doses (20 mg/L Fe). Fe(VI) reduced the formation of NDMA was observed in most of the samples after both ClO₂ and Fe(VI) pre-oxidation.

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1. Introduction

In water treatment, pre-oxidation is carried out for a variety of purposes including taste and odor control, algae removal, oxidation of manganese and iron, improving coagulation performance and disinfection. The pre-oxidizing chemicals often applied include chlorine, ozone, permanganate, and chlorine dioxide (ClO₂). Ferrate(VI) is another emerging water treatment chemical due to its dual functions as an oxidant and then a coagulant/precipitant as ferric hydroxide (Sharma,

2002; Filip et al., 2011; Prucek et al., 2013). ClO_2 is getting more uses in water treatment plants in China (Huang and Wu, 2010). Ferrate has the potential to be applied in water treatment due to the reduced cost (Alsheyab et al., 2009). Therefore, ClO_2 and ferrate are the target oxidants in this study.

Pre-oxidants can oxidize dissolved organic matter (DOM), which is ubiquitous in natural water and is a precursor of disinfection by-products (DBPs). Chlorine dioxide destroys the aromatic and conjugated structures of DOM and transforms large aromatic and long aliphatic chain organics to small and

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hydrophilic organics (Yang et al., 2013). Oxidation of DOM with ClO₂ also generates organic by-products such as aldehydes and long chain carboxylic acids (Swietlik et al., 2004). Ferrate oxidation enhances the levels of assimilable organic carbon and ferrate can partially oxidize DOM fluorophores (Ramseier et al., 2011; Horst et al., 2013). The changes in DOM properties then modify DBP formation during subsequent disinfection processes.

Trihalomethane (THMs) and haloacetic acid (HAAs) levels in drinking water are regulated in many countries around the world. The DBPs that have been observed in chlorinated water include haloacetonitriles (HANs), halonitromethanes (HNMs), haloketones (HKs) and chloral hydrate (CH) (Krasner et al., 2006; Bougeard et al., 2010). Nitrosamines such as N-nitrosodimethylamine (NDMA) are formed when monochloramine is used for disinfection (Mitch and Sedlak, 2001). Chlorine dioxide, however, does not produce significant amounts of halogenated organic DBPs (Hua and Reckhow, 2007). Chlorite and chlorate are the major products (Korn et al., 2002). Ferrate(VI) is also used for disinfection (Sharma et al., 2005). It does not form chlorinated byproducts, and its byproduct Fe(III) is non-toxic and can enhance coagulation processes (Jiang and Lloyd, 2002; Sharma, 2002).

DBP formation during ClO₂ pretreatment has been studied (Linder et al., 2006; Yang et al., 2013; Lee et al., 2007). It has been shown that ClO₂ oxidation prior to chlorination can reduce the formation of THM and total organic halogen (TOX) (Linder et al., 2006). A recent study has reported that ClO₂ preoxidation reduced THM, HAA, HAN and CH formation during subsequent chlorination (Yang et al., 2013). ClO₂ pre-oxidation reduces the NDMA formation potential (FP) of natural water sources by 32-94% (Lee et al., 2007). But enhanced NDMA formation after ClO₂ pre-oxidation has also been reported in wastewater-impacted waters (Shah et al., 2012). DBP formation after pre-oxidation with Fe(VI) has been reported in only a few studies. Pre-oxidation of several natural waters with a high dose of Fe(VI) (21 mg/L as Fe) has been reported to reduce NDMA's FP by 46-84% (Lee et al., 2008). Fe(VI) reduced UV absorbance and the carbon concentrations of DOM (Jiang and Lloyd, 2002). Fe(VI) reacts with amino acids, leading to the formation of aldehydes (Noorhasan et al., 2010; Sharma, 2010). Amino acids are precursors of HANs and aldehydes, generating HKs during chlorination (Yang et al., 2012). Tryptophan oxidation by ferrate led to the formation of intermediates including N-formylkynurenine, 4-hydroxyquinoline and kynurenic acid (Casbeer et al., 2013). Ferrate pre-oxidation may therefore likely affect DBP formation although the evidence to support its effect remains unclear.

The objective of this study was to investigate the formation of regulated and other DBPs during chlorination and chloramination after ClO_2 and Fe(VI) pre-oxidation. Water from 12 sources, including 9 surface waters, 1 groundwater, and 2 wastewater effluents were collected, characterized and their DBP formation potential was studied. Formation potential tests were applied since the results also reflect the amount of precursors. The surface waters and groundwater were the source water for 10 water treatment plants and most of them were located at different rivers or streams. The water characteristics varied regarding to dissolved organic carbon (DOC), dissolved organic nitrogen (DON) and bromide concentrations, which will significantly affect DBP formation during disinfection. The DBPs studied were four THMs (chloroform (CHCl₃), bromodichloromethane (CHCl₂Br), dibromochloromethane (CHClBr₂) and bromoform (CHBr₃)), four HANs (trichloroacetonitrile (TCAN), dichloroacetonitrile (DCAN), bromochloroacetonitrile (BCAN) and dibromoacetonitrile (DBAN)), two HKs (1,1dichloropropanone (1,1-DCP) and 1,1,1-trichloropropanone (1,1,1-TCP)), CH, trichloronitromethane (TCNM), and NDMA.

2. Materials and methods

2.1. Reagents and solutions

Calibration standards, internal standards and surrogate standards for the THM, CH, HK, HAN and TCNM analyses were purchased from Supelco (USA). NDMA was purchased from Supelco, and d₆-NDMA was purchased from Cambridge Isotope Laboratories (USA). HPLC-grade formic acid, acetonitrile, methanol, dichloromethane, DMA and n-hexamine were purchased from J&K (China). Benzenesulfonyl chloride was purchased from Sigma (USA). Resprep EPA Method 521 cartridges (6 mL/2 g) for NDMA extraction were purchased from Restek (USA).

A stock solution of free chlorine (HOCl) was prepared by diluting 5% sodium hypochlorite (NaOCl) (Sigma) to 1000 mg/L as Cl_2 . Monochloramine (NH₂Cl) solutions were prepared daily by reacting equal volumes of ammonium chloride and sodium hypochlorite solutions at a weight ratio of 4 mg/L Cl_2 to 1 mg/L $N-NH_4^+$. The resulting solutions were standardized by diethylp-phenylene diamine (DPD) ferrous titration (APHA et al., 1998). A solution of ClO_2 was prepared from gaseous ClO_2 by slowly adding dilute H_2SO_4 to a sodium chlorite (NaClO₂) solution according to the standard method (APHA et al., 1998).

The Fe(VI) was prepared in the laboratory as solid potassium ferrate (K₂FeO₄) with high purity (99%) by a previously optimized method based on the oxidation of ferric nitrate with hypochlorite (Li et al., 2005). Ferrate(VI) working solutions (1 g/L as Fe) were prepared by adding solid K₂FeO₄ to 2 M NaOH solution. They were then used within 30 min. A molar absorption coefficient of $\varepsilon_{505nm} = 1150 \text{ M}^{-1} \text{ cm}^{-1}$ was used to determine the ferrate(VI) concentration at pH 9.0 (Luo et al., 2011).

2.2. Sample collection

Water samples from 12 sources were collected, including 9 surface water, 1 groundwater, and 2 secondary wastewater effluent sources. The 10 surface water and groundwater sources were the water sources for 10 drinking water treatment plants. All the samples were filtered through 0.45- μ m membranes to remove any particulates. The concentrations of DOC, total nitrogen, ammonia, nitrite, nitrate and bromide, and the UV absorbance at 254 nm (UV₂₅₄) were measured. Specific UV absorbance (SUVA) was calculated as UV₂₅₄ divided by the DOC. The water properties are summarized in Table 1.

2.3. DBP formation potential

The DBP formation potential experiments were conduced in capped amber glass bottles at room temperature (22 \pm 1 °C) in the dark. Chlorine was dosed to evaluate the formation

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