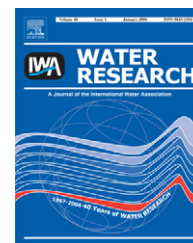


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The effect of ozonation on natural organic matter removal by alum coagulation

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ABSTRACT

Natural organic matter (NOM) was extracted from a moderately colored, eutrophic surface water source (Forge Pond, Granby, MA), and fractionated into quasi-homogeneous fractions. Fulvic acid (FA) and hydrophilic neutrals (HN) were the two most abundant NOM fractions that were isolated. Adsorption affinity of the isolated NOM fractions on preformed aluminum hydroxide flocs increased with increase in specific organic charge of the fractions, except for the two most highly charged fractions, FA and hydrophilic acids (HAA), which showed less adsorption affinity than expected based on their specific organic charge. Prior ozonation of FA and HN fractions resulted in a decline and an increase, respectively, in their adsorption affinity on aluminum hydroxide surface. Prior ozonation of Forge Pond raw water resulted in a progressive decline in dissolved organic carbon (DOC) removal by alum coagulation with increase in ozone dose. It appeared that ozone applied to raw water reacted preferentially with the humic fraction of NOM, resulting in the detrimental effects of ozonation on subsequent NOM removal by alum coagulation being magnified. Forge Pond raw water was pre-coagulated to remove humic substances. Ozonation of the pre-coagulated water demonstrated the beneficial effects of ozonation on the removal of non-humic NOM through alum coagulation. A strategy for staged coagulation with intermediate ozonation was proposed for waters containing both humic and non-humic NOM for maximum DOC and specific UV absorbance at 254 nm (SUVA) removal.

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1. Introduction

Concerns about disinfection by-products (DBPs) in potable water supplies led the US Environmental Protection Agency (USEPA) to promulgate the Stage 1 Disinfectants and Disinfection Byproducts Rule (Federal Register, 2001), which has set a maximum contaminant level (MCL) of $80 \mu\text{g L}^{-1}$ for Trihalo-methanes (THM) and $60 \mu\text{g L}^{-1}$ for five haloacetic acids (HAA5). Attainment of the above DBP levels in treated water supplies requires removal of DBP precursors, i.e., natural organic matter (NOM) present in water.

The suggested method for NOM removal during water treatment is enhanced coagulation or enhanced softening. Enhanced coagulation seeks to maximize dissolved organic carbon (DOC) removal by providing an excess of metal hydroxide precipitate surface for dissolved NOM adsorption during water treatment. Numerous literature reports suggest that enhanced coagulation can increase DBP precursor removals (Edzwald and Van Benschoten, 1990; Childress et al., 1999; Liu et al., 2005; Uyak and Toroz, 2005; Rizzo et al., 2005; Vrijenhoek et al., 1998; Freese et al., 2001; Cheng et al., 2003; Wang et al., 2002; Braul et al., 2001; Volk et al.,

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2000). In this context, Budd et al. (2004) reviewed the changes in the conventional coagulation process necessary for a switch-over to enhanced coagulation, including changes in pH and coagulant dose, use of alternative coagulant chemicals, and changes in the order of chemical addition. Considering the weight of empirical evidence concerning the effectiveness of enhanced coagulation, better mechanistic understanding of this process is desirable. For this purpose, the relevant properties of NOM must be elucidated and correlated with removal efficiencies observed during the enhanced coagulation process.

NOM is a conglomeration of diverse compounds with very different chemical properties. Krasner et al. (1996) reported that there are three approaches to NOM characterization; a non-perturbing approach, an approach based on isolation fractionation, and an approach based on calculations. Most NOM characterization studies seeking to relate NOM properties, with its removal by enhanced coagulation process, involve fractionation of NOM into several semi-homogeneous compound classes. A comprehensive method for NOM fractionation using the isolation-fractionation approach is the procedure outlined by Leenheer and Noyes (1984). By this procedure, NOM can be fractionated into eight compound classes: fulvic acid (FA), humic acid (HA), weak hydrophobic acids (WHYA), hydrophilic acids (HAA), hydrophobic neutrals (HYN), hydrophilic neutrals (HNs), hydrophobic bases (HYB), and hydrophilic bases (HB). Other NOM isolation-fractionation studies include those by Wong et al. (2002), MartinMouset et al. (1997), Marhaba and Van (1999), Marhaba et al. (2003), and Egeberg and Alberts (2003). Many researchers have carried out studies on removal of isolated NOM fractions by coagulation, based on which conclusions were drawn regarding NOM removal from natural waters by enhanced coagulation. Edzwald (1993) reported that a water rich in humic substances will probably show a DOC removal of 70% or more, while a water dominated by non-humic organics will show considerably lower removals. Fearing et al. (2004) investigated removal of NOM by conventional coagulation treatment using both bulk and fractionated NOM, and reported over 70% removal of the hydrophobic and HAA fractions, while only 16% of the hydrophilic non-acid fraction was removed. Kim and Yu (2005) reported that the hydrophobic fraction was removed more than the hydrophilic fraction through conventional water treatment. Chow et al. (2004, 2005) reported that HN fraction of NOM was hardly removed by alum treatment, while very hydrophobic acids, (VHAs) fraction of NOM was easily removed by alum treatment. Matilainen et al. (2005) observed that approximately 95% of high molar mass organic substances were removed in the coagulation process with both ferric and aluminum salts, while on average, only 10% of NOM with molar mass less than 1000 g mol^{-1} was removed.

Ozone has been used in water treatment for a variety of purposes. Its oxidative properties have been used for disinfection, iron and manganese oxidation, bleaching color, reducing taste and odor compounds, and controlling DBP formation (Langlais et al., 1991). However, results of empirical studies on the effect of ozone on NOM removal by enhanced coagulation are mixed and site specific (Reckhow and Singer, 1984; Farvardin and Collins, 1989; Colin et al., 1986; Singer et al., 2003; Marhaba and Pipada, 2000). In order to understand and

apply these findings in a generalized manner, fundamental studies must be carried out to ascertain the effects of ozone on isolated NOM fractions, and the impact of the consequent changes in NOM properties on its subsequent interaction with metal hydroxide surface. Such studies will determine applicability of and aid in optimization of the ozonation process prior to NOM removal by enhanced coagulation.

This study involved fractionation of NOM from a moderately eutrophic natural water source (Forge Pond, Granby, MA) into various fractions. The main objectives were: (1) determination of relevant properties of the isolated NOM fractions; (2) relating properties of the isolated NOM fractions with adsorption on aluminum hydroxide surface; (3) determination of the effect of ozonation on NOM fraction properties and, hence, on adsorption of ozonated NOM fractions on aluminum hydroxide surface; (4) effect of ozonation on NOM removal from Forge Pond raw water by enhanced coagulation; and (5) determination of a strategy for optimizing NOM removal from ozonated Forge Pond raw water.

2. Materials and methods

2.1. Experimental materials

Forge Pond water was moderately colored, eutrophic, and had moderately high NOM concentration. NOM in this water was fractionated into eight quasi-homogeneous compound classes; FA, HA, HAA, WHYA, HNs, HYNs, HBs, and HYBs. The extraction procedure used was described by Leenheer and Noyes (1984) and involved passing the water in series through three columns containing hydrophobic (XAD-8), cation (MSC-1), and anion exchange (Duolite A-7) resins, respectively. Various NOM fractions in water were adsorbed on the resins, except for the HN fraction, which passed through all three columns and was thus isolated in the column effluent. The adsorbed NOM fractions were isolated later by desorption from the resins under various conditions (see Bose, 1994 for details). Detailed NOM fractionation results are presented in Table 1. Humic materials contributed to 44.7% of NOM in this water (7.1% HA and 37.6% FA), while 22.2% were HNs.

2.2. Experimental methods

Some NOM fractions (FA, HA, and HN) were ozonated for the purpose of this study. Ozonated organic solutions were prepared in batches by adding ozone solution prepared in Milli-Q water at pH 3 to aliquots of various NOM fractions (see Bose et al., 1994 for details). Specific organic charge of the isolated NOM fractions were determined at various pH values. Specific organic charge of some of the fractions was also determined after prior ozonation (see Bose and Reckhow, 1998 for details).

Adsorption experiments involved contacting preformed aluminum hydroxide flocs with NOM aliquots. For preparation of preformed flocs, alum (from a $10,000 \text{ mg L}^{-1}$ stock solution at low pH) was added to buffered solution of deionized water at pH 7. The water was rapid mixed for 2 min and then slow mixed for 30 min using a jar test apparatus. Flocs formed after 30 min of slow mixing were

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