



# Applying the polarity rapid assessment method to characterize nitrosamine precursors and to understand their removal by drinking water treatment processes



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## ABSTRACT

Some *N*-nitrosamines (NAs) have been identified as emerging disinfection by-products during water treatment. Thus, it is essential to understand the characteristics of the NA precursors. In this study, the polarity rapid assessment method (PRAM) and the classical resin fractionation method were studied as methods to fractionate the NA precursors during drinking water treatment. The results showed that PRAM has much higher selectivity for NA precursors than the resin approach. The normalized *N*-nitrosodimethylamine formation potential (NDMA FP) and *N*-nitrosodiethylamine (NDEA) FP of four resin fractions was at the same level as the average yield of the bulk organic matter whereas that of the cationic fraction by PRAM showed 50 times the average. Thus, the cationic fraction was shown to be the most important NDMA precursor contributor. The PRAM method also helped understand which portions of the NA precursor were removed by different water treatment processes. Activated carbon (AC) adsorption removed over 90% of the non-polar PRAM fraction (that sorbs onto the C18 solid phase extraction [SPE] cartridge) of NDMA and NDEA precursors. Bio-treatment removed 80–90% of the cationic fraction of PRAM (that is retained on the cation exchange SPE cartridge) and 40–60% of the non-cationic fractions. Ozonation removed 50–60% of the non-polar PRAM fraction of NA precursors and transformed part of them into the polar fraction. Coagulation and sedimentation had very limited removal of various PRAM fractions of NA precursors.

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## 1. Introduction

Some *N*-nitrosoamines (NAs) have been identified as emerging disinfection by-products (DBPs) in drinking water (Krasner et al., 2013). NAs present much higher carcinogenicity than the traditional carbonaceous DBPs, such as trihalomethanes (THMs) and haloacetic acids (HAAs) (Richardson et al., 2007; Wagner et al., 2012). The *N*-nitrosodimethylamine (NDMA) regulatory and notification levels in Ontario, Canada is 9 ng/L, whereas in Massachusetts and California, U.S.A it is 10 ng/L. A low regulatory level presents major challenges for drinking water treatment plants,

especially for those with high concentration of NA precursors (Kristiana et al., 2012). Thus, it is essential to understand the characteristics of the NA precursors to develop and optimize water treatment removal processes.

Many studies have been conducted over the last decade to identify the NA precursors. The studies showed that NA precursors have various sources, including secondary amines (Choi and Valentine, 2002; Mitch et al., 2003; Mitch and Sedlak, 2002; Wang et al., 2011), certain water treatment polymers (Krasner et al., 2013; Padhye et al., 2011), certain pharmaceutical and personal care products (PPCPs) (Shen and Andrews, 2011), certain pesticides or their degradation products (Le Roux et al., 2011; Schmidt and Brauch, 2008), wastewater treatment plant effluent organic matter (EfOM) (Mitch and Sedlak, 2004; Wang et al., 2014), certain industrial chemicals (Kosaka et al., 2010; Wang et al., 2014),

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algal organic matter (Li et al., 2012) and natural organic matter (NOM) (Chen and Valentine, 2007; Gerecke and Sedlak, 2003). A general conclusion can be drawn from these investigations that NA precursors generally have the dialkylamine, tertiary amine or quaternary amine functional groups (Krasner et al., 2013; Chen et al., 2014). The long list of known NA precursor chemicals was site-specific and there were other NA precursors present that were not identified.

Thus, there is a great demand for efficient, highly selective methods to isolate, fractionate, and characterize the NA precursors because it is impossible for researchers to identify each kind of NA precursors from varying water sources. Formation potential (FP) tests are widely applied to determine the maximum amount of NA yield under the condition of sufficient nitrosation agents (e.g. monochloramine) and reaction time (Mitch et al., 2003). However, this method cannot provide information on the NA precursors' characteristic besides the amount.

The resin fractionation method has been widely applied to study NOM in water studies (Leenheer, 1981; Singer et al., 2007; Thurman and Malcolm, 1981). The method extracts humic and non-humic substances in series on different resins at basic pH and acid pH. Many liters of sample were used. According to the literature, hydrophilic (HPI) organic matter contributed more to nitrosamine precursors (Chen and Valentine, 2007; Wang et al., 2013; Doston et al., 2009) whereas hydrophobic (HPO) fraction contributed more to THM and HAA precursors (Chen et al., 2011, 2008). Analysis reproducibility was not evaluated because of the many liters of samples used and the requirement for extensive cleaning of the resin between samples.

A low-volume sample method and selective tool to characterize NOM in drinking water (Rosario-Ortiz et al., 2007) and subsequently NDMA precursors in wastewater (Chen et al., 2014) was developed, as referred to as the Polarity Rapid Assessment Method (PRAM). This method used 100 mL to liter samples. This method used solid-phase extraction (SPE) cartridges in parallel, where the strong cation exchange (SCX) and non-polar C<sub>18</sub> SPE cartridges isolated and fractionated the NA precursors at ambient pH and not under acidic or basic conditions as used in the resin fractionation method. A SCX SPE cartridge isolated the amine-type functional group of NDMA precursors and the C<sub>18</sub> SPE cartridge sorbed their non-polar part. Quality assurance was easily conducted during the PRAM operation with the surrogate indicators of dissolved organic carbon (DOC) and ultraviolet absorbance at 254 nm (UV<sub>254</sub>), and less than 5% relative standard deviation (RSD) was observed for replicate SPE cartridges and for the re-loading of water samples.

The objective of this study was to compare the use of PRAM with the classical resin fractionation method to understand the characteristics of the NA precursors. Then the chosen fractionation method was applied to help understanding NA precursor removal by different drinking water treatment processes, including coagulation and sedimentation, ozonation, activated carbon (AC) adsorption, and bio-treatment.

## 2. Materials and methods

### 2.1. Raw water quality

Water samples were taken from one aquaculture-impacted lake water in the Yangtze River Delta, China. The water quality of the raw water for this investigation is shown in Table 1. RSDs were determined by triplicate tests of the samples. The specific UV absorbance (SUVA) was low, which indicated that the water was low in humic substances. The DOC/DON ratio was less than 10 mg C/mg N, indicating that the water was nitrogen-rich (the average DOC/DON ratio in a U.S. survey was 18 mg C/mg N [Lee et al., 2006]).

### 2.2. PRAM method

The PRAM procedure included a syringe pump (Longer Pump Corp., Baoding, China), glass syringes, SPE cartridges, connecting tips, and 40-mL vials as filtrate collectors. Two SPE cartridges types were used for PRAM measurements; the C<sub>18</sub> and SCX cartridges. The cartridges (Extract Clean SPE kit catalog #210100) were purchased from Alltech Associates (Deerfield, IL, U.S.A.).

The matrix of the C<sub>18</sub> cartridge was octadecyl-silyl. The organic matter adsorbed by the C<sub>18</sub> cartridge was defined as the non-polar fraction and the filtrate was defined as the polar fraction. The matrix of the SCX cartridge was benzenesulfonic acid-silyl. The organic matter adsorbed by the SCX cartridge was defined as the cationic fraction and the filtrate was defined as the non-cationic fraction. In PRAM, the quantitative difference in organic matter concentrations between the influent and effluent of the cartridges was determined. More details of the PRAM operation were presented by Rosario-Ortiz et al. (2007) and Chen et al. (2014), which are discussed in the Supplementary Information (SI). A UV–Vis photometer (UV 2700, Shimadzu Corp., Kyoto, Japan) was used to monitor the UV absorbance of the filtrate in real time as a quality assurance procedure.

### 2.3. Resin fractionation method

A modified resin fractionation method was applied to separate the bulk organic matter into the HPO and HPI fractions referring to previous studies (Chen et al., 2008; Leenheer, 1981). According to the literature, the organic matter adsorbed by the XAD-8 resin (with a matrix of acrylic ester) was defined as the HPO NOM and the filtrate was defined as the HPI fraction. The HPO fraction was further divided into hydrophobic acid (HPOA), hydrophobic base (HPOB), and hydrophobic neutral (HPON) NOM by 0.1 M hydrochloric acid, 0.1 M sodium hydroxide or methanol elution, respectively. The HPI fraction was not further fractionated into acid, base, and neutral sub-fractions due to the fairly limited organic matter within this fraction in this water.

### 2.4. Tests of different unit processes

#### 2.4.1. Coagulation and sedimentation

A jar tester (ZR4-6, Zhongrun Corp, Shenzhen, China) was used for the coagulation and sedimentation test. An alum coagulant (20 mg/L as Al<sub>2</sub>O<sub>3</sub>) was used. The jar tester was set at the speed of 200 rpm for 1 min (rapid mix); 60 rpm for 6 min, 45 rpm for 6 min, and 20 rpm for 6 min (three stages of flocculation), which are conditions generally used in China (Lin et al., 2015; Yan et al., 2006). Then, the jar tester was stopped and sedimentation was allowed to occur for 20 min. The supernatant was further filtered with a 0.45- $\mu$ m membrane filter to remove the tiny flocs and particles. The residual concentration of NAFB and other water quality parameters were determined.

#### 2.4.2. Ozonation

An ozone generator (Model: CF-G-2-10G, Guolin Corp., Qingdao, China) with pure oxygen as the gas source was used in this study. The generated ozone was introduced into glass reactors filled with 2 L of test water. The ozone concentrations of input gas and output gas were determined by a portable ozone analyzer (PCII 58700-04, HACH Corp., U.S.A.). The flux of ozonated gas was carefully adjusted to get the ozone dose to 1 mg/L in the test waters, which is the common dose in the drinking water industry in China.

#### 2.4.3. AC adsorption

For adsorption tests, a powdered activated carbon (PAC)

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