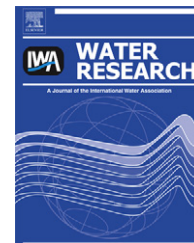


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Predicting disinfection by-product formation potential in water

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ABSTRACT

Formation of regulated and non-regulated disinfection by-products (DBPs) is an issue at both potable water and wastewater treatment plants (W/WWTs). Water samples from W/WWTs across the USA were collected and DBP formation potentials (DBFPs) in the presence of free chlorine and chloramine were obtained for trihalomethane (THM), haloacetic acid (HAA), haloacetonitrile (HAN), and N-nitrosodimethylamine (NDMA). With nearly 200 samples covering a range of dissolved organic carbon (0.6–23 mg/L), ultraviolet absorbance ($0.01\text{--}0.48\text{ cm}^{-1}$ at 254 nm wavelength), and bromide (0–1.0 mg/L) levels, power function models were developed to predict the carbonaceous DBP (C-DBP) and nitrogenous DBP (N-DBP) precursors spanning 3 orders of magnitudes. The predicted THM and HAA formation potentials fitted well with the measured data (analytical variance of less than 22%). Inclusion of dissolved organic nitrogen (DON) into the HANFP model improved the predictions. NDMAFP was the most difficult one to predict based upon the selected water quality parameters, perhaps suggesting that bulk measurements such as DOC or UVA₂₅₄ were not appropriate for tracking NDMAFP. These are the first such DBFP models for wastewater systems, and among the few models that consider both C-DBPs and N-DBPs formation potentials from the same water sources.

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1. Introduction

Disinfection via chlorination or chloramination has been critical processes for controlling pathogens in both potable water and treated wastewaters. However, both chlorination and chloramination form a variety of carcinogenic disinfection by-products (DBPs) of health and ecosystem concerns. The United States Environmental Protection Agency (USEPA) currently regulates two classes of carbonaceous DBPs (C-DBPs), trihalomethanes (THMs) and haloacetic acids (HAAs), under the Stage 2 Disinfection and DBP Rule. Nitrogenous DBPs (N-DBPs) are of emerging concern because N-nitrosodimethylamine (NDMA) and other N-DBPs may be more

toxic than C-DBPs (Plewa et al., 2004; Mitch et al., 2003; Richardson et al., 2008). The USEPA's Integrated Risk Information System (IRIS) database indicates that low ng/L levels of NDMA and seven other nitrosamines in water are associated with a 10^{-6} lifetime cancer risk. Because dissolved organic carbon (DOC), dissolved organic nitrogen (DON), and ammonia concentrations are higher at wastewater treatment plants (WWTs) than at drinking water treatment plants (DWTPs), increased concern over N-DBP formation has arisen.

DBP precursors are defined as a mixture of organic and inorganic compounds that can form DBPs under some level of disinfection. DBP formation potential (DBFPs) experiments are designed to maximize reactions between the precursors

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and disinfectant (e.g., Krasner et al., 2008). With knowledge of trends between precursors and DBPFP, unit processes capable of reducing more important precursors can be employed to reduce overall DBP levels in treated water. Extensive characterization of organic matter using resins and/or spectroscopy concluded that the hydrophobic fraction of organic matter, or aromatic organic matter, produced more THM and HAA than other organic fractions (Kitis et al., 2002). Ultraviolet absorbance at 254 nm wavelength (UVA_{254}), a parameter of aromatic and/or hydrophobic matter, was related with THM and HAA formation (Hwang et al., 2002). The hydrophilic bases were associated with haloacetonitrile (HAN) formation (Westerhoff and Mash, 2002). The nitrogen-enriched transphilic fractions of natural organic matter (NOM) were associated with the highest NDMA formation yield (up to 27 ng-NDMA/mg-DOC) (Luo et al., 2005). In addition, bromide ion was found to be an adjuster of DBPs speciation for THM and HAA (Liang and Singer, 2003).

Power function models developed for many C-DBPs have emerged as valuable tools for assessing means of limiting DBP formation through improved control of the precursors in DWTPs (Harrington et al., 1992; Solarik et al., 2000; Sohn et al., 2001; Sadiq and Rodriguez, 2004). While many of these models take into account both precursors (DOC, bromide) and disinfection conditions (dose, reaction time, pH, temperature, etc) as the modeling parameters, an important first step in the development of such models is to identify the trends among the key precursors. These experimental conditions can be held constant when performing formation potential (FP) tests to streamline the precursors. However, such DBP precursor models have not been developed for WWTP samples, and comparisons of DBPFP models across different water sources (e.g., DWTP versus WWTP) also do not exist. This paper hence was intended to develop models that correlate the suspected precursors and DBPFP.

The purpose of this paper was to evaluate a mathematical approach to predict four types of DBP precursors in five different water sources. The method employed a multivariable power equation (Equation (1)) based on three water quality parameters: DOC, UVA_{254} , and bromide (Equation (1A)). DOC represented the relative amount of precursor material, UVA_{254} represented the precursors' relative reactivity toward chlorine-based disinfectants, and bromide represented a control for the distribution among chlorinated and brominated species. Bromide was expressed as $Br + 1$ to avoid nil DBPFP estimation when bromide was absent. In addition, nitrogenous species (ammonia, nitrite, nitrate, and DON) were incorporated into the models of nitrogenous DBP precursors (Equation (1B)).

$$DBPFP = a \times DOC^b \times UVA_{254}^c \times (Br + 1)^d \quad (1A)$$

$$DBPFP = a \times DOC^b \times UVA_{254}^c \times (Br + 1)^d (N + 1)^e \quad (1B)$$

2. Materials and methods

2.1. Sampling campaign

Based upon a prior USA nationwide survey from 2004 to 2005, 168 water samples were collected for four consecutive seasons

within two years (Krasner et al., 2008), and the DBPFPs results were employed herein for calibration of models (Equation (1)). THM, HAA, and HAN precursors were analyzed by chlorination FP tests; NDMA precursors were analyzed by chloramination FP tests. The DBPFP experiments were conducted under disinfectant dosage in excess, long reaction time (24 h for chlorination and 72 h for chloramination tests), pH \sim 8.2, and 25 °C. During chlorination test, the chlorine dose was added based on the DOC and ammonia demand plus 10 mg/L extra chlorine: $3 \times DOC + 8 \times NH_3-N + 10$, in weight basis (Krasner et al., 2004). For chloramination test, chlorine was added to high-ammonia waters according to the level of DOC (i.e., $3 \times DOC$, in weight basis); but for low-ammonia ($NH_3-N < 5$ mg/L) waters in chloramination test, certain amount of ammonia was spiked to reach a chlorine-to-nitrogen ratio (Cl_2/N) $< 4:1$ in weight ratio. Chlorine residuals were quenched by ammonia chloride, and chloramine residuals were quenched by sodium sulfite. The details of DBPFP experimental conditions, DBP analytical methods (USEPA 551.1 and 552.2; Taguchi et al., 1994; Yoo et al., 2000; Cheng et al., 2004), and water quality parameter analytical methods (APHA method 5310 and 4500; USEPA 300.0; Lee and Westerhoff, 2005) were reported elsewhere where site specific observations and general nationwide occurrences were presented (Krasner et al., 2008). The samples represented a range of DOC (0.6–23 mg/L), DON (0.03–2.44 mg/L), UVA_{254} (0.01–0.48 cm^{-1}), and bromide levels (0–1.0 mg/L) (Table 1), which included a) 73 effluents from 24 wastewater treatment plants (WWTPs) after various types of physical/chemical/biological treatment (e.g., activated carbon, lagoon, activated sludge, nitrification, denitrification, membrane bioreactor, reverse osmosis, etc); b) 27 grab samples from 9 drinking water treatment plants (DWTPs) equipped with coagulation, softening, and ozonation processes; c) 50 samples from 11 surface rivers, and d) 18 samples from 10 monitoring wells undergoing soil aquifer treatment processes. To test the accuracy of the calibrated models, a laboratory database including 42 jar-tested wastewater effluents was used to exemplify an independent data source. These jar-test samples were obtained from six different effluents of WWTPs (Krasner et al., 2008), followed by varying degrees of batch-scale coagulation, softening, or activated carbon adsorption treatment (Westerhoff et al., 2005). The numbers of samples used for THMFP, HAAFP, HANFP, and NDMAFP precursor modeling were 210, 207, 166, and 134, respectively. Table 1 provides a summary of the water qualities and DBPFP levels.

2.2. Model calibration and statistical analysis

Calibrations of models were accomplished by a computer program Solver as embedded in Excel 2007 via trial-and-error method to obtain the best statistical results. Statistical analyses of predicted and measured data were conducted via Excel functions, including F-test, Student's T-test, linear correlation coefficient (R^2), analytical variance (AV, Equation (3)), standard error (SE, Equation (4)), and sensitivity analysis (EPA/100/K-09/003).

The statistical comparison started with the F-test analysis, which determined the variance similarity between observed values and predicted values. If the F-test value was >0.5 , the

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