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Evaluation of the relationship between bulk organic precursors and disinfection byproduct formation for advanced oxidation processes



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HIGHLIGHTS

G R A P H I C A L A B S T R A C T

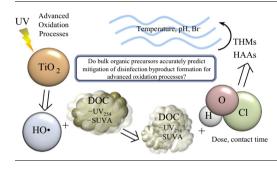
- Bulk DBP indicators may not be good predictors when using limited-input AOPs.
- Bulk measures were poor indicators of DBP formation for AOP and non-AOP scenarios.
- DBP models are of limited use as datasets rarely satisfy all boundary conditions.

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ABSTRACT

Advanced oxidation processes (AOPs) are gaining traction as they offer mineralization potential rather than transferring contaminants between media. However, AOPs operated with limited energy and/or chemical inputs can exacerbate disinfection byproduct (DBP) formation, even as precursors such as dissolved organic carbon, UV₂₅₄, and specific UV absorbance (SUVA) decrease. This study examined the relationship between DBP precursors and formation using TiO₂ photocatalysis experiments, external AOP and non-AOP data, and predictive DBP models. The top-performing indicator, SUVA, generally correlated positively with trihalomethanes and haloacetic acids, but limited-energy photocatalysis yielded contrasting negative correlations. The accuracy of predicted DBP values from models based on bulk parameters was generally poor, regardless of use and extent of AOP treatment and type of source water. Though performance improved for scenarios bounded by conditions used in model development, only 0.5% of the model/dataset pairings satisfied all measured parameter boundary conditions, thereby introducing skepticism toward model usefulness. Study findings suggest that caution should be employed when using bulk indicators and/or models as a metric for AOP mitigation of DBP formation potential, particularly for limited-energy/chemical inputs.

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1. Introduction

Increasing implementation of advanced oxidation processes (AOPs) makes it imperative to improve understanding of their * Corresponding author at: Department of Civil, Construction and Environmental Engineering, Marquette University, P.O. Box 1881, Milwaukee, WI 53201-1881, United States. Tel.: +1 414 288 2161; fax: +1 414 288 7521.

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disinfection byproduct (DBP) mitigation potential, including the extent to which traditional DBP indicators and the predictive DBP formation models based on them reliably predict AOP performance. This study evaluated the relationship between traditional bulk DBP precursors and DBP formation following AOP treatment using pilot-scale TiO₂ photocatalysis experiments, external DBP data (both AOP and non-AOP), and



published predictive models for total trihalomethanes (TTHMs), haloacetic acids (HAA5s), and subspecies thereof.

Since their discovery in 1974 (Bellar et al., 1974; Rook, 1974), the production of over 600 potentially harmful DBPs has attracted considerable attention (Krasner et al., 2006). Drinking water DBPs are widely regulated, as described in the Supplementary Information (SI), and increasing interest in water reclamation and reuse has sparked recognition of the occurrence and impacts of DBPs in wastewater (Wei et al., 2009). Chlorine disinfection alternatives such as chloramines, chlorine dioxide, ozone, or ultraviolet (UV) may reduce the formation of trihalomethanes (THMs) and haloacetic acids (HAAs); however, they may still form DBPs, and can shift DBP production toward species of greater health concern (Richardson, 2005). Additionally, alternative disinfectants may be ineffective against certain pathogens, increase nitrification and corrosion in the distribution system. and/or lack disinfectant residual (USEPA, 2006; Zhang et al., 2008). Thus, the most common DBP reduction strategy is removal of precursor natural organic matter (NOM) prior to disinfection (Kulkarni and Chellam, 2010). The best available techniques - enhanced coagulation or softening, granular activated carbon (GAC), or membrane filtration (Liu et al., 2008b; USEPA, 2006) – are often sufficient to control DBP formation; however, alternatives such as AOPs are also being investigated (Mayer et al., 2014; WHO, 2000).

The highly reactive, nonspecific radical species, such as HO, produced by AOPs are capable of mineralizing NOM, oxidizing trace emerging contaminants, and inactivating microorganisms (Gerrity et al., 2009). Given their potential to mineralize NOM rather than capturing and transferring it to another phase, implementation of AOPs is becoming increasingly attractive (Gerrity et al., 2009; Mayer et al., 2014), making it critically important to examine their effect on DBP formation (Dotson et al., 2010). Photocatalysis provides one pathway by which HO[•] radicals are produced using ultraviolet (UV) light (λ < 387 nm) to irradiate semiconductors such as TiO₂. However, AOPs such as TiO₂ photocatalysis are unlikely to be operated at sufficiently high energy and/or chemical inputs to mineralize NOM. Instead, under more practical operating conditions, AOPs are likely to incompletely oxidize organics, resulting in a shift toward smaller, less aromatic, and more hydrophilic moieties (Liu et al., 2008a; Mayer et al., 2014; Sarathy and Mohseni, 2010), which can exacerbate DBP formation beyond initial levels (Dotson et al., 2010; Gerrity et al., 2009; Liu et al., 2008a; Mayer et al., 2014). For example, 5 kW h m⁻³ TiO₂/UV AOP treatment increased TTHMs up to 119% and HAA5s up to 299% when applied to waters of varying quality, ranging from untreated to finished drinking water (Gerrity et al., 2009; Mayer et al., 2014). When higher AOP energy/chemical inputs are used, more complete NOM mineralization follows, resulting in decreased DBP formation, e.g., reductions of up to 95% TTHMs and 96% HAA5s (Gerrity et al., 2009; Mayer et al., 2014). The initial trend of increasing DBP production using limited-input AOP treatments contrasts with decreases in traditional bulk DBP precursors such as dissolved organic carbon (DOC), ultraviolet absorbance at a wavelength of 254 nm (UV₂₅₄), and specific UV absorbance (SUVA). This suggests that traditional bulk indicators of DBPs may not provide suitable performance metrics for AOPs, which fundamentally augment NOM characteristics rather than physically removing it.

2. Materials and methods

2.1. TiO₂ photocatalysis

Samples were collected from several locations in Central Arizona drinking water treatment trains using a mixture of surface and groundwater, including following sedimentation (SW), following GAC filtration (GAC), and from the distribution system (DS). Details of the treatment trains are provided in the SI. The samples were used as the influent water for pilot-scale TiO₂ photocatalysis experiments.

Photocatalysis was performed using the Purifics (London, ON) Photo-Cat[®] Lab reactor at an optimized concentration of 1 g L⁻¹ (Gerrity et al., 2009; Liu et al., 2008b) suspended reagent-grade Degussa P25 TiO₂ (Dusseldorf, Germany). The reactor was operated in batch configuration, as described previously (Gerrity et al., 2009; Mayer et al., 2014). For each source water, dark adsorption (no UV) samples were collected to provide a measure of physical removal due to adsorption and retention on the Photo-Cat's® submicronpore-size ceramic membrane filter. Additionally, three levels of treatment were tested for each water: limited (5 kW h m^{-3}) , $(80 \text{ kW h m}^{-3}),$ photocatalysis medium and extended $(160 \text{ kW h m}^{-3})$, providing a spectrum of partial, incomplete, and near-complete NOM oxidation (Mayer et al., 2014).

2.2. Water quality parameters

Following photocatalysis, turbidity was measured using a Hach (Loveland, CO) model 2100P turbidimeter, and a Mettler meter (Columbus, OH) was used for pH. Traditional DBP indicators quantified included DOC, UV_{254} , SUVA, and bromide (Br⁻). Particulate matter was removed from untreated samples using 0.45 μ m Pall (Port Washington, NY) Acrodisc[®] GHP membrane filters. DOC samples were acidified with 1 M HCl, and analyzed using a Shimadzu (Kyoto, Japan) 5050A Total Organic Carbon Analyzer. A Hach DR 5000 spectrophotometer was used to measure UV_{254} , and SUVA was calculated by normalizing UV_{254} values with respect to DOC. A Dionex (Sunnyvale, CA) DX-120 ion chromatograph was used to measure Br⁻.

2.3. Disinfection byproduct formation

Treated samples were chlorinated in accordance with a modified simulated distribution system test (SDS-THM), which targeted a chlorine residual of 1 mg L⁻¹ after 24 h at 28 °C (APHA, 2005; Mayer et al., 2014). A Hach D4/4000U spectrophotometer and DPD free chlorine reagent powder pillows were used to analyze free chlorine. The dominant weight-based classes of chlorine-DBPs, TTHMs and HAA5s (Krasner et al., 2006), were measured as described by Mayer et al. (2014). To gauge the evolution of DBP formation over time, samples were collected at 10–15 min, 1 h, 2 h, and 6 h post-chlorination. 24 h DBP formation was reported by Mayer et al. (2014).

2.4. External DBP datasets

In addition to internal DBP data from this study, external AOP and non-AOP datasets of TTHMs, HAA5s, and/or subspecies were collected from published literature to provide additional data to investigate the relationship between precursors and DBPs and to evaluate DBP model performance. This external data also functioned as a control to ensure that study findings were not artifacts of the laboratory protocol used in the current study. Twenty external datasets encompassing a range of experimental parameters were selected, as detailed in the SI.

2.5. DBP models

Internal and external DBP data were compared with DBP predictions using 149 published chlorine-DBP models, as described in the SI. These models predicted mass concentrations of TTHMs, HAA5s, and/or subspecies using traditional DBP indicators: DOC, UV_{254} , SUVA, chlorine dose (D), pH, temperature (*T*), Br⁻, and

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