



Photolytic dehalogenation of disinfection byproducts in water by natural sunlight irradiation



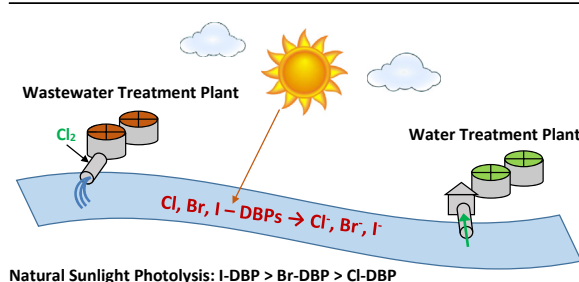
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HIGHLIGHTS

- Sunlight photolytic dehalogenation kinetics of disinfection byproducts were determined.
- Photolytic dehalogenation of DBPs increased with increasing number and size of halogens.
- Iodoform, triiodoacetic acid, and diiodoacetic acid degraded rapidly under sunlight irradiation.
- Typical levels of nitrate, nitrite, and pH had little impact on photolytic dehalogenation of DBPs.
- Natural organic matter inhibited photolytic dehalogenation of DBPs.

GRAPHICAL ABSTRACT



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ABSTRACT

The aqueous photolysis of halogenated disinfection byproducts (DBPs) by natural sunlight irradiation was studied to determine their photolytic dehalogenation kinetics. Total organic halogen analysis was used to quantify the dehalogenation extents of DBPs during outdoor photolysis experiments. Dichloroacetamide, chloral hydrate, chloroform, dichloroacetonitrile, monochloro-, monobromo-, dichloro-, dibromo-, and trichloroacetic acids were generally resistant to photolytic dehalogenation and showed less than 10% reduction after 6 h sunlight irradiation. Monoiodoacetic acid, tribromoacetic acid, bromoform, dibromoacetonitrile, and trichloronitromethane showed moderate to high dehalogenation degrees with half-lives of 4.0–19.3 h. Diiodoacetic acid, triiodoacetic acid, and iodoform degraded rapidly under the sunlight irradiation and exhibited half-lives of 5.3–10.2 min. In general, the photosensitive cleavage of carbon-halogen bonds of DBPs increased with increasing number of halogens (tri- > di- > mono-halogenated) and size of the substituted halogens (I > Br > Cl). Nitrate, nitrite, and pH had little impact on the photodehalogenation of DBPs under typical levels in surface waters. The presence of natural organic matter (NOM) inhibited the photodehalogenation of DBPs by light screening. The NOM inhibiting effects were more pronounced for the fast degrading iodinated DBPs. The results of this study improve our understanding about the photolytic dehalogenation of wastewater-derived DBPs in surface waters during water reuse.

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1. Introduction

Disinfection is a critical process in the treatment of wastewater to inactivate pathogenic organisms and prevent the spread of waterborne diseases to the environment. Chlorine is the most

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widely used chemical disinfectant for municipal wastewater disinfection prior to effluent discharge. When chlorine is added to the treated effluent, it can react with effluent organic matter to form a suite of harmful disinfection byproducts (DBPs) including trihalomethanes (THMs), haloacetic acids (HAAs), trihaloacetaldehydes (THAs), haloacetonitriles (HANs), halonitromethanes (HNMs), haloacetamides, nitrosamines, and others (Mitch and Sedlak, 2002; Yang et al., 2005; Krasner et al., 2009; Huang et al., 2016). It has been shown that the formation of DBPs during wastewater chlorination is highly influenced by the level of wastewater treatment and the concentrations of effluent ammonia. Based on a survey of wastewater treatment plants (WWTPs) in the United States, the DBPs formed at the WWTPs that achieved breakpoint chlorination consisted of high levels of THMs, HAAs, THAs and HANs, which were similar to those detected in drinking waters. For WWTPs disinfected with chloramines, high levels of N-nitrosodimethylamine (NDMA) in the effluent were identified (Krasner et al., 2009).

Municipal WWTP effluents typically contain relatively high levels of bromide and iodide ions compared with drinking water due to wastewater discharges from industrial facilities, hospitals, oil and gas production, and other anthropogenic activities (Krasner et al., 2009; Gong and Zhang, 2013; Hladik et al., 2014; Harkness et al., 2015). The bromide and iodide ions present in the treated effluent can be oxidized by chlorine to bromine and iodine, which subsequently react with effluent organic matter to form brominated and iodinated DBPs in a way analogous to chlorine (Hua et al., 2006). Bromine and iodine containing DBPs have been shown to be more cytotoxic and genotoxic than their corresponding chlorinated DBPs according to toxicological studies (Richardson et al., 2007; Pals et al., 2013; Yang and Zhang, 2013). This has raised concerns about the potential adverse impacts of these highly toxic DBPs on the aquatic life and human health.

In recent years, treated wastewater has been increasingly used as an alternative water resource to augment drinking water supplies through reclamation, recycling, and reuse. These wastewater recycling and reuse programs are expected to continue to increase in the future to mitigate the water scarcity due to population and economic growth. Indirect potable reuse (intentional and incidental) is one of the water recycling applications that has been in operation in many areas for decades (Rodriguez et al., 2009). During this water reuse practice, DBPs discharged into surface waters from upstream WWTPs can occur in the source water of downstream drinking water treatment plants. Therefore, it is important to understand the fate and transport of DBPs in surface waters to protect aquatic ecosystems and public health during water reuse.

Sunlight photolysis is one of the major natural processes that can affect the concentrations of wastewater-derived DBPs in surface waters. However, limited studies have evaluated the photodegradation of DBPs under natural sunlight irradiation conditions (Lekkas and Nikolaou, 2004; Lifongo et al., 2004; Chen et al., 2010). It has been shown that NDMA and other nitrosamines were highly photosensitive and the half-lives were less than 16 min under natural and simulated sunlight conditions (Plumlee and Reinhard, 2007; Chen et al., 2010). Chlorine and bromine containing HNMs and HANs typically exhibited higher photolytic losses than THMs and HAAs. Solar photolysis experimental results and quantitative structure–activity relationship (QSAR) analysis suggest that bromine and iodine substituted DBP species were more photosensitive than their chlorinated analogues (Lekkas and Nikolaou, 2004; Chen et al., 2010).

Photodegradation of halogenated DBPs can proceed by stepwise pathways which may result in intermediate products. For example, Castro and Belser (1981) conducted photolysis experiments of trichloronitromethane in aqueous solutions and found that the

production rate of chloride was not equal to 3 times that of trichloronitromethane disappearance, suggesting that chlorinated intermediates were produced. The halogenated intermediates and final products from DBP photolysis may still possess toxicity. It is necessary to determine complete dehalogenation extents of DBPs in water by sunlight irradiation in order to provide more accurate information for toxicity and health risk analysis. Moreover, the impact of water matrix (e.g., pH, organic matter, and nitrate) on the solar photolysis of DBPs has not been carefully investigated. Several emerging nitrogenous and iodinated DBPs including dichloroacetamide, iodoform and iodoacids have been identified in treated wastewater effluents. However, information about the impact of natural sunlight irradiation on these highly toxic DBPs is lacking in the literature. The objective of this study was to determine the dehalogenation kinetics of selected chlorine, bromine and iodine containing DBPs under natural sunlight irradiation conditions. Total organic halogen (TOX) was used as an analytical tool to quantify the dehalogenation extents of selected DBPs during outdoor photolysis experiments. The impact of pH, dissolved organic matter, nitrate, and nitrite on solar photolysis of DBPs was also evaluated. The results of this study provide quantitative information about the dehalogenation kinetics of DBPs including several emerging nitrogenous and iodinated compounds under the natural sunlight irradiation, which can help evaluate the fate of chlorinated, brominated and iodinated DBPs in surface waters.

2. Experimental methods

2.1. Preparation of DBP samples

A total of 17 DBPs were selected for the solar photolysis experiments. These DBPs included three THMs (chloroform, bromoform, iodoform), nine HAAs (monochloro-, monobromo-, monoiodo-, dichloro-, dibromo-, diiodo-, trichloro-, tribromo-, and triiodoacetic acid (MCAA, MBAA, MIAA, DCAA, DBAA, DIAA, TCAA, TBAA, and TIAA, respectively)), two HANs (dichloro- and dibromoacetonitrile (DCAN, and DBAN)), trichloronitromethane (TCNM), chloral hydrate (CH), and dichloroacetamide (DCAcAm). DIAA (98%), and TIAA (90%) were obtained from Toronto Research Chemicals, Inc. (Toronto, Ontario). All other DBPs were purchased from Sigma Aldrich (St Luis, MO). Each DBP was dissolved in ultrapure water at an initial concentration of $300 \pm 10 \mu\text{g/L}$ as Cl, Br, or I for chlorinated, brominated, and iodinated compounds, respectively, before photolysis experiments. Bromoform, chloroform, DBAN, DCAN, iodoform, and TCNM have low solubility in water. Those DBPs were first dissolved in acetone before being diluted in ultrapure water. The Suwannee River humic acid (SRHA) and the Pony Lake fulvic acid (PLFA) were obtained from the International Humic Substances Society. These natural organic matter (NOM) extracts were used to spike the DBP solutions to investigate the NOM impact on photolysis. Potassium nitrate and potassium nitrite (American Chemical Society reagent grade) were purchased from Fisher Scientific (Fairlawn, NJ) and used as nitrate and nitrite sources for the photolysis experiments. All solutions used in this study were prepared with ultrapure water (18 M Ω -cm) produced by a Barnstead NANOpure system.

2.2. Natural solar photolysis experiments

Natural solar photolysis experiments were performed using 60 mL quartz tubes with an outside diameter of 25 mm and a length of 150 mm. The thickness of the tube wall was 1.5 mm. The quartz tubes were capped to prevent the volatilization and evaporation of each DBP sample during the photolysis experiments. The sample tubes were placed on fabricated platforms at a 30° angle. The

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