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Decomposition of 1,4-dioxane by vacuum ultraviolet irradiation: Study of economic feasibility and by-product formation

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ABSTRACT

We report the first use of vacuum ultraviolet (VUV) treatment to decompose 1,4-dioxane, a persistent organic contaminant that is difficult to remove by conventional drinking water treatment processes. The efficiency of VUV treatment was compared to that of VUV- and UV-based advanced oxidation processes (AOPs) (VUV/TiO_2, VUV/H_2O_2, UV/TiO_2, and UV/H_2O_2), and by-product formation was investigated. VUV treatment decomposed 1,4-dioxane more rapidly than did UV and UV/TiO_2 treatments. The decomposition rate was enhanced when VUV irradiation was combined with TiO_2 or H_2O_2. VUV/H_2O_2 decomposed 1,4-dioxane more rapidly than UV/H_2O_2 at a low H_2O_2 dose (1 mg/L), but the rate difference became small at a high H_2O_2 dose (5 mg/L). Electrical energy per order analysis revealed that VUV treatment, and the VUV- and UV-based AOPs, were economically feasible for 1,4-dioxane decomposition. Using raw water samples, we investigated by-product formation during VUV treatment and the effect of VUV irradiation on chlorinated disinfection by-product formation potential. Although the samples contained high concentrations of bromide, no bromate was produced by VUV treatment. VUV treatment slightly decreased trihalomethane formation potential (THMFP), whereas haloacetic acid formation potential (HAAFP) was unchanged, and total aldehyde concentration increased. The trend in HAAFP agreed with that had been reported for the VUV irradiation with much higher dose (Buchanan et al., 2006), whereas the trend in THMFP was different from that with much higher dose. THMFP, HAAFP, and aldehyde concentration were reduced by subsequent treatment with granular activated carbon (GAC) or biological activated carbon (BAC). Nitrite was produced by VUV treatment but disappeared after subsequent BAC treatment. These results suggest that VUV treatment should be combined with GAC or BAC treatment to suppress by-product formation.
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Abbreviations: 2-MIB, 2-methylisoborneol; AOP, advanced oxidation process; BAA, bromoacetic acid; BAC, biological activated carbon; BCAA, bromochloroacetic acid; BDCAA, bromodichloroacetic acid; BDCM, bromodichloromethane; CAA, chloroacetic acid; CDBP, chlorination disinfection by-products; CDBPFP, formation potential of CDBP; DBAA, dibromoacetic acid; DBCAA, dibromochloroacetic acid; DBCM, dibromochloromethane; DCAA, dichloroacetic acid; DTW, dechlorinated tap water; EE/O, amount of electrical energy required for 1 m3 of contaminated water to bring about a reduction by one order of magnitude in target compound concentration; ES1, experimental setup 1; ES2, experimental setup 2; GAC, granular activated carbon; GC/MS, gas chromatograph/mass spectrometer; HAAFP, formation potential of HAAs; HAAs, haloacetic acids; JDWQS, Japanese drinking water quality standard; NOM, natural organic matter; SIM, selected ion monitoring; TBAA, tribromoacetic acid; TCAA, trichloroacetic acid; THMs, trihalomethanes; THMFP, formation potential of THMs; UV, ultraviolet; VUV, vacuum ultraviolet; VUVBP, VUV by-product; WHO, World Health Organization.

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1. Introduction

1,4-Dioxane, which is classified in Group 2B (possibly carcinogenic) by the International Agency for Research on Cancer, has been widely used as a stabilizing agent and solvent (Zenker et al., 2003) in chemical engineering applications, drug manufacturing, and the textile industry. In addition, 1,4-dioxane is present in many consumer products as a by-product of polyester synthesis (Black et al., 1983; Zenker et al., 2003), as well as in many household detergents and shampoos (Fuh et al., 2005; Tanabe and Kawata, 2008) as an impurity of surfactants (Guo and Brodowsky, 2000; Zenker et al., 2003). Industrial and domestic waste water contaminated with 1,4-dioxane flows into sewage treatment systems, where it is difficult to remove by means of conventional biological treatment processes (i.e., activated sludge treatment): no removal of 1,4-dioxane concentration was reported in a laboratory-scale experiment (Adams et al., 1994), and no or limited removal (0–30%) was observed in sewage treatment plants (Abe, 1999; Tanabe et al., 2006). Therefore, 1,4-dioxane is expected to be released into environmental waters after sewage treatment. In addition, 1,4-dioxane has been reported to be eluted from landfill sites and to contaminate leachates at concentrations of several to thousands of micrograms per liter, depending on the nature of the discarded materials (Isaacson et al., 2006; Lesage et al., 1990). Owing to its high hydrophilicity, 1,4-dioxane does not interact with soil particles and easily penetrates the ground (Zenker et al., 2003), which leads to contamination of groundwater rather than surface water.

The World Health Organization (WHO) has established guideline values for various chemical contaminants in drinking water (Guidelines for Drinking-water Quality), and the recommended value for 1,4-dioxane is <50 μg/L. However, 1,4-dioxane is difficult to remove from drinking water; it persists not only after conventional treatment consisting of coagulation, sedimentation, and sand filtration but also after activated carbon adsorption (McGuire et al., 1978). Oxidation processes such as ozonation (Adams et al., 1994) and chlorination (Klečka and Gonsior, 1986; McGuire et al., 1978) are also ineffective. Advanced oxidation processes (AOPs), such as a combination of ozone and hydrogen peroxide (H_2O_2), have been reported to decompose 1,4-dioxane (Adams et al., 1994; Suh and Mohseni, 2004). In addition, ultraviolet (UV) irradiation in the presence of a photocatalyst may be another option (Coleman et al., 2007; Hill et al., 1997), but the powdered photocatalyst studied for this process is so fine (particle diameter, 20–25 nm) that the necessity of recovering the fine powder makes the process impractical.

AOP consisting of UV and H_2O_2 has proven to be effective process for decomposition of organic contaminants, and has been already applied to full-scale drinking water treatment plants (Kruithof et al., 2007). In contrast, to our knowledge, vacuum ultraviolet (VUV) irradiation, which dissociates water molecules into hydroxyl radicals (Oppenländer and Sosnin, 2005), is a developing method that has not been applied to actual treatment plants, but is reported to be a powerful method for rapid decomposition of organic substances in water, including earthy–musty odor compounds (Kutschera et al., 2009), pharmaceuticals (Szabó et al., 2011), and pesticides (Imoberdorf and Mohseni, 2012). Moreover, the process is operationally simple and requires no chemicals that must be transported or stored. Therefore, it has the potential to be practical for use in urban water utilities that rely on a contaminated groundwater source for drinking water.

However, the nature of the reaction products and by-products formed from compounds of interest during VUV treatment must be considered, as is the case for other oxidation processes. During UV-based AOPs, 1,4-dioxane is reported to be transformed into various compounds, including 1,2-ethanediol monoformate (Stefan and Bolton, 1998), 1,2-ethanediol diformate (Maurino et al., 1997; Mehrvar et al., 2000; Stefan and Bolton, 1998), and methoxyacetic acid (Stefan and Bolton, 1998). These compounds are reportedly then transformed into acetic acid (Mehrvar et al., 2000) and formic acid (Mehrvar et al., 2000; Stefan and Bolton, 1998), which are eventually mineralized (Stefan and Bolton, 1998). In addition, during VUV treatment of raw water, by-products (VUVBPs) are formed from natural organic matter (NOM) present in the water, and some of these by-products may act as precursors of chlorination disinfection by-products (CDBPs) such as trihalomethanes (THMs) and haloacetic acids (HAAs). CDBPs are toxic to humans (Hebert et al., 2010), and for some of them, guidelines and standards for their concentrations in drinking water have been established.

Detailed information about VUVBPs and VUVBP-derived CDBPs is limited. Buchanan et al. investigated changes in CDBP formation potential (CDBPFP) during VUV treatment and after treatment with biological activated carbon (BAC) and found that the THM and HAA formation potentials (THMFP and HAAFP, respectively) tend to decrease with VUV irradiation (Buchanan et al., 2006) and that THMFP and HAAFP tend to decrease further upon subsequent BAC treatment (Buchanan et al., 2008). However, because the main target of these investigators was NOM at markedly high concentrations (8–10 mg-DOC/L), VUV dose used in their studies were 16–186 J/cm^2, which is several orders of magnitude higher than the doses required for the decomposition of persistent organic contaminants in raw water at trace levels (e.g., 0.1, 0.2, and 0.6 J/cm^2 for 2,4-dichlorophenoxyacetic acid (Imoberdorf and Mohseni, 2012), anatoxin-a (Afzal et al., 2010), and earthy–musty odor compounds (Kutschera et al., 2009)). The effect of low VUV doses on CDBPFP has not yet been investigated. The fact that THMFP initially increases upon O_3/UV treatment of surface water samples at a low UV dose and then decreases with increasing UV dose (Glaze et al., 1982) highlights the need for further research.

In the present study, we investigated the use of VUV treatment for 1,4-dioxane removal from water, paying particular attention to decomposition efficiency and by-product formation. In addition to VUV alone, we investigated VUV-based AOPs (VUV/TiO_2 and VUV/H_2O_2), UV-based AOPs (UV/TiO_2 and UV/H_2O_2), and UV alone, and we compared the performances of various treatments. For the VUV/TiO_2 and UV/TiO_2 treatments, we introduced a new photocatalyst consisting of TiO_2 immobilized on a nonwoven fabric, which meant that no catalyst separation was required after the photocatalytic reactions. Furthermore, to investigate the economic feasibility of the treatments, we performed electric energy per order (EE/O) analysis of the decomposition of 1,4-dioxane. We also compared the removal of 1,4-dioxane with that of 2 earthy-musty odor compounds, 2-methylisoborneol (2-MIB) and geosmin, whose behaviors in AOPs have been well studied. For the second purpose, we used two natural water samples (a lake water sample and a groundwater sample) to investigate the formation of VUVBPs from NOM and inorganic ions at a VUV dose high enough for the decomposition of trace-level organic contaminants and to investigate CDBPFP. The target VUVBPs included 10 aldehydes, as well as nitrite and bromate; and the

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