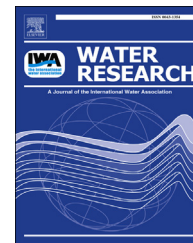




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Fate of anthropogenic cyclic volatile methylsiloxanes in a wastewater treatment plant

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ABSTRACT

The fate of cyclic volatile methylsiloxanes (cVMS) – octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6) – was evaluated in a typical secondary activated sludge wastewater treatment plant (WWTP). Water samples (influent, primary effluent, and final effluent) and sludge (primary sludge and waste activated sludge) samples were collected at overnight low, morning high, afternoon low, and evening high flows. Concentrations of cVMS in influents fluctuated with the influent flows, ranging from 0.166 to 1.13 $\mu\text{g L}^{-1}$, 3.47–19.3 $\mu\text{g L}^{-1}$, and 0.446–3.87 $\mu\text{g L}^{-1}$ for D4, D5, and D6, respectively. Mass balance analysis of cVMS showed the average mass of D4, D5, and D6 entering and exiting the plant in influent and effluent, respectively, were 109 g d^{-1} , 2050 g d^{-1} , 280 g d^{-1} , and 1.41 g d^{-1} , 27.0 g d^{-1} , 1.90 g d^{-1} . The total removal efficiency of cVMS was >96%. To elucidate their detailed removal mechanisms, Mackay's fugacity-based treatment plant model was used to simulate the fate of cVMS through the WWTP. Due to the unusual combination of high hydrophobicity and volatility of cVMS, volatilization in the aeration tank and adsorption to sludge were the two main pathways of cVMS removal from water in this WWTP based on the experimental and modeled results. The morning and evening high influent mass flows contributed almost equally at approximately 40% of the total daily cVMS mass, with D5 accounting for the majority of this daily loading.

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1. Introduction

Cyclic volatile methylsiloxanes (cVMS) – octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6) – are used in a number

of commercial applications such as intermediates for the polymerization of polyorganosiloxanes, cosmetic and personal care products, defoamers, sealants, adhesives and coatings (Wang et al., 2013a; Alaei et al., 2013). In 2006, the U.S. annual production of D4, D5, and D6 ranged between 100 and 500, 50 and 100, and 1 and 10 million pounds, respectively (US

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EPA, 2002). D4, D5 and D6 are not manufactured in Canada, but they are imported to make polymers and other commercial products. As a result of their high production volume and widespread use, cVMS were reported in different environmental matrices including air (Genualdi et al., 2011; McLachlan et al., 2010), river water (Sparham et al., 2008), sediment (Kaj et al., 2005a), soil (Sanchez-Brunete et al., 2010), and biota (Kaj et al., 2005a; Kala et al., 1997).

cVMS have combinatorial properties of hydrophobicity ($\log K_{OW} > 6$) and volatility ($\log K_{AW} > 2$) (Xu and Kropscott, 2012), resulting in unusual environmental fate (Whelan and Breivik, 2013; Whelan, 2013). cVMS have high long-range atmospheric transport (LRAT) potential (Genualdi et al., 2011), but they remain in the atmosphere even under Arctic temperature conditions (Wania, 2003). Consequently, they have little influence on remote aquatic ecosystems through LRAT (Kierkegaard et al., 2010; Warner et al., 2010). cVMS are not biodegradable but undergo degradation with radicals in atmosphere (Atkinson, 1991) and hydrolysis in aquatic and soil environments (Durham, 2005, 2006; Xu, 1999; Xu and Chandra, 1999). According to Canadian regulations (Canada, 1999), they are persistent in air (half-lives ≥ 2 days) and sediment (half-lives ≥ 365 days) (Environment Canada and Health Canada, 2008a,b,c). D4 is also toxic to aquatic organisms with a chronic no observed effect concentration (NOEC) of $4.4 \mu\text{g L}^{-1}$ (Sousa et al., 1995). Recently, toxicity of the D5 was also observed for plant growth, survival, and reproduction (Velicogna et al., 2012).

Wastewater treatment plants (WWTPs) have been identified as a conduit for cVMS to enter the environment (Environment Canada and Health Canada, 2008a,b,c). Biogas, biosolids, and influent and effluent water were reported to have high concentrations of cVMS (Kaj et al., 2005a; Badjagbo et al., 2009; Schweigkofler and Niessner, 1999; McBean, 2008; Schlabach et al., 2007; Dewil et al., 2007; Xu et al., 2013; Sanchís et al., 2013). High concentrations in biogas even have influence on the air concentrations around the WWTP (Wang et al., 2001). Typically, previous studies on WWTPs have been completed to demonstrate the presence or absence of cVMS in influent, effluent, biogas, and sludge (Xu et al., 2013; Wang et al., 2013b; Liu et al., 2014). All the studies have demonstrated that cVMS can be well removed ($>80\%$) by these WWTPs using secondary activated sludge processes (Wang et al., 2013a). The details of removal mechanisms on the influence of the operational parameters in WWTPs are not always clearly understood. However, limited studies have explored the partitioning of compounds into municipal wastewater residuals and their subsequent fate during activated sludge process (Bletsou et al., 2013). This lack of information was due in part to the difficulty of accurately detecting and quantifying cVMS in challenging matrices of municipal sludge and wastewater. As a result of all these factors, a study of cVMS emissions to the aquatic and terrestrial environment through wastewater treatment was undertaken.

Environment Canada's laboratory developed and employed isotope dilution gradient with large volume injection GC–MS to analyze cVMS in influent, effluent, receiving water, and biosolid from 11 Canadian WWTPs in 2010 (Wang et al., 2013b, 2013c). Using these new methods, it is possible to analyze concentrations in water and sludge samples for cVMS in a

WWTP. Then we can calculate the air concentrations based on a mass balance method. The objective of this study was to determine the mass flows of D4, D5, and D6 in a municipal WWTP using previously reported analytical methods for cVMS in wastewater and biosolids. Fluctuations of cVMS concentrations in the aqueous and sludge phases were measured at low- and high-flows over a 36-h period. To elucidate their detailed removal mechanisms, the fugacity-based treatment plant model (STP-EX) (Clark et al., 1995) was used to simulate their fate in the WWTP.

2. Materials and methods

2.1. Study site and sampling

The municipal WWTP selected for this study discharges into Lake Ontario, Canada. The WWTP currently serves a population of approximately 285,900 and has a capacity of $165,000 \text{ m}^3 \text{ d}^{-1}$. The plant treats wastewater from homes (70%) and businesses (30%) including shopping malls, restaurants, hotels, supermarkets, and banks, which drains from toilets, showers, and sinks. The WWTP uses a typical activated sludge process containing intake screens, grit tanks, primary and secondary settling tanks, fine-bubble aeration tanks, and an outfall pipe to the lake (Figure S1). The operational conditions of this WWTP include the mixed liquor suspended solids (MLSS) concentration (2500 g m^{-3}), solids retention time (SRT) (5–7 days), and hydraulic retention time (HRT) (6.23 h).

Two sets of samples were collected in January 7th and March 8th and 9th, 2011 under normal weather conditions and no precipitation. Winter sampling was conducted to obtain the worst-case scenario as previous analyses indicated high removal rates in the summer (Wang et al., 2013b). Concentrations of 24-h composite samples were commonly reported in studies on pharmaceuticals and personal care products in WWTPs (Ort et al., 2010), but a very high background of cVMS was observed using an on-line automatic sampler in the preliminary study; therefore, a single time grab sampling method was used instead. During the first sampling event in January, only morning high flow influent and effluent samples were collected at 10AM. During the second sampling in March, both aqueous (influent, primary effluent, and final effluent) and sludge (primary sludge and waste activated sludge (WAS) samples were obtained (Figure S1). The influent sampling times were 7AM, 10AM, 4PM, and 10PM for overnight low, morning high, afternoon low, and evening high flows, respectively. The times were selected based on recent flow patterns at this WWTP and are representative of typical high and low variations in domestic wastewater flow. In order to track the same influent mass flow, the primary and final effluent and sludge samples were collected according to the influent sampling time, influent flow, and calculated hydraulic retention time except the overnight low flow (Table 1). In addition, we missed the effluent and WAS samples in the afternoon low flow.

Water samples (total 600 mL) were collected without headspace in 100-mL serum bottles and quickly crimp sealed with Teflon coated butyl septa and aluminum seals. Triplicate water samples were analyzed for cVMS and the other three

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