



The impact of antibacterial handsoap constituents on the dynamics of triclosan dissolution from dry sand



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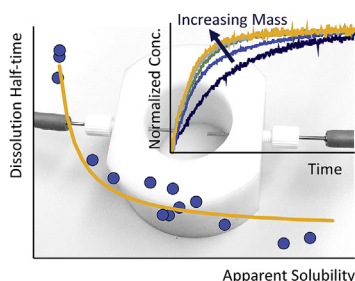
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HIGHLIGHTS

- Surfactants in antibacterial handsoap accelerate dissolution of triclosan.
- Small increases in solubility correspond to large decreases in dissolution half-time.
- Dissolution dynamics are consistent with a parallel micellar solubilization model.

GRAPHICAL ABSTRACT



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ABSTRACT

Triclosan has been widely used as an antibacterial agent in consumer and industrial products, and large quantities continue to be discharged to natural waters annually. The focus of this work was on studying the dynamics of triclosan dissolution following evaporative drying. Warm weather can cause the water in intermittent streams or the unsaturated zone to evaporate, causing nonvolatile compounds to form solid precipitates. Because dissolution of precipitates is a relatively slow process, the dynamics of dissolution following evaporation may play an important role in controlling the release of contaminants to the environment. The specific purpose of the work was to explore the effects of surfactant co-contaminants from an industrial antibiotic handsoap on the dissolution dynamics of triclosan. The work used a fiber optic-based optical cell to conduct stirred-batch dissolution experiments for sands coated with different mass loadings of triclosan. Results show that the presence of surfactants from the hand soap not only increase the apparent equilibrium solubility, but also increase the rate of approach to equilibrium. A model describing the dissolution process was developed, and was found to be consistent with experimental data. Results of the work suggest that even small solubility enhancement by surfactant co-contaminants may have a significant impact on dissolution dynamics. Because waters containing significant quantities of triclosan are also among those most likely to contain surfactant co-contaminants, it is likely that the release of triclosan to the environment following evaporation may be faster in many cases than would be predicted from experiments based on pure triclosan.

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1. Introduction

Emerging contaminants, such as pharmaceuticals and personal care products (PPCPs) or suspected endocrine disrupting chemicals

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(EDCs) can potentially enter the environment through a wide range of pathways, including discharge of incompletely treated wastewater, and land application of biosolids. Analyses have found emerging contaminants to be pervasive in natural waters and other environmental media worldwide (Daughton and Ternes, 1999; Kolpin et al., 2002; Snyder et al., 2003; Benotti et al., 2009; Sherburne et al., 2016). Triclosan (5-Chloro-2-(2,4-dichlorophenoxy)phenol), an antibacterial compound, is one emerging contaminant that has seen extensive use in consumer and industrial products, and has been widely detected in environmental waters. For example, it has been reported that more than 100,000 kg of triclosan are discharged by wastewater treatment plants in the United States annually (Carey and McNamara, 2015). Environmental concerns about triclosan stem from suspicions that it may be an EDC, as well as concerns about its potential to create antibiotic resistance in bacteria (Aiello et al., 2007; Kibbey et al., 2009, 2010; Carey and McNamara, 2015; Gao et al., 2015; Wang and Tian, 2015). While the use of triclosan in over-the-counter consumer wash products was recently banned in the United States effective September 2017, triclosan is still widely used in consumer wash products internationally, and is present in other consumer products, such as toothpastes and antiseptic wipes both in the United States and elsewhere. Triclosan is also a major component of antiseptic washes and other products used in health care, laboratory, and industrial settings in the United States and elsewhere (FDA, 2016).

One significant reason that high quantities of triclosan have entered the environment is that high concentrations have been used in products. For example, triclosan-based antibacterial handsoaps often contain triclosan at concentrations ranging from 0.1 to 0.5% w/v, i.e., 1000–5000 mg/L. The inclusion of triclosan at these concentrations means that even if nearly all is removed by treatment or reduced by dilution, readily detectable concentrations can remain in natural waters. Furthermore, the presence of triclosan at these concentrations in antibacterial products highlights the important fact that other product constituents must accompany triclosan in the formulations for triclosan to be soluble at these levels. Note that the solubility of triclosan in pure water is typically reported to be approx. 10 mg/L or less, depending on conditions. As such, concentrations in antiseptic wash products can be three orders of magnitude higher than the solubility in pure water. While liquid handsoaps are often comprised of 80% water or more, the remainder can consist of various anionic or nonionic surfactants, as well as different polymers, such as polyethylene glycol. The presence of the surfactants and other additives allow for the creation of stable formulations containing high concentrations of triclosan. However, the presence of these constituents, typically at an order of magnitude higher in concentration than triclosan itself, also means that they are likely to be co-contaminants in the environment with triclosan, and may ultimately influence the release of triclosan to the environment.

Previous studies have examined the effects of surfactants and other surface-active compounds on the fate of emerging contaminants in environmental media. For example, Hari et al. (2005) studied the effect of cationic and nonionic surfactants on the adsorption of several pharmaceutical compounds to aquifer materials. Studies with triclosan have observed effects from a bio-surfactant (Wu et al., 2015) and humic acid (Behera et al., 2010) on adsorption to natural environmental media. Furthermore, work has been reported examining methods of increasing the aqueous solubility of triclosan in formulations by inclusion of surfactants and other additives (e.g. (Grove et al., 2003),). However, little is known about the effect of surface-active co-contaminants on the dynamics of dispersal of triclosan into the environment following evaporative drying of triclosan-contaminated water. This topic is the focus of

the current manuscript.

Intermittent streams, where flow is seasonal, and ephemeral streams, where flow only occurs following rain events, are widespread. In the continental United States, it has been reported that 81% of all streams in arid regions are intermittent or ephemeral (Levick et al., 2008), a fact that suggests repeated wetting-drying cycles may play an especially important role in the environmental dispersal of aqueous contaminants. Wetting and drying cycles are also important in the unsaturated zone, where cycles of drought and rainfall infiltration can cause drastic changes in soil moisture content.

When evaporation occurs, any non-volatile constituents present in water will increase in concentration, and ultimately form a precipitate on solid surfaces. Because dissolution is a slow process, an important area of study is to understand how the formation of chemical precipitates ultimately affects the dynamics of release upon rewetting. Furthermore, it is essential to understand how common non-volatile co-contaminants, such as surfactants, may affect the release of contaminants to the environment.

The focus of this work is on understanding how surface-active co-contaminants influence the dynamics of release of precipitated triclosan from sand during rewetting following drying. The work uses real-time triclosan dissolution measurements in a stirred reactor to quantify dissolution rates, and numerical simulations to explore the observed phenomena.

2. Materials and methods

2.1. Materials and sample preparation

Triclosan was purchased from Sigma-Aldrich (St. Louis, Mo) with a reported purity of >97%, and was used as received. This triclosan was used for calibration of detection methods, as well as for dissolution experiments conducted in the absence of surface-active constituents. For sake of discussion, this triclosan will be referred to as pure triclosan in this paper.

An industrial antiseptic liquid handsoap (trade name Bacdown; Deacon Laboratories) was purchased from Fisher Scientific, and was used as received. The handsoap had a triclosan concentration of 0.5%. The major surfactant constituent of the handsoap is reported to be the surfactant sodium laureth sulfate ($n \geq 3$). Sodium laureth sulfate surfactants are ethoxylated anionic surfactants, and are among the most common surfactants in liquid handsoaps. Other minor constituents are reported to include propylene glycol, PEG 75 lanolin, cocamide diethanolamine (a foaming agent), and ethoxylated nonylphenol nonionic surfactant.

Sand used for dissolution experiments was U.S. Silica (Berkeley Springs, WV) F-65 fine sand. F-65 sand is a high purity foundry-grade sand, and has a mean grain size of 0.21 mm. Triclosan-containing sand samples for analyses were prepared by creating aqueous dilutions of the handsoap, mixing them with sand to saturate the sand, and then oven-drying the mixture at 60 °C for 24 h. For comparison, sand samples were also prepared with the pure triclosan to test dissolution in the absence of surfactant constituents. The pure triclosan was deposited on sand by preparing a high-concentration triclosan solution in HPLC-grade acetonitrile (Fisher Scientific; Waltham, MA), mixing it with the sand to saturate the sand, and then air-drying for 24 h. Note that acetonitrile was used for the pure triclosan because the low aqueous solubility of triclosan precluded creating comparable mass loadings for dissolution experiments from an aqueous stock solution. Triclosan mass loadings in experiments ranged from approx. 40 to 125 mg/kg, and were designed to provide excess triclosan mass on solid surfaces to facilitate quantitative interpretation of dynamic results. For the samples prepared with handsoap, mass loadings of

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