Chemosphere 90 (2013) 581-587

Contents lists available at SciVerse ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Effects of monorhamnolipid and dirhamnolipid on sorption and desorption of triclosan in sediment-water system

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HIGHLIGHTS

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

- ► The solubilization enhancement of trilosan (TCS) by rhamnolipids was investigated.
- ► The sorption of rhamnolipids by the sediments was adequately described by Langmuir-type isotherm.
- The sorption and desorption of TCS mainly depend on the sediment composition and rhamnolipid structure.
- Dirhamnolipid was more effective than monorhamnolipid for TCS desorption from sediments.

ARTICLE INFO

Article history: Received 14 April 2012 Received in revised form 13 August 2012 Accepted 20 August 2012 Available online 6 October 2012

Keywords: Rhamnolipid Triclosan Sorption Desorption Sediment

ABSTRACT

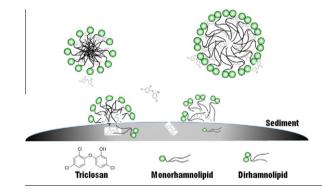
The effects of monorhamnolipid (RL-F1) and dirhamnolipid (RL-F2) on the sorption and desorption of triclosan (TCS) in sediment–water system were investigated in this study. Results of the bath equilibrium experiments showed that RL-F2 provided much higher solubilization enhancement for TCS than RL-F1. Sorption of both rhamnolipids by the sediments was highly correlated with the sediment clay content. Moreover, the apparent distribution coefficients of TCS (K_d^*) decreased with the increase of rhamnolipid concentration (0.05–7.5 mM), and RL-F2 presented a larger distribution capacity of TCS into the aqueous phase at relatively higher concentrations (>2.5 mM). Further results also indicated that the release of TCS from sediment could be enhanced by both rhamnolipids. RL-F2 was more efficient than RL-F1 in desorbing TCS from the sediment with low clay content. The TCS desorption percentages (R_d^*) of RL-F2 (5 mM) was 1.8–2.4 times that of RL-F1. These findings could provide useful guidelines for the application of rhamnolipid-enhanced remediation technologies for TCS contaminated sediment.

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1. Introduction

Triclosan [TCS, 5-chloro-2-(2,4-dichlorophenoxy)-phenol], one of the most common antimicrobial agents, is widely used in house-

hold and personal care products. Owing to its relatively low aqueous solubility and hydrophobic property with a log K_{ow} of 4.7 (Ying et al., 2007), TCS has a tendency to accumulate in sludge (Heidler et al., 2006; Ying and Kookana, 2007) and sediment (Zhao et al.,







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^{0045-6535/\$ -} see front matter @ 2012 Elsevier Ltd. All rights reserved. http://dx.doi.org/10.1016/j.chemosphere.2012.08.036

2010) where it can persist with concentrations at mg kg⁻¹ levels. Recently, TCS has been globally detected in surface water (Lindstrom et al., 2002; Donna et al., 2004; Hua et al., 2005; Wu et al., 2007). Studies on the occurrence and fate of TCS in aquatic environment indicate that TCS is potentially toxic to various organisms (Orvos et al., 2002; Chalew and Halden, 2009) and was found to be accumulated in human milk and blood (Adolfsson-Erici et al., 2002; Allmyr et al., 2006). Laboratory studies also showed that TCS can be degraded by microbial processes in soil under aerobic conditions, but it was highly resistant to biodegradation in soil anaerobic conditions (Ying et al., 2007). Therefore, it is extremely necessary to investigate the distribution of TCS in solid–water system for the elimination of TCS.

Due to the high sorption affinity to the solid phase, TCS is very difficult to remove by conventional pump-and-treat technologies (Kommalapati et al., 1997). It is well known that surfactant can increase the solubility of hydrophobic organic compounds (HOCs) or lower the interfacial tension to enhance the mobility of HOCs. Surfactant-enhanced remediation (SER) is suggested as a promising technology that has recently been extensively exploited for the remediation of organic contaminated soil and water (groundwater or surface water) (West and Harwell, 1992; Mulligan et al., 2001; Paria, 2008). The interactions of surfactants with organic contaminants in soil are very complex and are influenced by various factors including CMC, sorption behavior of the surfactant and contaminant, solubility of the contaminant and the soil composition (Haigh, 1996). As a result of lower cellular toxicity and ready biodegradability, biosurfactants have recently gained more attention and are extensively used in many environmental applications including remediation of PAHs, pesticides, heavy metals and so on (Mulligan, 2005). It has been suggested to be more attractive from an environmental perspective for SER processes (Clifford et al., 2007). However, compared with many investigations of the distribution of PAHs or pesticides in soil-water-surfactant systems (Edwards et al., 1994; Zhou and Zhu, 2005, 2007; Wang and Keller, 2008, 2009; Zhu and Zhou, 2008), much less information is known about that of TCS within a soil-water system containing surfactant or biosurfactant (Lin et al., 2011).

As one of the typical biosurfactant, rhamnolipid has been well defined in pollutant remediation, and as many as 28 different homologues of rhamnolipid have been reported (Nitschke et al., 2005). Rhamnolipid dosage is an important parameter in mobilization of HOCs in contaminated soil. It has been reported that pesticide desorption from soil was only enhanced when rhamnolipid concentrations were high enough to reach soil saturation and form micelles in aqueous phase (Mata-Sandoval et al., 2002). In another study (Chen et al., 2005), PAHs had linear sorption isotherms on silica sand with partition coefficients decreasing with the increase of rhamnolipid concentration until the CMC, while the partition coefficients increased with increasing rhamnolipid concentration above the CMC. Moreover, the different structures of biosurfactants may have different surface and micelle properties (Zhang et al., 1997). While most current research has focused on the applications of rhamnolipid mixtures, little knowledge is available on the effects of different forms of rhamnolipid for remediation. Thus, more information about the effect of rhamnolipid structure and concentration on solubilization and distribution of HOCs in soilwater system are certainly needed for the development of effective biosurfactant-enhanced remediation technologies.

The present study was based on previous investigations of TCS sorption onto sediments and its distribution in sediment–water system by a rhamnolipid mixture (Lin et al., 2011). It reported that the rhamnolipid mixture was more efficient in distributing TCS into aqueous phase in sediments with large volume of pores and low organic matter. The rhamnolipid mixture could be further purified into two fractions, monorhamnolipid (RL-F1) and dirhamnolipid

(RL-F2). The purpose of this study was to further quantify the effect of both RL-F1 and RL-F2 on TCS sorption and desorption in sediment–water systems. A series of batch experiments was performed to determine TCS solubilization in aqueous solutions by both rhamnolipids. The portioning of TCS within sediment– water–rhamnolipid system is also discussed. The results of this study could provide a better understanding on the distribution behavior of TCS in sediment–water–rhamnolipid system, and offer valuable information for the application of rhamnolipid in the remediation of organic contaminated sediment.

2. Materials and methods

2.1. Materials

Triclosan, with a purity of 99%, was purchased from Alfa Aesar (Ward Hill, MA). Sodium azide was obtained from Solarbio. All analytical solvents including methanol and acetonitrile (HPLC grade) were obtained from Merck (Hannover, Germany).

RL-F1 and RL-F2 were produced and purified from *Pseudomonas aeruginosa* mutant strain MIG-N146 as described previously (Guo et al., 2009). The average molecular weight of RL-F1 is 509, and the CMC is 0.09 mM. RL-F2 has an average molecular weight of 656 and CMC of 0.18 mM.

2.2. Sediment preparation

Three sediments were collected from different sites of Pearl River Delta in southern China. Sediment samples were air-dried and sieved to obtain particles less than 1.0 mm. Organic matter in sediment was determined by combustion method of loss on ignition at 450 °C for 4 h. Particle size analysis was determined by hydrometer method. The physic-chemical properties of sediment samples are shown in Table 1 (Lin et al., 2011).

2.3. Rhamnolipid solubilization of TCS

The solubility enhancement of TCS by rhamnolipids was conducted in duplicate. 10 mL RL-F1 and RL-F2 solutions each with a series of concentrations were placed in 15 mL glass centrifuge tubes with Teflon-lined screw cap. TCS was subsequently added to each tube in an amount slightly greater than required to saturate the solution. These tubes were shaken in the dark at 22 ± 0.1 °C, 225 rpm for 48 h and then centrifuged at 2000 rpm for 30 min. An appropriate aliquot of supernatant was then carefully taken and the concentrations of TCS were analyzed by HPLC.

2.4. Sorption of rhamnolipids onto sediments

Batch sorption experiments were conducted in duplicate to determine rhamnolipid equilibrium sorption isotherms in a series of 15 mL glass centrifuge tubes. A set of RL-F1 and RL-F2 solutions with varying concentrations (0.05-10 mM) were prepared in deionized water with 0.01 M NaCl to minimize ionic strength change and 0.01% (w/v) NaN₃ to inhibit microbial growth. All solutions were buffered at pH 6.8 with 15 mM NaHCO₃. A mass of 0.2 g sediments and 10 mL rhamnolipid solutions were added to each tube. The tubes were then equilibrated at 22 ± 1 °C and 225 rpm for 48 h

Table 1

Select physic-chemical properties of sediment samples in this study.

Sediment samples	Organic content (%)	Clay/silt/sand (%)
Sediment A	2.4	26.4/46.4/27.2
Sediment B	3.0	19.5/35.3/45.20
Sediment C	1.1	13.3/14.9/71.8

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