



# Effect of cations on the solubilization/deposition of triclosan in sediment-water-rhamnolipid system



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## HIGHLIGHTS

- The migration of TCS in sediment-water was regulated by RL with cations.
- TCS was deposited in sediment by RL with  $\text{Ca}^{2+}/\text{Mg}^{2+}$ .
- Solubilization/deposition of TCS was related to self-assembly of RL with cations.
- Deposition-ex-situ remediation could be a new insight for the application of RL.

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## ABSTRACT

Cations had great influence on the self-assembly of rhamnolipid, which in turn affected the fate of triclosan. The migration of triclosan from sediment to water benefited its biodegradation but it could be transformed into more toxic compounds. To regulate the fate of triclosan and reduce environmental risks extremely, the effect of four common cations in surface water ( $\text{Na}^+/\text{K}^+/\text{Ca}^{2+}/\text{Mg}^{2+}$ ) on the solubilization/deposition of triclosan in sediment-water-rhamnolipid system was investigated. The interaction among cations, triclosan and rhamnolipid was explored based on self-assembly of rhamnolipid and water solubility of triclosan in rhamnolipid solutions. Results showed that cations had little influence on the fate of triclosan in the absence of rhamnolipid. Cations, especially  $\text{Ca}^{2+}/\text{Mg}^{2+}$ , reduced the critical micelle concentration, micellar size and zeta potential of rhamnolipid solutions. The changes in self-assembly of rhamnolipid with different cations led to the difference of residual rhamnolipid concentration in water, which was nearly invariant with 0.01 M  $\text{Na}^+/\text{K}^+$  while decreased significantly with 0.01 M  $\text{Ca}^{2+}/\text{Mg}^{2+}$ . Consequently, water solubility of triclosan in rhamnolipid solutions increased with the addition of  $\text{Na}^+/\text{K}^+$  whereas decreased with  $\text{Ca}^{2+}/\text{Mg}^{2+}$ . In sediment-water-rhamnolipid system, triclosan was slightly solubilized from sediment to water with  $\text{Na}^+/\text{K}^+$  while deposited in sediment with  $\text{Ca}^{2+}/\text{Mg}^{2+}$ . These findings provided an alternative application of rhamnolipid for the remediation of triclosan-polluted sediment.

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## 1. Introduction

Triclosan (TCS, 5-chloro-2-(2,4-dichlorophenoxy)-phenol) is a widely used broad-spectrum antimicrobial agent in personal care and consumer products. For the steadily increasing usage of these products, TCS is frequently detected in sediment/soil, surface water and even in human-related biological samples (Dann and Hontela, 2011; Mehmet et al., 2015; Stéphane et al., 2013). TCS has been

demonstrated to be highly toxic to various organisms and may cause development of resistant bacteria and changes in microbial community structure (Daughton and Ternes, 1999; Necip et al., 2015; Orvos et al., 2002). Due to its high hydrophobicity, TCS is expected to be absorbed onto particulate materials and then accumulated in sediments (Zhao et al., 2010). On one hand, the biodegradation of TCS on sediment is restricted because of the anaerobic condition, while TCS in water can be removed/transformed by aerobic biodegradation (Ying et al., 2007). On the other hand, TCS in water can be transformed into chlorinated dibenzodioxins after photooxidation, and chlorinated phenols after chlorination, which are potentially more toxic and persistent

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compounds (Bedoux et al., 2012). So the deposition of TCS onto sediment is needed under the condition where these hazards may be produced. In view of the two-sided effect of TCS remediation in water, it is important to regulate the fate of TCS in water/sediment system according to the real ambient conditions.

Recently, biosurfactant based remediation technology for organic contaminated sediment/soil and water is of increasing importance (Behera et al., 2010; Lin et al., 2011). Critical micelle concentration (CMC) is a characteristic parameter of biosurfactant. Below the biosurfactant's CMC, the biosurfactants exist as monomers and have little effects in the solubility of HOCs. Micellar solubilization occurs when the biosurfactant concentration exceeds the CMC, where the solubility of HOCs is enhanced by the incorporation of hydrophobic molecules into biosurfactant micelles. The effect of biosurfactant on the fate of hydrophobic organic compounds (HOCs) in sediment/soil-water system is focused on two mechanisms: (i) the micellar solubilization which results in the facilitated transport of HOCs to water (Cao et al., 2008; Shi et al., 2015) and (ii) the adsorption of biosurfactant onto sediment/soil which leads to HOCs partitioning onto sediment (Paria and Yuet, 2007; Zhou and Zhu, 2007). Therefore, it is an alternative way for the application of biosurfactant to enhance the remediation of HOCs. As one of the most representative biosurfactant, rhamnolipid (RL) has been studied extensively in pollutant remediation for its high surface activity, low toxicity, excellent biodegradability and high thermal/chemical stability (Wu et al., 2015). The improved remediation efficiency of HOCs contaminated sediment/soil by RL has been described in several studies (Zhang et al., 2013). Some attempts have been reported focusing on the effect of sediment/soil composition, RL dosage, RL structure, solubility of HOCs, environmental factors such as pH, ionic strength and dissolved organic matter (DOM) on the solubilization of HOCs (ElSayed and Prasher, 2014; Wu et al., 2015; Yu et al., 2011). Cations, especially  $\text{Na}^+/\text{K}^+/\text{Ca}^{2+}/\text{Mg}^{2+}$ , are ubiquitous in surface water and the hardness of surface water are various. However, few studies have investigated the effect of cations on the solubilization/deposition of HOCs concerning both the advantage of biodegradation and disadvantage of toxic transformed compounds.

Importantly, solutions with different cations can significantly affect the self-assembly behavior of surfactants, which in turn affect the fate of HOCs (Arutchelvi et al., 2014; Alargova et al., 2003; Chen et al., 2013; Helvacı et al., 2004; Sammalkorpi et al., 2009). Helvacı et al. (2004) found that the addition of NaCl could cause a decrease in CMC and surface tension of RL solution. Researchers discovered that the strong binding between divalent cations and anionic surfactant could cause the precipitation of RL (Alargova et al., 2003; Yu et al., 2008). However, Chen et al. (2013) found that  $\text{Ca}^{2+}$  had little effect upon the self-assembly of mono-rhamnolipid/di-rhamnolipid while it had a great impact on the self-assembly of RL/sodium dodecylbenzene-sulfonate (LAS). In addition, Bai et al. (1998) found that the effect of cations ( $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ) on the solubilization of hydrocarbon in RL solutions was different. The addition of  $\text{Na}^+$  and  $\text{K}^+$  could significantly increase the solubility of hydrocarbon in RL solution. But there was little effect on hexadecane's solubility for  $\text{Ca}^{2+}$  concentration merely greater than 0.2 mM. Moreover, the interaction among RL, sediment and TCS molecules with different cations is still not clear.

In order to regulate the fate of TCS and reduce environmental risks extremely, the effect of cations on the solubilization/deposition of TCS in sediment-water-RL systems was investigated by controlled batch experiments. The self-assembly of RL and the solubility of TCS in RL solutions were tested to explore the interaction among cations, TCS and RL. The influence of cations on the self-assembly of RL was monitored in terms of CMC, micellar size and zeta potential of RL solutions. The experimental data provided

meaningful information for the effectively controlled distribution of TCS by RL based on different cations and provided preliminary guide to the subsequent RL-enhanced remediation of TCS in sediment-water system.

## 2. Materials and methods

### 2.1. Materials

Triclosan (99%) and RL rude (90%, the mixture of mono- and di-RL,  $M = 541.1$  g/mol) were obtained from Alfa Aesar (Ward Hill, MA) and biological company (Huzhou, China), respectively. Analytical grade sodium chloride, potassium chloride, calcium chloride, and magnesium chloride were purchased from Sinopharm Chemical Reagent. Analytical grade sodium azide was provided by Solarbio. HPLC grade methanol was supplied by Merck (Germany). Background solutions were made by 0.01 M different cations ( $\text{NaCl}/\text{KCl}/\text{CaCl}_2/\text{MgCl}_2$ ) (the ionic strengths were within RL's tolerance range) (Wang et al., 2005) and 100 mg/L  $\text{NaN}_3$  was added to suppress microbial growth. Cations were maintained at a constant ionic strength in order to focus on the effect of cation type. Preliminary experiments showed that anion type had little effect on the solubilization/deposition of TCS in sediment-water-RL system. Influence of  $\text{NaN}_3$  on the solubility of TCS and the solubilization/deposition of TCS in sediment-water-RL system could be ignored. TCS stock solutions were prepared at 100 mg/L in methanol and stored in amber glass bottles at 4 °C.

### 2.2. Sediment preparation

The sediment sample was obtained from an alluvial estuarine site in Pearl River near the north shore of Yingzhou Ecological Park, Guangzhou China. The sediment was air-dried, gently disaggregated, sieved to a particle size of  $\leq 1$  mm and then stored in sealed amber glass jars prior to use. The selected physicochemical properties of the sediment were shown in Table 1. Preliminary experiments showed that the background concentration of TCS in sediment sample was below the detection limit (0.005 mg/L).

### 2.3. Determination of CMC

CMC was a characteristic feature of a given surfactant. The CMC values of RL with different cations were determined by the surface tension method. At the start of each set of experiments, surface tension of water was measured to calibrate the tensiometer. The measurements were carried out at  $25 \pm 1$  °C by the Wilhelmy plate technique with a BZY-1 model tensiometer (Shanghai Hengpin Instrument Co., Ltd., China). The CMC values were determined by plotting the surface tension versus the logarithm of RL concentration.

### 2.4. Determination of size distribution and zeta potential of RL solutions

The size distribution and zeta potential of RL solutions with different cations were determined by using Malvern NANO-ZS90 (Worcestershire, UK). The samples were made by dissolving RL (1250 mg/L, which was 25 times of pure RL CMC) in different background solutions as mentioned in section 2.1. The size distribution of RL aggregates in the presence and absence of cations were determined by the dynamic light scattering (DLS) at an angle of 90° at 298 K. Zeta potential of the samples was the measurement of RL micelles particles. It was measured based on the velocity of the suspended particles under an applied electric field (electrophoretic mobility) using laser Doppler velocimetry (Arutchelvi et al., 2014).

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