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Sorption and degradation of triclosan in sediments and its effect on microbes

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ABSTRACT

Sorption and degradation behavior of triclosan (TCS) and its effect on microbes were studied in three sediments spiked at different concentration levels (1, 10, and 100 μg g^{-1}). TCS showed a strong affiliation to all the sediments with linear adsorption coefficients (K_d) that varied from 220 to 1092 L g^{-1}, and the adsorption capacity is related to the total organic carbon (TOC) contents of the sediments. The half-lives of TCS varied from 55 to 239 days, and were longer in sediment with higher K_d. TCS showed minor effect on the activities of fluorescein diacetate hydrolase, dehydrogenase, alkaline phosphatase, and urease in the 1 μg g^{-1} treatment, but at higher levels, a short-term effect was observed followed by a rapid recovery except the urease activity in sediment with the lowest adsorption capacity. PCA plots of phospholipid fatty acid showed that the phenotypic community in sediments with low TOC were more sensitive to TCS. A positive relation between bacterial biomass and total microbial biomass suggests that changes of bacteria biomass were responsible for changes of total microbial biomass in treatments. Denaturing gradient gel electrophoresis analysis of the 16 S rDNA showed that the bacterial community structure deviated further away from the control at higher TCS concentration levels, with similarity coefficients in Un-weighted Pair Group Mathematics Average clustering between control and 100 μg g^{-1} treatment varied from 0.38 to 0.73. Both degradation rate and toxic effects of TCS decreased in sediment with higher sorption capacity, which can be attributed to a reduced bioavailablity.

1. Introduction

Triclosan (TCS) is a broad-spectrum antibacterial agent widely used in many personal care products. It has been frequently detected in the environment worldwide, especially in waters receiving effluent discharge (Bester, 2005). Although TCS can be efficiently removed from the wastewater with overall removal rates up to 94%, TCS residuals were still detected in wastewater effluent (42–213 ng L^{-1}) and in rivers receiving effluent discharge (11–98 ng L^{-1}) (Singer et al., 2002). Sediment shows great capacity for TCS accumulation due to a hydrophobic property of TCS, and therefore can be an important sink for TCS. In sediment from the Shijing River (Pearl River system) in China, TCS varied from 345 to 1329 ng g^{-1} with an average concentration of 739 ng g^{-1} (Zhao et al., 2010). In a tributary of the Rine River, TCS was detected up to 450 ng g^{-1} in sediment (Kronimus et al., 2004). Study on the dated sediment cores from urbanized estuaries indicated the preservation and accumulation of TCS (Cantwell et al., 2010). Therefore, it is important to understand the fate and effect of TCS end up into sediments.

TCS inhibits bacterial growth by blocking lipid biosynthesis via inhibition of the enzyme enoyl-ACP reductase (Surolia and Surolia, 2001). The ubiquitous presence of TCS in the environment arouses growing concern on its potential ecological effect on microbes. Previously, TCS can cause a decline in bacterial population in river biofilms at environmentally relevant concentrations (Ricart et al., 2010). Following TCS addition, inhibition of microbial respiration in a sandy soil was reported (Waller and Kookana, 2009), and alteration of the fungal to bacterial ratio was also observed in soils (Butler et al., 2012). In anaerobic digesters, TCS amendment decreased the methane production and selected resistant bacteria (McNamara et al., 2014). However, effects of TCS on microbes in sediments have not been well characterized.

In sediments, the fate and effect of organic pollutants are affected by their bioavailabilities. Organic compounds become progressively less available for degradation and for exerting inhibitory effects when sorb to soils or sediments. For instance, the effect of

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ciprofloxacin on microbial communities was inversely correlated to the degree of sorption to the sediments (Cordova-Kreylos and Scow, 2007). Mineralization of 2-methyl-4-chlorophenoxyacetic acid (MCPA) showed an inverse relationship with sorption capacity, and only occurred in the water extractable pool of MCPA (Jensen et al., 2004). The aims of this work are to assess the adsorption, desorption and degradation of TCS, and its effects on microbes in sediments. The influence of sorption on degradation and effects of TCS are discussed.

2. Materials and methods

2.1. Sediment and water samples

Three sediments with different properties were used in this study. Surface sediments and associated water were collected from Donghu Lake, Xiangxi River and Hewangmiao River, China. Donghu Lake (DL) is the largest urban lake in China with a total surface area of 32 km^2 located in Wuhan City. Xiangxi River (XXR) is one of the longest Yangtze River tributary in the lower reach of the Three Gorges Reservoir (TGR). Hewangmiao River (HWM) is an old course of Yangtze River located in Jinzhou city with a total length of about 33 km. The surface sediment samples (0–11 cm) were obtained with a Petersen Grab sampler and sealed in polyethylene bags. Their associated water samples (0.5 m in depth) were collected from the same site by a 5 L acrylic hydrophore and stored in 1 L amber glass bottles. Samples were transported back to the laboratory in a cooler as soon as possible. The sediment samples were passed through a 2 mm sieve to remove large debris, plant litter, and zoobenthos. Water samples were filtered through Whatman GF/C glass fiber filters to remove suspended materials. Portions of the sediments were used for physicochemical property measurement. The rest of samples were stored at 4 °C for no more than 15 days before used in the experiment.

2.2. Adsorption and desorption experiments

Adsorption and desorption of TCS onto the sediments were performed in triplicate using a batch equilibrium method with reference to the Organization for Economic Co-Operation and Development Test Guidelines 106 (OECD, 2000). Aliquots of 0.5 g freeze-dried sediment samples were put in 50 mL Teflon tubes, filled with 40 mL of 0.01 M $CaCl_2$ (including 0.01% (w v^{-1}) NaN_3), and equilibrated overnight by shaking on a reciprocal shaker at 200 rpm at 25 °C in dark. Then, 40 μL of TCS standard solutions in methanol were added to each tube to achieve desired concentrations ranging from 0.25 to 3 mg L^{-1}. The samples were equilibrated for another 36 h and were centrifuged at 4000 rpm for 20 min, 1 mL supernatant was transferred to a 2 mL amber glass vial for the analysis, the remaining supernatant was decanted. Another 40 mL 0.01 M $CaCl_2$ solution was added to the tubes and single step desorption was carried out by shaking for 36 h in dark. Then, the samples were centrifuged as in the adsorption experiment. Preliminary equilibrium experiments were performed for 120 h and results indicated that no significant change of TCS residual in liquid phase occurred after 36 h for both adsorption and desorption, suggesting that 36 h was sufficient for the apparent equilibrium. TCS concentrations in liquid phase were analyzed by high-performance liquid chromatography (HPLC) connected to a photodiode array detector (DAD).

2.3. Microcosm setup

Sediment and associated water were acclimated for 2 weeks in a 3 L beaker at 25 °C. After removing the overlying water, the sediment was spiked with TCS in dimethylsulfoxide (DMSO) to achieve a concentration of 1, 10, and 100 μg g^{-1} in the sediments. The same volume of DMSO was added to the control. Spiked sediments were thoroughly mixed by hand using a spatula, and aliquots of 9 g sediment (on dry weight basis) were transferred into the 100 mL serum bottles. The samples were equilibrated in the dark for 2 h at 25 °C and the overlying water was added slowly by a siphon. All bottles were wrapped with aluminum foil, and were covered with membranes contain a 0.2 μm filter (Zhentai, Beijing, China) to allow air exchange. During the incubation, the concentration of dissolved oxygen (DO) in the overlying water was monitored in every fourth day using a Mettler (Columbus, OH, USA) SevenGo Pro meter. When necessary, the oxygen in the overlying water was replenished by aeration with a stone air diffuser about 1.5 cm above the surface of sediment to ensure that the DO level was above 3 mg L^{-1}. The disturbance of the water–sediment interface was minimum. Three bottles were withdrawn from each group on day 0, 2, 4, 8, 13, 27 and 48. After thorough mixing, 1 g (fresh weight) of sediment was used for the measurement of each enzyme activity immediately, and the remaining sediments were freeze-dried and stored at −20 °C for phospholipid fatty acids (PLFA), denaturing gradient gel electrophoresis (DGGE) and TCS analysis.

2.4. Enzyme activities

Four enzymes activities (fluorescein diacetate hydrolase, dehydrogenase, alkaline phosphatase and urease) were measured in treated sediment. Fluorescein diacetate (FDA) hydrolysis assay are used to estimate the total microbial activity, because of the ubiquitous presence of lipase, protease, and esterase, which involve in the hydrolysis of FDA. Phosphatase catalyzes the hydrolysis of organic phosphorus (P) to inorganic phosphorus, and urease catalyzes hydrolysis of organic nitrogen (N) into ammonia. Thus, alkaline phosphatase (APA) and urease (UA) are responsible for P and N cycles in sediments. Dehydrogenase (DHA) reflects the total oxidative activity of microbes, which play a significant role in the biological oxidation of organic matter.

FDA hydrolysis assay was measured as described by Adam and Duncan (2001). DHA and APA activity were measured with reference to Hadas and Pinkas (1997) and Mosher et al. (2003). UA activity was determined using the method described by Kandeler and Gerber (1988). Two controls, one without the addition of substrate and another one containing only buffer and substrate, were used for each enzyme activity. The unit of enzyme activity was expressed as the μg of substrate hydrolyzed by 1 g of dried sediment per hour.

2.5. TCS extraction and analysis

An aliquot of 1 g freeze-dried sediment was extracted three times using ultrasonication with acetone. Each time 4 mL acetone was used and extracted for 15 min. The extract from each time was combined and evaporated to dryness under a gentle nitrogen stream, reconstituted to 1 mL with methanol after adding 0.1 mL simatone (1 mg L^{-1}) as internal standard. TCS in the liquid phase was analyzed directly without any treatment. TCS was analyzed using a Waters (Milford, MA, USA) 2695 series HPLC connected to a 2996 series DAD. A Phenomenex (Torrance, CA, USA) Luna C8 column (100 × 4.6 mm × 3 μm) was used for the separation. The instrumental conditions and quantification method are described in detail elsewhere (Huang et al., 2014).

2.6. PLFA analysis

On day 48, sediment samples from three replicates were