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Anaerobic degradation of tetrachlorobisphenol-A in river sediment

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ABSTRACT

This study investigated the anaerobic degradation of tetrachlorobisphenol-A (TCBPA) in sediment samples collected at three sites along the Erren River in southern Taiwan. TCBPA anaerobic degradation half-lives ($t_{1/2}$) in the sediment were 12.6, 16.9 and 21.7 d at concentrations of 50, 100, and 250 μg g^{-1}, respectively. TCBPA (50 μg g^{-1}) anaerobic degradation half-lives ($t_{1/2}$) in the sediment were 10.1, 11.8, 11.0, 11.6, 10.8, 9.1, 8.5, 18.2, 19.3, and 16.1 d by the addition of yeast extract (5 mg l^{-1}), cellulose (0.96 mg l^{-1}), sodium chloride (1%), brij 30 (130 mg l^{-1}), brij 35 (43 mg l^{-1}), rhamnolipid (55 μM), surfactin (91 μM), phthalic esters (2 mg l^{-1}), nonylphenol (2 mg l^{-1}), and heavy metals (2 mg l^{-1}), respectively. The degradation rate of TCBPA was enhanced by the addition of yeast extract, cellulose, sodium chloride, brij 30, brij 35, rhamnolipid, or surfactin. However, it was inhibited by the addition of phthalic esters, nonylphenol, or heavy metals. Also noted was the presence of dichlorobisphenol-A and bisphenol-A, two intermediate products resulting from the anaerobic degradation of TCBPA accumulated in the sediments.

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1. Introduction

Tetrachlorobisphenol-A (4,4-isopropylidenebis [2,6-dichlorophenol], TCBPA) is used as an additive in polymers, epoxy and polycarbonate resins, high impact polystyrene, phenolic resins, and adhesives (Horikoshi et al. 2008). Due to its hydrophobicity, it tends to adsorb onto surface water particles and sediments and accumulate in aquatic organisms (Blanco et al. 2006). It has shown significant thyroid hormonal activities as well as oestrogenic activity (Kitamura et al. 2002; Sun et al. 2009), and it is suspected to be carcinogenic (Meerts et al. 2000).

Possible fates for TCBPA released into the environment include volatilization, photo-oxidation, chemical oxidation, bioaccumulation, and adsorption on to sediment particles. The principal processes for its successful removal are currently believed to be microbial transformation and degradation. Although TCBPA is toxic to the environment, detailed studies on the fate of TCBPA in aquatic environments are lacking. There are only two other studies on this specific area. Voordeckers et al. (2002) examined the biotransformation of TCBPA in estuarine sediments. Yuan et al. (2010) investigated the effects of various factors on the aerobic degradation of TCBPA in the sediment.

The fate of TCBPA in anoxic sediment, where it may be subject to anaerobic reductive dehalogenation, is thus of great interest. In order to enhance the efficiency of biodegradation, three remedial strategies, namely natural attenuation, bioaugmentation, and biostimulation have been proposed (Yu et al. 2005). The addition of yeast extract, surfactant, cellulose, or sodium chloride has been shown to influence the anaerobic degradation of organic pollutants in the sediment (Chang et al. 2004, 2005, 2006, 2009). However, little is known about the effects of various factors on the anaerobic degradation of TCBPA in river sediment.

The Erren River in southern Taiwan has been subjected to continuous pollution, with pollution sources including municipal sewage, industrial waste, and emissions from pig farms. The biodegradation of nonylphenol, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and phthalate esters (PAEs) was investigated in the river (Chang et al. 2004, 2005, 2006; Yuan and Chang 2007). The aims of this study were to investigate the anaerobic degradation of TCBPA in Erren River sediment.

2. Materials and methods

2.1. Chemicals

TCBPA of 98.0% analytical standard was purchased from the Sigma Chemical Co. (St. Louis, MO). Solvents were purchased from Mallinckrodt, Inc. (Paris, KY). All other chemicals were purchased from the Sigma Chemical Co. (St. Louis, MO). Individual stock solutions of TCBPA dissolved in chloroform were established at a concentration of 10,000 mg l^{-1}, and then diluted to 500 mg l^{-1} before use.

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2.2. Production of biosurfactants

The biosurfactants used in this study were surfactin and rhamnolipid. The surfactin was produced using *Bacillus subtilis* ATCC 21332 grown on an iron-enriched mineral salt medium at 30 °C. The details of surfactin production can be found in Yeh et al. (2005). The rhamnolipid was produced using *Pseudomonas aeruginosa* J4, an indigenous bacteria isolated from petrochemical wastewater. The isolate was grown in Luria–Bertani (LB) medium at 30 °C for rhamnolipid production. The details of rhamnolipid production can be found in Wei et al. (2005).

2.3. Sampling and medium

In July of 2008, sediment samples were taken from the Erren River — one of the most heavily contaminated rivers in southern Taiwan. Fig. 1 shows the locations of the three sample collection sites. The three sampling sites, A (22.55°10.98′N, 120.11° 3.51′E, B (22.55°14.32′N, 120.11°12.9′E) and C (22.54°51.13′N, 120.13°27.01′E) are downriver and are well known from previous studies of aquatic pollutants (Chang et al. 2004). The deep sediments (>15 cm) were collected during low tides using a soil core. All sediment samples were taken randomly, in triplicate, from an area of around 1 m^2 at the centre of each sediment site. For the sediment samples from sites A, B, and C, the organic carbon was 0.86, 1.30, and 0.65%, respectively; the pH was 7.2, 6.6, and 6.8, respectively; and the anaerobic count was 3.21×10^5, 6.7×10^7, and 6.7×10^6 CFU g^{-1}, respectively; TCBPA concentration was 460.8 ng g^{-1}, 542.6 ng g^{-1}, and 238.9 ng g^{-1}, respectively (Yuan et al. 2010).

The experimental medium consisted of (all concentrations in g l^{-1}): NH_4Cl, 2.7; $MgCl_2 \cdot 6H_2O$, 0.1; $CaCl_2 \cdot 2H_2O$, 0.1; $FeCl_2 \cdot 4H_2O$, 0.02; K_2HPO_4, 0.27; KH_2PO_4, 0.35; and resazurin, 0.001. The pH was adjusted to 7.0 following autoclaving; 0.9 mM titanium citrate was added as a reducing reagent.

2.4. Experimental design

All the experiments were performed using 125 ml serum bottles containing 45 ml of medium, 5 g of river sediment and 50 μg g^{-1} of TCBPA. TCBPA concentration and anaerobic degradation were first measured in the sediment samples collected from sites A, B, and C. The effects of the following factors on anaerobic degradation in sediment collected from site B were then measured: substrate concentration (50, 100, and 250 μg g^{-1}); yeast extract (5 mg l^{-1}); sodium chloride (1%, wt/vol); cellulose (0.96 mg l^{-1}); the phthalic esters (PAEs) di-*n*-butyl phthalate (DBP) and di-(2-ethylhexyl) phthalate (DEHP) (2 mg l^{-1}); nonylphenol (2 mg l^{-1}); a mixture of four heavy metals (2 mg l^{-1} Cd, 2 mg l^{-1} Cu, 2 mg l^{-1} Pb, and 2 mg l^{-1} Zn); the surfactants rhamnolipid, surfactin, brij 30, and brij 35 at a concentration of 1 CMC (the CMC values for brij 30, brij 35, rhamnolipid, and surfactin were determined to be 55 μM, 91 μM, 130 mg l^{-1}, and 43 mg l^{-1}, respectively (Wei et al. 2005; Yeh et al. 2005; Chang et al. 2009); and sodium hydrogen carbonate (20 mM) to create methanogenic conditions, sodium sulfate (20 mM) to create sulfate-reducing conditions, or sodium nitrate (20 mM) to create nitrate-reducing conditions (Chang et al. 2009). Inoculated control samples (without sodium hydrogen carbonate, sodium sulfate, or sodium nitrate), which were considered as non-sterile sediment, were shaken prior to incubation at 30 °C and pH 7.0 in the dark. Sterile controls were autoclaved at 121 °C for 30 min on two consecutive days.

All the experiments were conducted in an anaerobic glove box (Forma Scientific, USA) filled with N_2 (85%), H_2 (10%), and CO_2 (5%) gases. 125 ml serum bottles were capped with butyl rubber stoppers, wrapped in aluminium foil to prevent photolysis, and incubated without shaking at 30 °C in the dark. Each treatment was performed in triplicate. Bottles were vigorously shaken to ensure mixing, and a sediment sample (2 ml) was withdrawn by use of a deoxygenated syringe fitted with an 18-gauge needle. Samples were collected every 1 or 2 d to measure residual TCBPA, pH, and ORP values. Methane was sampled from the headspace of serum bottles.

2.5. Analytical methods

TCBPA was extracted twice from sediment samples using hexane and acetone (9:1), then extracted again over 20 min at 30 °C with a Branson 5200 ultrasonic cleaner (Branson, USA). Extracts were analyzed using a gas chromatograph (Hewlett Packard 6820) equipped with an electron capture detector and HP-5 capillary column. The initial column temperature was set at 170 °C, increased by 6 °C min^{-1} to 220 °C, and increased by 7 °C min^{-1} to

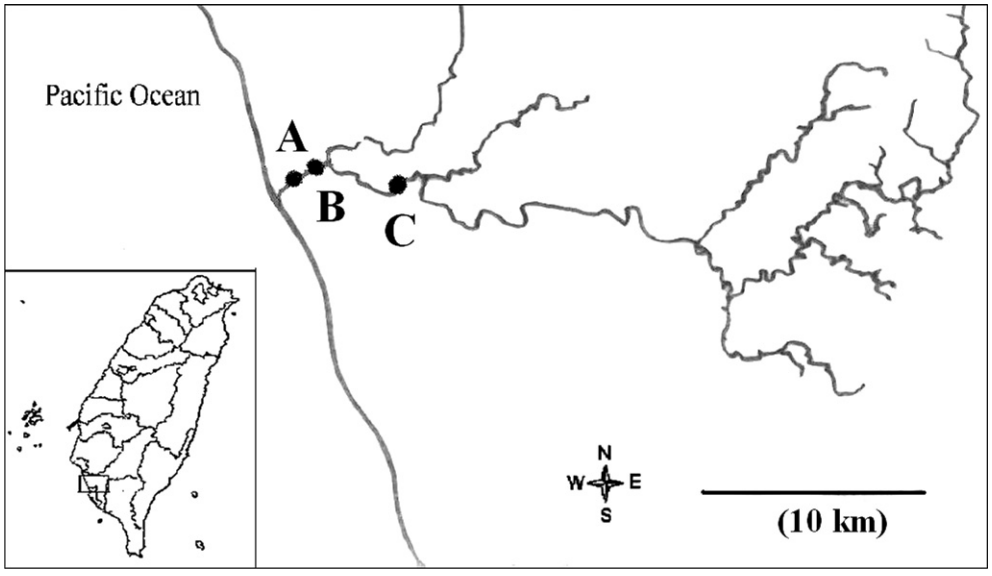

Fig. 1. Erren River sediment sampling sites in Taiwan.

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