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Effects of root exudates on the mobility of pyrene in mangrove sedimentwater system

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ABSTRACT

Mangrove ecosystems receive substantial polycyclic aromatic hydrocarbons (PAHs) from human activities. The mechanism of transport and translocation of PAHs within mangrove sediment is not well understood. In the present study, the mobility of pyrene in two typical mangrove sediments was studied using soil thin layer chromatography (TLC). The mobility of pyrene in the sediments displayed an inverse trend with organic carbon (f_{oc}) content. As one of the important fraction in sediment organic matter pool, the influence of root exudates on adsorption of pyrene within the tested sediments was also evaluated. In particular, root exudates significantly enhanced the bioavailability and mobility of pyrene in the sediments. The effectiveness of root exudates in promotion of pyrene mobility follows an order: citric acid > oxalic acid > malic acid > artificial root exudates (ARE). In addition, mobility factors of pyrene from root exudate treatments were significantly higher than the value determined in the control, and the magnitude of mobility factor (R_f) showed a positive correlation with root exudate concentrations. Results of this study could promote our understanding of the mobility of PAHs in contaminated mangrove sediments.

1. Introduction

Hydrophobic organic pollutants such as polynuclear aromatic hydrocarbons (PAHs), are ubiquitous in coastal ecosystems derived from a series of human activities, such as oil spills, urban runoff and industrial processes [\(Wei et al., 2014; Zhang et al., 2017\)](#page--1-0). Mangrove ecosystems are important habitats, located in tropical and subtropical inter-tidal regions. Due to the high productivity of mangrove forests, mangrove sediments usually have high organic matter content and show a low redox potential ([Jiang et al., 2016\)](#page--1-1). As the interaction zone between land and ocean, mangrove systems receive substantial PAHs, reaching > 10 mg kg⁻¹ in the heavily polluted zone ([Tam et al., 2001](#page--1-2)). PAHs are carcinogenic, mutagenic and teratogenic, they can be concentrated in marine animals through the food chains ([Jia et al., 2016c](#page--1-3)), eventually getting into human body and cause disease ([Dodor et al.,](#page--1-4) [2004\)](#page--1-4). Recently, a growing interest in the distribution and toxicity of PAHs to mangrove sediment are widely reported [\(Ahammed et al.,](#page--1-5) [2012; Lu et al., 2011; Wang et al., 2014](#page--1-5)).

In comparison, the mechanism of transport and translocation of PAHs within mangrove sediments, especially interaction between the sediment and pore water is not well studied. Due to the high hydrophobicity and low water soluble of PAHs, bioavailability of these organic pollutants have a limit act on solid to the liquid phase mass transfer in soil - water system [\(An et al., 2011\)](#page--1-6). Previous researches have shown that the process of mobility mechanisms is complex, and our knowledge on the mobility of soil pollutants were affected by soil properties, soil-water solution properties, and the inherent nature of the pollutants [\(Ding et al., 2011; Majumdar and Singh, 2007\)](#page--1-7). In addition, recent studies showed that mobility of PAHs in mangrove sediment can be significantly influenced by plant roots exudates ([Jia et al., 2016a,](#page--1-8) [2016b\)](#page--1-8).

Plant roots secrete a wide range of organic compounds into the rhizosphere, known as root exudates, which are often divided into two groups: low and high-molecular weight exudates ([Haichar et al., 2014](#page--1-9)). Low-molecular-weight organic acids (LMWOAs) are chemically active. They can be easily involved in a series of rhizospheric processes ([Lu](#page--1-10) [et al., 2007\)](#page--1-10). For example, they can be substrates for microbial metabolism [\(Shukla et al., 2011](#page--1-11)), regulate plant growth, detoxify of harmful elements and increase the number of dissolved cations by complexing of metal ions [\(Jones, 1998; Tao et al., 2003; Lu et al., 2007\)](#page--1-12). The most frequently reported LMWOAs exuded from mangrove plant roots are citric, malic, acetic and oxalic acids [\(Lu et al., 2007; Wang et al., 2014](#page--1-10)). Published information on the mobility and availability of pollutants in soils [\(Schwab et al., 2008; Ding et al., 2011; Abdel-Shafy and Mansour,](#page--1-13)

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[2016; Cornelissen et al., 1998; Thorsen et al., 2004](#page--1-13)), less research interests are available which refers particularly to the mobility of PAH in rhizosphere environments, particularly the significance of mangrove plants.

Because the adsorption dynamics act as the fundamental support to the remediation process, investigations of the transport and translocation of organic pollutants and evaluation of factors that influence these procedures are critical. In the present study, we measured the mobility of pyrene (a high-ring PAH compound, which commonly observed in coastal zones and kept longer retention time in sediment/soil than lowring PAH) in two mangrove sediments, using soil thin-layer chromatography (TLC), developed by [Helling and Turner \(1968\)](#page--1-14) in order to analyse the relationship between PAHs mobility and organic matter (OM) content. We also assessed the influence of artificial root exudates (AREs) and LMWOAs on the mobility of pyrene in the sediments. Rhizosphere model systems, where sediment is given ARE have previously been used to study PAHs mobility in the rhizosphere ([Gao et al., 2010](#page--1-15)). ARE used in this study contained 12.5 mM amino acids (arginine, serine and cysteine), 25 mM organic acids (succinic and malic acid), 50 mM sugars (glucose, fructose and sucrose) [\(Joner et al., 2002](#page--1-16)). The objectives of the study are (i) to compare the distribution and mobilization of total pyrene among two sediments with different OM content by root exudates, (ii) to examine the activation and bioavailability of pyrene in contaminated sediments at different root exudate concentrations.

2. Materials and methods

2.1. Materials

Pyrene with a purity of 99%, acetone, methanol, dichloromethane and acetonitrile (HPLC grade) obtained from Sigma Aldrich Chemie BV. Other chemical reagents were analytical reagents and purchased from the Sinopharm Co. Ltd. in Shanghai, China.

The two typical mangrove sediments are silty in the Jiulong Estuary and sandy in the Dongzhai Harbour sediments, both of them low in pyrene, were collected during low tides using soil sample barrels form Fujian (24°24′N, 117°55′E) and Hainan (19°55′N,110°36′E), respectively [\(Table 1](#page-1-0)). The sampling depth was 0-20 cm from the surface layer of the sediment.

Some properties of ARE stock solution are listed in [Table 2,](#page-1-1) ARE used in this study was adapted from [Gao et al. \(2010\)](#page--1-15) and [Joner et al.](#page--1-16) [\(2002\).](#page--1-16) In this paper, we focused on the investigation of pyrene mobility in sediments using ARE, citric, oxalic and acetic acid.

2.2. Experimental design

The mobility of pyrene in mangrove sediments was measured through a thin-layer chromatography (TLC) experiment ([Fig. 1](#page--1-17)). Mixture of sediments (50 g) and distilled water (50 mL) were spread onto a 20×10 cm clean glass plate, then dried at room temperature. The thick layer of the mixture was 0.5 ± 0.05 mm. Aliquot (100 μL) of experimental spotting solution of pyrene dissolved in methanol were placed onto the plates. Plates were developed in root exudates solution

Physicochemical properties of the sediments studied.

Two sediments were collected from the Jiulong Estuary in Fujian and the Dongzhai Harbour in Hainan during low tides using soil sample barrels, respectively (this table).

Some properties of ARE stock solution.

Rhizosphere model systems - artificial root exudates (ARE) used in this study were shown in this table.

(0, 10, 160 mmol⁻¹) taking 0.005 mmol L⁻¹ CaCl₂ as spreader ([Flessner et al., 2014](#page--1-18)), afterwards, the plate was carried out and dried at 22 ± 2 °C without changing the slop until the excess of solvent evaporated. Distance from the baseline (2 cm) to the foreline (18 cm) was divided into eight partitions, and the concentration of pyrene detected in each partition was analyzed separately.

The pyrene concentrations in the glass plates at different partitions indicated the dynamic variations of pyrene in the glass plates during developed by root exudates. In order to elucidate the effects of root exudates on the mobility of PAHs in a sediment-water system, we partitioned the sediment column into eight partitions, and see how it changed with the addition of root exudates. This experiment provides a representation of breakthrough gained from multiple samples in distance and integrating physicochemical aspects of transporting behavior.

To investigate these changes, the mobility factor (R_f) was calculated according to [Jia et al. \(2016a\)](#page--1-8).

$$
\overline{Z_p} = \frac{\sum Z_i \times M_i}{\sum M_i} \tag{1}
$$

$$
R_f = \frac{\overline{Z_p}}{Z_w} = \frac{\sum Z_i \times M_i}{Z_w \times \sum M_i}
$$
\n(2)

where, \overline{Z}_p was the average distance that pyrene transported from the start point; Z_w was the distance moved by the developing solvent from the start point; i was the number of segments; Z_i was the distance of the segment *i* from the start point, and M_i was pyrene content in segment *i*.

2.3. PAH analysis

The concentration of pyrene in sediments was determined according to our previous studies ([Jia et al., 2016a, 2016b\)](#page--1-8). Sediment samples (0.5 g) were treated using an ultrasonic extraction with 15 mL dichloromethane for 2 h. After extraction, the mixture was centrifuged, and then the supernatant liquid (2 mL) was evaporated in a vacuum to dry by rotary evaporation. Finally, the extract was dissolved with 2 mL acetonitrile, and filtered through a 0.22 μm membrane prior to analysis. An external standard was used for quantification of PAHs. The extraction efficiency of PAH was about 90% from freshly spiked sample.

The pyrene level in extracted solution was quantified using a high performance liquid chromatography (HPLC, Shimadzu, Japan) equipped with fluorescence detector. Chromatographic separation was performed on a reversed phase C_{18} column (4.6 \times 250 mm) with methanol: water (85:15). The excitation and emission wavelength was 270 and 390 nm, respectively.

2.4. Fourier transform infrared spectroscopy (FT-IR) analysis

The functional groups of the two sediment samples were recorded in

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