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Interactive effects of cadmium and pyrene on contaminant removal from co-contaminated sediment planted with mangrove *Kandelia obovata* (S., L.) Yong seedlings

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ABSTRACT

The interactive effects of cadmium (Cd) and pyrene (Pyr) on contaminant removal from co-contaminated sediment planted with *Kandelia obovata* were investigated by a pot experiment. We found that dry weight of plant was significantly decreased under high level of Cd–Pyr combined stress. High Pyr caused the increase of Cd toxicity to *K. obovata* under high Cd stress because more Cd translocated to the plant tissues. Cd toxicity inhibited Pyr degradation in co-contaminated sediments and higher Pyr degradation was found in the rhizosphere than that in the non-rhizosphere sediment under high Cd treatment. The total number of microorganisms in sediments tended to decrease with increasing Cd under Cd–Pyr combined stress and more amount existed in the rhizosphere sediment. In conclusion, Cd and Pyr removal by *K. obovata* can influence interactions between these two pollutants in co-contaminated sediment. This suggests that this mangrove can effectively remedy sites co-contaminated with these two types of contamination.

1. Introduction

Sites polluted with heavy metals are frequently affected by different types of organic pollutants. It has been reported that 40% of hazardous waste sites on the U.S. Environmental Protection Agency's national priority list (NPL) are co-contaminated with organic and heavy metal pollutants (Sandrin and Maier, 2002; Cheng, 2003). Heavy metals and polycyclic aromatic hydrocarbons (PAHs) are frequently becoming hazards in soil environments as a result of wastewater irrigation, solid waste disposal, sludge applications, automobile exhausts, and industrial activities (Shi et al., 2005). Soil contamination by heavy metals and PAHs has been also accelerated in China during the past decade because of rapid urbanization and industrialization (Sun et al., 2011; Yang et al., 2011). In particular, these two pollutants may be accumulated in the substrate when wetlands come into contact with wastewater over extended periods of time (Batty and Younger, 2004; Srogi, 2007). Recently, there have been increasing concerns over co-contaminated heavy metals and PAHs (Singer et al., 2007; Almeida et al., 2008; Lin et al., 2008; Batty and Anslow, 2008; Yang et al., 2011; Wang et al., 2012, 2013) due to their persistence, carcinogenic potential, mutagenic and teratogenic properties and their ubiquitous occurrence in the environment (Murakami et al., 2009; Teng et al., 2011).

The combined presence of different pollutants might influence remediation processes, because different compounds may interact among themselves and/or with plants and their rhizosphere biota (Almeida et al., 2008). Previous studies have shown that the presence of PAHs may change the mechanisms of metal uptake, thus influencing kinetics, extent of metal phytoextraction and tolerance mechanisms between plants and heavy metals. For instance, the presence of Pyr in copper (Cu) contaminated soils decreased phytoextraction efficiency of Cu by *Zea mays* L. (Lin et al., 2008). PAHs may modify Cu solubility and sorption by *Halimione portulacoides*, and/or the passive penetration of Cu into the root cells (Almeida et al., 2008). The uptake of zinc (Zn) by Indian mustard (*Brassica juncea*) was greater from a soil co-contaminated by Pyr and Zn as compared to that from Zn-only contaminated soil (Batty and Anslow, 2008). Cd phytoextraction would be inhibited under contamination of Pyr (Zhang et al., 2009). Mixture of low Cd and low PAH increased Cd accumulation in plant tissues of *Juncus subsecundus*, thus enhancing Cd removal by this species (Zhang et al., 2011). Wang et al. (2012) demonstrated that both phenanthrene (Phe) and Pyr exhibited negative effects on phytoextraction of Cd from Cd spiked soil. In the other hand, in heavy metal–organic pollutant combined systems, previous studies have shown that biodegradation of organic contaminants is often severely inhibited

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by toxic metals in both aerobic and anaerobic co-contaminated systems (Maslin and Maier, 2000; Sandrin and Maier, 2003). The occurrence of Cd, Cu and Pb had inhibitive effects on benzo[a]pyrene uptake and accumulation by *Tagetes patula*, simultaneously (Sun et al., 2011). In some cases, however, the addition of metals has been observed to stimulate biodegradation of organic contaminants (Kuo and Genthner, 1996). Nevertheless, the effects of metal toxicity on organic pollutant biodegradation and/or organic pollutants on the phytoextraction of metal are poorly understood.

Mangroves are an important intertidal habitat distributing in the tropics and subtropics and playing a significant role in protecting embankments and maintaining coastal ecological balance and species diversity. As a result of high organic matter content and sulphur and low redox potential in the sediments (Harbison, 1986; Silva et al., 1990), as well as high concentrations of clay and organic clastic fine particles and slow rate of tide current (Preda and Cox, 2002), heavy metals are abundantly retained in mangrove forests which act as sinks for pollutants from river and marine sources (Harbison, 1986; Tam and Wong, 1996). Despite potential exposure of mangrove plants to metal contaminated sediments, mangroves seem to possess a great tolerance to high levels of heavy metal pollution (MacFarlane et al., 2007) due to they have developed various mechanisms such as exclusion, chelation, compartmentalization and sequestration for heavy-metal tolerance. Mangrove ecosystems are closely tied to human activities and are subject to PAHs contamination (Tam et al., 2002). PAHs in mangrove ecosystems are introduced from a number of routes including tidal water, river water and land-based sources (Klekowski et al., 1994), run-off and erosion, effluent discharges, atmospheric deposition, naturally occurring biological processes, as well as from shipping and oil spills (Zhang et al., 2004). Anoxic conditions and high levels of organic carbon are preferential for uptake and conservation of PAHs (Bernard et al., 1996), thus PAHs in marine sediments have a strong tendency to concentrate in marine food webs and cause potential health risks to humans. Mangrove plants can enhance metal removal and/or stabilization (Ye et al., 2012) and may also facilitate biodegradation of organic pollutants such as PAHs directly in the rhizosphere by the release of root exudates, and indirectly by improving soil biology via buildup of organic carbon (Lu et al., 2011; Fang et al., 2012). Plant root-promoted dissipation was found to be the predominant contribution to the remediation enhancement for sediment Phe and Pyr in the presence of mangrove plant *Kandelia obovata* (Lu et al., 2011).

Questions to be addressed are: can PAHs influence heavy metal accumulation by mangrove plants? Or can heavy metals influence the degradation of PAHs in mangrove sediments? So far, although there is some existing literature on the efficiency of metal phytoextraction in soils contaminated with mixture of inorganic and organic pollutants, studies concerning mangrove are to our knowledge, non-existent. In this investigation, *K. obovata* was applied as the test material because it is widely distributed in mangrove wetlands along the south China coastline and is quite adaptive to adverse environmental conditions (Lu et al., 2007; Qin et al., 2007), and this plant can produce strong rhizosphere effect because of its well-developed root system. Pyr was used as the model organic species in this study because it represents a class of organic compounds, polyaromatic hydrocarbons, with carcinogenic potential that are present at many USA superfund sites. Cd was chosen as the metal as it is the second most common metal found at superfund sites and is one of the ten high priority pollutants. The objectives of this study were to investigate (1) the effect of Pyr on growth of *K. obovata* treated with Cd; (2) the effect of Pyr on Cd removal by *K. obovata* treated with Cd; and (3) the effect of Cd on the Pyr degradation in the sediments treated with Pyr; and (4) the effect of Cd on the total number of microorganisms in sediments treated with Pyr.

2. Materials and methods

2.1. Plant materials and culture conditions

Mature *K. obovata* propagules and sediments were collected from Jiulong River mangrove natural reserve (24°24′N, 117°55′E), Fujian China, in April 2011. Complete undamaged propagules with testa intact and high vitality were chosen for planting. Every three *K. obovata* propagules were planted in a seedling cultivation pot and every three cultivation pots were placed in a rubber pot (35 cm in diameter, 15 cm in depth) filled with washed clean sand. The water level was kept at 2 L added with 1/4 Hoagland's nutrient solution which was replaced once a week. These seedlings were prepared for planting in Cd–Pyr co-contaminated sediment. The experiment was conducted in a greenhouse with natural light, day/night temperatures, and relative humidity of 33/25 °C and 65/8 5%, respectively.

2.2. Sediment treatment

Sediment was collected from the top layer (0–20 cm) of mangrove forest at the Jiulong River mangrove natural reserve (24°24′N, 117°55′E), Fujian China. Their physicochemical properties are presented in Table 1. The sediment was air-dried and sieved through a 3 mm mesh. Levels of cadmium and pyrene mixed into the sediment were 0, 5, 10, 20, 40 mg Cd kg^{-1} sediment and 0, 10, 50 mg Pyr kg^{-1} sediment. Bulk sediment was first mixed thoroughly with Cd (as $CdCl_2 \cdot 2.5\ H_2O$) in an aqueous solution and incubated under moist conditions for 30 days. Subsamples containing Pyr were prepared with the above material. Sediment was taken and spiked with Pyr (99.9% purity, which was obtained from Sigma–Aldrich Co. Ltd.) as follows: a portion of the sediment was accurately weighed in the vessel. A volume of the Pyr dissolved in acetone was added and allowed to equilibrate with the matrix, stored in the dark and allowed to dry. The acetone was evaporated for 12 h and the portion of spiked sediment was first mixed with near 25% of total sediment, and then mixed with the remaining 75% of wet sediment followed by mechanical mixing (Lu et al., 2011). After Pyr ageing for 7 days, the sediment was used for the pot experiment.

2.3. Experimental design

After 37 days, the similar apparent health seedlings which planted in sand cultivation pot were transplanted into rhizobag. Each selected seedling had one pairs of leaves. In order to separate the rhizosphere from non-rhizosphere sediment, a cylindrical

Table 1
Physicochemical properties of sediment collected from the Jiulong River mangrove natural reserve, Fujian China ($n = 3$).

Parameter	Mean value ± S.D.
pH	6.68 ± 0.03
Total organic matter (%)	11.21 ± 0.12
Moisture content (%)	49.5 ± 0.53
Total N (mg kg^{-1} DW)	0.76 ± 0.02
Total P(mg kg^{-1} DW)	0.83 ± 0.01
Pyrene (mg kg^{-1} DW)	0.038 ± 0.00
Cd (mg kg^{-1} DW)	0.79 ± 0.01
Cr (mg kg^{-1} DW)	64.36 ± 0.87
Cu (mg kg^{-1} DW)	34.38 ± 0.98
Zn (mg kg^{-1} DW)	187.66 ± 4.32
Pb (mg kg^{-1} DW)	101.49 ± 2.56
Mn (g kg^{-1} DW)	1.51 ± 0.03
Total Fe%	4.87 ± 0.18

Note: Dates are shown as mean ± S.D. ($n = 3$).