

Bioconcentration of polycyclic aromatic hydrocarbons in roots of three mangrove species in Jiulong River Estuary

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Abstract: The polycyclic aromatic hydrocarbons (PAHs) concentrations were determined in the root of three mangrove species (*Kandelia candel*, *Avicennia marina* and *Bruguiera gymnorrhiza*) and their growing environment (sediment) in mangrove wetlands of Jiulong River Estuary, Fujian, China. The total PAHs (16 parent PAHs) in mangrove sediments ranged from 193.44 to 270.53 ng/g dw, with a mean value of 231.76 ± 31.78 ng/g dw. Compared with other mangrove and coastal marine sediments, the PAHs concentrations of all the sampling areas in this study were at relatively lower level. The total PAHs (13 parent PAHs) values varied from 30.83 to 62.73 ng/g dw in mangrove roots. Benzo[a]pyrene (five-ring), fluoranthene (four-ring) and pyrene (four-ring) dominated in mangrove sediments. Based on ratios of phenanthrene/anthracene, fluoranthene/pyrene and fluoranthene/pyrene + fluoranthene, the main possible sources of surface sediment PAHs were identified as grass, wood or coal combustion for mangrove wetlands of Jiulong River Estuary. Naphthalene (two-ring) and phenanthrene (three-ring) were the most abundant compounds in mangrove roots. Sediment-to-vegetation bioconcentration factors (BCF_{sv} s) were calculated and their relationships with PAHs' physico-chemical properties were investigated. The average BCF_{sv} s of PAHs for three mangrove species roots were almost all under the level of 1 except for naphthalene. Good linear relationship between BCF_{sv} values for mangrove roots and PAHs water solubility, octanol-water partitioning coefficients was derived in present study. The solubility and the octanol-water partition coefficient were proved to be good predictors for the accumulation of PAHs in mangrove roots, respectively.

Keywords: mangrove; PAHs; Jiulong River Estuary; bioconcentration factors

Introduction

Mangroves are intertidal wetlands, common in tropical and subtropical coastal regions, especially in bays and river estuaries. They are among the most biologically important nature resources on earth, promoting the diversities of terrestrial and aquatic organisms. As the primary producer, mangroves supply food for marine animals and also provide habitats for birds, insects, fishes, algae and bacteria. Estuary mangrove ecosystems are suffering from increasing environmental stresses due to the discharge of many pollutants caused by the rapid urbanization and industrialization of coastal regions. Mangrove pollution ecology therefore has aroused more and more public and scientific attentions (Lin, 1997; Zheng, 1997; Tam, 2001; MacFarlane, 2003).

Polycyclic aromatic hydrocarbons (PAHs) are widespread environmental contaminants, some of which may exhibit toxic, carcinogenic and mutagenic effects. Sixteen individual PAHs have been identified as priority pollutants by the United States Environmental Protection Agency (USEPA). PAHs arise from incomplete combustion and pyrolysis of fossil fuels (coal and petroleum) and other organic materials in flames, engines and industrial processes and from emissions of noncombustion derived petrogenic oil.

Mangroves' unique features such as high primary productivity, abundant detritus, rich organic matter and anoxic/reduced conditions possibly make them a preferential

site for uptake and preservation of PAHs from anthropogenic inputs (Bernard, 1996). Elevated concentrations of PAHs have been found in mangrove sediments (Klekowski, 1994). During the last decades, there has been considerable interest in understanding the uptake of PAHs by plants (Ryan, 1988; Jones, 1989; Wild, 1992; Kipopoulou, 1999). However, few study on bioconcentration of PAHs in mangroves have been reported (Lu, 2002). In the present paper, we made an attempt to analyze the PAHs concentration and distribution in three mangrove species *Bruguiera gymnorrhiza*, *Avicennia marina* and *Kandelia candel* roots and sediments of mangrove wetlands in Jiulong River Estuary and evaluate the possible sources of PAHs contamination in the wetland. Sediment-to-vegetation bioconcentration factors were calculated, and their relationships with the physico-chemical properties of PAHs were also investigated. The aim is to reveal the relationships between PAHs accumulated in mangroves roots and their concentrations in sediments.

1 Materials and methods

1.1 Study area

The study area is located in Mangrove Nature Reserve of Jiulong River Estuary (24°24'N, 117°55'E), Fujian. Three sample sites were selected in *B. gymnorrhiza* forest in Fugong Town (FG), *A. marina* forest on the north shore of Haimen Island (HMa) and *K. candel* forest on the south shore of Haimen Island (HMb; Fig. 1).

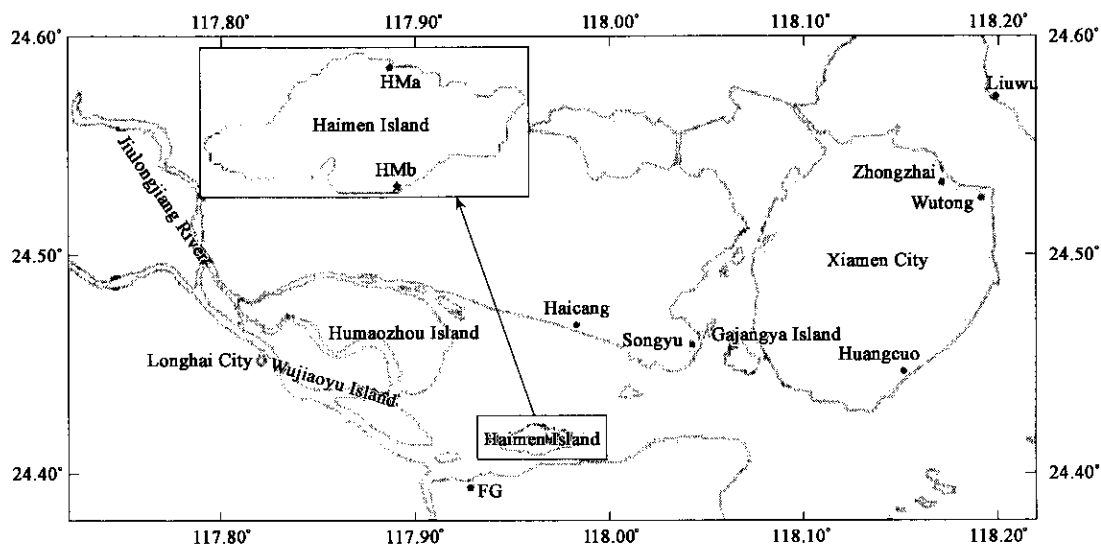


Fig. 1 Map showing the sampling sites in the Jiulong River Estuary from this study

K. candel, *A. marina* and *B. gymnorrhiza* root samples were randomly collected from three sample sites, respectively. Surface sediments (up to 30 cm depth) were sampled with a stainless steel grab sampler from the same locations simultaneously with the mangrove roots. The characteristics of the mangrove communities have been reported by Lin *et al.* (Lin, 1987). All the samples were transported to the laboratory immediately.

1.2 Methods of analysis

The mangrove root samples were carefully washed with running distilled water to remove surface silt. All samples were freeze-dried, grounded into a fine powder and filtered through 60 mesh screen sieve, then stored in dark glass bottles and kept dry prior to PAHs analysis.

Ten gram dried sediment or mangrove root sample were placed into the lined extraction vessel in Microwave Extraction System (MSP-1000 CEM Co., USA) for microwave sample preparation. The samples were spiked with 5 μ l deuterium-labelled PAHs surrogate standards (A. R., Aldrich Co., USA) prior to extraction, then covered by 20 ml hexane and acetone solvent mixture (1:1 v/v for sediments and 3:2 v/v for roots) and extracted for 10 min at a temperature depending on the procedure. After extraction, the vessels were allowed to cool to room temperature before opening. The extract was carefully filtered through an anhydrous sodium sulphate layer into a 250 ml flat-bottomed flask, then evaporated to around 2 ml by placing the flat-bottomed flask in a water bath (60°C). The PAHs fraction was eluted with a chromatographic column consisting of 9 g activated Al_2O_3 and 18 g activated silica gel, topped with 1 g activated copper powder to remove sulphides in sediment with dichloromethane and pentane solvent mixture (1:1 v/v). The purified extracts were evaporated to around 5 ml in water bath (60°C), subsequently concentrated just to dryness with a stream of pure nitrogen. The residue was immediately

dissolved in 0.5 ml of hexane and the solution was transferred into a brown vial, spiked with surrogate internal standard (deuterium-labeled pyrene) and capped.

A Hewlett-Packard 6890 Gas Chromatography with quadruple Hewlett-Packard 5973 Mass Selective Detector was used for determining the levels of PAHs in the sediment and mangrove root samples. Quartz capillary column (HP-5MS) was 30 m long, 0.25 mm internal diameter, coated with 0.25 μ m thickness film. Splitless injection volume was 2 μ l at 320°C; the oven temperature for analyses was programmed from 60°C to 150°C at a rate of 15°C/min, 120–220°C, at a rate of 5°C/min, 220–300°C at a rate of 10°C/min, and held at 300°C for 10 min. Helium was used as the carrier gas. Mass Selective Detector was operated in Selective Ion Mode (SIM) at 70 eV. Ion source temperature was 230°C.

Sixteen EPA PAHs were identified and quantified based on GC retention time and by GC-MS analyses. Sixteen PAHs were naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo [a] anthracene (B[a]A), chrysene (Chr), benzo [b] fluoranthene (B[b]F), benzo [k] fluoranthene (B[k]F), benzo [a] pyrene (B[a]P), indeno [c, d] pyrene (I[c, d]P), dibenzo [a, h] anthracene (DB[a, h]A) and benzo [g, h, i] perylene (B[g, h, i]P).

All analytical data were subject to strict quality control. The procedural blank value was 27 ng/g. Surrogate recoveries ranged from 68% to 117% for the entire procedure. The limit of detection for an individual PAH compound was 10 ng/g.

2 Results and discussion

2.1 PAHs in surface sediments

Almost all 16 EPA PAHs were quantified in the surface sediment of mangrove wetlands in Jiulong River Estuary. The concentrations of individual PAH compounds and total PAHs

(Σ PAHs) are shown in Table 1. The Σ PAHs concentrations in surface sediments did not vary significantly among the mangrove wetlands, ranging from 193.44 ng/g dw at FG to 270.53 ng/g dw at HMa, with a mean value of 231.76 ± 31.78 ng/g dw. The Σ PAHs concentrations in mangrove sediments from the study area were similar to the levels found in marine sediments in Jiulong River Estuary and Xiamen Harbour (Table 2). The levels in the study area were lower than those found in other mangrove sediments, and one order of magnitude lower than those found in many other coastal environments such as Pearl River Estuary and San Francisco Bay (Table 2). The results were different from those reported by previous workers.

The Σ PAHs concentrations in mangrove sediments were generally higher than those in marine sediments in Hong Kong (Connell, 1998; Tam, 2001). Compared with other mangrove and coastal sediments, the Σ PAHs concentrations recorded in present study were at relatively lower level.

The PAHs composition was very similar among three sampling stations. The dominant PAHs were B[a]P (five-ring), Flt (four-ring) and pyr (four-ring) (Table 1). The percentages of the four- and five-ring to total PAHs were 28.3%—37.5% and 27.8%—48.5%, respectively.

These findings were in good consistent with those reported by previous workers. It was reported that the contaminated mangrove sediments in Caribbean Island of Guadeloupe were dominated by Flt, Pyr, B[e]P and perylene.

Table 1 Concentration and distribution of PAHs in surface sediment of mangrove wetlands in Jiulong River Estuary (ng/g dw)

PAHs	FG	HMa	HMb
Nap	10.96 (5.7) ^a	14.79 (5.5)	2.54 (1.2)
Acy	ND ^b	1.25 (0.5)	9.03 (4.3)
Ace	4.16 (2.2)	2.15 (0.8)	1.11(0.5)
Flu	8.50 (4.4)	9.11(3.4)	6.69 (3.2)
Phe	20.43 (10.6)	26.56 (9.8)	11.13 (5.3)
Ant	3.91 (2.0)	7.17 (2.7)	3.11(1.5)
Flt	27.63 (14.3)	37.58 (13.9)	23.65 (11.2)
Pyr	22.60 (11.7)	34.26 (12.7)	23.35 (11.1)
B[a]A	ND	11.33 (4.2)	10.44 (4.9)
Chr	14.1 (47.3)	18.36 (6.8)	2.34 (1.1)
B[b]F	14.7 (47.6)	20.39 (7.5)	26.05 (12.3)
B[k]F	9.45 (4.9)	12.75 (4.7)	9.44 (4.5)
B[a]P	28.97 (15.0)	42.18 (15.6)	63.22 (30.0)
I[c,d]P	11.87 (6.1)	11.50 (4.3)	10.05 (4.8)
DB[a,h]A	2.20 (1.1)	ND	3.50 (1.7)
B[g,h,i]P	13.88 (7.2)	21.15 (7.8)	5.29 (2.5)
Σ PAHs	193.44 (100.0)	270.53 (100.0)	210.94(100.0)

Notes: ^a Percentage to Σ PAHs (%); ^b ND. not detected

Table 2 Total PAHs concentration in surface sediment of mangrove wetlands of the Jiulong River Estuary, other mangrove wetlands as well as coastal areas (ng/g dw)

Sample sites	Mean \pm S.E	Range	Reference
Mangrove sediments in Puerto Rico	1820 \pm 2394	500—6000	Klekowski, 1994
Mangrove sediments in Caribbean Island	502 \pm 554	103—1675	Bernard, 1996
Mai Po mangrove sediments in Hong Kong	610 \pm 236	212—1042	Zheng, 2000
Mangrove sediments in Jiulong River Estuary	232 \pm 26	193—271	This study
Marine sediments in Jiulong River Estuary	334 \pm 337	59—1177	Maskaoui, 2002
Marine sediments in Xiamen Harbour	367 \pm 87	247—480	Zhou, 2000
Marine sediments in Pearl River Estuary	2196 \pm 3078	408—10811	Mai, 2001
Marine sediments in San Francisco Bay	7457 \pm 6908	2635—27680	Pereira, 1996

Yang (Yang, 2000) found that PAHs in South China Sea sediments were dominated by the four- and five-ring PAHs. The five-ring PAHs were also found at relatively high level in sediments collected from Jiulong River Estuary and Western Xiamen Sea (Maskaoui, 2002).

The sources of PAHs in sediments may be assessed by the ratios of individual PAH concentration such as phenanthrene/anthracene (Phe/Ant) and fluoranthene/pyrene (Flt/Pyr) (Gschwend, 1981; Sicre, 1987; Budzinski, 1997). Low Phe/Ant ratios (< 10) suggested that PAHs mainly generated by the combustion of fossil fuel (Gschwend, 1981). For the Flt/Pyr ratios, values greater than 1 are obviously related to pyrolytic origins, whereas values less

than 1 are attributed to petrogenic source (Sicre, 1987). As shown in Table 3, all the mangrove sediments examined in this study exhibited Phe/Ant ratios lower than 10, and Flt/Pyr ratios greater than 1, suggesting strong pyrolytic input. The Flt/Pyr + Flt ratio could also be used to identify PAHs sources. The Flt/Pyr + Flt ratio between 0.40 and 0.50 are more characteristic of liquid fossil fuel (vehicle and crude oil) combustion whereas ratio > 0.50 are characteristic of grass, wood or coal combustion (Zheng, 2002; Yunker, 2002). In the present study, the Flt/Pyr + Flt ratio in mangrove sediments ranged from 0.50 to 0.55, suggesting that the major PAHs input came from grass, wood or coal combustion process.

Table 3 Ratios of different individual PAHs in surface sediment of mangrove wetlands in Jiulong River Estuary

Sample sites	Percentages of the 2—6-ring ^a to total PAHs					Phe/Ant	Flt/Pyr	Flt/Pyr + Flt
	2-ring	3-ring	4-ring	5-ring	6-ring			
FG	5.7	19.1	33.3	28.6	13.3	5.23	1.22	0.55
HMa	5.5	17.1	37.5	27.8	12.1	3.70	1.10	0.52
HMb	1.2	14.7	28.3	48.5	7.3	3.58	1.01	0.50

Notes: ^a 2-ring PAHs include Nap; 3-ring PAHs include Acy, Ace, Flu, Phe and Ant; 4-ring PAHs include Flt, Pyr, B[a]A and Chr; 5-ring PAHs include B[b]F, B[k]F, B[a]P and DB[a,h]A; 6-ring PAHs include I[c,d]P and B[g,h,i]P

2.2 PAHs in mangrove roots

Table 4 shows the concentration and distribution of PAHs in roots of three mangrove species in Jiulong River Estuary. The Σ PAHs (13 EPA PAHs) concentrations in roots did not vary significantly among three mangrove species, ranging from 30.83 to 48.36 ng/g dw. It was reported that Σ PAHs in carrot roots grown in an industrial area of Thessaloniki in Northern Greece ranged from 48 to 94 ng/g dw (Kipopoulou, 1999). The PAHs range was very similar to the values recorded in present study. The lower molecular weight PAHs such as Nap (two-ring) and Phe (three-ring) were the most abundant compounds. Nap, the simplest low molecular weight PAHs, accounted for 19.2%—22.4% of Σ PAHs. The percentages of Phe to Σ PAHs ranged from 14.1% to 16.8%. Higher abundance of the lower molecular weight PAHs in plants has been reported before (Larsson, 1981; Wang, 1981; Wild, 1992; 1994) and was attributed to their higher aqueous solubility and volatility.

Table 4 Concentration and distribution of PAHs in roots of three mangrove species in Jiulong River Estuary (ng/g dw)

PAHs	<i>B. gymnorrhiza</i> (FG)	<i>A. marina</i> (HMa)	<i>K. candel</i> (HMb)
Nap	6.92 (22.4)	9.29 (19.2)	6.30 (19.9)
Acy	0.73 (2.4)	0.70 (1.4)	1.57 (5.0)
Ace	0.52 (1.7)	0.76 (1.6)	0.58 (1.8)
Flu	1.37 (4.4)	1.71 (3.5)	2.30 (7.3)
Phe	5.17 (16.8)	6.80 (14.1)	5.18 (16.4)
Ant	0.71 (2.3)	1.06 (2.2)	1.13 (3.6)
Flt	3.25 (10.5)	5.00 (10.3)	2.86 (9.0)
Pyr	3.79 (12.3)	3.40 (7.0)	3.35 (10.6)
B[a]A	1.20 (3.9)	1.35 (2.8)	1.38 (4.4)
Chr	1.96 (6.4)	2.71 (5.6)	0.77 (2.4)
B[b]F	1.90 (6.2)	3.57 (7.4)	1.99 (6.3)
B[k]F	1.25 (4.1)	3.32 (6.9)	1.87 (5.9)
B[a]P	2.06 (6.7)	8.69 (18.0)	2.39 (7.5)
Σ PAHs	30.83 (100.0)	48.36 (100.0)	31.67 (100.0)

2.3 Bioconcentration factors and PAHs properties

Sediment-to-vegetation bioconcentration factors (BCF_{SV} s) could be defined as the ratio of compound concentrations in various parts of the plant relative to concentrations in the sediments. The BCF_{SV} s of PAHs for three mangrove species roots were calculated using the data of Table 1 and Table 3. As shown in Table 5, the average BCF_{SV} of PAHs were almost all under the level of 1 except for Nap, thus suggesting relatively lower accumulation ability for PAHs in mangroves. For example, the BCF_{SV} s of B[a]P in this study were 0.038, 0.206 and 0.071 for *K. candel*, *A. marina* and *B. gymnorrhiza*, respectively. Concentrations of PAHs in vegetation are generally less than concentrations in the soil where they grow (Edwards, 1983). Vegetation-soil concentration ratios ranging between 0.002 and 0.64 were reported for B[a]P in an urban industrial area (Kolar, 1975). Median values of vegetable-soil B[a]P concentration ratios in an industrial area of Thessaloniki in

Northern Greece were found equal to 0.055, 0.065, 0.079, 0.091 and 0.125 for cabbage, carrot, leek, endive and lettuce, respectively (Kipopoulou, 1999). The BCF_{SV} values were almost at the same level as those recorded in this study.

Table 5 Sediment-to-vegetation bioconcentration factors of PAHs in different mangrove in nature habitats

PAHs	<i>B. gymnorrhiza</i> (FG)	<i>A. marina</i> (HMa)	<i>K. Candel</i> (HMb)
Nap	0.631	0.628	2.480
Acy	ND	0.560	0.174
Ace	0.125	0.353	0.523
Flu	0.161	0.188	0.344
Phe	0.253	0.256	0.465
Ant	0.182	0.148	0.363
Flt	0.118	0.133	0.121
Pyr	0.168	0.099	0.143
B[a]A	ND	0.119	0.132
Chr	0.139	0.148	0.329
B[b]F	0.129	0.175	0.076
B[k]F	0.132	0.260	0.198
B[a]P	0.071	0.206	0.038

The relationships between BCFs and the physico-chemical properties of pollutants, such as lipophilicity, water solubility, vapour pressure and Henry's Law constant, have

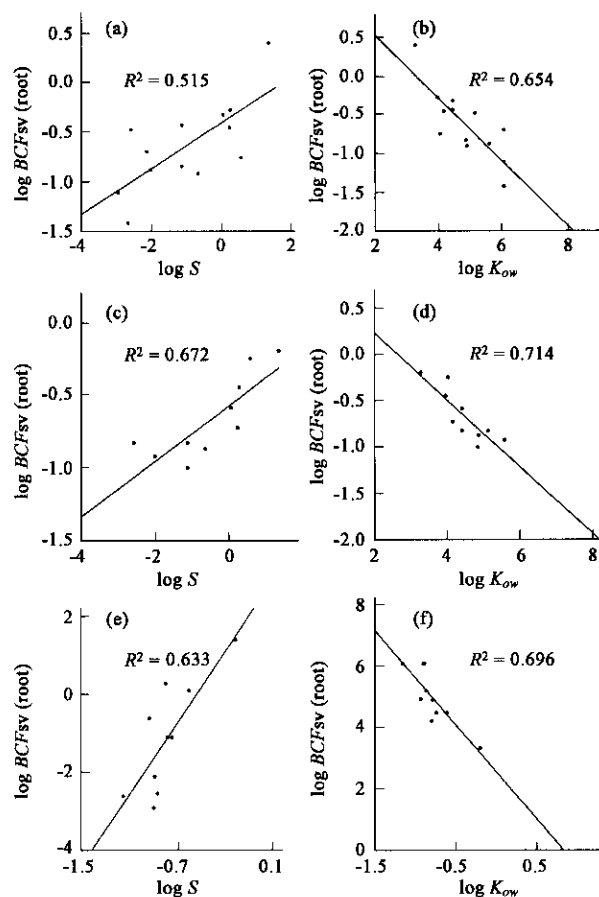


Fig. 2 Correlation between sediment-to-vegetation bioconcentration factors for *K. candel* (HMb) (a, b), *A. marina* (HMa) (c, d) and *B. gymnorrhiza* (FG) (e, f) roots and PAHs water solubility, octanol-water partitioning coefficient

been examined by several workers (Tolls, 1994; Wang, 1994; Simonich, 1995; Kipopoulou, 1999). Good linear relationship between BCF_{sv} values for mangrove roots and PAHs water solubility (S), octanol-water partitioning coefficient (K_{ow}) was derived in present study. Positive linear relationship between $\log S$ and $\log BCF_{sv}$ (root), negative linear relationship between $\log K_{ow}$ and $\log BCF_{sv}$ were found in *K. candel* roots (Fig. 2a, b). The similar correlation was also found in *A. marina* and *B. gymnorhiza* roots (Fig. 2c, d, e and f). Solubility is indicative of compound mobility in soil, while K_{ow} indicated the propensity for PAHs sorption onto the organic phase (Kipopoulou, 1999). It is evident that both properties are good predictors of PAHs accumulation in mangroves. The highest BCF_{sv} values occurred for Nap as a result of the highest water solubility and the lowest K_{ow} among the compounds tested. This suggested that Nap was the most likely of the PAHs to be taken up from contaminated sediment through roots, to move from the outer to the inner root, and to probably be distributed within the plant.

Acknowledgments: The authors are very grateful to Prof. YUAN Dong-xing and CAI Li-zhe for their valuable advice and Dr. ZHANG Jun for his contribution to PAHs analysis.

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