



UNIVERSITI PUTRA MALAYSIA

***DISTRIBUTION AND SOURCE IDENTIFICATION OF POLYCYCLIC
AROMATIC HYDROCARBONS IN MANGROVE SEDIMENTS AND
PNEUMATOPHORES IN REMBAU-LINGGI ESTUARY, MALAYSIA***

MUHAMMAD RAZA

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

DISTRIBUTION AND SOURCE IDENTIFICATION OF POLYCYCLIC AROMATIC HYDROCARBONS IN MANGROVE SEDIMENTS AND PNEUMATOPHORES IN REMBAU-LINGGI ESTUARY, MALAYSIA

By

MUHAMMAD RAZA

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Chairman : Prof. Mohamad Pauzi bin Zakaria, PhD

Faculty : Environmental Studies

Polycyclic aromatic hydrocarbons (PAHs) are one of the foremost widespread classes of pollutants which can accumulate in sediments for a long period of time due to their low water solubility and hydrophobicity properties. Mangrove sediment acts as a sink for organic pollutants especially in an estuary environment. The aims of this study are to determine the composition, distribution and sources of PAHs in mangrove sediments and pneumatophores of *Sonneratia* in Rembau-Linggi estuary. The samples were collected from nine stations along Rembau-Linggi estuary, Negeri Sembilan. The samples were then extracted using a soxhlet extractor with dichloromethane followed by silica gel column chromatography to separate the hydrocarbons into fractions. The 17 PAHs were analyzed and identified using GC-MS based on the response factor of a PAH standard. Low molecular weight PAHs were abundant in pneumatophores while high molecular weight PAHs were abundant in sediments. Total PAHs in pneumatophores were significantly higher ($p < 0.05$) than in sediments in all eight stations. There is no significant correlation between PAHs and organic carbon which means that the distribution of PAHs was

not affected by the organic carbon content. Total methylphenanthrenes (MP) shows weak correlation with total PAHs ($p < 0.05$) implying that petrogenic input has some contribution to the sedimentary PAHs but not a major control over distribution of PAHs within the estuary. CombPAH shows a positive correlation with total PAHs with $R^2 = 0.9368$ ($p < 0.05$) suggesting that pyrogenic input has significant contribution to the sedimentary PAHs. It is suggested that the combustion of petroleum, wood and coal were the main sources of PAHs which have been brought by long-range atmospheric transport and thus, deposited into this estuary. The PAH concentrations increased with distance from upstream of the estuary to the coastal area of Straits of Malacca which implied that river discharge and urban runoff flowing from residential areas centred near the coastline of the Straits of Malacca were the probable source of PAHs. There is a moderate relationship between PAHs of sediments and pneumatophores ($p < 0.05$) which implied that sedimentary PAHs have moderate influence on the PAH distribution in pneumatophores. None of individual PAH compounds exceeded the values of ERL-ERM and TEL-PEL guidelines thus PAH level in sediments in Rembau-Linggi estuary are unlikely to cause any adverse biological effects to aquatic organisms.

Keywords: Mangrove Sediments, Pneumatophores, Pyrogenic PAHs, Petrogenic PAHs, Anthropogenic activities, Long-range transport

Abstrak ini yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

**TABURAN DAN PENGENALPASTIAN SUMBER HIDROKARBON
AROMATIK POLISIKLIK DALAM TANAH BAKAU DAN
PNEUMATOPHORES DI KUALA REMBAU-LINGGI, MALAYSIA**

Oleh

MUHAMMAD RAZA

April 2012

Pengerusi : Prof. Mohamad Pauzi bin Zakaria, PhD

Fakulti : Pengajian Alam Sekitar

Hidrokarbon aromatik polisiklik (PAH) ialah salah satu daripada bahan pencemar utama yang tersebar meluas di mana ianya mampu berkumpul di dalam tanah untuk jangka masa yang lama disebabkan tahap kelarutan dalam air yang rendah dan bersifat hidrofobik. Tanah bakau bertindak sebagai tempat berkumpul bagi bahan pencemar organik terutamanya di kawasan kuala sungai. Objektif kajian ini ialah mengenal pasti komposisi, taburan dan sumber pencemaran PAH dalam tanah bakau dan pneumatophores *Sonneratia* di kuala Rembau-Linggi. Sampel diambil daripada sembilan stesen di sepanjang kuala Rembau-Linggi, Negeri Sembilan. Sampel diekstrak dengan menggunakan pengekstrak Soxhlet menggunakan diklorometana dan diikuti dengan kolum kromatografi gel silika untuk memisahkan hidrokarbon kepada beberapa bahagian kecil. Semua 17 PAH dianalisis dengan GC-MS berdasarkan faktor respon. PAH jisim molekul rendah adalah dominan di dalam pneumatophores manakala PAH jisim molekul tinggi adalah dominan di dalam tanah. Jumlah PAH di dalam pneumatophores adalah tinggi secara signifikan

($p < 0.05$) daripada tanah di semua lapan stesen. Didapati bahawa tiada hubungan yang signifikan diantara PAH dan karbon organik, yang menunjukkan bahawa taburan PAH tidak dipengaruhi oleh kandungan karbon organik. Jumlah metilphenanthrena (MP) menunjukkan hubungan yang lemah dengan jumlah PAH ($p < 0.05$) menandakan bahawa input petrogenik mempunyai pengaruh terhadap PAH dalam tanah tetapi bukanlah pengaruh yang besar kepada taburan PAH di Kuala Lumpur. CombPAH menunjukkan hubungan positif dengan jumlah PAH dengan $R^2 = 0.9368$ ($p < 0.05$) menandakan bahawa input pirogenik mempunyai sumbangan yang penting kepada PAH dalam tanah. Adalah dicadangkan bahawa pembakaran petroleum, kayu dan arang adalah sumber utama PAH yang mana telah dibawa oleh pengangkutan atmosfera jarak jauh dan termendap di Kuala Lumpur. Kepekatan PAH adalah meningkat dengan jarak dari hulu Kuala Lumpur sehingga ke kawasan pesisir pantai Selat Melaka yang mana menunjukkan bahawa sisa buangan sungai dan larian air dari kawasan berpenduduk yang berpusat di kawasan pesisir pantai Selat Melaka adalah berpotensi menjadi sumber PAH. Adalah didapati hubungan sederhana diantara PAH di dalam tanah dan pneumatophores ($p < 0.05$) menunjukkan bahawa PAH di dalam tanah mempunyai pengaruh yang sederhana terhadap taburan PAH di dalam pneumatophores. Tiada PAH sebatian individu yang melebihi nilai garis panduan ERL-ERM dan TEL-PEL dan seterusnya menunjukkan bahawa tahap PAH di dalam tanah di Kuala Lumpur-Linggi adalah tidak akan menyebabkan kesan biologiikal yang buruk kepada haiwan akuatik.

Kata kunci: Tanah bakau, Pneumatophores, PAH pirogenik, PAH petrogenik, Aktiviti anthropogenik, Pengangkutan jarak jauh

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LIST OF ABBREVIATIONS

Ant	Anthracene
Ant/178	Anthracene / (Phenanthrene + Anthracene)
B[a]A	Benzo[a]anthracene
B[a]A/228	Benzo[a]anthracene / (Benzo[a]anthracene + Chrysene)
B[a]P	Benzo[a]pyrene
B[e]P	Benzo[e]pyrene
B[k]F	Benzo[k]fluoranthene
B[g,h,i]P	Benzo[g,h,i]perylene
Chr	Chrysene
Chr-d ₁₂	Chrysene-deuterated-12
DB[a,h]A	Dibenzo[a,h]anthracene
DCM	Dichloromethane
dry wt.	Dry weight
ERL	Effect Range Low
ERM	Effect Range Median
Flt	Fluoranthene
Flu/202	Fluoranthene / (Fluoranthene + Pyrene)
GC-MS	Gas Chromatography-Mass Spectrometry
LMW/HMW	Low Molecular Weight PAH/High Molecular Weight PAH
LOD	Limit of Detection
LOQ	Limit of Quantification
Hex	Hexane
IIS	Internal Injection Standard
I[c,d]P	Indeno[1,2,3-cd]pyrene

I[c,d]P/276	Indeno[1,2,3-cd]pyrene / (Indeno[1,2,3cd]pyrene + Benzo[g,h,i]perylene)
MeOH	Methanol
MP/P	Methylphenanthrene/Phenanthrene
Na ₂ SO ₄	Anhydrous Sodium Sulphate
PAHs	Polycyclic Aromatic Hydrocarbons
PEL	Probable Effects Level
Phe	Phenanthrene
ppm	part per million
ppb	part per billion
Pyr	Pyrene
S/N	Signal to Noise Ratio
SIS	Surrogate Internal Standard
SQG	Sediment Quality Guidelines
spp.	Species (plural)
TEL	Threshold Effects Level
TOC	Total Organic Carbon
UPM	Universiti Putra Malaysia

CHAPTER 1

INTRODUCTION

1.1 Background of the Study

Southeast Asia's mangroves are known internationally as being the world's largest and best-developed mangrove forests (Giesen and Wulffraat, 1998; Giesen *et al.*, 2007). In the western part of Peninsular Malaysia, mangroves are a continuous belt of forest area facing the Straits of Malacca which cover the muddy shores of sheltered coasts and river estuaries. Located between land and the sea, mangroves are consistently under threat from both sea-based and land-based pollutants. Studies have shown that mangroves are vulnerable to oil spills which are the main source of the hydrocarbon contamination in marine environments (Dicks 1986; Burns *et al.*, 1993; Burns and Yelle-Simmons 1994; Burns *et al.*, 1994; Levings *et al.*, 1994; Tam *et al.*, 2005; Farias *et al.*, 2006; Farias *et al.*, 2008; Dsikowitzky *et al.*, 2011). Furthermore, the location of mangrove forests in West of Peninsular Malaysia facing the Straits of Malacca and Sumatra, Indonesia regularly experience haze due to biomass burning in southern Sumatra (Radzi *et al.*, 2004; He *et al.*, 2010).

Due to the increasing trend of industrialization and urbanization in Malaysia, mangroves are facing the threat of hydrocarbon accumulation. Polycyclic aromatic hydrocarbons (PAHs) are given more attention among the hydrocarbons since they are one of the most widespread organic pollutants. PAHs are derived from land-based and sea-based sources where most are the result of anthropogenic activities.

Most mangroves occur in estuaries and act as sink for various pollutants where they tend to adsorb to surface sediments.

Anthropogenic activities resulting from industrialization and urbanization i.e. fossil fuel combustion, discharge of domestic sewage and incineration of industrial waste give a great contribution to pollution generation especially in marine and coastal environments. This study aims to investigate hydrocarbon contamination in mangrove sediments and pneumatophores with special focus on polycyclic aromatic hydrocarbons.

The results and findings of the study enhance the knowledge and awareness about hydrocarbons contamination, in directions that are not only meant for conservation of the mangrove forests, but also for the protection of the aquatic organisms that are closely associated with mangrove ecosystems.

1.2 Problem Statement

Mangrove ecosystems are exposed to the threat of hydrocarbon accumulation due to the increase in human activities along the coastline facing the Straits of Malacca. This has led to pollutions to the marine and coastal environments including the mangrove forests, estuaries and wetlands. The aromatic group of hydrocarbons, PAHs, is able to accumulate and persist in sediments for a long period of time due to their low water solubility and hydrophobic properties. As a result, PAHs tend to adsorb strongly onto sediments and sediment particles in water. Moreover, the unique characteristics of mangrove environments such as rich in organic carbon

content, generally anaerobic condition below the surface sediments layers due to high bacterial activities (Bernard *et al.*, 1996) and the presents of brackish water, result in PAH biodegradation occurs very slowly (Suess, 1976; Hensel *et al.*, 2002). In this instance, PAHs are likely to accumulate and persist in the mangrove environments (Neff, 1979) thus poses threat to the ecosystems.

There is lack of study on PAHs in mangrove ecosystems in Malaysia. There is only one scientific publication available on PAHs study on Malaysian mangroves (Tahir *et al.*, 2011), however, the study was carried out in the eastern part of Peninsular Malaysia in 2001. Hence, there is need recent PAH studies in mangroves especially for the western part of Peninsular Malaysia. What is more, there is no scientific publication available on PAH study on mangrove plant of *Sonneratia* species. Most of the PAH studies in other countries tested on other mangrove species such as *Aegiceras corniculatum*, *Avicennia marina*, *Bruguiera gymnorhiz*, *Kandelia candel* and *Rhizophora mangle* (Garrity *et al.*, 1994; Ke *et al.*, 2003a; Ke *et al.*, 2003b; Zhi-qiang *et al.*, 2005; Tam and Wong, 2008). It is important to study the distribution of PAHs in *Sonneratia* species since this species is the most abundant species in the mangrove ecosystem in Malaysia. Therefore, a study on PAH contamination in mangrove sediments and pneumatophores of *Sonneratia* species is required in order to determine the current level of PAHs in Peninsular Malaysia especially the mangrove regions located along the Straits of Malacca such as the chosen site, the Rembau-Linggi estuary.

1.3 Objectives of the Study

- i. To determine the compositional patterns of PAHs in mangrove sediments and in pneumatophores of *Sonneratia* species
- ii. To determine the relationship between sedimentary PAHs and TOC content
- iii. To identify the sources of PAHs in mangrove sediments and pneumatophores
- iv. To compare the sedimentary PAH concentrations with sediment quality guidelines

1.4 Hypotheses

The hypotheses of this study were built based on the findings and conclusion made from previous studies in the literature review:

- i. Sediment is abundant with HMW PAHs and pneumatophores is abundant with LMW PAHs
- ii. There is a significant positive correlation between sedimentary PAHs and TOC content
- iii. PAHs in mangrove sediments and pneumatophores are mainly originate from petrogenic sources
- iv. Sedimentary PAH levels in Rembau-Linggi estuary do not exceed the recommended PAH levels of sediment quality guidelines

1.5 Significance of the Study

Study on PAHs in mangrove sediments and pneumatophores will significantly help to quantify the current level and distribution of these compounds in typical Malaysian mangrove ecosystems. Moreover, it is believed that PAHs have the potential to be major pollutants deposited and accumulated in the mangrove estuaries. Rembau-Linggi estuary was chosen as it is located in the western part of Peninsular Malaysia facing the Straits of Malacca as well as Sumatra, Indonesia. These two regions i.e. Straits of Malacca and Sumatra, Indonesia have great potential to be significant sources in Malaysian mangrove ecosystems. In this case, pyrogenic PAHs originate from biomass burning in Sumatra, Indonesia transferred by long range transport through air masses and wind (Radzi *et al.*, 2004; He *et al.*, 2010), while the petrogenic PAHs derived from ships and tankers in the Straits of Malacca brought by sea tides and wave to these mangrove estuaries.

Moreover, based on previous study by Nazli and Hashim (2010), *Sonneratia caseolaris* in Rembau-Linggi estuary possess the capacity to take up selected heavy metals via its pneumatophores. Thus, further study is required in order to prove the capability of mangrove plants, specifically *Sonneratia* species, to take up and accumulate the PAHs in the pneumatophores. For those reasons, this study is required in order to fill the gap of information on PAH study in Malaysian mangrove ecosystems.

The results of this study will be compared with the proposed PAH levels of sediment quality guidelines in order to evaluate the biological effects of sedimentary PAHs to

the aquatic organisms, especially the ones that are closely associated with mangrove ecosystems. Finally, a number of new findings on PAHs fate in Malaysian mangrove ecosystems will be scientifically shown through this study.



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