DISTRIBUTION AND SOURCES OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN SURFACE SEDIMENT SAMPLES COLLECTED IN SOUTH CHINA SEA

NORAZIDA MANAN

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MASTER OF SCIENCE

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DISTRIBUTION AND SOURCES OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN SURFACE SEDIMENT SAMPLES COLLECTED IN SOUTH CHINA SEA

By

NORAZIDA MANAN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the degree of Master Science

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Special dedication to my parents, Manan bin Mohamad and Zaliha Binti Awang; all my family members; Norzelawatie and Azmi, Norhazlini and Azwan Hazle, Norhazliza, Norhazlina and Rosdillah, Mohd Ridzwan and Rosbazillah, Mohd Rizatulfaiz, Norazihah, Muhammad Rizalman; and last but not least, my nephews and nieces; Adly Qusyairi, Adam Harith, Alif Haikal, Afif Hazriq, Nur Amani Humairah dan Nur Rania Qaisara.

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

DISTRIBUTION AND SOURCES OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN SURFACE SEDIMENT SAMPLES COLLECTED IN SOUTH CHINA SEA

By

Norazida Manan

April 2012

Chairman: Prof Mohamad Pauzi Zakaria, PhD

Faculty: Faculty of Environmental Studies

Petroleum pollution is one of the pollution that needs serious attention from world. Petroleum is worldwide used and introduced to aquatic environment through accidental oil spill, discharge from routine tanker operations. In other case, land based pollutants are contributed by rainfall and runoff waters. Polycyclic aromatic hydrocarbons (PAHs) are one of the major compounds in petroleum known as the most widespread organic pollutants. PAHs diagnostic rations applied in this study to identify the anthropogenic sources of PAH; petrogenic and pyrogenic. Thirty surface sediment samples were collected from South China Sea adjacent to the

coast of Peninsular Malaysia using Ekcman Dredge sampler. The samples were extracted and fractionated prior to the analysis using gas chromatography- mass spectrometry (GC-MS) to quantify the concentration of PAHs. The total PAHs concentration of the marine sediment ranged from $0.0315x10^3$ and $1.48x10^3$ ng/g, dry weight. The concentration level of PAHs in the South China Sea is indicated as moderate to high contamination based on the global PAHs sedimentation records. Principal Component Analysis (PCA) in associated with Multiple Linear Regressions (MLR) has been performed to apportion PAHs sources in sediment samples. Seven factors and their percentages of contributions were identified based on PCA loadings of PAHs and were attributed to the following sources; (1) oil exploitation , 8.1%; (2) traffic emission, 43.8%; (3) road dust, 8.9%; (4) the spill of oil products, 3.6%; (5) unburned fossil fuels, 2.1%; (6) coal combustion, 10.2%; (7) oil combustion, 23.3%. The major source of anthropogenic PAHs pollution in the South China Sea is pyrogenic PAHs. This study should be conducted from time to time to monitor the concentration of PAHs in sediments.

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

TABURAN DAN SUMBER HIDROKARBON AROMATIK POLISIKLIK (PAHS) DALAM PERMUKAAN SEDIMEN DARI LAUT CHINA SELATAN

Oleh

Norazida Manan

April 2012

Pengerusi: Prof Mohamad Pauzi Zakaria, PhD

Fakulti: Fakulti Pengajian Alam Sekitar

Pencemaran petroleum merupakan salah satu pencemaran yang memerlukan perhatian yang serius dari dunia. Petroluem digunakan secara meluas dan ia terdedah kepada persekitaran akuatik melalui tumpahan minyak yang tidak sengaja, sisa dari operasi kapal tangka rutin. Dalam kes lain, pencemar yang dari tanah disumbangkan oleh hujan dan air larian. Hidrokabon aromatik polisiklik (PAH) adalah salah satu sebatian utama dalam petroleum dan dikenali sebagai bahan pencemar organik yang tersebar secara meluas. Nisbah diagnostik PAH digunakan dalam kajian ini untuk mengenal pasti sumber- sumber antropogenik PAH; petrogenik dan pyrogenik. Tiga puluh sample permukaan sedimen telah

diambil dari Laut China Selatan bersebelahan dengan Pantai Timur Semenanjung Malaysia menggunakan pensampel Eckman. Sampel yang telah diekstrak dan dipertingkatkan sebelum dianalisis menggunakan gas kromatografi spektrometri jisim (GC-MS) untuk mengukur kandungan PAH. Jumlah kandungan PAH di dalam sedimen adalah diantara $0.0315x10^3$ dan $1.48x10^3$ ng/g berat kering. Tahap kandungan PAH di Laut China Selatan ditunjukkan sebagai pencemaran sederhana kepada yang tinggi berdasarkan rekod pemendapan global PAH. Analisis komponen principal (PCA) dengan regrasi kepelbagaian garis lurus (MLR) telah dijalankan untuk mendapatkan potensi sumber PAH di dalam sampel sedimen. Tujuh factor dan peratusan sumbangan mereka telah dikenalpasti berdasarkan pembebanan PCA daripada PAH iaitu disebabkan oleh sumber-sumber berikut; (1) ekploitasi minyak, 8.1%; (2) pelepasan trafik, 43.8%; (3) debu jalan, 8.9%; (4) tumpahan produk minyak, 3.6%; (5) bahan api fosil yang tidak terbakar, 2.1%; (6) pembakaran arang batu, 10.2%; (7) pembakaran minyak, 23.3%. Punca utama pencemaran antropogenik PAH di Laut China Selatan ialat pyrogenic PAHs. Kajian perlu dijalankan dari semasa ke semasa untuk memantau kandungan PAH dalam sedimen.

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I certify that an Examination Committee has met on 05 April 2012 to conduct the final examination of **Norazida Manan** on her Master Science thesis entitled **"Distribution and Sources of Polycyclic Aromatic Hydrocarbons (PAHs) in Surface Sediment Samples Collected from South China Sea"** in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the student be awarded the *Master Science Marine and Aquatic Studies.*

Members of the Examination Committee were as follows:

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Master Science. The members of the Supervisory committee were as follows:

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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and its not concurrently, submitted for any other degree at University Putra Malaysia or at any other institution.

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(NORAZIDA MANAN)

Date: 05 April 2012

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LIST OF ABBREVIATIONS

1-MP	1-methylphenanthrene
1-MPy	1-methylpyrene
2-MA	2-methylanthrancene
2-MP	2-methylphenanthrene
3-MP	3-methylphenanthrene
9-MP	9-methylphenanthrene
Ant	Anthrancene
Ant/ (Ant + Phe)	Anthrancene/ (Anthrancene + Phenanthrene)
BAnt	Benzo(a) anthrancene
BaP	Benzo(a) pyrene
BeAce	Benzo(e) acephenanthrene
BeP	Benzo(e) pyrene
BkFlu	Benzo(k) fluoranthene
Chry	Chrysene
DBahAnt	Dibenzo(a,h) anthrancene
DBT	Dibenzothiophene
DCM	Dichloromethane
Flu	Fluoranthene
GC-MS	Gas Chromatography Mass Spectrometry
IARC	Internal Agency for Research on Cancer
IIS	Internal Injection Standards

LABs	Linear alkylbenzenes
LMW/HMW	Low Molecular Weight/ High Molecular Weight
LOD	Limit of Detection
m/z	Mass to change ratio
MLR	Multiple Linear Regressions
Nap	Naphthalene
PAHs	Polycyclic Aromatic Hydrocarbons
PC	Principle Component
PCA	Principle Component Analysis
PEC	Potency Equivalent Concentration
Phe	Phenanthrene
Phe/ Ant	Phenanthrene/ Anthrancene
Py	Pyrene
SIM	Selected Ion Monitoring
USEPA	United State Environmental Potency Agency
v/v	Volume per volume
VF	Varimax Factor

LIST OF EQUATIONS

CHAPTER ONE

INTRODUCTION

1.1. General Introduction

In last few decades, Malaysia experienced fast urbanization, rapid population growth, industrialization, and motorization. Malaysia is also well-known for resources such as agriculture, forestry minerals and petroleum. On the east coast of peninsular Malaysia facing South China Sea, there are a variety of potential sources and activities of petroleum. Furthermore, this coastal area is a very active place for fishing activities, marine activities and tourism activities consequently due to various threats by petroleum pollution including routine and accidental oil spills from tankers, spillage of crude oils from inland and offshore oil fields, as well as run-off from land-based human activities (Chandru *et al.,* 2008).

The marine ecosystem is very important to Malaysia as transport paths, source of food, revenue and defense. Sources of pollution either from land, atmosphere or sea will put this priceless resource to threat. Many studies have been carried out in the Malaysian marine ecosystem, however limited information gathered on the east coast of Malaysia, especially in the South China Sea. Marine began impacting mankind since industrial evolution and since been an inevitable

phenomena in any country. Many industrial processes and modern technologies have introduced pollutants into the environment and destructing the natural processes especially in the developing country like Malaysia. Our marine environment is currently under great stress from the anthropogenic activities carried out in the mainland and sea.

Petroleum or crude oil is universally used either as a raw material or refined products consists of a complex mixture of hydrocarbons of various molecular weight and other liquid organic compounds (Sakari et al., 2011). There are many possible sources of petroleum input to aquatic environments including land-based discharge of untreated or insufficiently treated municipal sewage and storm water (urban runoff), discharge into rivers, discharges of untreated or insufficiently treated waste water from coastal industries; accidental or operational discharges of oil from coastal refineries, oil storage facilities; oil terminals and reception facilities, emission of gaseous hydrocarbons from oil-handling onshore facilities; sea based discharged (accidental oil spill from tanker, emissions of volatile organic carbon) and natural input (natural seeps). Polycyclic aromatic hydrocarbons (PAHs) are one of component petroleum (Blumer, 1976; Sakari *et al.,* 2011). PAHs are ubiquitous organic pollutants and they continuous released to the environment since they have a variety of formation PAHs (Countway *et al,* 2003; Chen *et al,* 2004; Cheollee *et al.,* 2005). PAHs have been detected in trace quantities in a wide variety of environmental samples including air, sediment, water and soil. The occurrence of PAHs in

environmental samples at various locations around the world has been reported (Laflamme and Hites, 1977; Chen *et al.,* 2004)

Some PAHs have been found to show strong mutagenicity, carcinogenicity. The fate of PAHs in the environment has attracted much attention around the world (Bucheli *et al.,* 2004; Tao *et al.,* 2004; Ma *et al.,* 2005; Zhang *et al.,* 2006). Since PAHs are hydrophobic and tend to partition on organic matters (Tolosa *et al.,* 2004), study on sedimentary PAHs and identification of PAHs sources are required to determine the concentration level of PAHs in the South China Sea. However, identification of PAH sources frequently were analyzed by using diagnostic ratio even though the increasing formation of PAHs are expected to be presented in this region due to rapid industrial development in Malaysia. Chemometric methods (such as principal component analysis and multiple linear regressions) were used in this study to identify and apportion the anthropogenic PAH sources in surface sediment collected from South China Sea.

1.2. Significance of the Study

There are many possible sources polluted the South China Sea since South China Sea dense with a various activities such as fishing, tourism and petroleum activities and human activities at the coastlines. Sometimes, accidental spills, discharged from routine tanker, oil activities occurred at the South China Sea.

Day after day it became polluted by many contaminants especially petroleum. Petroleum pollution was caused by either from sea based, land based or natural sources. In this study, the major anthropogenic sources of PAHs can be investigated thus the possible sources of anthropogenic sources can be apportioned (Hupp *et al.,* 2000; Lee and Kim, 2007). The level of PAHs also can be explained by determining the concentration of PAHs in selected surface sediment of South China Sea. From this study, hopefully it will help further research to monitor Malaysian marine aquatic.

1.3. Hypotheses

i) PAHs are abundant in surface sediment.

ii) PAHs introduced to marine aquatic by a variety routes.

1.4. Problem Statements

This research is important to Malaysia due to lack of data and information of PAHs in the South China Sea. Many possible sources can contribute to the petroleum pollution. Well documented petroleum pollution especially PAHs are needed in South China Sea due to their widespread occurrence in the environment and concerns due to toxicity and their persistence features (Nauss, 1995; Marr *et al.,* 1999).

1.5. Objectives of the Study

i. To determine the concentration of PAHs in surface sediments of South China Sea

ii. To identify the source of PAHs in surface sediments using diagnostic ratios and Principal Component Analysis (PCA)

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LIST OF PUBLICATIONS

1. **Publication In Journal Article**

   Norazida Manan, Muhammad Raza, Yen Shen Yuh, Loo Woan Theng, Mohamad Pauzi Zakaria, 2011. Distribution of Petroleum Hydrocarbons in Aquaculture Fish from Selected Location in the Straits of Malacca, Malaysia. *(Accepted by World Applied Science Journal)*

2. **Submitting to Journal Article**

   Norazida Manan, Mohamad Pauzi Zakaria, Hafizan Juahir, Abdul Halim Abdullah, Che Abdul Rahim, Lee Chiow Yee, Norliza Ismail, Najat Masood, 2011. Chememetrial Pattern Recognition and Source Apportionment of Polycyclic Aromatic Hydrocarbons in Surface Sediments Collected form southwest corner of South China Sea.
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   Norazida Manan, Mohamad Pauzi Zakaria, Muhammad Raza, Norliza Ismail, Abdul Halim Abdullah, Che Abdul Rahim Mohamad, 2012. Distribution and Sources of Polycyclic Aromatic Hydrocarbons in Sediments of South China Sea: A Major Input of Anthropogenic PAHs. *(Submitted to UMT Journal, Special Issue)*

3. Exhibition

Norazida Manan, Mohamad Pauzi Zakaria, Abdul Halim Abdullah, Hafizan Juahir, Norliza Ismail, Lee Chiow Yee, Che Abdul Rahim Mohamed and Munirah Abdul Zali, 2010. Application of Chemometric Technique in Identifying Source apportionment of PAHs in Sediment Sample Collecte form South China Sea. *Pameran Reka Cipta, Penyelidikan dan Inovasi UPM 2010.* (Silver)

Norazida Manan, Mohamad Pauzi Zakaria, Lee Chiow Yee, Che Abdul Rahim Mohamed and Abdul Halim Abdullah, 2009. Distribution and Sources of Polycyclic Aromatic Hydrocarbon (PAHs) in Sediment Samples Collected from South China Sea. *Pameran Reka Cipta, Penyelidikan dan Inovasi UPM 2009.* (Bronze)