

UNIVERSITI PUTRA MALAYSIA

CHARACTERIZATION OF POLYCYCLIC AROMATIC HYDROCARBONS IN ATMOSPHERIC AEROSOLS COLLECTED FROM SELECTED LOCATIONS IN PENINSULAR MALAYSIA

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By

POURYA SHAHPOURY BAHRY

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

December 2007



DEDICATION

To my dear mother, close friends, my family, and my supervisor who have been the most important reasons of hopefulness during my study.



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

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Chairman: Associate Professor Mohamad Pauzi Zakaria, PhD

Faculty: Environmental Studies

With rapid modernization and development of Malaysia during the recent years, atmospheric hydrocarbons have been increasing due to increase in industrialization, motorization, biomass burning and deforestation in this country. One of the most important classes of hydrocarbons is polycyclic aromatic hydrocarbon (PAHs) that has long been of interest in the field of environmental forensic. It is very important to characterize the Polycyclic Aromatic Hydrocarbons in the environment because of the known carcinogenic and mutagenic effect of theses compounds to human health especially on the endocrine system. Generally, anthropogenic PAHs are released from both pyrogenic and petrogenic sources. Particulate phase PAHs have significant contribution from the total concentration of PAHs in the atmosphere. This study specifically focuses on characteristics of this group of compounds in total suspended particulates at nine sampling stations in peninsular Malaysia. The objectives of this project are to determine the sources, distribution and concentrations of compound specific PAHs in selected locations. For this



purpose, atmospheric aerosols are collected using high volume air samplers. The samples are further soxhlet extracted using high-grade dichloromethane then purified and fractionated by a two-step column chromatography. Subsequently, PAHs fraction with 3-5 benzene rings is analyzed by gas chromatography coupled with mass spectrometer (GC-MS). The results of this study revealed that concentration of PAHs ranged from 0.28 to 13.02 ng/m³ with the mean value of 2.73 ng/m^3 . The ratio of the sum of methylphenanthrenes to phenanthrene (MP/P) was under unity for 16 samples from the 18 samples analyzed. This result indicated that the atmospheric PAHs are from pyrogenic sources. Interestingly samples from Tanah Rata station MP/P ratio were found to be above unity that may come from petrogenic input of PAHs. Application of lower molecular weight (LMW) to higher molecular weight (HMW) PAHs proportion provided very useful supportive data to identify the origin of PAHs. The results indicated that distribution of compound specific PAHs during the sampling period are strongly controlled by dominance of higher molecular weight PAHs, which is consistent with results of MP/P ratio. The only exceptions consist of two samples from Alor Setar and Tanah Rata stations that LMW/HMW and MP/P ratios do not relate to each other, suggesting unique source of PAHs in the study area that contains both pyrogenic and petrogenic PAHs. Finally, it has concluded that the atmospheric environment of peninsular Malaysia during the period of sampling has influenced by pyrogenic sources of PAHs.



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

PENCIRIAN HIDROKARBON AROMATIK POLISIKLIK DALAM AEROSOL ATMOSFERA DARI BEBERAPA LOKASI TERPILIH DALAM SEMENANJUNG MALAYSIA

Oleh

POURYA SHAHPOURY BAHRY

December 2007

Pengerusi : Profesor Madya Mohamad Pauzi Zakaria, PhD

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Pembangunan yang pesat di Malaysia dalam beberapa tahun kebelakangan ini telah meningkatkan kepekatan hidrokarbon dalam atmosfera. Peningkatan tersebut adalah berpunca daripada pengindustrian, pemotoran, pembakaran biojisim dan penebangan hutan di negara ini. Salah satu kelas hidrokarbon yang penting ialah hidrokarbon polisiklik beraromatik yang telah mendapat perhatian dalam bidang alam sekitar forensik. Mengenalpasti hidrokarbon polisiklik beraromatik dalam alam sekitar adalah penting kerana ia bersifat karsinogenik dan mutagenik. Ia juga memberi kesan kepada kesihatan manusia dan boleh menjejaskan sistem endokrin. Pada kebiasaannya, hidrokarbon polisiklik beraromatik yang dihasilkan daripada aktiviti manusia terdiri daripada dua jenis iaitu sumber pirogenik dan petrogenik. Hidrokarbon polisiklik beraromatik wujud dalam fasa partikulat secara siknifikan di dalam atmosfera berbanding dengan fasa lain. Oleh itu kajian ini memfokus sifat sebatian tersebut dalam jumlah partikulat terampai dari sembilan stesen persampelan di semenanjung Malaysia. Objektif bagi projek ini termasuk



mengenalpasti sumber, taburan dan kepekatan sebatian spesifik hidrokarbon polisiklik beraromatik. Untuk tujuan tersebut, aerosol atmosfera telah diambil dengan menggunakan alat pemungut udara sampel isipadu tinggi. Sampel yang diambil seterusnya diekstrak secara sokslet dengan menggunakan diklorometana bergred tinggi. Seterusnya, langkah pemurnian dan pengasingan dijalankan dengan kaedah dua langkah kromatografi kolum. Seterusnya, fraksi hidrokarbon polisiklik beraromatik dengan cincin benziena 3-5 dianalisis dengan menggunakan kromatografi gas yang digabungkan dengan pengesan jisim selektif (GC-MS). Keputusan kajian menunjukkan kepekatan julat hidrokarbon polisiklik beraromatik dari 0.28 hingga 13.02 ng/m³ dengan nilai min adalah 2.73 ng/m³. Nisbah jumlah metilfenantrina bahagi fenantrina adalah kurang daripada uniti untuk 16 daripada 18 sampel. Keputusan tersebut menunjukkan sumber hidrokarbon polisiklik beraromatik adalah pirogenik. Menariknya, nisbah MP/P bagi sampel yang diambil di stesyen Tanah Rata menunjukkan nilai yang lebih tinggi daripada nilai uniti yang mungkin terhasil daripada sumber hidrokarbon polisiklik beraromatik dari petrogenik. Aplikasi nisbah berat molikul rendah bahagi berat molikul tinggi boleh memberikan data sokongan untuk mengenalpasti sumber hidrokarbon polisiklik beraromatik. Keputusan kajian ini menunjukkan taburan hidrokarbon polisiklik beraromatik didominasikan oleh berat molikul tinggi. Keputusan tersebut adalah konsisten dengan nisbah metilfenantrina bahagi fenantrina. Dua stesen yang menunjukkan keputusan nisbah LMW/HMW dan MP/P yang berbeza membuktikan sumber hidrokarbon polisiklik beraromatik yang bercampur diantara petrogenik dan pirogenik. Akhir sekali, kesimpulan boleh dibuat



bahawa atmosfera di semenanjung Malaysia pada bulan Julai 2006 dipengaruhi oleh sumber pirogenik.



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I certify that an Examination Committee has met on 18th December 2007 to conduct the final examination of Pourya Shahpoury Bahry on his Master of Science thesis entitled "Characterization of Polycyclic Aromatic Hydrocarbons in Atmospheric Aerosols Collected from Selected Locations in Peninsular Malaysia" 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 degree of Master of Science.

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DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been published previously or concurrently submitted for any other degree at UPM or other institutions.

POURYA SHAHPOURY BAHRY

Date: 28 January 2008



TABLE OF CONTENTS

DEDICATION	ii
ABSTRACT	iii
ABSTRAK	V
ACKNOWLEDGEMENTS	viii
APPROVAL	ix
DECLARATION	xi
LIST OF TABLES	XV
LIST OF FIGURES	xvi
LIST OF ABBREVIATIONS	xviii

CHAPTER

1.

INTRODUCTION			
1.1	Background of Study	1	
1.2	Significance of Study	6	
1.3	The Objectives of the Study	8	
	1.1 1.2	INTRODUCTION1.1Background of Study1.2Significance of Study1.3The Objectives of the Study	

2. LITERATURE REVIEW

2.1	Sources of Hydrocarbon Pollution in Malaysia	9
2.2	Sources of Atmospheric Hydrocarbons in Malaysia	10
2.3	Atmospheric Particles	12
2.4	Application of Molecular Marker in Pollution Studies	16
	2.4.1 Polycyclic Aromatic Hydrocarbons	18
2.5	Why Focus on PAHs	19
2.6	Transport Pathway of PAHs in the Atmosphere	20
2.7	Fate of PAHs in the Atmospheric Environment	23
2.8	Environmental Health Impact of PAHs	25

3. METHODOLOGY

Sampling Sites		
1 6		
1		36
3.3.1	Surrogate Internal Standard (SIS), Internal	36
Injecti	ion Standard (IIS) and PAHs Standard Mixture	
-		37
e		
1 1		38
2		38
3.5.2	1 st Step Column Chromatography	39
		39
3.5.4		40
PAHs Concentration		41
	Sampl Chem 3.3.1 Injecti 3.3.2 Glass Analy 3.5.1 3.5.2 3.5.3 3.5.4	Sample Collection Chemicals 3.3.1 Surrogate Internal Standard (SIS), Internal Injection Standard (IIS) and PAHs Standard Mixture 3.3.2 Organic Solvent Glass Equipment Analytical Procedure 3.5.1 Soxhlet Extraction 3.5.2 1 st Step Column Chromatography 3.5.3 2 nd Step Column Chromatography 3.5.4 Analysis of PAHs with GC-MS



RESUI	LTS AND DISCUSSION	
4.1	Concentration and Composition of PAHs in	48
	Alor Setar Station	
	4.1.1 Source and Distribution of PAHs in	49
	Alor Setar Station	
4.2	Concentration and Composition of PAHs in	56
	Bukit Kledang Station	
	4.2.1 Source and Distribution of PAHs in	57
	Bukit Kledang Station	
4.3	Concentration and Composition of PAHs in	61
	Bayan Lepas Station	()
	4.3.1 Source and Distribution of PAHs in	62
4 4	Bayan Lepas Station	66
4.4	Concentration and Composition of PAHs in	00
	Kuala Terengganu Station 4.4.1 Source and Distribution of PAHs in	67
	Kuala Terengganu Station	07
4.5	Concentration and Composition of PAHs in	71
ч.5	Melaka Station	/ 1
	4.5.1 Source and Distribution of PAHs in	71
	Melaka Station	/1
4.6	Concentration and Composition of PAHs in	75
	Petaling Jaya Station	
	4.6.1 Source and Distribution of PAHs in	77
	Petaling Jaya Station	
4.7	Concentration and Composition of PAHs in	81
	Perai Station	
	4.7.1 Source and Distribution of PAHs in	83
	Perai Station	- -
4.8	Concentration and Composition of PAHs in	87
	Senai Station	07
	4.8.1 Source and Distribution of PAHs in	87
4.0	Senai Station	91
4.9	Concentration and Composition of PAHs in Tanah Rata Station	91
	4.9.1 Source and Distribution of PAHs in	92
	Tanah Rata Station)2
4.10	Statistical Analysis	96
1.10	4.10.1 Comparison of Total PAHs Concentrations	96
	in the Study Area Using Descriptive Analysis	
	4.10.2 Consideration of B(a)P Concentrations	98
	with Permissible Risk Unit at nine Stations Using	
	Dunnett Method	
	4.10.3 Regression between Total PAHs and TSP	101
	Concentrations	
	4.10.4 Correlation between Total PAHs, B(e)P	102
	and B(k)F Concentrations	

4.



5. CONCLUSIONS AND RECOMMENDATIONS			
	5.1	Conclusion	104
	5.2	Recommendation	106
RE	FEREN	ICES	108
AP	PENDI	CES	115
BIC)DATA	OF THE AUTHOR	120



LIST OF TABLES

Table		Page
3.1	Description of Sampling Stations in Peninsular Malaysia	30
3.2	Meteorological condition during the sampling period	35
3.3	The Analyzed PAHs Compounds, Internal Injection Standard (IIS) and Surrogate Internal Standards in the Samples	44
3.4	The Corresponding Deuterated Surrogates Used to Quantify Each Target PAHs Compounds Recovery Rates	45
4.1	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Alor Setar Station	53
4.2	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Bukit Kledang Station	59
4.3	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Bayan Lepas Station	64
4.4	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Kuala Terengganu Station	69
4.5	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Melaka Station	73
4.6	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Petaling Jaya Station	79
4.7	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Perai Station	85
4.8	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Senai Station	89
4.9	PAHs Concentration and Source-Identifier in Atmospheric Aerosols from Tanah Rata Station	94



LIST OF FIGURES

Figure		Page
3.1	Sampling Stations in Peninsular Malaysia	28
3.2	Analytical Scheme of PAHs	46
3.3	Molecular Structure of PAHs Analyzed in this Study	47
4.1	PAHs Profile of (a) Alor Setar-1; (b) Alor Setar-2; (c) PAHs Diagnostic Ratios	55
4.2	PAHs Profile of (a) Bukit Kledang-1; (b) Bukit Kledang-2; (c) PAHs Diagnostic Ratios	60
4.3	PAHs Profile of (a) Bayan Lepas-1; (b) Bayan Lepas-2; (c) PAHs Diagnostic Ratios	65
4.4	PAHs Profile of (a) Kuala Terengganu-1; (b) Kuala Terengganu-2; (c) PAHs Diagnostic Ratios	70
4.5	PAHs Profile of (a) Melaka-1; (b) Melaka-2; (c) PAHs Diagnostic Ratios	74
4.6	PAHs Profile of (a) Petaling Jaya-1; (b) Petaling Jaya-2; (c) PAHs Diagnostic Ratios	80
4.7	PAHs Profile of (a) Perai-1; (b) Perai-2; (c) PAHs Diagnostic Ratios	86
4.8	PAHs Profile of (a) Senai-1; (b) Senai-2; (c) PAHs Diagnostic Ratios	90
4.9	PAHs Profile of (a) Tanah Rata-1; (b) Tanah Rata-2; (c) PAHs Diagnostic Ratios	95
4.10.1	 (a) Consideration of Total PAHs Concentrations at Nine Stations Using Descriptive Analysis (b) Mean Values of Total PAHs Concentrations in Sampling Stations and their Related Average 	97
4.10.2	Concentrations of B(a)P at Nine Stations and the Respected Mean Concentration Against Permissible Risk Level	100



4.10.3	Regression between total PAHs and TSP concentrations	101
4.10.4	(a) Correlation between benzo(e)pyrene and total PAHs concentrations(b) Correlation between benzo(k)fluoranthene and total PAHs concentrations	103
A-1	High volume air sampler	115
A-2	Filter paper containing aerosols	115
A-3	Soxhlet Extraction	115
A-4	Rotary Evaporator	115
A-5	Column chromatography	116
A-6	GC-MS analysis	116
B-1	Typical GC-MS chromatogram of PAHs standard mixture	117
B-2	Example of GC-MS chromatogram in aerosols sample	118
B-3	Specific diagnostic ratio of MP/P used in this study	119



LIST OF ABBREVIATIONS

Ant	Anthracene
Ant-d10	Anthracene-d10
BaA	Benzo(a)anthracene
B(a)P	Benzo(a)pyrene
B(e)Acep	Benzo(e)acephenanthrylene
B(e)P	Benzo(e)pyrene
B(k)F	Benzo(k)fluoranthene
Chry	Chrysene
Chry-d12	Chrysene-d12
DBahA	Dibenzo(a,h)anthracene
DBT	Dibenzothiophene
DCM	Dichloromethane
Flu	Fluoranthene
GC/MS	Gas Chromatography Mass Spectrometry
LMW/HMW-PAHs	Lower Molecular Weight PAHs / Higher Molecular Weight PAHs



Hex	Hexane
HMW	Higher Molecular Weight
IIS	Internal Injection Standard
LMW	Lower Molecular Weight
2-MA	2-methylanthracene
МеОН	Methanol
1-MP	1-methylphenanthrene
2-MP	2-methylphenanthrene
3-MP	3-methylphenanthrene
9-MP	9-methylphenanthrene
MP/P	Methylphenanthrene/ Phenanthrene
1MPyr	1-methylpyrene
Naph-d8	Naphthalene-d8
P-terph-d14	P-terphenyl-d14
PAHs	Polycyclic Aromatic Hydrocarbons
Pery-d12	Perylene-d12



Phe Phenanthrene

Pyr Pyrene

SIS Surrogate Internal Standard



CHAPTER 1

INTRODUCTION

1.1 Background of Study

There is a strong relation between life on earth and the nature of earth's atmosphere that determines its suitability for life. After discovery of fire, due to its special dynamic character, atmosphere has been transformed into a dumping ground to dispose pollutant materials by human being. After the industrial revolution in the late 1800's, industries confronted rapid development in various point of views especially by invention of internal combustion engines that played a particular role in human history. Considering that these engines have used fossil fuels, the interest of human regarding this new invention led them in a search for more supplies of fossil fuels especially petroleum. Nowadays, exploration of petroleum is obvious in every corner of the world and this is the trigger point that releases the petroleum pollution to the environment. Usually utilization of petroleum as source of energy can emit various types of chemicals from the exploration points and the refining process to combustion in industrial power plants and engines. Some of these chemicals may be non-hazardous but the rest could remain in the environment as persistent organic chemicals and cause carcinogenic and mutagenic effect on organisms. Signs of petroleum pollution produced by human activities are obvious in different phases of the environment, especially in the atmosphere.



Atmospheric pollution is one of the most important concerns humankind faces these days. The air contamination causes damage on human health and harm to wildlife and vegetation. Among the released chemical to the atmosphere by utilization of petroleum, hydrocarbons has a share of more than 50-98% of the total composition. This truth is another proof that crude oil contains thousands of chemicals of which hydrocarbons are the most abundant compounds depending on the specific locations where the oil was found.

Hydrocarbon contamination (HC) in atmosphere has increased globally due to growth in urbanization, industrialization, motorization and deforestation. Natural sources are the most important contributors of organics in the atmosphere, and hydrocarbons generated and released by human activities constitute only about one seventh of the total hydrocarbons in the atmosphere. On the other hand, more than 80% of petroleum hydrocarbons are produced by anthropogenic sources and the other 20% are released naturally through oil seeps and biogenic sources. Among petroleum hydrocarbons, the major compounds are straight chain alkanes (*n*-alkanes), cycloalkanes and aromatics. The petroleum hydrocarbons of greatest concern are PAHs that are widespread in the environment. Polycyclic aromatic hydrocarbons (PAHs) are one of the most important classes of micro organic pollutants produced mostly by means of incomplete combustion of organic material containing carbon, hydrogen and other minor compounds.



Until the last two decades, it was generally presumed that PAHs were formed only during the high temperature pyrolysis of organic materials. The discovery of complex mixture of PAHs spanning a wide molecular weight range in fossil fuels such as coal and crude petroleum has led to the conclusion that given significant time, pyrolysis of organic materials at temperature as low as 100-150° C can cause production of PAHs. Besides, there has been considerable speculation in recent years and some experimental evidence in its favor that PAHs are synthesized by bacteria and plants. Thus, PAHs are probably formed in three ways, high temperature pyrolysis of organic materials, low to moderate temperature of diagenesis of sedimentary organic material to form fossil fuels or direct biosynthesis by microbes and plants.

Although PAHs could be produced naturally, a wide variety of human activities increases the environmental load of these substances. Incineration of industrial and domestic wastes, forest and grass fires, power generation from fossil fuels, and the combustion of fuels in internal combustion engines also produce emission rich in PAHs. These anthropogenic PAHs may reach the aquatic environment in industrial and domestic sewage effluents, surface runoffs from land, deposition of air born particulates and spillage of petroleum and petroleum products into water bodies.

Polycyclic aromatic hydrocarbons in atmospheric particles have received a great deal of attention due to the known carcinogenic effects of some of these compounds. Prominent among these compounds are benzo(a)pyrene,

benzo(a)anthracene, chrysene, benzo(e)pyrene, benzo(e)acephenanthrylene, benzo(j)fluoranthene, and indenol. The most often cited example of a PAHs compounds is benzo(a)pyrene, a compound that the body can metabolize to a carcinogenic form.

Over 120 PAHs compound have been identified in urban pollution and are produced by the combustion of organic matter. Organic compounds in combustion exhaust consist mainly of unburned alkanes and series of PAHs. Individual PAHs compounds from combustion sources enter the ambient atmosphere as gases or associated with the particles. Airborne particulate matters consists of a complex mixture of solid and liquid that contains PAHs and are emitted to the atmosphere from a variety of sources. Typically 2 to 3 rings condensed PAHs are partitioned with the gas phase while 4 to 6 ring PAHs are partitioned on particles.

What is the nature of particulate matter? Particle in the atmosphere that range from about 0.5 mm down to molecular dimensions, are made up of an amazing variety of materials and discrete objects. Particulates are believed to cause the most visible and obvious form of air pollution. Atmospheric aerosols are solid or liquid particles smaller than 100 μ m in diameter. Pollutant particles in the 0.001 to 10 μ m range are commonly suspended in the air near sources of pollution such as the urban atmosphere, industrial plants, highways and power plants.

