UNIVERSITI PUTRA MALAYSIA

DISTRIBUTION AND SOURCES OF POLYCYCLIC AROMATIC HYDROCARBONS IN SELECTED LANDFILL SITES

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By

KHO HIAW GEIK

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

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DEDICATION

To my dear family, my parents, my sisters and brothers, my supervisor who have been my source of inspiration, wisdom and strength through the most difficult times of my life.

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

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Chairman : Associate Professor Mohamad Pauzi Zakaria, PhD

Faculty : Environmental Studies

Polycyclic aromatic hydrocarbons are one of the most important classes of anthropogenic micro-organic pollutants that had long been of interest in the field of environmental chemistry due to the fact that a small fraction of PAHs generated and released to environment by human activities had been shown to be carcinogenic and mutagenic to mammals. Polycyclic aromatic hydrocarbons have also been reported to disrupt endocrine system in humans. Upon entering the aquatic system, PAHs partition into few different phases namely truly dissolved, colloids, suspended particulate matter, surface sediments and biota. Landfilling and disposing of waste in open dumpsite had been expected to remain the most common and significant method for disposal of municipal solid wastes in Malaysia in near future. This study focuses on 3 landfill sites in Malaysia. The objectives of this study were to understand the distribution and sources of compound-specific PAHs in the landfill leachates and to determine their transport pathway to surrounding water bodies. The distribution of PAHs between various phases was the fundamental in the control of

their movement and impact to the environment. Results from this study revealed that most of the particulate phase samples showed the higher PAHs concentration as compared to dissolved phase indicating the hydrophobicity characteristic of individual PAHs. High abundance of higher molecular weight (HMW) PAHs in particulate phase river water from Ulu Maasop Landfill had indicated the origin of the PAHs was from pyrogenic source which could be attributed to the illegal waste burning. Particulate phase leachate from Taman Beringin Landfill shows a mixture of petrogenic and pyrogenic signature although there was dominance in petrogenic signature as evidenced in their lower molecular weight/higher molecular weight ratio (LMW/HMW) of 2.21, 2.17 and 2.60. Illegal dumping of waste petroleum products could be one of the petrogenic sources entering the river. Therefore, it can be concluded that the sources of PAHs among all of the landfills in this study was a mixture of petrogenic and pyrogenic origin. Similarities in PAHs distribution profile in leachate, groundwater and river water for all landfills studied was a grave concern due to the fact that leachate may had been transported to those aquatic environment. The flow of leachates into water bodies will have to be stopped immediately in order to protect the health of our environments.

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

TABURAN DAN SUMBER POLISIKLIK AROMATIK HIDROKARBONS DARI KAWASAN TAPAK PELUPUSAN SAMPAH TERTENTU

Oleh

KHO HIAW GEIK

Julai 2005

Pengerusi : Profesor Madya Mohamad Pauzi Zakaria, PhD

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Polisiklik aromatik hidrokarbons adalah sejenis bahan pencemar antropogenik dan telah menjadi tumpuan utama dalam bidang kimia alam sekitar oleh kerana walaupun terdapat sedikit bahan pencemar tersebut yang dilepaskan dari aktiviti manusia akan memberikan kesan mutasi dan karsinogenik kepada mamalia. Polisiklik aromatik hidrokarbons juga didapati boleh merosakkan sistem endokrin dalam tubuh manusia. Polisiklik aromatik hidrokarbons yang dilepaskan ke dalam sistem akuatik akan terpisah kepada beberapa fasa yang diantaranya ialah larutan mutlak, bahagian koloid, gabungan di permukaan apungan partikulat serta di permukaan sedimen dan biota. Aktiviti pembuangan pelupusan sampah di kawasan terbuka adalah satu cara yang paling umum yang terdapat di Malaysia dan dijangka akan menjadi lebih penting pada masa hadapan. Kajian ini menumpukan perhatian di tiga tapak pelupusan sampah di Malaysia. Objektif kajian ini adalah untuk memahami dengan lebih lanjut tentang taburan dan sumber PAHs dalam 'leachate' dari tapak pelupusan sampah serta cara pengalirannya ke sumber air yang terdapat di sekeliling tapak pelupusan.

Taburan PAHs dalam pelbagai fasa merupakan asas yang penting dalam kawalan pengaliran PAHs di tapak pelupusan dan kesannya terhadap alam sekitar. Keputusan kajian ini menunjukkan bahawa kebanyakan gabungan PAHs dengan bahan apungan partikulat dalam air didapati mengadungi kandungan PAHs yang lebih tinggi berbanding dengan fasa larutan mutlak PAHs dan ini dengan jelas mempamerkan hidrofobisiti PAHs tersebut secara individu. Kandungan PAHs bermolikul berat yang bergabung dengan apungan partikulat dari air sungai di tapak pelupusan Ulu Maasop menunjukkan sumber 'pyrogenic' yang mungkin disebabkan oleh pembakaran sampah secara berleluasa. Kandungan PAHs yang bergabung dengan apungan partikulat dari 'leachate' di tapak pelupusan Taman Beringin menunjukkan sumber 'petrogenic' dan 'pyrogenic' tetapi sumber petrogenik lebih dominan berdasarkan nilai nisbah LMW/HMW iaitu 2.21, 2.17 and 2.60. Pembuangan sisa petroleum secara haram menjadi salah satu daripada sumber 'petrogenic'. Kesimpulannya, PAHs yang terkandung dalam tapak pelupusan sampah adalah berpunca dari percampuran sumber 'petrogenic' dan 'pyrogenic'. Kesamaan dalam taburan profil PAHs dalam 'leachate', air bawah tanah dan air sungai di semua tapak pelupusan sampah menunjukkan kemungkinan 'leachate' akan mengalir ke sumber air yang berhampiran. Pengaliran 'leachate' ke sumber air yang berhampiran harus dielakkan dengan serta-merta untuk menjamin kesihatan alam sekeliling.

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LIST OF ABBREVIATIONS AND GLOSSARY

PAHs	Polycyclic aromatic hydrocarbons
GC-MS	Gas chromatography mass spectrometry
IISTD	Internal injection standard
SIS	Surrogate internal standard
SIM	Selective ion monitoring
DBT	Dibenzothiophene
Phe	Phenanthrene
Ant	Anthracene
3MPhe	3-methylphenanthrene
2MPhe	2-methylphenanthrene
2MAnt	2-methylanthracene
9MPhe	9-methylphenanthrene
1MPhe	1-methylphenanthrene
Fluo	Fluoranthene
Pyr	Pyrene
1MPyr	1-methylpyrene
BaAnt	Benz(a)anthracene
Chry	Chrysene
BkFluo	Benzo(k)fluoranthene
BeAcep	Benz(e)acephenanthrylene
BePyr	Benzo(e)pyrene
BaPyr	Benzo(a)pyrene
DBahAnt	Dibenz(a,h)anthracene
LMW	Lower molecular weight
HMW	Higher molecular weight

MP/P ratio	A ratio of the sum of 3-methylphenanthrene, 2-methylphenanthrene, 9-methylphenanthrene, 1-methylphenanthrene to phenanthrene
LMW/HMW ratio	A ratio of the sum of dibenzothiophene + phenanthrene + anthracene + 3-methylphenanthrene + 2-methylphenanthrene + 2-methylanthracene + 9-methylphenanthrene + 1-methylphenanthrene + fluoranthene + pyrene relative to sum of 1-methylpyrene + benzo(a)anthracene + chrysene + benzo(k)fluoranthene + benz(a)acephenanthrylene + benzo(e)pyrene + benzo(a)pyrene + dibenz(a,h)anthracene
Phe/Ant ratio	A ratio of phenanthrene relative to anthracene
Fluo/Pyr ratio	A ratio of fluoranthene relative to pyrene
BaAnt/Chry ratio	A ratio of benz(a)anthracene relative to chrysene
Chry/BaAnt ratio	A ratio of chrysene relative to benz(a)anthracene
Pyrogenic	High temperature anthropogenic combustion process such as combustion of oil, coal, wood, natural fire, etc.
Pyrolytic	Same as pyrogenic
Petrogenic	Low temperature maturation of hydrocarbon product or anthropogenic petroleum inputs from municipal discharge, tanker accident, storm water runoff, etc.
Source-identifier	MP/P ratio, LMW/HMW ratio, Phe/Ant ratio, Fluo/Pyr ratio, BaAnt/Chry ratio and Chry/BaAnt ratio.